(Unfinished) Notes on spin-orbit interaction, response functions, and more

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Introduction

These exhaustive and extensive notes have been written during the course of about a decade. They include calculations of most of everything I have ever done. The steps are as much detailed as I could possibly write, to avoid the previously common thought "what the hell have I done here?" This document is continuously growing, and probably never shrinking.

As I believe open science is the way we should move forward, I have distributed it to friends and colleagues indiscriminately. I hope they can be made good use, with future developments also made available in the same way.

Please note that some of the calculations done here are unfinished, others are probably not efficiently done, and sill some are probably wrong. Use it with moderation and proceed with caution. Above all: Don't panic.

Chapter 1

Hamiltonian

Let's calculate the transverse susceptibility in systems where the spin-orbit coupling (SOC) is present. In this case, the spin angular momentum is coupled to the orbital angular momentum and the system hamiltonian is

$$\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}} + \hat{H}_{\text{Z}} + \hat{H}_{\text{so}} \ . \tag{1.1}$$

Here, $\hat{H}_0$ is the tight-binding hamiltonian

$$\hat{H}_0 = \sum_{\substack{mn \\ s}} \sum_{\alpha\beta} t^{\alpha\beta}_{mn} c^\dagger_{m\alpha s} c_{n\beta s} \ . \tag{1.2}$$

The screened Coulomb interaction is given by the Hubbard's term

$$\hat{H}_{\text{int}} = \frac{1}{2} \sum_{\substack{m \\ s,s'}} \sum_{\alpha\beta\gamma\xi} U^m_{\alpha\beta\gamma\xi} c^\dagger_{m\alpha s} c^\dagger_{m\beta s'} c_{m\xi s'} c_{m\gamma s}, \tag{1.3}$$

where

$$U^m_{\alpha\beta\gamma\xi} = \iint d\mathbf{r} d\mathbf{r}' \phi^*_\alpha(\mathbf{r} - \mathbf{R}_m) \phi^*_\beta(\mathbf{r}' - \mathbf{R}_m) \frac{e}{|\mathbf{r} - \mathbf{r}'|} \phi_\gamma(\mathbf{r} - \mathbf{R}_m) \phi_\xi(\mathbf{r}' - \mathbf{R}_m) = U^m_{\beta\alpha\xi\gamma} \ . \tag{1.4}$$

1.1 Zeeman interaction

$\hat{H}_{\text{Z}}$ is the Zeeman interaction with an external magnetic field and it has a spin and an orbital momentum contribution, i.e., $\hat{H}_{\text{Z}} = \hat{H}_{\text{ZS}} + \hat{H}_{\text{ZL}}$. If we assume that the external magnetic field is given in the spin coordinates, $\mathbf{B}' = B'^x \hat{x} + B'^y \hat{y} + B'^z \hat{z}$, the spin contribution is given by

$$
\begin{aligned}
\hat{H}_{\text{ZS}} =& g_S\mu_B \sum_m \hat{\mathbf{S}}_m \cdot \mathbf{B}' \\
=& g_S\mu_B \sum_m \left\{ \hat{S}^x_m B'^x + \hat{S}^y_m B'^y + \hat{S}^z_m B'^z \right\} \\
=& \frac{g_S\mu_B}{2} \sum_{\substack{m,\mu \\ \alpha\beta}} \left\{ c^\dagger_{m\mu\alpha} \hat{\sigma}^x_{\alpha\beta} c_{m\mu\beta} B'^x + c^\dagger_{m\mu\alpha} \hat{\sigma}^y_{\alpha\beta} c_{m\mu\beta} B'^y + c^\dagger_{m\mu\alpha} \hat{\sigma}^z_{\alpha\beta} c_{m\mu\beta} B'^z \right\} \\
=& \frac{1}{2} \sum_{m,\mu} \left\{ \left(c^\dagger_{m\mu\uparrow} c_{m\mu\downarrow} + c^\dagger_{m\mu\downarrow} c_{m\mu\uparrow} \right) \hbar\omega_x - i \left(c^\dagger_{m\mu\uparrow} c_{m\mu\downarrow} - c^\dagger_{m\mu\downarrow} c_{m\mu\uparrow} \right) \hbar\omega_y \right. \\
& \left. + \left(c^\dagger_{m\mu\uparrow} c_{m\mu\uparrow} - c^\dagger_{m\mu\downarrow} c_{m\mu\downarrow} \right) \hbar\omega_z \right\} \\
=& \frac{1}{2} \sum_{m,\mu} \left\{ (\hbar\omega_x - i\hbar\omega_y)\, c^\dagger_{m\mu\uparrow} c_{m\mu\downarrow} + (\hbar\omega_x + i\hbar\omega_y)\, c^\dagger_{m\mu\downarrow} c_{m\mu\uparrow} + \hbar\omega_z \left(c^\dagger_{m\mu\uparrow} c_{m\mu\uparrow} - c^\dagger_{m\mu\downarrow} c_{m\mu\downarrow} \right) \right\} \ ,
\end{aligned}
\tag{1.5}
$$

where $\hbar\omega_\alpha = g_S\mu_B B'^\alpha$. It is important to remember that, since $\mathbf{S}$ is antiparallel to $\mathbf{m}$, we need to consider negative values of $\hbar\omega_\alpha$ to obtain peaks on positive energies in the transverse dynamic spin susceptibility.

The orbital contribution of the Zeeman interaction is obtained writing the scalar product in the cubic coordinates, as

$$
\begin{aligned}
\hat{H}_{\text{ZL}} =& g_L\mu_B \sum_m \hat{\mathbf{L}}_m \cdot \mathbf{B} \\
=& \mu_B \sum_m \left\{ \hat{L}^x_m B^x + \hat{L}^y_m B^y + \hat{L}^z_m B^z \right\} \ ,
\end{aligned}
\tag{1.6}
$$

since $g_L = 1$. We can use the results obtained in Eq. (D.19) of appendix D to obtain the magnetic field in the cubic coordinates as

$$
\mathbf{B} = \begin{pmatrix} B'^x \cos(\theta)\cos(\phi) - B'^y \sin(\phi) + B'^z \sin(\theta)\cos(\phi) \\ B'^x \cos(\theta)\sin(\phi) + B'^y \cos(\phi) + B'^z \sin(\theta)\sin(\phi) \\ -B'^x \sin(\theta) + B'^z \cos(\theta) \end{pmatrix} \ . \tag{1.7}
$$

The hamiltonian $\hat{H}_{\text{ZL}}$ becomes

$$
\begin{aligned}
\hat{H}_{\text{ZL}} = \mu_B \sum_m \Big\{ & \hat{L}^x_m \left[B'^x \cos(\theta)\cos(\phi) - B'^y \sin(\phi) + B'^z \sin(\theta)\cos(\phi) \right] \\
& + \hat{L}^y_m \left[B'^x \cos(\theta)\sin(\phi) + B'^y \cos(\phi) + B'^z \sin(\theta)\sin(\phi) \right] \\
& + \hat{L}^z_m \left[-B'^x \sin(\theta) + B'^z \cos(\theta) \right] \Big\} \\
= \sum_{\substack{m,\mu\nu \\ \sigma}} \Bigg\{ & L^x_{\mu\nu} \left[\frac{\hbar\omega^x}{2} \cos(\theta)\cos(\phi) - \frac{\hbar\omega^y}{2} \sin(\phi) + \frac{\hbar\omega^z}{2} \sin(\theta)\cos(\phi) \right] \\
& + L^y_{\mu\nu} \left[\frac{\hbar\omega^x}{2} \cos(\theta)\sin(\phi) + \frac{\hbar\omega^y}{2} \cos(\phi) + \frac{\hbar\omega^z}{2} \sin(\theta)\sin(\phi) \right] \\
& + L^z_{\mu\nu} \left[-\frac{\hbar\omega^x}{2} \sin(\theta) + \frac{\hbar\omega^z}{2} \cos(\theta) \right] \Bigg\} c^\dagger_{m\mu\sigma} c_{m\nu\sigma} \ .
\end{aligned}
\tag{1.8}
$$

1.2 Spin-orbit coupling

The spin-orbit interaction is obtained from

$$
\hat{H}_{\text{so}} = \lambda \hat{\mathbf{L}} \cdot \hat{\mathbf{S}} \ . \tag{1.9}
$$

$\hat{\mathbf{L}}$ and $\hat{\mathbf{S}}$ are the orbital and spin angular momentum operators, respectively. We include the $\hbar$ of both angular momenta in the λ coefficient, so it has the units of energy. We are going to consider a local spin-orbit interaction. In this case, we can write $\hat{H}_{\text{so}}$ as

$$
\begin{aligned}
\hat{H}_{\text{so}} &= \sum_m \lambda_m \hat{\mathbf{L}}_m \cdot \hat{\mathbf{S}}_m = \sum_{mn} \lambda_m \hat{L}^n_m \hat{S}^n_m \\
&= \sum_m \lambda_m \left(\hat{L}^x_m \hat{S}^x_m + \hat{L}^y_m \hat{S}^y_m + \hat{L}^z_m \hat{S}^z_m \right) \\
&= \sum_m \frac{\lambda_m}{2} \left(\hat{L}^+_m \hat{S}^-_m + \hat{L}^-_m \hat{S}^+_m + 2\hat{L}^z_m \hat{S}^z_m \right) \ ,
\end{aligned}
\tag{1.10}
$$

where the scalar product is calculated in the cubic or in the spin system of coordinates. In the spin system of coordinates, we can write this operator in second quantization as

$$
\begin{aligned}
\hat{H}_{\text{so}} &= \sum_{\substack{mn\alpha\beta \\ ss'}} \lambda_m \left\langle m\alpha s \left| \hat{L}'^n_m \hat{S}'^n_m \right| m\beta s' \right\rangle c^\dagger_{m\alpha s} c_{m\beta s'} \\
&= \sum_{\substack{mn\alpha\beta \\ ss'}} \frac{\lambda_m}{2} \left(L'^n_{\alpha\beta} \sigma^n_{ss'} \right) c^\dagger_{m\alpha s} c_{m\beta s'} \ .
\end{aligned}
\tag{1.11}
$$

or

$$\hat{H}_{\text{so}} = \sum_{\substack{m\alpha\beta \\ ss'}} \frac{\lambda_m}{2} \left\langle m\alpha s \left| \hat{L}'^{+}_m \hat{S}'^{-}_m + \hat{L}'^{-}_m \hat{S}'^{+}_m + 2\hat{L}'^{z}_m \hat{S}'^{z}_m \right| m\beta s' \right\rangle c^{\dagger}_{m\alpha s} c_{m\beta s'}$$
$$= \sum_{\substack{m\alpha\beta \\ ss'}} \frac{\lambda_m}{4} \left(L'^{+}_{\alpha\beta} \sigma^{-}_{ss'} + L'^{-}_{\alpha\beta} \sigma^{+}_{ss'} + 2L'^{z}_{\alpha\beta} \hat{\sigma}^{z}_{ss'} \right) c^{\dagger}_{m\alpha s} c_{m\beta s'} \ . \tag{1.12}$$

Using Pauli matrices

$$\sigma^+ = \begin{pmatrix} 0 & 2 \\ 0 & 0 \end{pmatrix} \quad , \quad \sigma^- = \begin{pmatrix} 0 & 0 \\ 2 & 0 \end{pmatrix} \quad , \quad \sigma^z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} , \tag{1.13}$$

We obtain

$$\hat{H}_{\text{so}} = \sum_{m\alpha\beta} \frac{\lambda_m}{2} \left[L'^{+}_{\alpha\beta} c^{\dagger}_{m\alpha\downarrow} c_{m\beta\uparrow} + L'^{-}_{\alpha\beta} c^{\dagger}_{m\alpha\uparrow} c_{m\beta\downarrow} + L'^{z}_{\alpha\beta} \left(c^{\dagger}_{m\alpha\uparrow} c_{m\beta\uparrow} - c^{\dagger}_{m\alpha\downarrow} c_{m\beta\downarrow} \right) \right] \ . \tag{1.14}$$

The orbital angular momentum matrices are calculated in appendix D.

1.3 Hartree-Fock Hamiltonian

Before we proceed with the calculation, we will write the Hartree-Fock hamiltonian in a convenient way. The total hamiltonian is given by Eq. (1.1). To obtain $\hat{H}^{\text{HF}}_{\text{int}}$, we are going to use the approximation

$$c^{\dagger}_{m\alpha s} c^{\dagger}_{m\beta s'} c_{m\xi s'} c_{m\gamma s} \rightarrow \langle c^{\dagger}_{m\beta s'} c_{m\xi s'} \rangle c^{\dagger}_{m\alpha s} c_{m\gamma s} + \langle c^{\dagger}_{m\alpha s} c_{m\gamma s} \rangle c^{\dagger}_{m\beta s'} c_{m\xi s'}$$
$$- \langle c^{\dagger}_{m\alpha s} c_{m\xi s'} \rangle c^{\dagger}_{m\beta s'} c_{m\gamma s} - \langle c^{\dagger}_{m\beta s'} c_{m\gamma s} \rangle c^{\dagger}_{m\alpha s} c_{m\xi s'} \ . \tag{1.15}$$

Substituting back in Eq. (1.3), we have

$$\hat{H}^{\text{HF}}_{\text{int}} = \frac{1}{2} \sum_{\substack{m \\ s,s'}} \sum_{\alpha\beta\gamma\xi} U^{m}_{\alpha\beta\gamma\xi} \Big\{ \langle c^{\dagger}_{m\beta s'} c_{m\xi s'} \rangle c^{\dagger}_{m\alpha s} c_{m\gamma s} + \langle c^{\dagger}_{m\alpha s} c_{m\gamma s} \rangle c^{\dagger}_{m\beta s'} c_{m\xi s'}$$
$$- \langle c^{\dagger}_{m\alpha s} c_{m\xi s'} \rangle c^{\dagger}_{m\beta s'} c_{m\gamma s} - \langle c^{\dagger}_{m\beta s'} c_{m\gamma s} \rangle c^{\dagger}_{m\alpha s} c_{m\xi s'} \Big\} \ . \tag{1.16}$$

Changing the orbital indices $\alpha \rightleftharpoons \beta$ and $\xi \rightleftharpoons \gamma$, as well as the spin index $s \rightleftharpoons s'$ in the second and fourth terms, we can rewrite the above equation as

$$\hat{H}^{\text{HF}}_{\text{int}} = \frac{1}{2} \sum_{\substack{m \\ s,s'}} \sum_{\alpha\beta\gamma\xi} \Big\{ U^{m}_{\alpha\beta\gamma\xi} \langle c^{\dagger}_{m\beta s'} c_{m\xi s'} \rangle c^{\dagger}_{m\alpha s} c_{m\gamma s} + U^{m}_{\beta\alpha\xi\gamma} \langle c^{\dagger}_{m\beta s'} c_{m\xi s'} \rangle c^{\dagger}_{m\alpha s} c_{m\gamma s}$$
$$- U^{m}_{\alpha\beta\gamma\xi} \langle c^{\dagger}_{m\alpha s} c_{m\xi s'} \rangle c^{\dagger}_{m\beta s'} c_{m\gamma s} - U^{m}_{\beta\alpha\xi\gamma} \langle c^{\dagger}_{m\alpha s} c_{m\xi s'} \rangle c^{\dagger}_{m\beta s'} c_{m\gamma s} \Big\} \ . \tag{1.17}$$

Using the symmetry of U in the second and fourth terms, we note that they become the same as the first and third, respectively. So the Hartree-Fock hamiltonian can be written as

$$\hat{H}^{\mathrm{HF}}_{\mathrm{int}} = \sum_{\substack{m \\ s,s'}} \sum_{\alpha\beta\gamma\xi} \Big\{ U^m_{\alpha\beta\gamma\xi} \langle c^\dagger_{m\beta s'} c_{m\xi s'} \rangle c^\dagger_{m\alpha s} c_{m\gamma s} - U^m_{\alpha\beta\gamma\xi} \langle c^\dagger_{m\alpha s} c_{m\xi s'} \rangle c^\dagger_{m\beta s'} c_{m\gamma s} \Big\}. \tag{1.18}$$

On this last term, we are going to separate the terms involving only creation and annihilation operators with the same spin, i.e.,

$$\begin{aligned}\hat{H}^{\mathrm{HF}}_{\mathrm{int}} = &\sum_{\substack{m \\ s}} \sum_{\alpha\beta\gamma\xi} \Big\{ U^m_{\alpha\beta\gamma\xi} \sum_{s'} \langle c^\dagger_{m\beta s'} c_{m\xi s'} \rangle c^\dagger_{m\alpha s} c_{m\gamma s} - U^m_{\alpha\beta\gamma\xi} \langle c^\dagger_{m\alpha s} c_{m\xi s} \rangle c^\dagger_{m\beta s} c_{m\gamma s} \Big\} \\ &- \sum_{m} \sum_{\alpha\beta\gamma\xi} U^m_{\alpha\beta\gamma\xi} \Big\{ \langle c^\dagger_{m\alpha\uparrow} c_{m\xi\downarrow} \rangle c^\dagger_{m\beta\downarrow} c_{m\gamma\uparrow} + \langle c^\dagger_{m\alpha\downarrow} c_{m\xi\uparrow} \rangle c^\dagger_{m\beta\uparrow} c_{m\gamma\downarrow} \Big\}.\end{aligned} \tag{1.19}$$

Change the indices, we can write

$$\begin{aligned}\hat{H}^{\mathrm{HF}}_{\mathrm{int}} = &\sum_{\substack{mn \\ s}} \sum_{\alpha\beta} \delta_{mn} \sum_{\gamma\xi} \Big\{ \sum_{s'} U^m_{\alpha\gamma\beta\xi} \langle c^\dagger_{m\gamma s'} c_{m\xi s'} \rangle - U^m_{\gamma\alpha\beta\xi} \langle c^\dagger_{m\gamma s} c_{m\xi s} \rangle \Big\} c^\dagger_{m\alpha s} c_{m\beta s} \\ &- \sum_{m\alpha\beta} \sum_{\gamma\xi} U^m_{\gamma\alpha\beta\xi} \Big\{ \langle c^\dagger_{m\gamma\uparrow} c_{m\xi\downarrow} \rangle c^\dagger_{m\alpha\downarrow} c_{m\beta\uparrow} + \langle c^\dagger_{m\gamma\downarrow} c_{m\xi\uparrow} \rangle c^\dagger_{m\alpha\uparrow} c_{m\beta\downarrow} \Big\}.\end{aligned} \tag{1.20}$$

The first line in the equation above, and also the L^z terms of $\hat{H}_{\mathrm{so}}$, can be put together into the on-site term of $\hat{H}_0$. The other terms of $\hat{H}_{\mathrm{so}}$ and $\hat{H}^{\mathrm{HF}}_{\mathrm{int}}$ can also be grouped in a "spin-flip" term, and we end up with

$$\begin{aligned}\hat{H}^{\mathrm{HF}} = &\sum_{\substack{mn \\ s}} \sum_{\alpha\beta} t^{s\,\alpha\beta}_{mn} c^\dagger_{m\alpha s} c_{n\beta s} + \sum_{m} \sum_{\alpha\beta} \Big\{ a_{m,\alpha\beta} c^\dagger_{m\alpha\uparrow} c_{m\beta\downarrow} + a^*_{m,\beta\alpha} c^\dagger_{m\alpha\downarrow} c_{m\beta\uparrow} \Big\} \\ = &\hat{H}'_0 + \hat{H}_{\mathrm{sf}} \ ,\end{aligned} \tag{1.21}$$

where

$$t^{s\,\alpha\beta}_{mn} = t^{\alpha\beta}_{mn} + \delta_{mn} \sum_{\gamma\xi} \Big\{ \sum_{s'} U^m_{\alpha\gamma\beta\xi} \langle c^\dagger_{m\gamma s'} c_{m\xi s'} \rangle - U^m_{\gamma\alpha\beta\xi} \langle c^\dagger_{m\gamma s} c_{m\xi s} \rangle \Big\} \pm \delta_{mn} \frac{\lambda_m}{2} L'^z_{\alpha\beta} \pm \delta_{mn} \delta_{\alpha\beta} \frac{\hbar\omega_z}{2} \ . \tag{1.22}$$

Here, $+$ and $-$ signs corresponds to $\uparrow$ and $\downarrow$ spins, respectively, and $\hbar\omega_\alpha = g_S \mu_B B'^{\mathrm{ext}\,\alpha}$ is the external magnetic field with respect to the spin system of coordinates. For the spin flip term, we have

$$a_{m,\alpha\beta} = \frac{\lambda_m}{2} L'^-_{\alpha\beta} - \sum_{\gamma\xi} U^m_{\gamma\alpha\beta\xi} \langle c^\dagger_{m\gamma\downarrow} c_{m\xi\uparrow} \rangle + \delta_{\alpha\beta} \frac{\hbar\omega_-}{2} \ . \tag{1.23}$$

With this definition,

$$\begin{aligned}a^*_{m,\alpha\beta} = &\frac{\lambda_m}{2} \left(L'^-_{\alpha\beta} \right)^* - \sum_{\gamma\xi} \left(U^m_{\gamma\alpha\beta\xi} \right)^* \langle c^\dagger_{m\xi\uparrow} c_{m\gamma\downarrow} \rangle + \delta_{\alpha\beta} \frac{\hbar\omega_+}{2} \\ = &\frac{\lambda_m}{2} L'^+_{\beta\alpha} - \sum_{\gamma\xi} U^m_{\xi\beta\alpha\gamma} \langle c^\dagger_{m\xi\uparrow} c_{m\gamma\downarrow} \rangle + \delta_{\alpha\beta} \frac{\hbar\omega_+}{2} \ ,\end{aligned} \tag{1.24}$$

where we have used that $\left(U^m_{\gamma\alpha\beta\xi}\right)^* = U^m_{\beta\xi\gamma\alpha} = U^m_{\xi\beta\alpha\gamma}$.

The eigenstates of this hamiltonian are

$$\hat{H}^{\mathrm{HF}}|\phi_\ell\rangle = E_\ell|\phi_\ell\rangle \ . \tag{1.25}$$

Projecting the equation above in a state of orbital ν of site j with spin σ we have

$$\langle j\nu\sigma|\hat{H}^{\mathrm{HF}}|\phi_\ell\rangle = E_\ell\langle j\nu\sigma|\phi_\ell\rangle \ . \tag{1.26}$$

Substituting Eq. (1.21),

$$\begin{aligned}\sum_{n\beta s} t^{s\,\nu\beta}_{jn}\delta_{\sigma s}\langle n\beta s|\phi_\ell\rangle + \sum_{\beta}\left\{a_{j,\nu\beta}\delta_{\sigma\uparrow}\langle j\beta\downarrow| + a^*_{j,\beta\nu}\delta_{\sigma\downarrow}\langle j\beta\uparrow|\right\}|\phi_\ell\rangle =& E_\ell\langle j\nu\sigma|\phi_\ell\rangle \\ \sum_{n\beta s}\left\{t^{s\,\nu\beta}_{jn}\delta_{\sigma s} + a_{n,\nu\beta}\delta_{nj}\delta_{\sigma\uparrow}\delta_{s\downarrow} + a^*_{n,\beta\nu}\delta_{nj}\delta_{\sigma\downarrow}\delta_{s\uparrow}\right\}\langle n\beta s|\phi_\ell\rangle =& E_\ell\langle j\nu\sigma|\phi_\ell\rangle \ .\end{aligned} \tag{1.27}$$

We can also write the equation

$$\langle\phi_\ell|\hat{H}^{\mathrm{HF}} = \langle\phi_\ell|E_\ell \ . \tag{1.28}$$

Projecting in a state of orbital μ of site i with spin σ from the right side,

$$\langle\phi_\ell|\hat{H}^{\mathrm{HF}}|i\mu\sigma\rangle = E_\ell\langle\phi_\ell|i\mu\sigma\rangle \ . \tag{1.29}$$

Using Eq. (1.21),

$$\begin{aligned}\sum_{m\alpha s} t^{s\,\alpha\mu}_{mi}\delta_{\sigma s}\langle\phi_\ell|m\alpha s\rangle + \sum_{\alpha}\langle\phi_\ell|\left\{a_{i,\alpha\mu}\delta_{\sigma\downarrow}|i\alpha\uparrow\rangle + a^*_{i,\mu\alpha}\delta_{\sigma\uparrow}|i\alpha\downarrow\rangle\right\} =& E_\ell\langle\phi_\ell|i\mu\sigma\rangle \\ \sum_{m\alpha s}\left\{t^{s\,\alpha\mu}_{mi}\delta_{\sigma s} + a_{m,\alpha\mu}\delta_{mi}\delta_{\sigma\downarrow}\delta_{s\uparrow} + a^*_{m,\mu\alpha}\delta_{mi}\delta_{\sigma\uparrow}\delta_{s\downarrow}\right\}\langle\phi_\ell|m\alpha s\rangle =& E_\ell\langle\phi_\ell|i\mu\sigma\rangle \ .\end{aligned} \tag{1.30}$$

1.3.1 Lowde-Windsor approximation

Using Lowde-Windsor approximation for the local eletronic interaction, $U^m_{\gamma\beta\xi\alpha} = U^m_{\alpha\beta}\delta_{\gamma\alpha}\delta_{\beta\xi}$, in Eq. (1.21), we have

$$\hat{H}'_0 = \sum_{\substack{mn\\s}}\sum_{\alpha\beta} t^{s\,\alpha\beta}_{mn} c^\dagger_{m\alpha s} c_{n\beta s} \ , \tag{1.31}$$

with

$$
\begin{aligned}
t^{s\,\alpha\beta}_{mn} =& t^{\alpha\beta}_{mn} + \delta_{mn} \sum_{\gamma\xi} \Big\{ \sum_{s'} U^m_{\gamma\xi} \delta_{\alpha\xi} \delta_{\gamma\beta} \langle c^\dagger_{m\gamma s'} c_{m\xi s'} \rangle - U^m_{\alpha\xi} \delta_{\gamma\xi} \delta_{\alpha\beta} \langle c^\dagger_{m\gamma s} c_{m\xi s} \rangle \Big\} \pm \delta_{mn} \frac{\lambda_m}{2} L'^z_{\alpha\beta} \pm \delta_{mn} \delta_{\alpha\beta} \frac{\hbar\omega_z}{2} \\
=& t^{\alpha\beta}_{mn} + \delta_{mn} \Big\{ \sum_{s'} U^m_{\beta\alpha} \langle c^\dagger_{m\beta s'} c_{m\alpha s'} \rangle - \sum_{\gamma} U^m_{\alpha\gamma} \delta_{\alpha\beta} \langle c^\dagger_{m\gamma s} c_{m\gamma s} \rangle \Big\} \pm \delta_{mn} \frac{\lambda_m}{2} L'^z_{\alpha\beta} \pm \delta_{mn} \delta_{\alpha\beta} \frac{\hbar\omega_z}{2} \\
=& t^{\alpha\beta}_{mn} + \delta_{mn} \Big\{ \sum_{s'} U^m \langle c^\dagger_{m\beta s'} c_{m\alpha s'} \rangle - U^m \delta_{\alpha\beta} n^s_m \Big\} \pm \delta_{mn} \frac{\lambda_m}{2} L'^z_{\alpha\beta} \pm \delta_{mn} \delta_{\alpha\beta} \frac{\hbar\omega_z}{2} \\
=& t^{\alpha\beta}_{mn} + \delta_{mn} \Big\{ \sum_{s'} U^m \langle c^\dagger_{m\beta s'} c_{m\alpha s'} \rangle + U^m \delta_{\alpha\beta} \frac{n^{\bar{s}}_m - n^s_m}{2} - U^m \delta_{\alpha\beta} \frac{n^{\bar{s}}_m + n^s_m}{2} \Big\} \pm \delta_{mn} \frac{\lambda_m}{2} L'^z_{\alpha\beta} \pm \delta_{mn} \delta_{\alpha\beta} \frac{\hbar\omega_z}{2} \\
=& t^{\alpha\beta}_{mn} + \delta_{mn} \Big\{ \sum_{s'} U^m \langle c^\dagger_{m\beta s'} c_{m\alpha s'} \rangle \mp \delta_{\alpha\beta} \frac{U^m m^{d,z}_m}{2\mu_\mathrm{B}} - \delta_{\alpha\beta} \frac{U^m n^d_m}{2} \Big\} \pm \delta_{mn} \frac{\lambda_m}{2} L'^z_{\alpha\beta} \pm \delta_{mn} \delta_{\alpha\beta} \frac{\hbar\omega_z}{2} \ .
\end{aligned}
\tag{1.32}
$$

where the upper sign holds for spins $\uparrow$ and the lower for spins $\downarrow$. The magnetization is defined as $m^{d,z}_m = \mu_B \left(n^\uparrow_m - n^\downarrow_m \right) = -\frac{2\mu_\mathrm{B}}{\hbar} \langle S^z_m \rangle$, and the total occupation $n^d_m = n^\uparrow_m + n^\downarrow_m$. n^s_m is obtained by summing over the d orbitals only, for which $U^m \neq 0$. Taking into account orthogonal orbitals on the same site, $\langle c^\dagger_{m\beta s'} c_{m\alpha s'} \rangle = \delta_{\alpha\beta} n^{s'}_{m\alpha}$, where $n_{m\alpha} = n^\uparrow_{m\alpha} + n^\downarrow_{m\alpha}$ is the total occupation of orbital α in site m (α is non zero only for the d orbitals). So, the final form of the hamiltonian is

$$
\begin{aligned}
t^{s\,\alpha\beta}_{mn} =& t^{\alpha\beta}_{mn} + \delta_{mn} \delta_{\alpha\beta} \left\{ U^m n_{m\alpha} - \sum_{\gamma\in d} \frac{U^m n_{m\gamma}}{2} \mp \frac{U^m m^{d,z}_m}{2\mu_\mathrm{B}} \right\} \pm \delta_{mn} \frac{\lambda_m}{2} L'^z_{\alpha\beta} \pm \delta_{mn} \delta_{\alpha\beta} \frac{\hbar\omega_z}{2} \\
=& t^{\alpha\beta}_{mn} + \delta_{mn} \delta_{\alpha\beta} \left\{ \sum_{\gamma\in d} \left(U^m \delta_{\alpha\gamma} - \frac{U^m}{2} \right) n_{m\gamma} \mp \frac{U^m m^{d,z}_m}{2\mu_\mathrm{B}} \right\} \pm \delta_{mn} \frac{\lambda_m}{2} L'^z_{\alpha\beta} \pm \delta_{mn} \delta_{\alpha\beta} \frac{\hbar\omega_z}{2} \ .
\end{aligned}
\tag{1.33}
$$

The values of $n_{m\alpha}$ ($\alpha \in d$) and m^d_m are obtained self-consistently as explained in Appendix K (n^d_m is obtained by summing $n_{m\alpha}$ over d orbitals).

The second term of the hamiltonian is

$$
\hat{H}_\mathrm{sf} = \sum_m \sum_{\alpha\beta} \left\{ a_{m,\alpha\beta} c^\dagger_{m\alpha\uparrow} c_{m\beta\downarrow} + a^*_{m,\beta\alpha} c^\dagger_{m\alpha\downarrow} c_{m\beta\uparrow} \right\} \ . \tag{1.34}
$$

where

$$
\begin{aligned}
a_{m,\alpha\beta} =& \frac{\lambda_m}{2} L'^{-}_{\alpha\beta} - \sum_{\gamma\xi} U^m \delta_{\gamma\xi}\delta_{\alpha\beta}\langle c^\dagger_{m\gamma\downarrow} c_{m\xi\uparrow}\rangle + \delta_{\alpha\beta}\frac{\hbar\omega_-}{2} \\
=& \frac{\lambda_m}{2} L'^{-}_{\alpha\beta} - \sum_{\gamma} U^m \delta_{\alpha\beta}\langle c^\dagger_{m\gamma\downarrow} c_{m\gamma\uparrow}\rangle + \delta_{\alpha\beta}\frac{\hbar\omega_-}{2} \\
=& \frac{\lambda_m}{2} L'^{-}_{\alpha\beta} - \frac{U^m}{\hbar}\delta_{\alpha\beta}\langle S^-_m\rangle + \delta_{\alpha\beta}\frac{\hbar\omega_-}{2} \\
=& \frac{\lambda_m}{2} L'^{-}_{\alpha\beta} - \frac{U^m}{2\mu_\mathrm{B}}\delta_{\alpha\beta} m^-_m + \delta_{\alpha\beta}\frac{\hbar\omega_-}{2} \ .
\end{aligned}
\tag{1.35}
$$

and

$$
\begin{aligned}
a^*_{m,\beta\alpha} =& \frac{\lambda_m}{2} L'^{+}_{\alpha\beta} - \sum_{\gamma\xi} U^m \delta_{\xi\gamma}\delta_{\beta\alpha}\langle c^\dagger_{m\xi\uparrow} c_{m\gamma\downarrow}\rangle + \delta_{\alpha\beta}\frac{\hbar\omega_+}{2} \\
=& \frac{\lambda_m}{2} L'^{+}_{\alpha\beta} - \frac{U^m}{\hbar}\delta_{\beta\alpha}\langle S^+_m\rangle + \delta_{\alpha\beta}\frac{\hbar\omega_+}{2} \\
=& \frac{\lambda_m}{2} L'^{+}_{\alpha\beta} - \frac{U^m}{2\mu_\mathrm{B}}\delta_{\beta\alpha} m^+_m + \delta_{\alpha\beta}\frac{\hbar\omega_+}{2} \ ,
\end{aligned}
\tag{1.36}
$$

Substituting Eqs. (1.31) and (1.34) into Eq. (1.21), we can write the hamiltonian in spin space as

$$
\begin{aligned}
\hat{H}^{\mu\nu}_{ij} =& H^{0\,\mu\nu}_{ij}\sigma^0 + \delta_{ij}\delta_{\mu\nu}\left(-\frac{U^i}{2\mu_\mathrm{B}}\right)\left[\sigma^z m^z_i + \sigma^x m^x_i + \sigma^y m^y_i\right] + \delta_{ij}\frac{\lambda_i}{2}\left[\sigma^z L'^z_{\mu\nu} + \sigma^x L'^x_{\mu\nu} + \sigma^y L'^y_{\mu\nu}\right] \\
&+ \delta_{ij}\delta_{\mu\nu}\frac{g_S\mu_B}{2}\left[\sigma^z B'^z + \sigma^x B'^x + \sigma^y B'^y\right] \\
=& H^{0\,\mu\nu}_{ij}\sigma^0 + \boldsymbol{\sigma}\cdot\left[-\frac{U^i}{2\mu_\mathrm{B}}\delta_{\mu\nu}\mathbf{m}_i + \frac{\lambda_i}{2}\mathbf{L}'_{\mu\nu} + \mu_B\delta_{\mu\nu}\mathbf{B}'\right]\delta_{ij} \\
=& H^{0\,\mu\nu}_{ij}\sigma^0 + \boldsymbol{\sigma}\cdot\left[\mathbf{B}^{\mathrm{xc}\,\mu\nu}_i + \mathbf{B}^{\mathrm{soc}\,\mu\nu}_i + \mathbf{B}^{\mathrm{ext}\,\mu\nu}_i\right]\delta_{ij} \ .
\end{aligned}
\tag{1.37}
$$

Here, σ^0 is the identity matrix, H^0 is the spin-independent part of the hamiltonian, we took $g_S = 2$ and we have defined (in units of energy) $\mathbf{B}^{\mathrm{xc}\,\mu\nu}_i = -\delta_{\mu\nu}\frac{U^i}{2\mu_\mathrm{B}}\mathbf{m}_i$, $\mathbf{B}^{\mathrm{soc}\,\mu\nu}_i = \frac{\lambda_i}{2}\mathbf{L}'_{\mu\nu}$ and $\mathbf{B}^{\mathrm{ext}\,\mu\nu}_i = \delta_{\mu\nu}\mathbf{B}'$. In a simplified form,

$$
\hat{H} = \sum_{\substack{ij\\ \mu\nu}}\sum_{\alpha} c^\dagger_{i\mu\alpha} H^{0\,\mu\nu}_{ij} c_{j\nu\alpha} + \sum_{\substack{m,i\\ \mu\nu}}\sum_{\alpha\beta} c^\dagger_{i\mu\alpha}\sigma^m_{\alpha\beta} B^{m\,\mu\nu}_i c_{i\nu\beta} \ . \tag{1.38}
$$

1.3.2 Correlation functions

The monoelectronic Green functions are defined as

$$
G^{\sigma\sigma'}(i\mu; j\nu; t) = -\frac{i}{\hbar}\Theta(t)\langle\{\hat{c}_{i\mu\sigma}(t), \hat{c}^\dagger_{j\nu\sigma'}\}\rangle \ . \tag{1.39}
$$

In terms of the correlation functions,

$$G^{\sigma\sigma'}(i\mu;j\nu;t) = -\frac{i}{\hbar}\Theta(t)\{J^{\sigma\sigma'}(i\mu;j\nu;t) + \overline{J}^{\sigma'\sigma}(j\nu;i\mu;t)\} \ . \tag{1.40}$$

where the correlation functions are

$$\begin{aligned} J^{\sigma\sigma'}(i\mu;j\nu;t) =& \langle \hat{c}_{i\mu\sigma}(t)\hat{c}^{\dagger}_{j\nu\sigma'}\rangle \ , \text{e} \\ \overline{J}^{\sigma'\sigma}(j\nu;i\mu;t) =& \langle \hat{c}^{\dagger}_{j\nu\sigma'}\hat{c}_{i\mu\sigma}(t)\rangle \ . \end{aligned} \tag{1.41}$$

The integral representation for the step function is

$$\Theta(t) = \lim_{\eta\to 0^+}\left(\frac{-1}{2\pi i}\right)\int_{-\infty}^{\infty}\frac{e^{-i\omega t}}{\omega + i\eta}d\omega \ , \tag{1.42}$$

and the Fourier transform of the correlation functions

$$\begin{aligned} J^{\sigma\sigma'}(i\mu;j\nu;t) =& \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-i\omega t}\widetilde{J}^{\sigma\sigma'}(i\mu;j\nu;\omega)d\omega \ , \text{e} \\ \overline{J}^{\sigma'\sigma}(j\nu;i\mu;t) =& \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-i\omega t}\widetilde{\overline{J}}^{\sigma'\sigma}(j\nu;i\mu;\omega)d\omega \ . \end{aligned} \tag{1.43}$$

Using the relation

$$\widetilde{J}^{\sigma\sigma'}(i\mu;j\nu;\omega) = e^{\beta\hbar\omega}\widetilde{\overline{J}}^{\sigma'\sigma}(j\nu;i\mu;\omega) \ , \tag{1.44}$$

where $\beta = \frac{1}{k_B T}$, the Green function can now be written as

$$\begin{aligned} G^{\sigma\sigma'}(i\mu;j\nu;t) =& \frac{1}{2\pi\hbar}\int d\omega' \lim_{\eta\to 0^+}\frac{1}{2\pi}\int d\omega \frac{e^{-i(\omega+\omega')t}}{\omega + i\eta}\left\{e^{\beta\hbar\omega'}+1\right\}\widetilde{\overline{J}}^{\sigma'\sigma}(j\nu;i\mu;\omega') \\ =& \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega'' d\omega' \lim_{\eta\to 0^+}\frac{1}{2\pi\hbar}\frac{e^{-i\omega'' t}}{\omega'' - \omega' + i\eta}\left\{e^{\beta\hbar\omega'}+1\right\}\widetilde{\overline{J}}^{\sigma'\sigma}(j\nu;i\mu;\omega') \ , \end{aligned} \tag{1.45}$$

Rearranging this terms, we can write

$$\begin{aligned} G^{\sigma\sigma'}(i\mu;j\nu;t) =& \frac{1}{2\pi}\int d\omega e^{-i\omega t}\lim_{\eta\to 0^+}\frac{1}{2\pi\hbar}\int d\omega' \frac{e^{\beta\hbar\omega'}+1}{\omega - \omega' + i\eta}\widetilde{\overline{J}}^{\sigma'\sigma}(j\nu;i\mu;\omega') \\ =& \frac{1}{2\pi}\int d\omega e^{-i\omega t}\widetilde{G}^{\sigma\sigma'}(i\mu;j\nu;\omega) \ . \end{aligned} \tag{1.46}$$

So we identify the Fourier transform of the monoeletronic Green function as

$$\widetilde{G}^{\sigma\sigma'}(i\mu;j\nu;\omega) = \lim_{\eta\to 0^+}\frac{1}{2\pi\hbar}\int d\omega' \frac{e^{\beta\hbar\omega'}+1}{\omega - \omega' + i\eta}\widetilde{\overline{J}}^{\sigma'\sigma}(j\nu;i\mu;\omega') \ . \tag{1.47}$$

Doing the same steps for the advanced Green function, defined as

$$G^{-\sigma\sigma'}(i\mu;j\nu;t) = \frac{i}{\hbar}\Theta(-t)\{J^{\sigma\sigma'}(i\mu;j\nu;t) + \overline{J}^{\sigma'\sigma}(j\nu;i\mu;t)\} \ , \tag{1.48}$$

we find

$$\widetilde{G}^{-\sigma\sigma'}(i\mu;j\nu;\omega) = \lim_{\eta\to 0^+} \frac{1}{2\pi\hbar} \int d\omega' \frac{e^{\beta\hbar\omega'}+1}{\omega-\omega'-i\eta} \widetilde{\overline{J}}^{\sigma'\sigma}(j\nu;i\mu;\omega') \ . \tag{1.49}$$

The difference between retarded and advanced Green functions is given by

$$\begin{aligned}\widetilde{G}^{\sigma\sigma'}(i\mu;j\nu;\omega) - \widetilde{G}^{-\sigma\sigma'}(i\mu;j\nu;\omega) =& \frac{1}{2\pi\hbar} \int d\omega' \left(e^{\beta\hbar\omega'}+1\right) \widetilde{\overline{J}}^{\sigma'\sigma}(j\nu;i\mu;\omega') \\ & \times \lim_{\eta\to 0^+} \left\{ \frac{1}{\omega-\omega'+i\eta} - \frac{1}{\omega-\omega'-i\eta} \right\} \ .\end{aligned} \tag{1.50}$$

Using the relation

$$\lim_{\eta\to 0^+} \frac{1}{x\pm i\eta} = P\Big(\frac{1}{x}\Big) \mp i\pi\delta(x) \ , \tag{1.51}$$

we obtain

$$\begin{aligned}\widetilde{G}^{\sigma\sigma'}(i\mu;j\nu;\omega) - \widetilde{G}^{-\sigma\sigma'}(i\mu;j\nu;\omega) =& \frac{1}{2\pi\hbar} \int d\omega' \left(e^{\beta\hbar\omega'}+1\right) \widetilde{\overline{J}}^{\sigma'\sigma}(j\nu;i\mu;\omega') \left[-2\pi i\delta(\omega-\omega')\right] \\ =& -\frac{i}{\hbar} \left(e^{\beta\hbar\omega}+1\right) \widetilde{\overline{J}}^{\sigma'\sigma}(j\nu;i\mu;\omega) \ .\end{aligned} \tag{1.52}$$

This gives us the discontinuity of the Green function on the real axis.

Finally, we can write the correlation function as

$$\begin{aligned}\widetilde{\overline{J}}^{\sigma'\sigma}(j\nu;i\mu;\omega) =& i\hbar \frac{1}{e^{\beta\hbar\omega}+1} \left[\widetilde{G}^{\sigma\sigma'}(i\mu;j\nu;\omega) - \widetilde{G}^{-\sigma\sigma'}(i\mu;j\nu;\omega)\right] \\ =& i\hbar f(\omega) \left[\widetilde{G}^{\sigma\sigma'}(i\mu;j\nu;\omega) - \widetilde{G}^{-\sigma\sigma'}(i\mu;j\nu;\omega)\right] \ ,\end{aligned} \tag{1.53}$$

where $f(\omega)$ is the Fermi-Dirac distribution function. Substituting in the Fourier transform,

$$\begin{aligned}\overline{J}^{\sigma'\sigma}(j\nu;i\mu;t) = \langle \hat{c}^\dagger_{j\nu\sigma'} \hat{c}_{i\mu\sigma}(t) \rangle =& \frac{1}{2\pi} \int d\omega e^{-i\omega t} \widetilde{\overline{J}}^{\sigma'\sigma}(j\nu;i\mu;\omega) \\ =& \frac{i\hbar}{2\pi} \int d\omega e^{-i\omega t} f(\omega) \left[\widetilde{G}^{\sigma\sigma'}(i\mu;j\nu;\omega) - \widetilde{G}^{-\sigma\sigma'}(i\mu;j\nu;\omega)\right] \ .\end{aligned} \tag{1.54}$$

The quantities we need to calculate are in the fundamental state. So, for $t = 0$,

$$\langle \hat{c}^\dagger_{j\nu\sigma'} \hat{c}_{i\mu\sigma} \rangle = \frac{i\hbar}{2\pi} \int d\omega f(\omega) \left[\widetilde{G}^{\sigma\sigma'}(i\mu;j\nu;\omega) - \widetilde{G}^{-\sigma\sigma'}(i\mu;j\nu;\omega)\right] \ . \tag{1.55}$$

Also, for $T = 0K$,

$$\langle \hat{c}^\dagger_{j\nu\sigma'} \hat{c}_{i\mu\sigma} \rangle = \frac{i\hbar}{2\pi} \int_{-\infty}^{\omega_F} d\omega \left\{ \widetilde{G}^{\sigma\sigma'}(i\mu;j\nu;\omega) - \left[\widetilde{G}^{\sigma'\sigma}(j\nu;i\mu;\omega)\right]^* \right\} \ . \tag{1.56}$$

Using the spin operators, we can write

$$\langle \hat{S}^{m}_{ij}{}^{\mu\nu} \rangle = \frac{1}{2}\sum_{\alpha\beta}\langle \hat{c}^{\dagger}_{i\mu\alpha}\sigma^{m}_{\alpha\beta}\hat{c}_{j\nu\beta}\rangle = \frac{i\hbar}{4\pi}\int_{-\infty}^{\omega_F} d\omega \left\{ \widetilde{G}^{\beta\alpha}(j\nu;i\mu;\omega) - \left[\widetilde{G}^{\alpha\beta}(i\mu;j\nu;\omega)\right]^{*} \right\}$$
$$= -\frac{\hbar}{4i\pi}\int_{-\infty}^{\omega_F} d\omega \sigma^{m}_{\alpha\beta} \left\{ \widetilde{G}^{\beta\alpha}(j\nu;i\mu;\omega+i0^{+}) - \widetilde{G}^{\beta\alpha}(j\nu;i\mu;\omega-i0^{+}) \right\} \ . \quad (1.57)$$

Using Eq. (C.12), we have

$$\begin{aligned}\langle \hat{S}^{m}_{ij}{}^{\mu\nu} \rangle &= -\frac{\hbar}{2\pi}\int_{-\infty}^{\omega_F} d\omega \, \mathrm{Tr}\, \Im \boldsymbol{\sigma}^{m} \widetilde{\mathbf{G}}(j\nu;i\mu;\omega) \\ &= -\frac{1}{\pi}\int_{-\infty}^{\omega_F} d\omega \, \mathrm{Tr}\, \Im \hat{\boldsymbol{S}}^{m} \widetilde{\mathbf{G}}(j\nu;i\mu;\omega) \ .\end{aligned} \quad (1.58)$$

We need to calculate

$$\langle S^{+}_{m} \rangle = \sum_{\mu}\langle \hat{c}^{\dagger}_{m\mu\uparrow}\hat{c}_{m\mu\downarrow}\rangle = \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F} d\omega \left\{ \widetilde{G}^{\downarrow\uparrow}(m\mu;m\mu;\omega) - \left[\widetilde{G}^{\uparrow\downarrow}(m\mu;m\mu;\omega)\right]^{*} \right\} \ , \quad (1.59)$$

and

$$\langle S^{-}_{m} \rangle = \sum_{\mu}\langle \hat{c}^{\dagger}_{m\mu\downarrow}\hat{c}_{m\mu\uparrow}\rangle = \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F} d\omega \left\{ \widetilde{G}^{\uparrow\downarrow}(m\mu;m\mu;\omega) - \left[\widetilde{G}^{\downarrow\uparrow}(m\mu;m\mu;\omega)\right]^{*} \right\} \ . \quad (1.60)$$

Extending the first integration of Eq. (1.56) to the upper half-plane of the imaginary space, we have $I_r = -I_i - I_C$. So,

$$\frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F} dz \widetilde{G}^{\sigma\sigma'}(i\mu;j\nu;z) = \frac{\hbar}{2\pi}\int_{\eta}^{\infty} dy \widetilde{G}^{\sigma\sigma'}(i\mu;j\nu;\omega_F+iy) + \frac{\hbar}{4}\delta_{ij}\delta_{\mu\nu}\delta_{\sigma\sigma'} \ . \quad (1.61)$$

The second integration is closed on the lower half-plane, and we obtain

$$-\frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F} dz \widetilde{G}^{-\sigma\sigma'}(i\mu;j\nu;z) = \frac{\hbar}{2\pi}\int_{\eta}^{\infty} dy \widetilde{G}^{-\sigma\sigma'}(i\mu;j\nu;\omega_F-iy) + \frac{\hbar}{4}\delta_{ij}\delta_{\mu\nu}\delta_{\sigma\sigma'} \ . \quad (1.62)$$

Thus, the correlation function can be written as

$$\langle \hat{c}^{\dagger}_{j\nu\sigma'}\hat{c}_{i\mu\sigma}\rangle = \frac{\hbar}{2}\delta_{ij}\delta_{\mu\nu}\delta_{\sigma\sigma'} + \frac{\hbar}{2\pi}\int_{\eta}^{\infty} dy \left\{ \widetilde{G}^{\sigma\sigma'}(i\mu;j\nu;\omega_F+iy) + \left[\widetilde{G}^{\sigma'\sigma}(j\nu;i\mu;\omega_F+iy)\right]^{*} \right\} \ . \quad (1.63)$$

To calculate the Hartre-Fock Hamiltonian, we need

$$\langle S^{+}_{m} \rangle = \sum_{\mu}\langle \hat{c}^{\dagger}_{m\mu\uparrow}\hat{c}_{m\mu\downarrow}\rangle = \frac{\hbar}{2\pi}\int_{\eta}^{\infty} dy \sum_{\mu} \left\{ \widetilde{G}^{\downarrow\uparrow}(m\mu;m\mu;\omega_F+iy) + \left[\widetilde{G}^{\uparrow\downarrow}(m\mu;m\mu;\omega_F+iy)\right]^{*} \right\} \ . \quad (1.64)$$

$\langle S_m^- \rangle$ can be obtained by

$$\begin{aligned}\langle S_m^- \rangle =& \sum_\mu \langle \hat{c}^\dagger_{m\mu\downarrow} \hat{c}_{m\mu\uparrow} \rangle = \sum_\mu \left[\langle \hat{c}^\dagger_{m\mu\uparrow} \hat{c}_{m\mu\downarrow} \rangle \right]^* = \left[\langle S_m^+ \rangle \right]^* \\ =& \frac{\hbar}{2\pi} \int_\eta^\infty dy \sum_\mu \left\{ \widetilde{G}^{\uparrow\downarrow}(m\mu; m\mu; \omega_F + iy) + \left[\widetilde{G}^{\downarrow\uparrow}(m\mu; m\mu; \omega_F + iy) \right]^* \right\},\end{aligned} \tag{1.65}$$

and the last new correlation function we need is

$$\langle c^\dagger_{m\beta s'} c_{m\alpha s'} \rangle = \frac{\hbar}{2}\delta_{\alpha\beta} + \frac{\hbar}{2\pi} \int_\eta^\infty dy \left\{ \widetilde{G}^{s's'}(m\alpha; m\beta; \omega_F + iy) + \left[\widetilde{G}^{s's'}(m\beta; m\alpha; \omega_F + iy) \right]^* \right\} \ . \tag{1.66}$$

In multilayers, we can write

$$\langle \hat{c}^\dagger_{\ell'\nu\sigma'} \hat{c}_{\ell\mu\sigma} \rangle (\mathbf{R}_i - \mathbf{R}_j) = \frac{i\hbar}{2\pi} \int_{-\infty}^{\omega_F} d\omega \left\{ \widetilde{G}^{\sigma\sigma'}(\ell\mu; \ell'\nu; \mathbf{R}_j - \mathbf{R}_i; \omega) - \left[\widetilde{G}^{\sigma'\sigma}(\ell'\nu; \ell\mu; \mathbf{R}_i - \mathbf{R}_j; \omega) \right]^* \right\} \ . \tag{1.67}$$

Using the Fourier transformation of the Green functions,

$$\widetilde{G}^{\sigma\sigma'}(\ell\mu; \ell'\nu; \mathbf{R}_\parallel; \omega_F + iy) = \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel \cdot \mathbf{R}_\parallel} \widetilde{G}^{\sigma\sigma'}(\ell\mu; \ell'\nu; \mathbf{k}_\parallel; \omega_F + iy), \tag{1.68}$$

we have

$$\langle \hat{c}^\dagger_{\ell'\nu\sigma'} \hat{c}_{\ell\mu\sigma} \rangle (\mathbf{R}_i - \mathbf{R}_j) = \frac{i\hbar}{2\pi} \int_{-\infty}^{\omega_F} d\omega \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel \cdot (\mathbf{R}_j - \mathbf{R}_i)} \left\{ \widetilde{G}^{\sigma\sigma'}(\ell\mu; \ell'\nu; \mathbf{k}_\parallel; \omega) - \left[\widetilde{G}^{\sigma'\sigma}(\ell'\nu; \ell\mu; \mathbf{k}_\parallel; \omega) \right]^* \right\} \ . \tag{1.69}$$

In the imaginary axis,

$$\begin{aligned}\langle \hat{c}^\dagger_{\ell'\nu\sigma'} \hat{c}_{\ell\mu\sigma} \rangle (\mathbf{R}_i - \mathbf{R}_j) = \frac{\hbar}{2}\delta_{\ell\ell}\delta_{\mu\nu}\delta_{\sigma\sigma'}\delta(\mathbf{R}_j - \mathbf{R}_i) + \frac{\hbar}{2\pi} \int_\eta^\infty dy \Big\{ \widetilde{G}^{\sigma\sigma'}(\ell\mu; \ell'\nu; \mathbf{R}_j - \mathbf{R}_i; \omega_F + iy) \\ + \left[\widetilde{G}^{\sigma'\sigma}(\ell'\nu; \ell\mu; \mathbf{R}_i - \mathbf{R}_j; \omega_F + iy) \right]^* \Big\} \ .\end{aligned} \tag{1.70}$$

Once again we use the Fourier transformation to obtain

$$\begin{aligned}\langle \hat{c}^\dagger_{\ell'\nu\sigma'} \hat{c}_{\ell\mu\sigma} \rangle (\mathbf{R}_i - \mathbf{R}_j) =& \frac{\hbar}{2}\delta_{\ell\ell'}\delta_{\mu\nu}\delta_{\sigma\sigma'}\delta(\mathbf{R}_j - \mathbf{R}_i) \\ &+ \frac{\hbar}{2\pi} \int_\eta^\infty dy \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel \cdot (\mathbf{R}_j - \mathbf{R}_i)} \Big\{ \widetilde{G}^{\sigma\sigma'}(\ell\mu; \ell'\nu; \mathbf{k}_\parallel; \omega_F + iy) \\ &+ \left[\widetilde{G}^{\sigma'\sigma}(\ell'\nu; \ell\mu; \mathbf{k}_\parallel; \omega_F + iy) \right]^* \Big\} \ .\end{aligned} \tag{1.71}$$

Since we need only on-site correlation functions,

$$\begin{aligned}\langle\hat{c}^\dagger_{\ell\nu\sigma'}\hat{c}_{\ell\mu\sigma}\rangle(0) = \frac{\hbar}{2}\delta_{\mu\nu}\delta_{\sigma\sigma'} + \frac{\hbar}{2\pi}\int_\eta^\infty dy\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\Big\{&\widetilde{G}^{\sigma\sigma'}(\ell\mu;\ell\nu;\mathbf{k}_\parallel;\omega_F+iy)\\ &+\Big[\widetilde{G}^{\sigma'\sigma}(\ell\nu;\ell\mu;\mathbf{k}_\parallel;\omega_F+iy)\Big]^*\Big\}\ .\end{aligned}\tag{1.72}$$

or

$$\begin{aligned}\langle\hat{c}^\dagger_{\ell\mu\sigma}\hat{c}_{\ell\nu\sigma'}\rangle(0) = \frac{\hbar}{2}\delta_{\mu\nu}\delta_{\sigma\sigma'} + \frac{\hbar}{2\pi}\int_\eta^\infty dy\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\Big\{&\widetilde{G}^{\sigma'\sigma}(\ell\nu;\ell\mu;\mathbf{k}_\parallel;\omega_F+iy)\\ &+\Big[\widetilde{G}^{\sigma\sigma'}(\ell\mu;\ell\nu;\mathbf{k}_\parallel;\omega_F+iy)\Big]^*\Big\}\ .\end{aligned}\tag{1.73}$$

1.3.3 Non-perturbed Orbital Angular Momentum

To calculate the orbital angular momentum in the fundamental state, we can write the operator $\hat{L}^m$ in second quantization as

$$\begin{aligned}\hat{L}^m_i =&\sum_{\substack{\alpha\beta\\ s}}\left\langle i\alpha s\left|\hat{L}^m_i\right|i\beta s\right\rangle c^\dagger_{i\alpha s}c_{i\beta s}\\ =&\sum_{\substack{\alpha\beta\\ s}}\hat{L}^m_{\alpha\beta}c^\dagger_{i\alpha s}c_{i\beta s}\ .\end{aligned}\tag{1.74}$$

Its expectation value is given by

$$\begin{aligned}\langle\hat{L}^m_i\rangle =&\sum_{\substack{\alpha\beta\\ s}}\hat{L}^m_{\alpha\beta}\langle c^\dagger_{i\alpha s}c_{i\beta s}\rangle\\ =&\frac{i\hbar}{2\pi}\sum_{\substack{\alpha\beta\\ s}}\hat{L}^m_{\alpha\beta}\int_{-\infty}^{\omega_F}d\omega\left\{\widetilde{G}^{ss}(i\beta;i\alpha;\omega)-\Big[\widetilde{G}^{ss}(i\alpha;i\beta;\omega)\Big]^*\right\}\\ =&\frac{i\hbar}{2\pi}\sum_{\substack{\alpha\beta\\ s}}\int_{-\infty}^{\omega_F}d\omega\left\{\hat{L}^m_{\alpha\beta}\widetilde{G}^{ss}(i\beta;i\alpha;\omega)-\Big[\hat{L}^m_{\beta\alpha}\widetilde{G}^{ss}(i\alpha;i\beta;\omega)\Big]^*\right\}\\ =&\frac{i\hbar}{2\pi}\sum_{\substack{\alpha\beta\\ s}}\int_{-\infty}^{\omega_F}d\omega\left\{\hat{L}^m_{\alpha\beta}\widetilde{G}^{ss}(i\beta;i\alpha;\omega)-\Big[\hat{L}^m_{\alpha\beta}\widetilde{G}^{ss}(i\beta;i\alpha;\omega)\Big]^*\right\}\\ =&-\frac{\hbar}{\pi}\sum_{\alpha\beta}\int_{-\infty}^{\omega_F}d\omega\,\mathrm{Im}\left\{\hat{L}^m_{\alpha\beta}\Big[\widetilde{G}^{\uparrow\uparrow}(i\beta;i\alpha;\omega)+\widetilde{G}^{\downarrow\downarrow}(i\beta;i\alpha;\omega)\Big]\right\}\\ =&\frac{\hbar}{\pi}\sum_{\alpha\beta}\int_\eta^\infty dy\,\mathrm{Re}\left\{\hat{L}^m_{\alpha\beta}\Big[\widetilde{G}^{\uparrow\uparrow}(i\beta;i\alpha;\omega_F+iy)+\widetilde{G}^{\downarrow\downarrow}(i\beta;i\alpha;\omega_F+iy)\Big]\right\}\ ,\end{aligned}\tag{1.75}$$

where we have used Eqs. 1.56 and 1.63 and the fact that all diagonal elements of the matrices $L^m_{\alpha\beta}$ are zero.

For multilayers, this last result can be transformed to

$$\langle \hat{L}_\ell^m \rangle = \frac{\hbar}{\pi} \sum_{\alpha\beta} \int_\eta^\infty dy \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \mathrm{Re} \left\{ \hat{L}_{\alpha\beta}^m \left[\widetilde{G}^{\uparrow\uparrow}(\ell\beta; \ell\alpha; \mathbf{k}_\parallel; \omega_F + iy) + \widetilde{G}^{\downarrow\downarrow}(\ell\beta; \ell\alpha; \mathbf{k}_\parallel; \omega_F + iy) \right] \right\} \ . \tag{1.76}$$

The orbital angular momentum can be calculated in the cubic system of coordinates using the matrices $\hat{L}_{\alpha\beta}^m$ or in the spin system of coordinates using $\hat{L}_{\alpha\beta}'^m$ (both are obtained in appendix D).

1.3.4 Spin diffusion length

To estimate the spin diffusion length, we can calculate the probability density of an electron invert its spin after some time t as a function of the distance, i.e.,

$$\left| \sum_{\mu\nu} \langle \hat{c}_{i\mu\overline{\sigma}}(t) \hat{c}_{j\nu\sigma}^\dagger \rangle \right|^2 \ . \tag{1.77}$$

To obtain this function, we need the correlation

$$\sum_{\mu\nu} \langle \hat{c}_{i\mu\overline{\sigma}}(t) \hat{c}_{j\nu\sigma}^\dagger \rangle = \sum_{\mu\nu} J^{\overline{\sigma}\sigma}(i\mu; j\nu; t) \ . \tag{1.78}$$

We can also look at this behaviour in the frequency domain using the Fourier transform of the correlation function:

$$\begin{aligned}
\sum_{\mu\nu} \widetilde{J}^{\overline{\sigma}\sigma}(i\mu; j\nu; \omega) =& \sum_{\mu\nu} e^{\beta\hbar\omega} \widetilde{\widetilde{J}}^{\sigma\overline{\sigma}}(j\nu; i\mu; \omega) \\
=& i\hbar \sum_{\mu\nu} e^{\beta\hbar\omega} f(\omega) \left[\widetilde{G}^{\overline{\sigma}\sigma}(i\mu; j\nu; \omega) - \widetilde{G}^{-\overline{\sigma}\sigma}(i\mu; j\nu; \omega) \right] \\
=& i\hbar \sum_{\mu\nu} f(-\omega) \left[\widetilde{G}^{\overline{\sigma}\sigma}(i\mu; j\nu; \omega) - \widetilde{G}^{-\overline{\sigma}\sigma}(i\mu; j\nu; \omega) \right] \ .
\end{aligned} \tag{1.79}$$

where we have used Eqs. 1.44 and 1.53 and the fact that $e^{\beta\hbar\omega} f(\omega) = f(-\omega)$. For $T = 0K$, the spin or charge currents are carried by the electrons in the Fermi level. So, we can calculate

$$\sum_{\mu\nu} \widetilde{J}^{\overline{\sigma}\sigma}(i\mu; j\nu; \omega_F) = \sum_{\mu\nu} i\hbar \left[\widetilde{G}^{\overline{\sigma}\sigma}(i\mu; j\nu; \omega_F) - \widetilde{G}^{-\overline{\sigma}\sigma}(i\mu; j\nu; \omega_F) \right] \ . \tag{1.80}$$

In multilayers,

$$\begin{aligned}
\sum_{\mu\nu} \widetilde{J}^{\overline{\sigma}\sigma}(i\mu; j\nu; \omega_F) =& i\hbar \sum_{\mu\nu} \left\{ \widetilde{G}^{\overline{\sigma}\sigma}(\ell\mu; \ell'\nu; \mathbf{R}_j - \mathbf{R}_i; \omega_F) - \left[\widetilde{G}^{\sigma\overline{\sigma}}(\ell'\nu; \ell\mu; \mathbf{R}_i - \mathbf{R}_j; \omega_F) \right]^* \right\} \\
=& i\hbar \sum_{\mu\nu} \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel \cdot (\mathbf{R}_j - \mathbf{R}_i)} \left\{ \widetilde{G}^{\overline{\sigma}\sigma}(\ell\mu; \ell'\nu; \mathbf{k}_\parallel; \omega_F) - \left[\widetilde{G}^{\sigma\overline{\sigma}}(\ell'\nu; \ell\mu; \mathbf{k}_\parallel; \omega_F) \right]^* \right\} \ .
\end{aligned} \tag{1.81}$$

To obtain the behaviour of this correlation function as a function of the distance between sites of the same plane, we have

$$\sum_{\mu\nu} \widetilde{J}^{\overline{\sigma}\sigma}(i\mu;j\nu;\omega_F) = i\hbar\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)} \left\{\sum_{\mu\nu}\widetilde{G}^{\overline{\sigma}\sigma}(\ell\mu;\ell\nu;\mathbf{k}_\parallel;\omega_F) - \sum_{\mu\nu}\left[\widetilde{G}^{\sigma\overline{\sigma}}(\ell\nu;\ell\mu;\mathbf{k}_\parallel;\omega_F)\right]^*\right\} . \tag{1.82}$$

1.3.5 Superconducting terms

In Eq. 1.15, we assumed that the terms with expectation values of two creation or two annihilation operators were zero, since they involve orthogonal states. This is not the case in superconductors, where $\langle cc\rangle$ and $\langle c^\dagger c^\dagger\rangle$ give rise to the superconducting gap. In this case, the complete form of Eq. 1.15 is

$$\begin{aligned} c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s} \rightarrow & \langle c^\dagger_{m\beta s'}c_{m\xi s'}\rangle c^\dagger_{m\alpha s}c_{m\gamma s} + \langle c^\dagger_{m\alpha s}c_{m\gamma s}\rangle c^\dagger_{m\beta s'}c_{m\xi s'} \\ & - \langle c^\dagger_{m\alpha s}c_{m\xi s'}\rangle c^\dagger_{m\beta s'}c_{m\gamma s} - \langle c^\dagger_{m\beta s'}c_{m\gamma s}\rangle c^\dagger_{m\alpha s}c_{m\xi s'} \\ & + \langle c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}\rangle c_{m\xi s'}c_{m\gamma s} + \langle c_{m\xi s'}c_{m\gamma s}\rangle c^\dagger_{m\alpha s}c^\dagger_{m\beta s'} \ . \end{aligned} \tag{1.83}$$

The extra terms in the hamiltonian are

$$\hat{H}^{\mathrm{HF}}_{\mathrm{int}} = \frac{1}{2}\sum_{\substack{m\\ s,s'}}\sum_{\alpha\beta\gamma\xi} U^m_{\alpha\beta\gamma\xi}\left\{\langle c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}\rangle c_{m\xi s'}c_{m\gamma s} + \langle c_{m\xi s'}c_{m\gamma s}\rangle c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}\right\} . \tag{1.84}$$

Within Lowde-Windsor approximation, where $U^m_{\alpha\beta\gamma\xi} = U^m_{\alpha\beta}\delta_{\alpha\xi}\delta_{\beta\gamma}$,

$$\hat{H}^{\mathrm{HF}}_{\mathrm{int}} = \frac{1}{2}\sum_{\substack{m\\ s,s'}}\sum_{\alpha\beta} U^m_{\alpha\beta}\left\{\langle c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}\rangle c_{m\alpha s'}c_{m\beta s} + \langle c_{m\alpha s'}c_{m\beta s}\rangle c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}\right\} . \tag{1.85}$$

Due to time-reversal symmetry (i.e., conservation of angular momentum), $s \neq s'$, such that

$$\hat{H}^{\mathrm{HF}}_{\mathrm{int}} = \frac{1}{2}\sum_{\substack{m\\ s}}\sum_{\alpha\beta} U^m_{\alpha\beta}\left\{\langle c^\dagger_{m\alpha s}c^\dagger_{m\beta\bar{s}}\rangle c_{m\alpha\bar{s}}c_{m\beta s} + \langle c_{m\alpha\bar{s}}c_{m\beta s}\rangle c^\dagger_{m\alpha s}c^\dagger_{m\beta\bar{s}}\right\} . \tag{1.86}$$

Chapter 2

Monoeletronic Green function

We start by calculating the monoeletronic Green function, which is given by

$$G_{ij}^{\sigma\,\mu\nu}(t) = \langle i\mu\sigma|G(t)|j\nu\sigma\rangle = -\frac{i}{\hbar}\Theta(t)\langle\{c_{i\mu\sigma}(t), c_{j\nu\sigma}^{\dagger}\}\rangle \ . \tag{2.1}$$

We can obtain this propagator through the equation of motion

$$i\hbar\frac{dG_{ij}^{\sigma\sigma'\,\mu\nu}(t)}{dt} = \delta(t)\langle\{c_{i\mu\sigma}, c_{j\nu\sigma'}^{\dagger}\}\rangle - \frac{i}{\hbar}\Theta(t)\langle\{[c_{i\mu\sigma}(t), \hat{H}], c_{j\nu\sigma'}^{\dagger}\}\rangle \ . \tag{2.2}$$

The first commutator is trivially obtained from the definition of the anticommutators of Fermi operators,

$$\langle\{c_{i\mu\sigma}, c_{j\nu\sigma'}^{\dagger}\}\rangle = \delta_{ij}\delta_{\mu\nu}\delta_{\sigma\sigma'}. \tag{2.3}$$

Let's calculate the commutator with the hamiltonian using Eq. (1.21) for the total hamiltonian, as we are calculating it in the fundamental state within Hartree-Fock approximation. So, what we need to calculate is $[c_{i\mu\sigma}, \hat{H}^{\mathrm{HF}}]$. The first commutator is

$$\begin{aligned}
[c_{i\mu\sigma}, \hat{H}_0'] &= \sum_{\substack{mn\\s}}\sum_{\alpha\beta} t_{mn}^{s\,\alpha\beta}[c_{i\mu\sigma}, c_{m\alpha s}^{\dagger}c_{n\beta s}] \\
&= \sum_{\substack{mn\\s}}\sum_{\alpha\beta} t_{mn}^{s\,\alpha\beta}\{c_{i\mu\sigma}, c_{m\alpha s}^{\dagger}\}c_{n\beta s} \\
&= \sum_{\substack{mn\\s}}\sum_{\alpha\beta} t_{mn}^{s\,\alpha\beta}\delta_{im}\delta_{\mu\alpha}\delta_{\sigma s}c_{n\beta s} \\
&= \sum_{n\beta} t_{in}^{\sigma\,\mu\beta}c_{n\beta\sigma} \\
&= \sum_{n\beta s} t_{in}^{\sigma\,\mu\beta}\delta_{s\sigma}c_{n\beta s}
\end{aligned} \tag{2.4}$$

For the second term, we obtain

$$\begin{aligned}
[c_{i\mu\sigma}, \hat{H}^{\mathrm{HF}}_{\mathrm{int}}] &= \sum_m \sum_{\alpha\beta} \left\{ a_{\alpha\beta}[c_{i\mu\sigma}, c^\dagger_{m\alpha\uparrow} c_{m\beta\downarrow}] + a^*_{m,\beta\alpha}[c_{i\mu\sigma}, c^\dagger_{m\alpha\downarrow} c_{m\beta\uparrow}] \right\} \\
&= \sum_m \sum_{\alpha\beta} \left\{ a_{\alpha\beta}\{c_{i\mu\sigma}, c^\dagger_{m\alpha\uparrow}\} c_{m\beta\downarrow} + a^*_{m,\beta\alpha}\{c_{i\mu\sigma}, c^\dagger_{m\alpha\downarrow}\} c_{m\beta\uparrow} \right\} \\
&= \sum_m \sum_{\alpha\beta} \left\{ a_{\alpha\beta}\delta_{im}\delta_{\mu\alpha}\delta_{\sigma\uparrow} c_{m\beta\downarrow} + a^*_{m,\beta\alpha}\delta_{im}\delta_{\mu\alpha}\delta_{\sigma\downarrow} c_{m\beta\uparrow} \right\} \\
&= \sum_{n\beta s} \left\{ a_{n,\mu\beta}\delta_{in}\delta_{\sigma\uparrow}\delta_{s\downarrow} + a^*_{n,\beta\mu}\delta_{in}\delta_{\sigma\downarrow}\delta_{s\uparrow} \right\} c_{n\beta s}
\end{aligned} \tag{2.5}$$

Substituting the results obtained in Eqs. 2.3, 2.4 and 2.5 in Eq. (2.2)

$$\begin{aligned}
i\hbar \frac{dG^{\sigma\sigma'\,\mu\nu}_{ij}(t)}{dt} &= \delta(t)\delta_{ij}\delta_{\mu\nu}\delta_{\sigma\sigma'} \\
&- \sum_{n\beta s} \left\{ t^{\sigma\,\mu\beta}_{in}\delta_{s\sigma} + a_{n,\mu\beta}\delta_{in}\delta_{\sigma\uparrow}\delta_{s\downarrow} + a^*_{n,\beta\mu}\delta_{in}\delta_{\sigma\downarrow}\delta_{s\uparrow} \right\} \frac{i}{\hbar}\Theta(t)\langle\{c_{n\beta s}(t), c^\dagger_{j\nu\sigma'}\}\rangle \ .
\end{aligned} \tag{2.6}$$

Recognizing the monoeletronic propagator in the second line, and Fourier transforming the equation above, we get

$$\hbar\omega G^{\sigma\sigma'\,\mu\nu}_{ij}(\omega) = \delta_{ij}\delta_{\mu\nu}\delta_{\sigma\sigma'} + \sum_{n\beta s} \left\{ t^{\sigma\,\mu\beta}_{in}\delta_{s\sigma} + a_{n,\mu\beta}\delta_{in}\delta_{\sigma\uparrow}\delta_{s\downarrow} + a^*_{n,\beta\mu}\delta_{in}\delta_{\sigma\downarrow}\delta_{s\uparrow} \right\} G^{s\sigma'\,\beta\nu}_{nj}(\omega) \ , \tag{2.7}$$

or

$$\begin{aligned}
\hbar\omega\langle i\mu\sigma|\hat{G}(\omega)|j\nu\sigma'\rangle &= \delta_{ij}\delta_{\mu\nu}\delta_{\sigma\sigma'} \\
&+ \sum_{n\beta s} \left\{ t^{\sigma\,\mu\beta}_{in}\delta_{s\sigma} + a_{n,\mu\beta}\delta_{in}\delta_{\sigma\uparrow}\delta_{s\downarrow} + a^*_{n,\beta\mu}\delta_{in}\delta_{\sigma\downarrow}\delta_{s\uparrow} \right\} \langle n\beta s|\hat{G}(\omega)|j\nu\sigma'\rangle \ .
\end{aligned} \tag{2.8}$$

Comparing with Eq. (1.27), we can see that

$$\begin{aligned}
\hbar\omega\langle i\mu\sigma|\hat{G}(\omega)|j\nu\sigma'\rangle &= \delta_{ij}\delta_{\mu\nu}\delta_{\sigma\sigma'} + \langle i\mu\sigma|\hat{H}^{\mathrm{HF}}\hat{G}(\omega)|j\nu\sigma'\rangle \\
\langle i\mu\sigma| \left(\hbar\omega - \hat{H}^{\mathrm{HF}}\right) \hat{G}(\omega)|j\nu\sigma'\rangle &= \delta_{ij}\delta_{\mu\nu}\delta_{\sigma\sigma'} = \langle i\mu\sigma|j\nu\sigma'\rangle \ .
\end{aligned} \tag{2.9}$$

Adding the complete set of the hamiltonian eigenstates we have, for the (retarded) Green function

$$G^{\sigma\sigma'\,\mu\nu}_{ij}(\omega) = \sum_\ell \frac{\langle i\mu\sigma|\phi_\ell\rangle\langle\phi_\ell|j\nu\sigma'\rangle}{\hbar\omega - E_\ell + i\eta} \ , \tag{2.10}$$

where $\eta \to 0^+$. The advanced Green function is given by

$$G_{ij}^{-\sigma\sigma'\,\mu\nu}(\omega) = \sum_\ell \frac{\langle i\mu\sigma|\phi_\ell\rangle\langle\phi_\ell|j\nu\sigma'\rangle}{\hbar\omega - E_\ell - i\eta} = [G_{ji}^{\sigma'\sigma\,\nu\mu}(\omega)]^* \ . \tag{2.11}$$

We will also need the relation

$$\begin{aligned} G_{ij}^{\sigma\sigma'\,\mu\nu}(\omega) - G_{ij}^{-\sigma\sigma'\,\mu\nu}(\omega) &= \sum_\ell \langle i\mu\sigma|\phi_\ell\rangle\langle\phi_\ell|j\nu\sigma'\rangle \left(\frac{1}{\hbar\omega - E_\ell + i\eta} - \frac{1}{\hbar\omega - E_\ell - i\eta}\right) \\ &= \sum_\ell \langle i\mu\sigma|\phi_\ell\rangle\langle\phi_\ell|j\nu\sigma'\rangle \left[-2i\pi\delta(\hbar\omega - E_\ell)\right] \ , \end{aligned} \tag{2.12}$$

where we have used the identity $\frac{1}{x\pm i\epsilon} = P\left(\frac{1}{x}\right) \mp i\pi\delta(x)$, in the limit $\epsilon \to 0$. We can also define

$$\widetilde{\mathrm{Im}} G_{ij}^{\sigma\sigma'\,\mu\nu}(\omega) = \frac{1}{2i}\left\{G_{ij}^{\sigma\sigma'\,\mu\nu}(\omega) - [G_{ji}^{\sigma'\sigma\,\nu\mu}(\omega)]^*\right\} = -\pi\sum_\ell \langle i\mu\sigma|\phi_\ell\rangle\langle\phi_\ell|j\nu\sigma'\rangle\delta(\hbar\omega - E_\ell) \ . \tag{2.13}$$

Chapter 3

Susceptibility

The calculation of the susceptibility is analogous to the one already done when the SOC was not present. However, there are two fundamental differences that make the calculations more difficult. The first one is the inclusion of the spin-orbit interaction given by Eq. (1.14) in the commutator $[S^+, \hat{H}]$. The second one occurs due to the fact that the total spin is not conserved anymore. In this sense, the eigenstates of the hamiltonian are not, in general, eigenstates of $\hat{S}^z$. Thus, all the terms involving expectation values of creation and annihilation operators with different spin operators (that vanished when there was no SOC), now has to be taken into account.

When we include those terms in the χ^{+-} equation of motion, it does not close anymore and we have to calculate new susceptibilities. There will be a system of equations for χ^{+-}, $\chi^{\uparrow -}$, $\chi^{\downarrow -}$ and χ^{--}. Solving this linear system we obtain the four susceptibilities at once.

One way of making the work easier is to obtain a general equation of motion for the susceptibility

$$\chi_{ijkl}^{\sigma\sigma'\mu\nu\gamma\xi}(t) = -\frac{i}{\hbar}\Theta(t)\langle[c_{i\mu\sigma}^{\dagger}(t)c_{j\nu\sigma'}(t), S_{kl}^{-\gamma\xi}]\rangle \ , \tag{3.1}$$

where

$$S_{kl}^{m\gamma\xi} = \frac{1}{2}\sum_{\alpha\beta} c_{k\gamma\alpha}^{\dagger}\sigma_{\alpha\beta}^{m}c_{l\xi\beta} \ . \tag{3.2}$$

With this notation, we can specify each case by doing: $\sigma =\uparrow$ and $\sigma' =\downarrow$ for χ^{+-}; $\sigma = \sigma' =\uparrow$ and $\sigma = \sigma' =\downarrow$ for $\chi^{\uparrow -}$ and $\chi^{\downarrow -}$, respectively; and $\sigma =\downarrow$ and $\sigma' =\uparrow$ for χ^{--}. To work on the $0, x, y, z$ basis, there should be an extra factor $1/2$. In general, the susceptibility can be written as

$$\begin{aligned}\chi_{ijkl}^{\alpha\beta\mu\nu\gamma\xi}(t) =& -\frac{i}{\hbar}\Theta(t)\langle[S_{ij}^{\alpha\mu\nu}(t), S_{kl}^{\beta\gamma\xi}]\rangle \\ =& -\frac{i}{4\hbar}\Theta(t)\langle[c_{i\mu m}^{\dagger}(t)\sigma_{mn}^{\alpha}c_{j\nu n}(t), c_{k\gamma m'}^{\dagger}\sigma_{m'n'}^{\beta}c_{l\xi n'}]\rangle \ ,\end{aligned} \tag{3.3}$$

The equation of motion for this susceptibility can be written as

$$i\hbar\frac{d}{dt}\chi_{ijkl}^{\sigma\sigma'\mu\nu\gamma\xi}(t) = \delta(t)\langle[c_{i\mu\sigma}^{\dagger}c_{j\nu\sigma'}, S_{kl}^{-\gamma\xi}]\rangle + \langle\langle[c_{i\mu\sigma}^{\dagger}(t)c_{j\nu\sigma'}(t), \hat{H}]; S_{kl}^{-\gamma\xi}\rangle\rangle \ . \tag{3.4}$$

Calculating the first term of the right-hand-side of Eq. (3.4), we have

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}, S^{-\gamma\xi}_{kl}] =& c^\dagger_{i\mu\sigma}\underbrace{c_{j\nu\sigma'}c^\dagger_{k\gamma\downarrow}}c_{l\xi\uparrow} - c^\dagger_{k\gamma\downarrow}c_{l\xi\uparrow}c^\dagger_{i\mu\sigma}c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma}c_{l\xi\uparrow}\delta_{jk}\delta_{\gamma\nu}\delta_{\sigma'\downarrow} - \underbrace{c^\dagger_{i\mu\sigma}c^\dagger_{k\gamma\downarrow}}\underbrace{c_{j\nu\sigma'}c_{l\xi\uparrow}} - c^\dagger_{k\gamma\downarrow}c_{l\xi\uparrow}c^\dagger_{i\mu\sigma}c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma}c_{l\xi\uparrow}\delta_{jk}\delta_{\gamma\nu}\delta_{\sigma'\downarrow} - c^\dagger_{k\gamma\downarrow}\underbrace{c^\dagger_{i\mu\sigma}c_{l\xi\uparrow}}c_{j\nu\sigma'} - c^\dagger_{k\gamma\downarrow}c_{l\xi\uparrow}c^\dagger_{i\mu\sigma}c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma}c_{l\xi\uparrow}\delta_{jk}\delta_{\gamma\nu}\delta_{\sigma'\downarrow} - c^\dagger_{k\gamma\downarrow}c_{j\nu\sigma'}\delta_{il}\delta_{\mu\xi}\delta_{\sigma\uparrow} \ .
\end{aligned}
\tag{3.5}
$$

So, the first term of the equation above only appears when $\sigma' =\downarrow$ and the second one appears when $\sigma =\uparrow$.

For the calculation of the second term, we are going to use the Hartree-Fock and the Random Phase Approximation (RPA).

3.1 Hartree-Fock Susceptibility

We have already obtained the hamiltonian within the Hartree-Fock approximation in Eq. (1.21). Now we need to calculate

$$
[c^\dagger_{i\mu\sigma}(t)c_{j\nu\sigma'}(t), \hat{H}^{\mathrm{HF}}] = [c^\dagger_{i\mu\sigma}(t)c_{j\nu\sigma'}(t), \hat{H}'_0 + \hat{H}_{\mathrm{sf}}] \ . \tag{3.6}
$$

The first term is given by

$$
[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}, \hat{H}'_0] = \sum_{\substack{mn\\s}}\sum_{\alpha\beta} t^{s\,\alpha\beta}_{mn}[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}, c^\dagger_{m\alpha s}c_{n\beta s}] \ . \tag{3.7}
$$

The above commutator is

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}, c^\dagger_{m\alpha s}c_{n\beta s}] =& c^\dagger_{i\mu\sigma}\underbrace{c_{j\nu\sigma'}c^\dagger_{m\alpha s}}c_{n\beta s} - c^\dagger_{m\alpha s}c_{n\beta s}c^\dagger_{i\mu\sigma}c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma}c_{n\beta s}\delta_{jm}\delta_{\nu\alpha}\delta_{s\sigma'} - \underbrace{c^\dagger_{i\mu\sigma}c^\dagger_{m\alpha s}}\underbrace{c_{j\nu\sigma'}c_{n\beta s}} - c^\dagger_{m\alpha s}c_{n\beta s}c^\dagger_{i\mu\sigma}c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma}c_{n\beta s}\delta_{jm}\delta_{\nu\alpha}\delta_{s\sigma'} - c^\dagger_{m\alpha s}\underbrace{c^\dagger_{i\mu\sigma}c_{n\beta s}}c_{j\nu\sigma'} - c^\dagger_{m\alpha s}c_{n\beta s}c^\dagger_{i\mu\sigma}c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma}c_{n\beta s}\delta_{jm}\delta_{\nu\alpha}\delta_{s\sigma'} - c^\dagger_{m\alpha s}c_{j\nu\sigma'}\delta_{in}\delta_{\mu\beta}\delta_{s\sigma} \ .
\end{aligned}
\tag{3.8}
$$

Substituting back in Eq. (3.7) we obtain

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, \hat{H}'_0] &= \sum_{\substack{mn\\s}} \sum_{\alpha\beta} t^{s\,\alpha\beta}_{mn} \left(c^\dagger_{i\mu\sigma} c_{n\beta s} \delta_{jm} \delta_{\nu\alpha} \delta_{s\sigma'} - c^\dagger_{m\alpha s} c_{j\nu\sigma'} \delta_{in} \delta_{\mu\beta} \delta_{s\sigma} \right) \\
&= \sum_{mn} \sum_{\alpha\beta} \left(t^{\sigma'\,\alpha\beta}_{mn} c^\dagger_{i\mu\sigma} c_{n\beta\sigma'} \delta_{jm} \delta_{\nu\alpha} - t^{\sigma\,\alpha\beta}_{mn} c^\dagger_{m\alpha\sigma} c_{j\nu\sigma'} \delta_{in} \delta_{\mu\beta} \right) \\
&= \sum_{n\beta} t^{\sigma'\,\nu\beta}_{jn} c^\dagger_{i\mu\sigma} c_{n\beta\sigma'} - \sum_{m\alpha} t^{\sigma\,\alpha\mu}_{mi} c^\dagger_{m\alpha\sigma} c_{j\nu\sigma'} \\
&= \sum_{m\alpha} \left(t^{\sigma'\,\nu\alpha}_{jm} c^\dagger_{i\mu\sigma} c_{m\alpha\sigma'} - t^{\sigma\,\alpha\mu}_{mi} c^\dagger_{m\alpha\sigma} c_{j\nu\sigma'} \right) \;.
\end{aligned}
\tag{3.9}
$$

Consequently, in the equation of motion this term gives rise to

$$
\begin{aligned}
&\sum_{m\alpha} \langle\langle t^{\sigma'\,\nu\alpha}_{jm} c^\dagger_{i\mu\sigma} c_{m\alpha\sigma'} - t^{\sigma\,\alpha\mu}_{mi} c^\dagger_{m\alpha\sigma} c_{j\nu\sigma'} ; S^{-\,\gamma\xi}_{kl} \rangle\rangle \\
=& \sum_{\substack{mn\\ \alpha\beta}} \langle\langle t^{\sigma'\,\nu\alpha}_{jm} c^\dagger_{n\beta\sigma} c_{m\alpha\sigma'} \delta_{ni} \delta_{\mu\beta} - t^{\sigma\,\alpha\mu}_{mi} c^\dagger_{m\alpha\sigma} c_{n\beta\sigma'} \delta_{jn} \delta_{\nu\beta} ; S^{-\,\gamma\xi}_{kl} \rangle\rangle \;.
\end{aligned}
\tag{3.10}
$$

Changing $\alpha \rightleftharpoons \beta$ and $m \rightleftharpoons n$ in the first term, we have

$$
\begin{aligned}
&\sum_{\substack{mn\\ \alpha\beta}} \langle\langle t^{\sigma'\,\nu\beta}_{jn} c^\dagger_{m\alpha\sigma} c_{n\beta\sigma'} \delta_{mi} \delta_{\mu\alpha} - t^{\sigma\,\alpha\mu}_{mi} c^\dagger_{m\alpha\sigma} c_{n\beta\sigma'} \delta_{jn} \delta_{\nu\beta} ; S^{-\,\gamma\xi}_{kl} \rangle\rangle \\
=& \sum_{\substack{mn\\ \alpha\beta}} \langle\langle \left(t^{\sigma'\,\nu\beta}_{jn} \delta_{mi} \delta_{\mu\alpha} - t^{\sigma\,\alpha\mu}_{mi} \delta_{jn} \delta_{\nu\beta} \right) c^\dagger_{m\alpha\sigma} c_{n\beta\sigma'} ; S^{-\,\gamma\xi}_{kl} \rangle\rangle \\
=& \sum_{\substack{mn\\ \alpha\beta}} \left(t^{\sigma'\,\nu\beta}_{jn} \delta_{mi} \delta_{\mu\alpha} - t^{\sigma\,\alpha\mu}_{mi} \delta_{jn} \delta_{\nu\beta} \right) \langle\langle c^\dagger_{m\alpha\sigma} c_{n\beta\sigma'} ; S^{-\,\gamma\xi}_{kl} \rangle\rangle \\
=& \sum_{\substack{mn\\ \alpha\beta}} \left(t^{\sigma'\,\nu\beta}_{jn} \delta_{mi} \delta_{\mu\alpha} - t^{\sigma\,\alpha\mu}_{mi} \delta_{jn} \delta_{\nu\beta} \right) \chi^{\sigma\sigma'\,\alpha\beta\gamma\xi}_{mnkl} \;.
\end{aligned}
\tag{3.11}
$$

The spin-flip term is

$$
[c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, \hat{H}_{\mathrm{sf}}] = \sum_{m\alpha\beta} \left\{ a_{\alpha\beta} [c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, c^\dagger_{m\alpha\uparrow} c_{m\beta\downarrow}] + a^*_{m,\beta\alpha} [c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, c^\dagger_{m\alpha\downarrow} c_{m\beta\uparrow}] \right\} \;. \tag{3.12}
$$

Calculating the first commutator on the right-hand-side,

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, c^\dagger_{m\alpha\uparrow} c_{m\beta\downarrow}] =& c^\dagger_{i\mu\sigma} \underbrace{c_{j\nu\sigma'} c^\dagger_{m\alpha\uparrow}} c_{m\beta\downarrow} - c^\dagger_{m\alpha\uparrow} c_{m\beta\downarrow} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{m\beta\downarrow} \delta_{mj} \delta_{\alpha\nu} \delta_{\uparrow\sigma'} - \underbrace{c^\dagger_{i\mu\sigma} c^\dagger_{m\alpha\uparrow}} \underbrace{c_{j\nu\sigma'} c_{m\beta\downarrow}} - c^\dagger_{m\alpha\uparrow} c_{m\beta\downarrow} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{m\beta\downarrow} \delta_{mj} \delta_{\alpha\nu} \delta_{\uparrow\sigma'} - c^\dagger_{m\alpha\uparrow} \underbrace{c^\dagger_{i\mu\sigma} c_{m\beta\downarrow}} c_{j\nu\sigma'} - c^\dagger_{m\alpha\uparrow} c_{m\beta\downarrow} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{m\beta\downarrow} \delta_{mj} \delta_{\alpha\nu} \delta_{\uparrow\sigma'} - c^\dagger_{m\alpha\uparrow} c_{j\nu\sigma'} \delta_{mi} \delta_{\beta\mu} \delta_{\downarrow\sigma} \;.
\end{aligned}
\tag{3.13}
$$

The second commutator can be obtained inverting the spins of the result above. So,

$$[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'},c^\dagger_{m\alpha\downarrow}c_{m\beta\uparrow}] = c^\dagger_{i\mu\sigma}c_{m\beta\uparrow}\delta_{mj}\delta_{\alpha\nu}\delta_{\downarrow\sigma'} - c^\dagger_{m\alpha\downarrow}c_{j\nu\sigma'}\delta_{mi}\delta_{\beta\mu}\delta_{\uparrow\sigma}\ . \tag{3.14}$$

So, the commutator with $\hat{H}_{\rm sf}$ is

$$\begin{aligned}
[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'},\hat{H}_{\rm sf}] &= \sum_{m\alpha\beta}\Big\{a_{\alpha\beta}\left(c^\dagger_{i\mu\sigma}c_{m\beta\downarrow}\delta_{mj}\delta_{\alpha\nu}\delta_{\uparrow\sigma'} - c^\dagger_{m\alpha\uparrow}c_{j\nu\sigma'}\delta_{mi}\delta_{\beta\mu}\delta_{\downarrow\sigma}\right)\\
&\qquad + a^*_{m,\beta\alpha}\left(c^\dagger_{i\mu\sigma}c_{m\beta\uparrow}\delta_{mj}\delta_{\alpha\nu}\delta_{\downarrow\sigma'} - c^\dagger_{m\alpha\downarrow}c_{j\nu\sigma'}\delta_{mi}\delta_{\beta\mu}\delta_{\uparrow\sigma}\right)\Big\}\\
&= \sum_{\substack{mn\\ \alpha\beta}}\Big\{a_{m,\nu\beta}c^\dagger_{n\alpha\sigma}c_{m\beta\downarrow}\delta_{ni}\delta_{mj}\delta_{\alpha\mu}\delta_{\uparrow\sigma'} - a_{m,\alpha\mu}c^\dagger_{m\alpha\uparrow}c_{n\beta\sigma'}\delta_{mi}\delta_{nj}\delta_{\beta\nu}\delta_{\downarrow\sigma}\\
&\qquad + a^*_{m,\beta\nu}c^\dagger_{n\alpha\sigma}c_{m\beta\uparrow}\delta_{ni}\delta_{mj}\delta_{\alpha\mu}\delta_{\downarrow\sigma'} - a^*_{m,\mu\alpha}c^\dagger_{m\alpha\downarrow}c_{n\beta\sigma'}\delta_{mi}\delta_{nj}\delta_{\beta\nu}\delta_{\uparrow\sigma}\Big\}\ .
\end{aligned} \tag{3.15}$$

Changing $m \rightleftharpoons n$ in the first and third terms,

$$\begin{aligned}
[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'},\hat{H}_{\rm sf}] &= \sum_{\substack{mn\\ \alpha\beta}}\Big\{a_{n,\nu\beta}c^\dagger_{m\alpha\sigma}c_{n\beta\downarrow}\delta_{mi}\delta_{nj}\delta_{\alpha\mu}\delta_{\uparrow\sigma'} - a_{m,\alpha\mu}c^\dagger_{m\alpha\uparrow}c_{n\beta\sigma'}\delta_{mi}\delta_{nj}\delta_{\beta\nu}\delta_{\downarrow\sigma}\\
&\qquad + a^*_{n,\beta\nu}c^\dagger_{m\alpha\sigma}c_{n\beta\uparrow}\delta_{mi}\delta_{nj}\delta_{\alpha\mu}\delta_{\downarrow\sigma'} - a^*_{m,\mu\alpha}c^\dagger_{m\alpha\downarrow}c_{n\beta\sigma'}\delta_{mi}\delta_{nj}\delta_{\beta\nu}\delta_{\uparrow\sigma}\Big\}\ .
\end{aligned} \tag{3.16}$$

In the equation of motion,

$$\begin{aligned}
\langle\langle[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'},\hat{H}_{\rm sf}];S^{-\gamma\xi}_{kl}\rangle\rangle &= \sum_{\substack{mn\\ \alpha\beta}}\Big\{a_{n,\nu\beta}\delta_{mi}\delta_{nj}\delta_{\alpha\mu}\delta_{\uparrow\sigma'}\chi^{\sigma\downarrow\alpha\beta\gamma\xi}_{mnkl} - a_{m,\alpha\mu}\delta_{mi}\delta_{nj}\delta_{\beta\nu}\delta_{\downarrow\sigma}\chi^{\uparrow\sigma'\alpha\beta\gamma\xi}_{mnkl}\\
&\qquad + a^*_{n,\beta\nu}\delta_{mi}\delta_{nj}\delta_{\alpha\mu}\delta_{\downarrow\sigma'}\chi^{\sigma\uparrow\alpha\beta\gamma\xi}_{mnkl} - a^*_{m,\mu\alpha}\delta_{mi}\delta_{nj}\delta_{\beta\nu}\delta_{\uparrow\sigma}\chi^{\downarrow\sigma'\alpha\beta\gamma\xi}_{mnkl}\Big\}\ .
\end{aligned} \tag{3.17}$$

Remember that, here, the spin indices of the susceptibility are related to the first 2 creation and annihilation operators.

These are the terms that will couple each of the susceptibilities with two others. Now, we will particularize the values of σ and σ' for each case, and this will become even more clear.

3.1.1 Susceptibility χ^{+-}

First, we will particularize Eq. (3.4) for $\sigma =\uparrow$ and $\sigma' =\downarrow$. We begin by Fourier transforming it, and substituting results of Eqs. 3.5, 3.11 and 3.17 to obtain

$$\begin{aligned}\hbar\omega \chi^{+-\mu\nu\gamma\xi}_{ijkl}(\omega) =& \langle c^\dagger_{i\mu\uparrow} c_{l\xi\uparrow}\rangle \delta_{jk}\delta_{\gamma\nu} - \langle c^\dagger_{k\gamma\downarrow} c_{j\nu\downarrow}\rangle \delta_{il}\delta_{\mu\xi} \\ &+ \sum_{\substack{mn\\ \alpha\beta}} \left(t^{\downarrow\nu\beta}_{jn} \delta_{mi}\delta_{\mu\alpha} - t^{\uparrow\alpha\mu}_{mi}\delta_{jn}\delta_{\nu\beta}\right) \chi^{+-\alpha\beta\gamma\xi}_{mnkl} \\ &+ \sum_{\substack{mn\\ \alpha\beta}} \left\{ a^*_{n,\beta\nu}\delta_{mi}\delta_{nj}\delta_{\alpha\mu}\chi^{\uparrow-\alpha\beta\gamma\xi}_{mnkl} - a^*_{m,\mu\alpha}\delta_{mi}\delta_{nj}\delta_{\beta\nu}\chi^{\downarrow-\alpha\beta\gamma\xi}_{mnkl}\right\} \ .\end{aligned} \tag{3.18}$$

These results can be summarized in an equation of motion written in matrix form as

$$\left(\hbar\omega - B^{11}\right)\chi^{+-} - B^{12}\chi^{\uparrow-} - B^{13}\chi^{\downarrow-} = A^1 \ , \tag{3.19}$$

where

$$A^{1\,\mu\nu\gamma\xi}_{ijkl} = \langle c^\dagger_{i\mu\uparrow} c_{l\xi\uparrow}\rangle \delta_{jk}\delta_{\gamma\nu} - \langle c^\dagger_{k\gamma\downarrow} c_{j\nu\downarrow}\rangle \delta_{il}\delta_{\mu\xi} \ , \tag{3.20}$$

and the B elements are given by

$$B^{11\,\mu\nu\alpha\beta}_{ijmn} = t^{\downarrow\nu\beta}_{jn}\delta_{mi}\delta_{\mu\alpha} - t^{\uparrow\alpha\mu}_{mi}\delta_{jn}\delta_{\nu\beta} \tag{3.21}$$

$$B^{12\,\mu\nu\alpha\beta}_{ijmn} = a^*_{n,\beta\nu}\delta_{mi}\delta_{nj}\delta_{\alpha\mu} \tag{3.22}$$

$$B^{13\,\mu\nu\alpha\beta}_{ijmn} = -\, a^*_{m,\mu\alpha}\delta_{mi}\delta_{nj}\delta_{\beta\nu} \ . \tag{3.23}$$

3.1.2 Susceptibility $\chi^{\uparrow-}$

For $\sigma =\uparrow$ and $\sigma' =\uparrow$, we obtain

$$\begin{aligned}\hbar\omega \chi^{\uparrow-\mu\nu\gamma\xi}_{ijkl}(\omega) =& - \langle c^\dagger_{k\gamma\downarrow} c_{j\nu\uparrow}\rangle \delta_{il}\delta_{\mu\xi} \\ &+ \sum_{\substack{mn\\ \alpha\beta}} \left(t^{\uparrow\nu\beta}_{jn} \delta_{mi}\delta_{\mu\alpha} - t^{\uparrow\alpha\mu}_{mi}\delta_{jn}\delta_{\nu\beta}\right) \chi^{\uparrow-\alpha\beta\gamma\xi}_{mnkl} \\ &+ \sum_{\substack{mn\\ \alpha\beta}} \left\{ a_{n,\nu\beta}\delta_{mi}\delta_{nj}\delta_{\alpha\mu}\chi^{+-\alpha\beta\gamma\xi}_{mnkl} - a^*_{m,\mu\alpha}\delta_{mi}\delta_{nj}\delta_{\beta\nu}\chi^{--\alpha\beta\gamma\xi}_{mnkl}\right\} \ .\end{aligned} \tag{3.24}$$

In matrix form,

$$-B^{21}\chi^{+-} + \left(\hbar\omega - B^{22}\right)\chi^{\uparrow-} - B^{24}\chi^{--} = A^2 \ , \tag{3.25}$$

where

$$A^{2\,\mu\nu\gamma\xi}_{ijkl} = -\langle c^\dagger_{k\gamma\downarrow} c_{j\nu\uparrow}\rangle \delta_{il}\delta_{\mu\xi} \ , \tag{3.26}$$

and the elements of B matrices are given by

$$
\begin{aligned}
B^{21\,\mu\nu\alpha\beta}_{ijmn} &= a_{n,\nu\beta}\delta_{mi}\delta_{nj}\delta_{\alpha\mu} &(3.27)\\
B^{22\,\mu\nu\alpha\beta}_{ijmn} &= t^{\uparrow\nu\beta}_{jn}\delta_{mi}\delta_{\mu\alpha} - t^{\uparrow\alpha\mu}_{mi}\delta_{jn}\delta_{\nu\beta} &(3.28)\\
B^{24\,\mu\nu\alpha\beta}_{ijmn} &= \; - a^{*}_{m,\mu\alpha}\delta_{mi}\delta_{nj}\delta_{\beta\nu} \; . &(3.29)
\end{aligned}
$$

Note that $B^{24\,\mu\nu\alpha\beta}_{ijmn} = B^{13\,\mu\nu\alpha\beta}_{ijmn}$.

3.1.3 Susceptibility $\chi^{\downarrow-}$

Now, substituting $\sigma =\downarrow$ and $\sigma' =\downarrow$, we get

$$
\begin{aligned}
\hbar\omega\chi^{\downarrow-\,\mu\nu\gamma\xi}_{ijkl}(\omega) =& \langle c^{\dagger}_{i\mu\downarrow}c_{l\xi\uparrow}\rangle\delta_{jk}\delta_{\gamma\nu}\\
&+ \sum_{\substack{mn\\ \alpha\beta}} \left(t^{\downarrow\nu\beta}_{jn}\delta_{mi}\delta_{\mu\alpha} - t^{\downarrow\alpha\mu}_{mi}\delta_{jn}\delta_{\nu\beta}\right)\chi^{\downarrow-\,\alpha\beta\gamma\xi}_{mnkl}\\
&+ \sum_{\substack{mn\\ \alpha\beta}} \left\{-a_{m,\alpha\mu}\delta_{mi}\delta_{nj}\delta_{\beta\nu}\chi^{+-\,\alpha\beta\gamma\xi}_{mnkl} + a^{*}_{n,\beta\nu}\delta_{mi}\delta_{nj}\delta_{\alpha\mu}\chi^{--\,\alpha\beta\gamma\xi}_{mnkl}\right\} \; .
\end{aligned}
\tag{3.30}
$$

The equation of motion can be written as

$$
-B^{31}\chi^{+-} + \left(\hbar\omega - B^{33}\right)\chi^{\downarrow-} - B^{34}\chi^{--} = A^{3} \; . \tag{3.31}
$$

The matrices are given by

$$
\begin{aligned}
A^{3\,\mu\nu\gamma\xi}_{ijkl} &= \langle c^{\dagger}_{i\mu\downarrow}c_{l\xi\uparrow}\rangle\delta_{jk}\delta_{\gamma\nu} &(3.32)\\
B^{31\,\mu\nu\alpha\beta}_{ijmn} &= \; - a_{m,\alpha\mu}\delta_{mi}\delta_{nj}\delta_{\beta\nu} &(3.33)\\
B^{33\,\mu\nu\alpha\beta}_{ijmn} &= t^{\downarrow\nu\beta}_{jn}\delta_{mi}\delta_{\mu\alpha} - t^{\downarrow\alpha\mu}_{mi}\delta_{jn}\delta_{\nu\beta} &(3.34)\\
B^{34\,\mu\nu\alpha\beta}_{ijmn} &= a^{*}_{n,\beta\nu}\delta_{mi}\delta_{nj}\delta_{\alpha\mu} \; . &(3.35)
\end{aligned}
$$

Note that $B^{34\,\mu\nu\alpha\beta}_{ijmn} = B^{12\,\mu\nu\alpha\beta}_{ijmn}$.

3.1.4 Susceptibility χ^{--}

Finally, for $\sigma =\downarrow$ and $\sigma' =\uparrow$, we get

$$
\begin{aligned}
\hbar\omega\chi^{--\,\mu\nu\gamma\xi}_{ijkl}(\omega) =& \sum_{\substack{mn\\ \alpha\beta}} \left(t^{\uparrow\nu\beta}_{jn}\delta_{mi}\delta_{\mu\alpha} - t^{\downarrow\alpha\mu}_{mi}\delta_{jn}\delta_{\nu\beta}\right)\chi^{--\,\alpha\beta\gamma\xi}_{mnkl}\\
&+ \sum_{\substack{mn\\ \alpha\beta}} \left\{-a_{m,\alpha\mu}\delta_{mi}\delta_{nj}\delta_{\beta\nu}\chi^{\uparrow-\,\alpha\beta\gamma\xi}_{mnkl} + a_{n,\nu\beta}\delta_{mi}\delta_{nj}\delta_{\alpha\mu}\chi^{\downarrow-\,\alpha\beta\gamma\xi}_{mnkl}\right\} \; .
\end{aligned}
\tag{3.36}
$$

In matrix notation, it can be rewritten as

$$-B^{42}\chi^{\uparrow -} - B^{43}\chi^{\downarrow -} + \left(\hbar\omega - B^{44}\right)\chi^{--} = 0 \ . \tag{3.37}$$

Note that $A^4 = 0$ and

$$B^{42\,\mu\nu\alpha\beta}_{ijmn} = -\, a_{m,\alpha\mu}\delta_{mi}\delta_{nj}\delta_{\beta\nu} \tag{3.38}$$
$$B^{43\,\mu\nu\alpha\beta}_{ijmn} = a_{n,\nu\beta}\delta_{mi}\delta_{nj}\delta_{\alpha\mu} \tag{3.39}$$
$$B^{44\,\mu\nu\alpha\beta}_{ijmn} = t^{\uparrow\nu\beta}_{jn}\delta_{mi}\delta_{\mu\alpha} - t^{\downarrow\alpha\mu}_{mi}\delta_{jn}\delta_{\nu\beta} \ . \tag{3.40}$$

Here, $B^{42\,\mu\nu\alpha\beta}_{ijmn} = B^{31\,\mu\nu\alpha\beta}_{ijmn}$ and $B^{43\,\mu\nu\alpha\beta}_{ijmn} = B^{21\,\mu\nu\alpha\beta}_{ijmn}$.

3.1.5 Linear System

We have obtained a linear system composed of four equations, written in Eqs. 3.19, 3.25, 3.31 and 3.37. We can summarize this result as

$$\begin{pmatrix} \hbar\omega - B^{11} & -B^{12} & -B^{13} & 0 \\ -B^{21} & \hbar\omega - B^{22} & 0 & -B^{13} \\ -B^{31} & 0 & \hbar\omega - B^{33} & -B^{12} \\ 0 & -B^{31} & -B^{21} & \hbar\omega - B^{44} \end{pmatrix} \begin{pmatrix} \chi^{+-} \\ \chi^{\uparrow -} \\ \chi^{\downarrow -} \\ \chi^{--} \end{pmatrix} = \begin{pmatrix} A^1 \\ A^2 \\ A^3 \\ 0 \end{pmatrix} \ . \tag{3.41}$$

Simplifying even further,

$$(\hbar\omega - B)\chi^0 = \mathbf{A} \ . \tag{3.42}$$

We want to find the susceptibility matrix, which will be given by

$$\chi^0 = (\hbar\omega - B)^{-1}\mathbf{A} = \Gamma\mathbf{A} \ . \tag{3.43}$$

where $\Gamma = (\hbar\omega - B)^{-1}$. To obtain the solution of this system, we need to find the elements Γ^{ij} that satisfies

$$(\hbar\omega - B)\Gamma = 1 \ . \tag{3.44}$$

The equation above represents a linear system of 16 equations and 16 unknowns (the elements of Γ). Writing the matrices explicitly,

$$\begin{aligned} &\begin{pmatrix} \hbar\omega - B^{11} & -B^{12} & -B^{13} & 0 \\ -B^{21} & \hbar\omega - B^{22} & 0 & -B^{13} \\ -B^{31} & 0 & \hbar\omega - B^{33} & -B^{12} \\ 0 & -B^{31} & -B^{21} & \hbar\omega - B^{44} \end{pmatrix} \begin{pmatrix} \Gamma^{11} & \Gamma^{12} & \Gamma^{13} & \Gamma^{14} \\ \Gamma^{21} & \Gamma^{22} & \Gamma^{23} & \Gamma^{24} \\ \Gamma^{31} & \Gamma^{32} & \Gamma^{33} & \Gamma^{34} \\ \Gamma^{41} & \Gamma^{42} & \Gamma^{43} & \Gamma^{44} \end{pmatrix} \\ &\qquad = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \ . \end{aligned} \tag{3.45}$$

There will be 4 uncoupled systems, one for each column of Γ. The first one is given can be written as

$$\begin{pmatrix} \hbar\omega - B^{11} & -B^{12} & -B^{13} & 0 \\ -B^{21} & \hbar\omega - B^{22} & 0 & -B^{13} \\ -B^{31} & 0 & \hbar\omega - B^{33} & -B^{12} \\ 0 & -B^{31} & -B^{21} & \hbar\omega - B^{44} \end{pmatrix} \begin{pmatrix} \Gamma^{11} \\ \Gamma^{21} \\ \Gamma^{31} \\ \Gamma^{41} \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix} . \tag{3.46}$$

Analyzing the equations of motion and using the "Costa conjecture", we can see that the elements of Γ are given by

$$\Gamma^{11\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\uparrow\rangle\langle n\beta\downarrow|\phi_{p'}\rangle\langle k\gamma\uparrow|\phi_p\rangle\langle\phi_{p'}|l\xi\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} , \tag{3.47}$$

$$\Gamma^{21\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\uparrow\rangle\langle n\beta\uparrow|\phi_{p'}\rangle\langle k\gamma\uparrow|\phi_p\rangle\langle\phi_{p'}|l\xi\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} , \tag{3.48}$$

$$\Gamma^{31\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\downarrow\rangle\langle n\beta\downarrow|\phi_{p'}\rangle\langle k\gamma\uparrow|\phi_p\rangle\langle\phi_{p'}|l\xi\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} . \tag{3.49}$$

Using Eq. (1.27) and (1.30), it is easy to show that these expressions satisfy the equation

$$\left(\hbar\omega - B^{11}\right)\Gamma^{11} - B^{12}\Gamma^{21} - B^{13}\Gamma^{31} = 1 . \tag{3.50}$$

The same way, we can obtain

$$\Gamma^{12\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\uparrow\rangle\langle n\beta\downarrow|\phi_{p'}\rangle\langle k\gamma\uparrow|\phi_p\rangle\langle\phi_{p'}|l\xi\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} , \tag{3.51}$$

$$\Gamma^{22\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\uparrow\rangle\langle n\beta\uparrow|\phi_{p'}\rangle\langle k\gamma\uparrow|\phi_p\rangle\langle\phi_{p'}|l\xi\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} , \text{ and} \tag{3.52}$$

$$\Gamma^{42\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\downarrow\rangle\langle n\beta\uparrow|\phi_{p'}\rangle\langle k\gamma\uparrow|\phi_p\rangle\langle\phi_{p'}|l\xi\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} , \tag{3.53}$$

which satisfy

$$-B^{21}\Gamma^{12} + \left(\hbar\omega - B^{22}\right)\Gamma^{22} - B^{24}\Gamma^{42} = 1 ; \tag{3.54}$$

$$\Gamma^{13\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\uparrow\rangle\langle n\beta\downarrow|\phi_{p'}\rangle\langle k\gamma\downarrow|\phi_p\rangle\langle\phi_{p'}|l\xi\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} , \tag{3.55}$$

$$\Gamma^{33\,\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\downarrow\rangle\langle n\beta\downarrow|\phi_{p'}\rangle\langle k\gamma\downarrow|\phi_p\rangle\langle\phi_{p'}|l\xi\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \ , \text{ and} \tag{3.56}$$

$$\Gamma^{43\,\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\downarrow\rangle\langle n\beta\uparrow|\phi_{p'}\rangle\langle k\gamma\downarrow|\phi_p\rangle\langle\phi_{p'}|l\xi\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \ , \tag{3.57}$$

which satisfy

$$-B^{31}\Gamma^{13} + \left(\hbar\omega - B^{33}\right)\Gamma^{33} - B^{34}\Gamma^{43} = 1 \ ; \text{ and} \tag{3.58}$$

$$\Gamma^{24\,\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\uparrow\rangle\langle n\beta\uparrow|\phi_{p'}\rangle\langle k\gamma\downarrow|\phi_p\rangle\langle\phi_{p'}|l\xi\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \ , \tag{3.59}$$

$$\Gamma^{34\,\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\downarrow\rangle\langle n\beta\downarrow|\phi_{p'}\rangle\langle k\gamma\downarrow|\phi_p\rangle\langle\phi_{p'}|l\xi\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \ , \text{ and} \tag{3.60}$$

$$\Gamma^{44\,\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\downarrow\rangle\langle n\beta\uparrow|\phi_{p'}\rangle\langle k\gamma\downarrow|\phi_p\rangle\langle\phi_{p'}|l\xi\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \ , \tag{3.61}$$

which satisfy

$$-B^{42}\Gamma^{24} - B^{43}\Gamma^{34} + \left(\hbar\omega - B^{44}\right)\Gamma^{44} = 1 \ . \tag{3.62}$$

The elements of the secondary diagonal follows the same pattern, and can be obtained using the other equations of the system. The elements are given by

$$\Gamma^{14\,\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\uparrow\rangle\langle n\beta\downarrow|\phi_{p'}\rangle\langle k\gamma\downarrow|\phi_p\rangle\langle\phi_{p'}|l\xi\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \ , \tag{3.63}$$

$$\Gamma^{23\,\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\uparrow\rangle\langle n\beta\uparrow|\phi_{p'}\rangle\langle k\gamma\downarrow|\phi_p\rangle\langle\phi_{p'}|l\xi\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \ , \tag{3.64}$$

$$\Gamma^{32\,\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\downarrow\rangle\langle n\beta\downarrow|\phi_{p'}\rangle\langle k\gamma\uparrow|\phi_p\rangle\langle\phi_{p'}|l\xi\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \ , \text{ and} \tag{3.65}$$

$$\Gamma^{41\,\alpha\beta\gamma\xi}_{mnkl} = \sum_{pp'} \frac{\langle\phi_p|m\alpha\downarrow\rangle\langle n\beta\uparrow|\phi_{p'}\rangle\langle k\gamma\uparrow|\phi_p\rangle\langle\phi_{p'}|l\xi\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \ . \tag{3.66}$$

Now that we have the complete matrix Γ, we can use Eq. (3.43) to calculate the susceptibility within Hartree-Fock approximation.

3.1.6 Solution for $\chi^{+-}_{(0)}$

The susceptibility $\chi^{+-}_{(0)}$ is given by the first element of χ vector, i.e.,

$$\chi^{+-}_{(0)}{}^{\mu\nu\gamma\xi}_{ijkl}(\omega) = \chi^{1}_{(0)}{}^{\mu\nu\gamma\xi}_{ijkl}(\omega) = \left[\Gamma^{11}A^1 + \Gamma^{12}A^2 + \Gamma^{13}A^3\right]^{\mu\nu\gamma\xi}_{ijkl} \ . \tag{3.67}$$

The expectation value of the creation and annihilation operators product can be written as

$$\langle c^{\dagger}_{i\mu\sigma} c_{j\nu\sigma'}\rangle = \hbar \int d\omega' f(\omega') \sum_p \langle \phi_p | i\mu\sigma\rangle\langle j\nu\sigma'|\phi_p\rangle \delta(\hbar\omega' - E_p) \ . \tag{3.68}$$

So the first term of Eq. (3.67) is

$$\begin{aligned}
\sum_{\substack{mn\\ \alpha\beta}} \Gamma^{11}{}^{\mu\nu\alpha\beta}_{ijmn} A^{1}{}^{\alpha\beta\gamma\xi}_{mnkl} =& \hbar \sum_{\substack{m\\ \alpha}} \int d\omega' f(\omega') \sum_{pp'p''} \langle \phi_{p''}|m\alpha \uparrow\rangle\langle l\xi \uparrow|\phi_{p''}\rangle \delta(\hbar\omega' - E_{p''}) \\
&\times \frac{\langle \phi_p | i\mu \uparrow\rangle\langle j\nu \downarrow|\phi_{p'}\rangle\langle m\alpha \uparrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \\
&- \hbar \sum_{\substack{n\\ \beta}} \int d\omega' f(\omega') \sum_{pp'p''} \langle \phi_{p''}|k\gamma \downarrow\rangle\langle n\beta \downarrow|\phi_{p''}\rangle \delta(\hbar\omega' - E_{p''}) \\
&\times \frac{\langle \phi_p | i\mu \uparrow\rangle\langle j\nu \downarrow|\phi_{p'}\rangle\langle l\xi \uparrow|\phi_p\rangle\langle\phi_{p'}|n\beta\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \ .
\end{aligned} \tag{3.69}$$

Were we have used Eqs. 3.20 and 3.47. Similarly, the second and third term are

$$\begin{aligned}
\sum_{\substack{mn\\ \alpha\beta}} \Gamma^{12}{}^{\mu\nu\alpha\beta}_{ijmn} A^{2}{}^{\alpha\beta\gamma\xi}_{mnkl} =& -\hbar \sum_{\substack{n\\ \beta}} \int d\omega' f(\omega') \sum_{pp'p''} \langle \phi_{p''}|k\gamma \downarrow\rangle\langle n\beta \uparrow|\phi_{p''}\rangle \delta(\hbar\omega' - E_{p''}) \\
&\times \frac{\langle \phi_p | i\mu \uparrow\rangle\langle j\nu \downarrow|\phi_{p'}\rangle\langle l\xi \uparrow|\phi_p\rangle\langle\phi_{p'}|n\beta\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \ , \text{ and}
\end{aligned} \tag{3.70}$$

$$\begin{aligned}
\sum_{\substack{mn\\ \alpha\beta}} \Gamma^{13}{}^{\mu\nu\alpha\beta}_{ijmn} A^{3}{}^{\alpha\beta\gamma\xi}_{mnkl} =& \hbar \sum_{\substack{m\\ \alpha}} \int d\omega' f(\omega') \sum_{pp'p''} \langle \phi_{p''}|m\alpha \downarrow\rangle\langle l\xi \uparrow|\phi_{p''}\rangle \delta(\hbar\omega' - E_{p''}) \\
&\times \frac{\langle \phi_p | i\mu \uparrow\rangle\langle j\nu \downarrow|\phi_{p'}\rangle\langle m\alpha \downarrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \ .
\end{aligned} \tag{3.71}$$

Now we join these equations together using the relation $\sum_{m\alpha\sigma}\langle\phi_{p''}|m\alpha\sigma\rangle\langle m\alpha\sigma|\phi_p\rangle = \delta_{p''p}$ to obtain

$$\begin{aligned}
\chi^{+-}_{(0)}{}^{\mu\nu\gamma\xi}_{ijkl}(\omega) =& \hbar \int d\omega' f(\omega') \sum_{pp'} \delta(\hbar\omega' - E_p) \frac{\langle \phi_p | i\mu \uparrow\rangle\langle j\nu \downarrow|\phi_{p'}\rangle\langle l\xi \uparrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \\
&- \hbar \int d\omega' f(\omega') \sum_{pp'} \delta(\hbar\omega' - E_{p'}) \frac{\langle \phi_p | i\mu \uparrow\rangle\langle j\nu \downarrow|\phi_{p'}\rangle\langle l\xi \uparrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \ .
\end{aligned} \tag{3.72}$$

Note that the difference between the terms is in the delta function only. Using Eqs. (2.10) and (2.13), we can write Eq. (3.72) as

$$\begin{aligned}
\overset{+-}{\chi}{}^{\mu\nu\gamma\xi}_{(0)ijkl}(\omega) =&\hbar \int d\omega' f(\omega') \sum_{p'} \frac{\langle j\nu\downarrow|\phi_{p'}\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega+\hbar\omega'-E_{p'}} \sum_{p}\langle l\xi\uparrow|\phi_p\rangle\langle\phi_p|i\mu\uparrow\rangle\delta(\hbar\omega'-E_p) \\
&+\hbar \int d\omega' f(\omega') \sum_{p} \frac{\langle l\xi\uparrow|\phi_{p}\rangle\langle\phi_{p}|i\mu\uparrow\rangle}{\hbar\omega'-\hbar\omega-E_{p}} \sum_{p'}\langle j\nu\downarrow|\phi_{p'}\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle\delta(\hbar\omega'-E_{p'}) \\
=&\hbar \int d\omega' f(\omega') G^{\downarrow\downarrow\nu\gamma}_{jk}(\omega+\omega')\widetilde{\mathrm{Im}}G^{\uparrow\uparrow\xi\mu}_{li}(\omega') \\
&+\hbar \int d\omega' f(\omega') [G^{\uparrow\uparrow\mu\xi}_{il}(\omega'-\omega)]^*\widetilde{\mathrm{Im}}G^{\downarrow\downarrow\nu\gamma}_{jk}(\omega') \ .
\end{aligned} \tag{3.73}$$

So, the final result for the Hartree-Fock susceptibility $\overset{+-}{\chi}{}^{\mu\nu\gamma\xi}_{(0)ijkl}(\omega)$ in terms of the Green function can be written as

$$\begin{aligned}
\overset{+-}{\chi}{}^{\mu\nu\gamma\xi}_{(0)ijkl}(\omega) &= \hbar \int d\omega' f(\omega') \left\{ G^{\downarrow\downarrow\nu\gamma}_{jk}(\omega'+\omega)\widetilde{\mathrm{Im}}G^{\uparrow\uparrow\xi\mu}_{li}(\omega') + [G^{\uparrow\uparrow\mu\xi}_{il}(\omega'-\omega)]^*\widetilde{\mathrm{Im}}G^{\downarrow\downarrow\nu\gamma}_{jk}(\omega') \right\} \\
&= \frac{i\hbar}{2\pi} \int d\omega' f(\omega') \Big(G^{\downarrow\downarrow\nu\gamma}_{jk}(\omega'+\omega) \left\{ G^{\uparrow\uparrow\xi\mu}_{li}(\omega') - \left[G^{\uparrow\uparrow\mu\xi}_{il}(\omega')\right]^* \right\} \\
&\qquad + [G^{\uparrow\uparrow\mu\xi}_{il}(\omega'-\omega)]^* \left\{ G^{\downarrow\downarrow\nu\gamma}_{jk}(\omega') - \left[G^{\downarrow\downarrow\gamma\nu}_{kj}(\omega')\right]^* \right\} \Big) \ .
\end{aligned} \tag{3.74}$$

3.1.7 Solution for $\overset{\uparrow-}{\chi}_{(0)}$

The equations for the other susceptibilities are obtained analogously. The equation for $\overset{\uparrow-}{\chi}_{(0)}$ is given by

$$\overset{\uparrow-}{\chi}{}^{\mu\nu\gamma\xi}_{(0)ijkl}(\omega) = \overset{2}{\chi}{}^{\mu\nu\gamma\xi}_{(0)ijkl}(\omega) = \left[\Gamma^{21}A^1 + \Gamma^{22}A^2 + \Gamma^{23}A^3\right]^{\mu\nu\gamma\xi}_{ijkl} \ . \tag{3.75}$$

The first term of the equation above is given by

$$\begin{aligned}
\sum_{\substack{mn\\ \alpha\beta}} \overset{21}{\Gamma}{}^{\mu\nu\alpha\beta}_{ijmn} \overset{1}{A}{}^{\alpha\beta\gamma\xi}_{mnkl} =&\hbar \sum_{\substack{m\\ \alpha}} \int d\omega' f(\omega') \sum_{pp'p''} \langle\phi_{p''}|m\alpha\uparrow\rangle\langle l\xi\uparrow|\phi_{p''}\rangle\delta(\hbar\omega'-E_{p''}) \\
&\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\uparrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega-E_{p'}+E_p} \\
&-\hbar \sum_{\substack{n\\ \beta}} \int d\omega' f(\omega') \sum_{pp'p''} \langle\phi_{p''}|k\gamma\downarrow\rangle\langle n\beta\downarrow|\phi_{p''}\rangle\delta(\hbar\omega'-E_{p''}) \\
&\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\uparrow|\phi_{p'}\rangle\langle l\xi\uparrow|\phi_p\rangle\langle\phi_{p'}|n\beta\downarrow\rangle}{\hbar\omega-E_{p'}+E_p} \ .
\end{aligned} \tag{3.76}$$

Comparing with Eq. (3.69), we note that there's only a change in one of the spins. The same is valid for all the terms, and we have

$$\begin{aligned}\sum_{\substack{mn\\\alpha\beta}} \Gamma^{22\,\mu\nu\alpha\beta}_{ijmn} A^{2\,\alpha\beta\gamma\xi}_{mnkl} =& -\hbar \sum_{\substack{n\\\beta}} \int d\omega' f(\omega') \sum_{pp'p''} \langle\phi_{p''}|k\gamma\downarrow\rangle\langle n\beta\uparrow|\phi_{p''}\rangle\delta(\hbar\omega' - E_{p''}) \\ &\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\uparrow|\phi_{p'}\rangle\langle l\xi\uparrow|\phi_p\rangle\langle\phi_{p'}|n\beta\uparrow\rangle}{\hbar\omega - E_{p'} + E_p}\ , \text{ and}\end{aligned} \tag{3.77}$$

$$\begin{aligned}\sum_{\substack{mn\\\alpha\beta}} \Gamma^{23\,\mu\nu\alpha\beta}_{ijmn} A^{3\,\alpha\beta\gamma\xi}_{mnkl} =& \hbar \sum_{\substack{m\\\alpha}} \int d\omega' f(\omega') \sum_{pp'p''} \langle\phi_{p''}|m\alpha\downarrow\rangle\langle l\xi\uparrow|\phi_{p''}\rangle\delta(\hbar\omega' - E_{p''}) \\ &\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\uparrow|\phi_{p'}\rangle\langle m\alpha\downarrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega - E_{p'} + E_p}\ .\end{aligned} \tag{3.78}$$

Putting all the terms together, we obtain

$$\begin{aligned}\chi^{\uparrow-\,\mu\nu\gamma\xi}_{(0)ijkl}(\omega) =& \hbar \int d\omega' f(\omega') \sum_{pp'} \delta(\hbar\omega' - E_p) \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\uparrow|\phi_{p'}\rangle\langle l\xi\uparrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \\ &- \hbar \int d\omega' f(\omega') \sum_{pp'} \delta(\hbar\omega' - E_{p'}) \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\uparrow|\phi_{p'}\rangle\langle l\xi\uparrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega - E_{p'} + E_p}\ ,\end{aligned} \tag{3.79}$$

which can be written as

$$\begin{aligned}\chi^{\uparrow-\,\mu\nu\gamma\xi}_{(0)ijkl}(\omega) =& \hbar \int d\omega' f(\omega') \sum_{p'} \frac{\langle j\nu\uparrow|\phi_{p'}\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega + \hbar\omega' - E_{p'}} \sum_p \langle l\xi\uparrow|\phi_p\rangle\langle\phi_p|i\mu\uparrow\rangle\delta(\hbar\omega' - E_p) \\ &+ \hbar \int d\omega' f(\omega') \sum_p \frac{\langle l\xi\uparrow|\phi_p\rangle\langle\phi_p|i\mu\uparrow\rangle}{\hbar\omega' - \hbar\omega - E_p} \sum_{p'} \langle j\nu\uparrow|\phi_{p'}\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle\delta(\hbar\omega' - E_{p'}) \\ =& \hbar \int d\omega' f(\omega') G^{\uparrow\downarrow\,\nu\gamma}_{jk}(\omega + \omega') \widetilde{\mathrm{Im}} G^{\uparrow\uparrow\,\xi\mu}_{li}(\omega') \\ &+ \hbar \int d\omega' f(\omega') [G^{\uparrow\uparrow\,\mu\xi}_{il}(\omega' - \omega)]^* \widetilde{\mathrm{Im}} G^{\uparrow\downarrow\,\nu\gamma}_{jk}(\omega')\ .\end{aligned} \tag{3.80}$$

So the solution is given by

$$\begin{aligned}\chi^{\uparrow-\,\mu\nu\gamma\xi}_{(0)ijkl}(\omega) =& \hbar \int d\omega' f(\omega') \left\{ G^{\uparrow\downarrow\,\nu\gamma}_{jk}(\omega' + \omega) \widetilde{\mathrm{Im}} G^{\uparrow\uparrow\,\xi\mu}_{li}(\omega') + [G^{\uparrow\uparrow\,\mu\xi}_{il}(\omega' - \omega)]^* \widetilde{\mathrm{Im}} G^{\uparrow\downarrow\,\nu\gamma}_{jk}(\omega') \right\} \\ =& \frac{i\hbar}{2\pi} \int d\omega' f(\omega') \left(G^{\uparrow\downarrow\,\nu\gamma}_{jk}(\omega' + \omega) \left\{ G^{\uparrow\uparrow\,\xi\mu}_{li}(\omega') - \left[G^{\uparrow\uparrow\,\mu\xi}_{il}(\omega') \right]^* \right\} \right. \\ &\left. + [G^{\uparrow\uparrow\,\mu\xi}_{il}(\omega' - \omega)]^* \left\{ G^{\uparrow\downarrow\,\nu\gamma}_{jk}(\omega') - \left[G^{\downarrow\uparrow\,\gamma\nu}_{kj}(\omega') \right]^* \right\} \right)\ .\end{aligned} \tag{3.81}$$

3.1.8 Solution for $\chi_{(0)}^{\downarrow -}$

Since the calculations are straight-forward, we can skip the first steps and write

$$\begin{aligned}\chi_{(0)ijkl}^{\downarrow -\mu\nu\gamma\xi}(\omega) =&\hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega' - E_p)\frac{\langle\phi_p|i\mu\downarrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle l\xi\uparrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega - E_{p'} + E_p}\\ &-\hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega' - E_{p'})\frac{\langle\phi_p|i\mu\downarrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle l\xi\uparrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega - E_{p'} + E_p}\ ,\end{aligned} \tag{3.82}$$

or, in terms of the monoeletronic green functions

$$\begin{aligned}\chi_{(0)ijkl}^{\downarrow -\mu\nu\gamma\xi}(\omega) =&\hbar\int d\omega' f(\omega')\sum_{p'}\frac{\langle j\nu\downarrow|\phi_{p'}\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega + \hbar\omega' - E_{p'}}\sum_{p}\langle l\xi\uparrow|\phi_p\rangle\langle\phi_p|i\mu\downarrow\rangle\delta(\hbar\omega' - E_p)\\ &+\hbar\int d\omega' f(\omega')\sum_{p}\frac{\langle l\xi\uparrow|\phi_p\rangle\langle\phi_p|i\mu\downarrow\rangle}{\hbar\omega' - \hbar\omega - E_p}\sum_{p'}\langle j\nu\downarrow|\phi_{p'}\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle\delta(\hbar\omega' - E_{p'})\\ =&\hbar\int d\omega' f(\omega')G_{jk}^{\downarrow\downarrow\nu\gamma}(\omega+\omega')\widetilde{\mathrm{Im}}G_{li}^{\uparrow\downarrow\xi\mu}(\omega')\\ &+\hbar\int d\omega' f(\omega')[G_{il}^{\downarrow\uparrow\mu\xi}(\omega'-\omega)]^*\widetilde{\mathrm{Im}}G_{jk}^{\downarrow\downarrow\nu\gamma}(\omega')\ .\end{aligned} \tag{3.83}$$

And the final solution is

$$\begin{aligned}\chi_{(0)ijkl}^{\downarrow -\mu\nu\gamma\xi}(\omega) = \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\Big(&G_{jk}^{\downarrow\downarrow\nu\gamma}(\omega'+\omega)\left\{G_{li}^{\uparrow\downarrow\xi\mu}(\omega') - \left[G_{il}^{\downarrow\uparrow\mu\xi}(\omega')\right]^*\right\}\\ &+[G_{il}^{\downarrow\uparrow\mu\xi}(\omega'-\omega)]^*\left\{G_{jk}^{\downarrow\downarrow\nu\gamma}(\omega') - \left[G_{kj}^{\downarrow\downarrow\gamma\nu}(\omega')\right]^*\right\}\Big)\ .\end{aligned} \tag{3.84}$$

3.1.9 Solution for $\chi_{(0)}^{--}$

An easy way to obtain the solution is to note that the spin indices that accompany the state of i,μ and j,ν are related to the susceptibility we are calculating. In this last case, we have

$$\begin{aligned}\chi_{(0)ijkl}^{--\mu\nu\gamma\xi}(\omega) = \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\Big(&G_{jk}^{\uparrow\downarrow\nu\gamma}(\omega'+\omega)\left\{G_{li}^{\uparrow\downarrow\xi\mu}(\omega') - \left[G_{il}^{\downarrow\uparrow\mu\xi}(\omega')\right]^*\right\}\\ &+[G_{il}^{\downarrow\uparrow\mu\xi}(\omega'-\omega)]^*\left\{G_{jk}^{\uparrow\downarrow\nu\gamma}(\omega') - \left[G_{kj}^{\downarrow\uparrow\gamma\nu}(\omega')\right]^*\right\}\Big)\ .\end{aligned} \tag{3.85}$$

$$\begin{aligned}\chi_{(0)ijkl}^{+-\mu\nu\gamma\xi}(\omega) =&\hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega' - E_p)\frac{\langle\phi_p|i\mu\sigma_1\rangle\langle j\nu\sigma_2|\phi_{p'}\rangle\langle l\xi\uparrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega - E_{p'} + E_p}\\ &-\hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega' - E_{p'})\frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle l\xi\uparrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega - E_{p'} + E_p}\ .\end{aligned} \tag{3.86}$$

Note that the difference between the terms is in the delta function only. Using Eqs. 2.10 and 2.13, we can write Eq. (3.72) as

$$\begin{aligned}\chi^{+-\,\mu\nu\gamma\xi}_{(0)ijkl}(\omega) =&\hbar\int d\omega' f(\omega')\sum_{p'}\frac{\langle j\nu\downarrow|\phi_{p'}\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle}{\hbar\omega+\hbar\omega'-E_{p'}}\sum_{p}\langle l\xi\uparrow|\phi_p\rangle\langle\phi_p|i\mu\uparrow\rangle\delta(\hbar\omega'-E_p)\\ &+\hbar\int d\omega' f(\omega')\sum_{p}\frac{\langle l\xi\uparrow|\phi_p\rangle\langle\phi_p|i\mu\uparrow\rangle}{\hbar\omega'-\hbar\omega-E_p}\sum_{p'}\langle j\nu\downarrow|\phi_{p'}\rangle\langle\phi_{p'}|k\gamma\downarrow\rangle\delta(\hbar\omega'-E_{p'})\\ &=\hbar\int d\omega' f(\omega')G^{\downarrow\downarrow\nu\gamma}_{jk}(\omega+\omega')\widetilde{\mathrm{Im}}G^{\uparrow\uparrow\xi\mu}_{li}(\omega')\\ &+\hbar\int d\omega' f(\omega')[G^{\uparrow\uparrow\mu\xi}_{il}(\omega'-\omega)]^*\widetilde{\mathrm{Im}}G^{\downarrow\downarrow\nu\gamma}_{jk}(\omega')\ .\end{aligned} \tag{3.87}$$

So, the final result for the Hartree-Fock susceptibility $\chi^{+-\,\mu\nu\gamma\xi}_{(0)ijkl}(\omega)$ in terms of the Green function can be written as

$$\begin{aligned}\chi^{+-\,\mu\nu\gamma\xi}_{(0)ijkl}(\omega) &= \hbar\int d\omega' f(\omega')\left\{G^{\downarrow\downarrow\nu\gamma}_{jk}(\omega'+\omega)\widetilde{\mathrm{Im}}G^{\uparrow\uparrow\xi\mu}_{li}(\omega')+[G^{\uparrow\uparrow\mu\xi}_{il}(\omega'-\omega)]^*\widetilde{\mathrm{Im}}G^{\downarrow\downarrow\nu\gamma}_{jk}(\omega')\right\}\\ &=\frac{i\hbar}{2\pi}\int d\omega' f(\omega')\left(G^{\downarrow\downarrow\nu\gamma}_{jk}(\omega'+\omega)\left\{G^{\uparrow\uparrow\xi\mu}_{li}(\omega')-\left[G^{\uparrow\uparrow\mu\xi}_{il}(\omega')\right]^*\right\}\right.\\ &\qquad\left.+[G^{\uparrow\uparrow\mu\xi}_{il}(\omega'-\omega)]^*\left\{G^{\downarrow\downarrow\nu\gamma}_{jk}(\omega')-\left[G^{\downarrow\downarrow\gamma\nu}_{kj}(\omega')\right]^*\right\}\right)\ .\end{aligned} \tag{3.88}$$

3.1.10 Generic solution for $\chi_{(0)}$

The results obtained before can be summarized as

$$\begin{aligned}\langle i\mu\sigma_1, j\nu\sigma_2|\chi|k\gamma\sigma_3, l\xi\sigma_4\rangle(\omega) = \frac{i\hbar}{2\pi}\int d\omega' f(\omega')&\left(G^{\sigma_2\sigma_3\nu\gamma}_{jk}(\omega'+\omega)\left\{G^{\sigma_4\sigma_1\xi\mu}_{li}(\omega')-\left[G^{\sigma_1\sigma_4\mu\xi}_{il}(\omega')\right]^*\right\}\right.\\ &\left.+[G^{\sigma_1\sigma_4\mu\xi}_{il}(\omega'-\omega)]^*\left\{G^{\sigma_2\sigma_3\nu\gamma}_{jk}(\omega')-\left[G^{\sigma_3\sigma_2\gamma\nu}_{kj}(\omega')\right]^*\right\}\right)\ .\end{aligned} \tag{3.89}$$

3.2 RPA Susceptibility

To obtain the susceptibility within the Random Phase Approximation, we are going to calculate the equation of motion using the full hamiltonian given by Eq. (1.1), and then use the approximation showed in Eq. (1.15).

The equation of motion was written in Eq. (3.4), but instead of using $\hat{H}^{\mathrm{HF}}$ as we did before, we need to calculate

$$[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'},\hat{H}] = [c^\dagger_{i\mu\sigma}c_{j\nu\sigma'},\hat{H}_0+\hat{H}_{\mathrm{int}}+\hat{H}_{\mathrm{so}}]\ . \tag{3.90}$$

Although we have renormalized the hopping and written a “spin-flip” coefficient a, we can notice that the terms involving $\hat{H}_0$ and $\hat{H}_{\mathrm{so}}$ will be the same within RPA, since they are one-electron parts of the hamiltonian. The difference will come from the interaction term

$$[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'},\hat{H}_{\rm int}] = \frac{1}{2}\sum_{\substack{m\\ s,s'}}\sum_{\alpha\beta\gamma\xi} U^m_{\alpha\beta\gamma\xi}[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}, c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}]\ . \tag{3.91}$$

The commutator above, $C = [c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}, c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}]$ is

$$\begin{aligned}
C =& c^\dagger_{i\mu\sigma}\underbrace{c_{j\nu\sigma'}c^\dagger_{m\alpha s}}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s} - c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}\\
=& c^\dagger_{i\mu\sigma}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}\delta_{mj}\delta_{\alpha\nu}\delta_{s\sigma'} - \underbrace{c^\dagger_{i\mu\sigma}c^\dagger_{m\alpha s}}c_{j\nu\sigma'}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s} - c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}\\
=& c^\dagger_{i\mu\sigma}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}\delta_{mj}\delta_{\alpha\nu}\delta_{s\sigma'} + c^\dagger_{m\alpha s}c^\dagger_{i\mu\sigma}\underbrace{c_{j\nu\sigma'}c^\dagger_{m\beta s'}}c_{m\xi s'}c_{m\gamma s} - c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}\\
=& c^\dagger_{i\mu\sigma}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}\delta_{mj}\delta_{\alpha\nu}\delta_{s\sigma'} + c^\dagger_{m\alpha s}c^\dagger_{i\mu\sigma}c_{m\xi s'}c_{m\gamma s}\delta_{mj}\delta_{\beta\nu}\delta_{s'\sigma'} - c^\dagger_{m\alpha s}\underbrace{c^\dagger_{i\mu\sigma}c^\dagger_{m\beta s'}}\underbrace{c_{j\nu\sigma'}c_{m\xi s'}}c_{m\gamma s}\\
&- c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}\\
=& c^\dagger_{i\mu\sigma}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}\delta_{mj}\delta_{\alpha\nu}\delta_{s\sigma'} + c^\dagger_{m\alpha s}c^\dagger_{i\mu\sigma}c_{m\xi s'}c_{m\gamma s}\delta_{mj}\delta_{\beta\nu}\delta_{s'\sigma'} - c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}\underbrace{c^\dagger_{i\mu\sigma}c_{m\xi s'}}c_{j\nu\sigma'}c_{m\gamma s}\\
&- c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}\\
=& c^\dagger_{i\mu\sigma}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}\delta_{mj}\delta_{\alpha\nu}\delta_{s\sigma'} + c^\dagger_{m\alpha s}c^\dagger_{i\mu\sigma}c_{m\xi s'}c_{m\gamma s}\delta_{mj}\delta_{\beta\nu}\delta_{s'\sigma'} - c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{j\nu\sigma'}c_{m\gamma s}\delta_{mi}\delta_{\mu\xi}\delta_{s'\sigma}\\
&+ c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c^\dagger_{i\mu\sigma}\underbrace{c_{j\nu\sigma'}c_{m\gamma s}} - c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}\\
=& c^\dagger_{i\mu\sigma}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}\delta_{mj}\delta_{\alpha\nu}\delta_{s\sigma'} + c^\dagger_{m\alpha s}c^\dagger_{i\mu\sigma}c_{m\xi s'}c_{m\gamma s}\delta_{mj}\delta_{\beta\nu}\delta_{s'\sigma'} - c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{j\nu\sigma'}c_{m\gamma s}\delta_{mi}\delta_{\mu\xi}\delta_{s'\sigma}\\
&- c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}\underbrace{c^\dagger_{i\mu\sigma}c_{m\gamma s}}c_{j\nu\sigma'} - c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}\\
=& c^\dagger_{i\mu\sigma}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}\delta_{mj}\delta_{\alpha\nu}\delta_{s\sigma'} + c^\dagger_{m\alpha s}c^\dagger_{i\mu\sigma}c_{m\xi s'}c_{m\gamma s}\delta_{mj}\delta_{\beta\nu}\delta_{s'\sigma'} - c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{j\nu\sigma'}c_{m\gamma s}\delta_{mi}\delta_{\mu\xi}\delta_{s'\sigma}\\
&- c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{j\nu\sigma'}\delta_{mi}\delta_{\mu\gamma}\delta_{s\sigma}\ .
\end{aligned} \tag{3.92}$$

Taking this result back to Eq. (3.91) we end up with

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'},\hat{H}_{\text{int}}] =&\frac{1}{2}\sum_{\substack{m\\ s,s'}}\sum_{\alpha\beta\gamma\xi}U^m_{\alpha\beta\gamma\xi}c^\dagger_{i\mu\sigma}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}\delta_{mj}\delta_{\alpha\nu}\delta_{s\sigma'}\\
&+\frac{1}{2}\sum_{\substack{m\\ s,s'}}\sum_{\alpha\beta\gamma\xi}U^m_{\alpha\beta\gamma\xi}\underbrace{c^\dagger_{m\alpha s}c^\dagger_{i\mu\sigma}}\underbrace{c_{m\xi s'}c_{m\gamma s}}\delta_{mj}\delta_{\beta\nu}\delta_{s'\sigma'}\\
&-\frac{1}{2}\sum_{\substack{m\\ s,s'}}\sum_{\alpha\beta\gamma\xi}U^m_{\alpha\beta\gamma\xi}\underbrace{c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}}\underbrace{c_{j\nu\sigma'}c_{m\gamma s}}\delta_{mi}\delta_{\mu\xi}\delta_{s'\sigma}\\
&-\frac{1}{2}\sum_{\substack{m\\ s,s'}}\sum_{\alpha\beta\gamma\xi}U^m_{\alpha\beta\gamma\xi}c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{j\nu\sigma'}\delta_{mi}\delta_{\mu\gamma}\delta_{s\sigma}\\
=&\frac{1}{2}\sum_{\substack{\beta\gamma\xi\\ s'}}U^j_{\nu\beta\gamma\xi}c^\dagger_{i\mu\sigma}c^\dagger_{j\beta s'}c_{j\xi s'}c_{j\gamma\sigma'}+\frac{1}{2}\sum_{\substack{\alpha\gamma\xi\\ s}}U^j_{\alpha\nu\gamma\xi}c^\dagger_{i\mu\sigma}c^\dagger_{j\alpha s}c_{j\gamma s}c_{j\xi\sigma'}\\
&-\frac{1}{2}\sum_{\substack{\alpha\beta\gamma\\ s}}U^i_{\alpha\beta\gamma\mu}c^\dagger_{i\beta\sigma}c^\dagger_{i\alpha s}c_{i\gamma s}c_{j\nu\sigma'}-\frac{1}{2}\sum_{\substack{\alpha\beta\xi\\ s'}}U^i_{\alpha\beta\mu\xi}c^\dagger_{i\alpha\sigma}c^\dagger_{i\beta s'}c_{i\xi s'}c_{j\nu\sigma'}\ .
\end{aligned}
\tag{3.93}
$$

Changing the names of the variables to combine the sums ($\xi\to\alpha$ and $s'\to s$ in the first term, $\xi\to\beta$ in the second, and $\xi\to\gamma$ and $s'\to s$ in the fourth), we get

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'},\hat{H}_{\text{int}}] =&\frac{1}{2}\sum_{\substack{\alpha\beta\gamma\\ s}}\Big(U^j_{\nu\beta\gamma\alpha}c^\dagger_{i\mu\sigma}c^\dagger_{j\beta s}c_{j\alpha s}c_{j\gamma\sigma'}+U^j_{\alpha\nu\gamma\beta}c^\dagger_{i\mu\sigma}c^\dagger_{j\alpha s}c_{j\gamma s}c_{j\beta\sigma'}\\
&-U^i_{\alpha\beta\gamma\mu}c^\dagger_{i\beta\sigma}c^\dagger_{i\alpha s}c_{i\gamma s}c_{j\nu\sigma'}-U^i_{\alpha\beta\mu\gamma}c^\dagger_{i\alpha\sigma}c^\dagger_{i\beta s}c_{i\gamma s}c_{j\nu\sigma'}\Big)\ .
\end{aligned}
\tag{3.94}
$$

Now we change the index $\alpha\rightleftharpoons\beta$ in the first and third terms, as well as $\beta\rightleftharpoons\gamma$ in the second one, to obtain

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'},\hat{H}_{\text{int}}] =&\frac{1}{2}\sum_{\substack{\alpha\beta\gamma\\ s}}\Big(U^j_{\nu\alpha\gamma\beta}c^\dagger_{i\mu\sigma}c^\dagger_{j\alpha s}c_{j\beta s}c_{j\gamma\sigma'}+U^j_{\alpha\nu\beta\gamma}c^\dagger_{i\mu\sigma}c^\dagger_{j\alpha s}c_{j\beta s}c_{j\gamma\sigma'}\\
&-U^i_{\beta\alpha\gamma\mu}c^\dagger_{i\alpha\sigma}c^\dagger_{i\beta s}c_{i\gamma s}c_{j\nu\sigma'}-U^i_{\alpha\beta\mu\gamma}c^\dagger_{i\alpha\sigma}c^\dagger_{i\beta s}c_{i\gamma s}c_{j\nu\sigma'}\Big)\\
=&\frac{1}{2}\sum_{\substack{\alpha\beta\gamma\\ s}}\Big[(U^j_{\nu\alpha\gamma\beta}+U^j_{\alpha\nu\beta\gamma})c^\dagger_{i\mu\sigma}c^\dagger_{j\alpha s}c_{j\beta s}c_{j\gamma\sigma'}\\
&-(U^i_{\beta\alpha\gamma\mu}+U^i_{\alpha\beta\mu\gamma})c^\dagger_{i\alpha\sigma}c^\dagger_{i\beta s}c_{i\gamma s}c_{j\nu\sigma'}\Big]\ .
\end{aligned}
\tag{3.95}
$$

Using the symmetry of the U elements showed in Eq. (1.4) and changing the index $\gamma\to\lambda$, we find

$$
[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'},\hat{H}_{\text{int}}]=\sum_{\substack{\alpha\beta\lambda\\ s}}\Big(U^j_{\nu\alpha\lambda\beta}c^\dagger_{i\mu\sigma}c^\dagger_{j\alpha s}c_{j\beta s}c_{j\lambda\sigma'}-U^i_{\beta\alpha\lambda\mu}c^\dagger_{i\alpha\sigma}c^\dagger_{i\beta s}c_{i\lambda s}c_{j\nu\sigma'}\Big)\ . \tag{3.96}
$$

Rewriting this equation as

$$
\begin{aligned}
&\sum_{\substack{\alpha\beta\lambda\\ s}} U^j_{\nu\alpha\lambda\beta}\,\underbrace{c^\dagger_{i\mu\sigma}c^\dagger_{j\alpha s}}\,\underbrace{c_{j\beta s}c_{j\lambda\sigma'}} - U^i_{\beta\alpha\lambda\mu}\,\underbrace{c^\dagger_{i\alpha\sigma}c^\dagger_{i\beta s}}\,\underbrace{c_{i\lambda s}c_{j\nu\sigma'}} \\
=&\sum_{\substack{\alpha\beta\lambda\\ s}} U^j_{\nu\alpha\lambda\beta}c^\dagger_{j\alpha s}\,\underbrace{c^\dagger_{i\mu\sigma}c_{j\lambda\sigma'}}\,c_{j\beta s} - U^i_{\beta\alpha\lambda\mu}c^\dagger_{i\beta s}\,\underbrace{c^\dagger_{i\alpha\sigma}c_{j\nu\sigma'}}\,c_{i\lambda s} \\
=&\sum_{\substack{\alpha\beta\lambda\\ s}} U^j_{\nu\alpha\lambda\beta}c^\dagger_{j\alpha s}c_{j\beta s}\delta_{ij}\delta_{\lambda\mu}\delta_{\sigma\sigma'} - U^i_{\beta\alpha\lambda\mu}c^\dagger_{i\beta s}c_{i\lambda s}\delta_{ij}\delta_{\alpha\nu}\delta_{\sigma\sigma'} \\
&+\sum_{\substack{\alpha\beta\lambda\\ s}} U^i_{\beta\alpha\lambda\mu}c^\dagger_{i\beta s}c_{j\nu\sigma'}c^\dagger_{i\alpha\sigma}c_{i\lambda s} - U^j_{\nu\alpha\lambda\beta}c^\dagger_{j\alpha s}c_{j\lambda\sigma'}c^\dagger_{i\mu\sigma}c_{j\beta s}\ .
\end{aligned}
\tag{3.97}
$$

The first two terms can be calculated summing in orbitals to get rid of the δ functions, and then changing the index $\lambda \to \alpha$ in the second term so the sums can be combined; after that, we make the change $\alpha \rightleftharpoons \beta$ in the second term to use the symmetry of U:

$$
\begin{aligned}
&\sum_{\substack{\alpha\beta\lambda\\ s}} U^j_{\nu\alpha\lambda\beta}c^\dagger_{j\alpha s}c_{j\beta s}\delta_{ij}\delta_{\lambda\mu}\delta_{\sigma\sigma'} - U^i_{\beta\alpha\lambda\mu}c^\dagger_{i\beta s}c_{i\lambda s}\delta_{ij}\delta_{\alpha\nu}\delta_{\sigma\sigma'} \\
=&\sum_{\substack{\alpha\beta\\ s}} U^j_{\nu\alpha\mu\beta}c^\dagger_{j\alpha s}c_{j\beta s}\delta_{ij}\delta_{\sigma\sigma'} - U^i_{\beta\nu\alpha\mu}c^\dagger_{i\beta s}c_{i\alpha s}\delta_{ij}\delta_{\sigma\sigma'} \\
=&\sum_{\substack{\alpha\beta\\ s}} U^j_{\nu\alpha\mu\beta}c^\dagger_{j\alpha s}c_{j\beta s}\delta_{ij}\delta_{\sigma\sigma'} - U^i_{\alpha\nu\beta\mu}c^\dagger_{i\alpha s}c_{i\beta s}\delta_{ij}\delta_{\sigma\sigma'} \\
=&\sum_{\substack{\alpha\beta\\ s}} U^j_{\nu\alpha\mu\beta}c^\dagger_{j\alpha s}c_{j\beta s}\delta_{ij}\delta_{\sigma\sigma'} - U^i_{\nu\alpha\mu\beta}c^\dagger_{i\alpha s}c_{i\beta s}\delta_{ij}\delta_{\sigma\sigma'} = 0\ .
\end{aligned}
\tag{3.98}
$$

Within RPA, the two remaining terms with four operators product of Eq. (3.97) become

$$
\begin{aligned}
&\sum_{\substack{\alpha\beta\lambda\\ s}} U^i_{\beta\alpha\lambda\mu}c^\dagger_{i\beta s}c_{j\nu\sigma'}c^\dagger_{i\alpha\sigma}c_{i\lambda s} - U^j_{\nu\alpha\lambda\beta}c^\dagger_{j\alpha s}c_{j\lambda\sigma'}c^\dagger_{i\mu\sigma}c_{j\beta s} \\
\to&\sum_{\substack{\alpha\beta\lambda\\ s}} U^i_{\beta\alpha\lambda\mu}\Big\{\langle c^\dagger_{i\alpha\sigma}c_{i\lambda s}\rangle c^\dagger_{i\beta s}c_{j\nu\sigma'} - \langle c^\dagger_{i\beta s}c_{i\lambda s}\rangle c^\dagger_{i\alpha\sigma}c_{j\nu\sigma'} \\
&\qquad\qquad +\langle c^\dagger_{i\beta s}c_{j\nu\sigma'}\rangle c^\dagger_{i\alpha\sigma}c_{i\lambda s} - \langle c^\dagger_{i\alpha\sigma}c_{j\nu\sigma'}\rangle c^\dagger_{i\beta s}c_{i\lambda s}\Big\} \\
&\quad -U^j_{\nu\alpha\lambda\beta}\Big\{\langle c^\dagger_{j\alpha s}c_{j\lambda\sigma'}\rangle c^\dagger_{i\mu\sigma}c_{j\beta s} - \langle c^\dagger_{j\alpha s}c_{j\beta s}\rangle c^\dagger_{i\mu\sigma}c_{j\lambda\sigma'} \\
&\qquad\qquad +\langle c^\dagger_{i\mu\sigma}c_{j\beta s}\rangle c^\dagger_{j\alpha s}c_{j\lambda\sigma'} - \langle c^\dagger_{i\mu\sigma}c_{j\lambda\sigma'}\rangle c^\dagger_{j\alpha s}c_{j\beta s}\Big\}\ .
\end{aligned}
\tag{3.99}
$$

To compare the terms of this equation with the ones that are included in the renormalized hoppings and in the spin-flip terms, it's easier to calculate the same commutator of Eq. (3.91), but using the Hartree-Fock interaction hamiltonian given by Eq. (1.18).

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, \hat{H}^{\mathrm{HF}}_{\mathrm{int}}] = \sum_{\substack{m\\ s,s'}} \sum_{\eta\rho\gamma\xi} \Big\{ & U^m_{\eta\rho\gamma\xi} \langle c^\dagger_{m\rho s'} c_{m\xi s'} \rangle [c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, c^\dagger_{m\eta s} c_{m\gamma s}] \\
& - U^m_{\eta\rho\gamma\xi} \langle c^\dagger_{m\eta s} c_{m\xi s'} \rangle [c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, c^\dagger_{m\rho s'} c_{m\gamma s}] \Big\} \,.
\end{aligned}
\tag{3.100}
$$

Calculating the commutators, we have

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, c^\dagger_{m\eta s} c_{m\gamma s}] =& c^\dagger_{i\mu\sigma} \underbrace{c_{j\nu\sigma'} c^\dagger_{m\eta s}} c_{m\gamma s} - c^\dagger_{m\eta s} c_{m\gamma s} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{m\gamma s} \delta_{mj} \delta_{\eta\nu} \delta_{s\sigma'} - \underbrace{c^\dagger_{i\mu\sigma} c^\dagger_{m\eta s}} \underbrace{c_{j\nu\sigma'} c_{m\gamma s}} - c^\dagger_{m\eta s} c_{m\gamma s} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{m\gamma s} \delta_{mj} \delta_{\eta\nu} \delta_{s\sigma'} - c^\dagger_{m\eta s} \underbrace{c^\dagger_{i\mu\sigma} c_{m\gamma s}} c_{j\nu\sigma'} - c^\dagger_{m\eta s} c_{m\gamma s} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{m\gamma s} \delta_{mj} \delta_{\eta\nu} \delta_{s\sigma'} - c^\dagger_{m\eta s} c_{j\nu\sigma'} \delta_{mi} \delta_{\gamma\mu} \delta_{s\sigma}
\end{aligned}
\tag{3.101}
$$

and

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, c^\dagger_{m\rho s'} c_{m\gamma s}] =& c^\dagger_{i\mu\sigma} \underbrace{c_{j\nu\sigma'} c^\dagger_{m\rho s'}} c_{m\gamma s} - c^\dagger_{m\rho s'} c_{m\gamma s} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{m\gamma s} \delta_{mj} \delta_{\rho\nu} \delta_{s'\sigma'} - \underbrace{c^\dagger_{i\mu\sigma} c^\dagger_{m\rho s'}} \underbrace{c_{j\nu\sigma'} c_{m\gamma s}} - c^\dagger_{m\rho s'} c_{m\gamma s} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{m\gamma s} \delta_{mj} \delta_{\rho\nu} \delta_{s'\sigma'} - c^\dagger_{m\rho s'} \underbrace{c^\dagger_{i\mu\sigma} c_{m\gamma s}} c_{j\nu\sigma'} - c^\dagger_{m\rho s'} c_{m\gamma s} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{m\gamma s} \delta_{mj} \delta_{\rho\nu} \delta_{s'\sigma'} - c^\dagger_{m\rho s'} c_{j\nu\sigma'} \delta_{mi} \delta_{\gamma\mu} \delta_{s\sigma} \,.
\end{aligned}
\tag{3.102}
$$

Substituting back in Eq 3.100 we obtain

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, \hat{H}^{\mathrm{HF}}_{\mathrm{int}}] = \sum_{\substack{m\\ s,s'}} \sum_{\eta\rho\gamma\xi} \Big\{ & U^m_{\eta\rho\gamma\xi} \langle c^\dagger_{m\rho s'} c_{m\xi s'} \rangle \left(c^\dagger_{i\mu\sigma} c_{m\gamma s} \delta_{mj} \delta_{\eta\nu} \delta_{s\sigma'} - c^\dagger_{m\eta s} c_{j\nu\sigma'} \delta_{mi} \delta_{\gamma\mu} \delta_{s\sigma} \right) \\
& - U^m_{\eta\rho\gamma\xi} \langle c^\dagger_{m\eta s} c_{m\xi s'} \rangle \left(c^\dagger_{i\mu\sigma} c_{m\gamma s} \delta_{mj} \delta_{\rho\nu} \delta_{s'\sigma'} - c^\dagger_{m\rho s'} c_{j\nu\sigma'} \delta_{mi} \delta_{\gamma\mu} \delta_{s\sigma} \right) \Big\} \,.
\end{aligned}
\tag{3.103}
$$

Eliminating all the delta functions,

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, \hat{H}^{\mathrm{HF}}_{\mathrm{int}}] =& \sum_{\substack{\rho\gamma\xi\\ s}} U^j_{\nu\rho\gamma\xi} \langle c^\dagger_{j\rho s} c_{j\xi s} \rangle c^\dagger_{i\mu\sigma} c_{j\gamma\sigma'} - \sum_{\substack{\eta\rho\xi\\ s}} U^i_{\eta\rho\mu\xi} \langle c^\dagger_{i\rho s} c_{i\xi s} \rangle c^\dagger_{i\eta\sigma} c_{j\nu\sigma'} \\
& - \sum_{\substack{\eta\gamma\xi\\ s}} U^j_{\eta\nu\gamma\xi} \langle c^\dagger_{j\eta s} c_{j\xi\sigma'} \rangle c^\dagger_{i\mu\sigma} c_{j\gamma s} + \sum_{\substack{\eta\rho\xi\\ s}} U^i_{\eta\rho\mu\xi} \langle c^\dagger_{i\eta\sigma} c_{i\xi s} \rangle c^\dagger_{i\rho s} c_{j\nu\sigma'} \,.
\end{aligned}
\tag{3.104}
$$

The second and fourth tens can be combined in a single sum. To do the same thing with the first and third, let's use the symmetry of U in the third term and change the indices $\eta \rightarrow \rho$ and $\gamma \rightleftharpoons \xi$. Using the symmetry of U also in the other terms,

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}, \hat{H}^{\text{HF}}_{\text{int}}] = &\sum_{\substack{\eta\rho\xi\\ s}} U^i_{\rho\eta\xi\mu}\left\{\langle c^\dagger_{i\eta\sigma}c_{i\xi s}\rangle c^\dagger_{i\rho s}c_{j\nu\sigma'} - \langle c^\dagger_{i\rho s}c_{i\xi s}\rangle c^\dagger_{i\eta\sigma}c_{j\nu\sigma'}\right\}\\
&- \sum_{\substack{\rho\gamma\xi\\ s}} U^j_{\nu\rho\gamma\xi}\left\{\langle c^\dagger_{j\rho s}c_{j\gamma\sigma'}\rangle c^\dagger_{i\mu\sigma}c_{j\xi s} - \langle c^\dagger_{j\rho s}c_{j\xi s}\rangle c^\dagger_{i\mu\sigma}c_{j\gamma\sigma'}\right\} \ .
\end{aligned}
\tag{3.105}
$$

Finally, changing the indices once more,

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma}c_{j\nu\sigma'}, \hat{H}^{\text{HF}}_{\text{int}}] = \sum_{\substack{\alpha\beta\lambda\\ s}} &U^i_{\beta\alpha\lambda\mu}\left\{\langle c^\dagger_{i\alpha\sigma}c_{i\lambda s}\rangle c^\dagger_{i\beta s}c_{j\nu\sigma'} - \langle c^\dagger_{i\beta s}c_{i\lambda s}\rangle c^\dagger_{i\alpha\sigma}c_{j\nu\sigma'}\right\}\\
&- U^j_{\nu\alpha\lambda\beta}\left\{\langle c^\dagger_{j\alpha s}c_{j\lambda\sigma'}\rangle c^\dagger_{i\mu\sigma}c_{j\beta s} - \langle c^\dagger_{j\alpha s}c_{j\beta s}\rangle c^\dagger_{i\mu\sigma}c_{j\lambda\sigma'}\right\} \ .
\end{aligned}
\tag{3.106}
$$

Comparing Eq. (3.99) with this equation we can see that the first two terms inside each curly brackets of that equation are contained in the Hartree-Fock approximation. So the terms that we need to add in the Hartree-Fock equation of motions to obtain the RPA are

$$
\begin{aligned}
\rightarrow &\sum_{\substack{\alpha\beta\lambda\\ s}} U^i_{\beta\alpha\lambda\mu}\left\{\langle c^\dagger_{i\beta s}c_{j\nu\sigma'}\rangle \chi^{\sigma s\,\alpha\lambda\gamma\xi}_{iikl} - \langle c^\dagger_{i\alpha\sigma}c_{j\nu\sigma'}\rangle \chi^{ss\,\beta\lambda\gamma\xi}_{iikl}\right\}\\
&\quad - U^j_{\nu\alpha\lambda\beta}\left\{\langle c^\dagger_{i\mu\sigma}c_{j\beta s}\rangle \chi^{s\sigma'\,\alpha\lambda\gamma\xi}_{jjkl} - \langle c^\dagger_{i\mu\sigma}c_{j\lambda\sigma'}\rangle \chi^{ss\,\alpha\beta\gamma\xi}_{jjkl}\right\}\\
= &\sum_{\substack{mn\\ \alpha\beta}}\sum_{\substack{\lambda\\ s}}\Big\{U^i_{\lambda\alpha\beta\mu}\langle c^\dagger_{i\lambda s}c_{j\nu\sigma'}\rangle\delta_{mi}\delta_{ni}\chi^{\sigma s\,\alpha\beta\gamma\xi}_{mnkl} - U^j_{\nu\alpha\beta\lambda}\langle c^\dagger_{i\mu\sigma}c_{j\lambda s}\rangle\delta_{mj}\delta_{nj}\chi^{s\sigma'\,\alpha\beta\gamma\xi}_{mnkl}\\
&\quad + \left[U^j_{\nu\alpha\lambda\beta}\langle c^\dagger_{i\mu\sigma}c_{j\lambda\sigma'}\rangle\delta_{mj}\delta_{nj} - U^i_{\alpha\lambda\beta\mu}\langle c^\dagger_{i\lambda\sigma}c_{j\nu\sigma'}\rangle\delta_{mi}\delta_{ni}\right]\chi^{ss\,\alpha\beta\gamma\xi}_{mnkl}\Big\} \ .
\end{aligned}
\tag{3.107}
$$

Including this terms, we can write the equation of motion the same way we did in Eq. (3.42) and find

$$
(\hbar\omega - B - \overline{B})\chi = \mathbf{A} \ , \tag{3.108}
$$

where $\overline{B}$ is the matrix form of the terms given by Eq. (3.107). Rewritting the equation above,

$$
\begin{aligned}
(\hbar\omega - B)\chi - \overline{B}\chi =& \mathbf{A}\\
\chi - \Gamma\overline{B}\chi =& \Gamma\mathbf{A}\\
\chi =& \chi^0 + \Gamma\overline{B}\chi = \chi^0 + \Lambda\chi \ ,
\end{aligned}
\tag{3.109}
$$

where we have used Eq. (3.43) to write $\Gamma = (\hbar\omega - B)^{-1}$ and $\chi^0 = \Gamma\mathbf{A}$, and also defined the matrix $\Lambda = \Gamma\overline{B}$.

Now we need to obtain $\overline{B}$. To do so, we need to particularize Eq. (3.107) for each susceptibility.

3.2.1 Susceptibility χ^{+-}

We begin by substituting $\sigma =\uparrow$ and $\sigma' =\downarrow$ in Eq. (3.107) to obtain

$$
\begin{aligned}
\sum_{\substack{mn\\ \alpha\beta}} \sum_{\lambda} \Big\{ & U^{i}_{\lambda\alpha\beta\mu} \langle c^{\dagger}_{i\lambda\uparrow} c_{j\nu\downarrow} \rangle \delta_{mi}\delta_{ni} \overset{\uparrow-}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} + U^{i}_{\lambda\alpha\beta\mu} \langle c^{\dagger}_{i\lambda\downarrow} c_{j\nu\downarrow} \rangle \delta_{mi}\delta_{ni} \overset{+-}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} \\
& - U^{j}_{\nu\alpha\beta\lambda} \langle c^{\dagger}_{i\mu\uparrow} c_{j\lambda\uparrow} \rangle \delta_{mj}\delta_{nj} \overset{+-}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} - U^{j}_{\nu\alpha\beta\lambda} \langle c^{\dagger}_{i\mu\uparrow} c_{j\lambda\downarrow} \rangle \delta_{mj}\delta_{nj} \overset{\downarrow-}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} \\
& + \sum_{s} \Big[U^{j}_{\nu\alpha\lambda\beta} \langle c^{\dagger}_{i\mu\uparrow} c_{j\lambda\downarrow} \rangle \delta_{mj}\delta_{nj} - U^{i}_{\alpha\lambda\beta\mu} \langle c^{\dagger}_{i\lambda\uparrow} c_{j\nu\downarrow} \rangle \delta_{mi}\delta_{ni} \Big] \overset{s-}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} \Big\} \ .
\end{aligned}
\tag{3.110}
$$

Now, we can extract

$$
\overline{B}^{11\,\mu\nu\alpha\beta}_{ijmn} = \sum_{\lambda} \Big\{ U^{i}_{\lambda\alpha\beta\mu} \langle c^{\dagger}_{i\lambda\downarrow} c_{j\nu\downarrow} \rangle \delta_{mi}\delta_{ni} - U^{j}_{\nu\alpha\beta\lambda} \langle c^{\dagger}_{i\mu\uparrow} c_{j\lambda\uparrow} \rangle \delta_{mj}\delta_{nj} \Big\} \tag{3.111}
$$

$$
\overline{B}^{12\,\mu\nu\alpha\beta}_{ijmn} = \sum_{\lambda} \Big\{ U^{j}_{\nu\alpha\lambda\beta} \langle c^{\dagger}_{i\mu\uparrow} c_{j\lambda\downarrow} \rangle \delta_{mj}\delta_{nj} + \Big(U^{i}_{\lambda\alpha\beta\mu} - U^{i}_{\alpha\lambda\beta\mu} \Big) \langle c^{\dagger}_{i\lambda\uparrow} c_{j\nu\downarrow} \rangle \delta_{mi}\delta_{ni} \Big\} \tag{3.112}
$$

$$
\overline{B}^{13\,\mu\nu\alpha\beta}_{ijmn} = \sum_{\lambda} \Big\{ \Big(U^{j}_{\nu\alpha\lambda\beta} - U^{j}_{\nu\alpha\beta\lambda} \Big) \langle c^{\dagger}_{i\mu\uparrow} c_{j\lambda\downarrow} \rangle \delta_{mj}\delta_{nj} - U^{i}_{\alpha\lambda\beta\mu} \langle c^{\dagger}_{i\lambda\uparrow} c_{j\nu\downarrow} \rangle \delta_{mi}\delta_{ni} \Big\} \tag{3.113}
$$

$$
\overline{B}^{14\,\mu\nu\alpha\beta}_{ijmn} = 0 \ . \tag{3.114}
$$

3.2.2 Susceptibility $\chi^{\uparrow-}$

The second equation of motion is obtained by substituting $\sigma = \sigma' =\uparrow$. We have

$$
\begin{aligned}
\sum_{\substack{mn\\ \alpha\beta}} \sum_{\lambda} \Big\{ & U^{i}_{\lambda\alpha\beta\mu} \langle c^{\dagger}_{i\lambda\uparrow} c_{j\nu\uparrow} \rangle \delta_{mi}\delta_{ni} \overset{\uparrow-}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} + U^{i}_{\lambda\alpha\beta\mu} \langle c^{\dagger}_{i\lambda\downarrow} c_{j\nu\uparrow} \rangle \delta_{mi}\delta_{ni} \overset{+-}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} \\
& - U^{j}_{\nu\alpha\beta\lambda} \langle c^{\dagger}_{i\mu\uparrow} c_{j\lambda\uparrow} \rangle \delta_{mj}\delta_{nj} \overset{\uparrow-}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} - U^{j}_{\nu\alpha\beta\lambda} \langle c^{\dagger}_{i\mu\uparrow} c_{j\lambda\downarrow} \rangle \delta_{mj}\delta_{nj} \overset{--}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} \\
& + \sum_{s} \Big[U^{j}_{\nu\alpha\lambda\beta} \langle c^{\dagger}_{i\mu\uparrow} c_{j\lambda\uparrow} \rangle \delta_{mj}\delta_{nj} - U^{i}_{\alpha\lambda\beta\mu} \langle c^{\dagger}_{i\lambda\uparrow} c_{j\nu\uparrow} \rangle \delta_{mi}\delta_{ni} \Big] \overset{s-}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} \Big\} \ ,
\end{aligned}
\tag{3.115}
$$

from which we obtain

$$
\overline{B}^{21\,\mu\nu\alpha\beta}_{ijmn} = \sum_{\lambda} U^{i}_{\lambda\alpha\beta\mu} \langle c^{\dagger}_{i\lambda\downarrow} c_{j\nu\uparrow} \rangle \delta_{mi}\delta_{ni} \tag{3.116}
$$

$$
\overline{B}^{22\,\mu\nu\alpha\beta}_{ijmn} = \sum_{\lambda} \Big\{ \Big(U^{i}_{\lambda\alpha\beta\mu} - U^{i}_{\alpha\lambda\beta\mu} \Big) \langle c^{\dagger}_{i\lambda\uparrow} c_{j\nu\uparrow} \rangle \delta_{mi}\delta_{ni} + \Big(U^{j}_{\nu\alpha\lambda\beta} - U^{j}_{\nu\alpha\beta\lambda} \Big) \langle c^{\dagger}_{i\mu\uparrow} c_{j\lambda\uparrow} \rangle \delta_{mj}\delta_{nj} \Big\} \tag{3.117}
$$

$$
\overline{B}^{23\,\mu\nu\alpha\beta}_{ijmn} = \sum_{\lambda} \Big\{ U^{j}_{\nu\alpha\lambda\beta} \langle c^{\dagger}_{i\mu\uparrow} c_{j\lambda\uparrow} \rangle \delta_{mj}\delta_{nj} - U^{i}_{\alpha\lambda\beta\mu} \langle c^{\dagger}_{i\lambda\uparrow} c_{j\nu\uparrow} \rangle \delta_{mi}\delta_{ni} \Big\} \tag{3.118}
$$

$$
\overline{B}^{24\,\mu\nu\alpha\beta}_{ijmn} = - \sum_{\lambda} U^{j}_{\nu\alpha\beta\lambda} \langle c^{\dagger}_{i\mu\uparrow} c_{j\lambda\downarrow} \rangle \delta_{mj}\delta_{nj} \ . \tag{3.119}
$$

3.2.3 Susceptibility $\chi^{\downarrow -}$

For $\sigma = \sigma' = \downarrow$, we obtain

$$\begin{aligned}\sum_{\substack{mn\\ \alpha\beta}} \sum_{\lambda} \Big\{ & U^i_{\lambda\alpha\beta\mu} \langle c^\dagger_{i\lambda\uparrow} c_{j\nu\downarrow} \rangle \delta_{mi}\delta_{ni} \overset{--}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} + U^i_{\lambda\alpha\beta\mu} \langle c^\dagger_{i\lambda\downarrow} c_{j\nu\downarrow} \rangle \delta_{mi}\delta_{ni} \overset{\downarrow -}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} \\ & - U^j_{\nu\alpha\beta\lambda} \langle c^\dagger_{i\mu\downarrow} c_{j\lambda\uparrow} \rangle \delta_{mj}\delta_{nj} \overset{+-}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} - U^j_{\nu\alpha\beta\lambda} \langle c^\dagger_{i\mu\downarrow} c_{j\lambda\downarrow} \rangle \delta_{mj}\delta_{nj} \overset{\downarrow -}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} \\ & + \sum_s \Big[U^j_{\nu\alpha\lambda\beta} \langle c^\dagger_{i\mu\downarrow} c_{j\lambda\downarrow} \rangle \delta_{mj}\delta_{nj} - U^i_{\alpha\lambda\beta\mu} \langle c^\dagger_{i\lambda\downarrow} c_{j\nu\downarrow} \rangle \delta_{mi}\delta_{ni} \Big] \overset{s-}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} \Big\} \ . \end{aligned} \tag{3.120}$$

The elements of $\overline{B}$ are

$$\overline{B}{}^{31\,\mu\nu\alpha\beta}_{ijmn} = -\sum_\lambda U^j_{\nu\alpha\beta\lambda} \langle c^\dagger_{i\mu\downarrow} c_{j\lambda\uparrow} \rangle \delta_{mj}\delta_{nj} \tag{3.121}$$

$$\overline{B}{}^{32\,\mu\nu\alpha\beta}_{ijmn} = \sum_\lambda \Big\{ U^j_{\nu\alpha\lambda\beta} \langle c^\dagger_{i\mu\downarrow} c_{j\lambda\downarrow} \rangle \delta_{mj}\delta_{nj} - U^i_{\alpha\lambda\beta\mu} \langle c^\dagger_{i\lambda\downarrow} c_{j\nu\downarrow} \rangle \delta_{mi}\delta_{ni} \Big\} \tag{3.122}$$

$$\overline{B}{}^{33\,\mu\nu\alpha\beta}_{ijmn} = \sum_\lambda \Big\{ \Big(U^i_{\lambda\alpha\beta\mu} - U^i_{\alpha\lambda\beta\mu} \Big) \langle c^\dagger_{i\lambda\downarrow} c_{j\nu\downarrow} \rangle \delta_{mi}\delta_{ni} + \Big(U^j_{\nu\alpha\lambda\beta} - U^j_{\nu\alpha\beta\lambda} \Big) \langle c^\dagger_{i\mu\downarrow} c_{j\lambda\downarrow} \rangle \delta_{mj}\delta_{nj} \Big\} \tag{3.123}$$

$$\overline{B}{}^{34\,\mu\nu\alpha\beta}_{ijmn} = \sum_\lambda U^i_{\lambda\alpha\beta\mu} \langle c^\dagger_{i\lambda\uparrow} c_{j\nu\downarrow} \rangle \delta_{mi}\delta_{ni} \ . \tag{3.124}$$

3.2.4 Susceptibility χ^{--}

And last, we substitute $\sigma = \downarrow$ and $\sigma' = \uparrow$:

$$\begin{aligned}\sum_{\substack{mn\\ \alpha\beta}} \sum_{\lambda} \Big\{ & U^i_{\lambda\alpha\beta\mu} \langle c^\dagger_{i\lambda\uparrow} c_{j\nu\uparrow} \rangle \delta_{mi}\delta_{ni} \overset{--}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} + U^i_{\lambda\alpha\beta\mu} \langle c^\dagger_{i\lambda\downarrow} c_{j\nu\uparrow} \rangle \delta_{mi}\delta_{ni} \overset{\downarrow -}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} \\ & - U^j_{\nu\alpha\beta\lambda} \langle c^\dagger_{i\mu\downarrow} c_{j\lambda\uparrow} \rangle \delta_{mj}\delta_{nj} \overset{\uparrow -}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} - U^j_{\nu\alpha\beta\lambda} \langle c^\dagger_{i\mu\downarrow} c_{j\lambda\downarrow} \rangle \delta_{mj}\delta_{nj} \overset{--}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} \\ & + \sum_s \Big[U^j_{\nu\alpha\lambda\beta} \langle c^\dagger_{i\mu\downarrow} c_{j\lambda\uparrow} \rangle \delta_{mj}\delta_{nj} - U^i_{\alpha\lambda\beta\mu} \langle c^\dagger_{i\lambda\downarrow} c_{j\nu\uparrow} \rangle \delta_{mi}\delta_{ni} \Big] \overset{s-}{\chi}{}^{\alpha\beta\gamma\xi}_{mnkl} \Big\} \ . \end{aligned} \tag{3.125}$$

The last elements of $\overline{B}$ are

$$\overline{B}{}^{41\,\mu\nu\alpha\beta}_{ijmn} = 0 \tag{3.126}$$

$$\overline{B}{}^{42\,\mu\nu\alpha\beta}_{ijmn} = \sum_\lambda \Big\{ \Big(U^j_{\nu\alpha\lambda\beta} - U^j_{\nu\alpha\beta\lambda} \Big) \langle c^\dagger_{i\mu\downarrow} c_{j\lambda\uparrow} \rangle \delta_{mj}\delta_{nj} - U^i_{\alpha\lambda\beta\mu} \langle c^\dagger_{i\lambda\downarrow} c_{j\nu\uparrow} \rangle \delta_{mi}\delta_{ni} \Big\} \tag{3.127}$$

$$\overline{B}{}^{43\,\mu\nu\alpha\beta}_{ijmn} = \sum_\lambda \Big\{ U^j_{\nu\alpha\lambda\beta} \langle c^\dagger_{i\mu\downarrow} c_{j\lambda\uparrow} \rangle \delta_{mj}\delta_{nj} + \Big(U^i_{\lambda\alpha\beta\mu} - U^i_{\alpha\lambda\beta\mu} \Big) \langle c^\dagger_{i\lambda\downarrow} c_{j\nu\uparrow} \rangle \delta_{mi}\delta_{ni} \Big\} \tag{3.128}$$

$$\overline{B}{}^{44\,\mu\nu\alpha\beta}_{ijmn} = \sum_\lambda \Big\{ U^i_{\lambda\alpha\beta\mu} \langle c^\dagger_{i\lambda\uparrow} c_{j\nu\uparrow} \rangle \delta_{mi}\delta_{ni} - U^j_{\nu\alpha\beta\lambda} \langle c^\dagger_{i\mu\downarrow} c_{j\lambda\downarrow} \rangle \delta_{mj}\delta_{nj} \Big\} \ . \tag{3.129}$$

3.2.5 Λ matrix

With all the elements of Γ and $\overline{B}$ in hand, we can now proceed to calculate the matrix $\Lambda = \Gamma\overline{B}$. The calculation is analogous the one we set fourth to obtain χ^0. The first element is given by

$$\Lambda^{11\,\mu\nu\gamma\xi}_{ijkl} = \left[\Gamma^{11}\overline{B}^{11} + \Gamma^{12}\overline{B}^{21} + \Gamma^{13}\overline{B}^{31}\right]^{\mu\nu\gamma\xi}_{ijkl} \quad , \tag{3.130}$$

since $\overline{B}^{41} = 0$. These terms are given by

$$\begin{aligned}\sum_{\substack{mn\\ \alpha\beta}} \Gamma^{11\,\mu\nu\alpha\beta}_{ijmn}\overline{B}^{11\,\alpha\beta\gamma\xi}_{mnkl} =& \hbar\int d\omega' f(\omega') \sum_{\substack{mn\\ \alpha\beta}} \sum_{pp'p''} \sum_{\lambda} U^{m}_{\lambda\gamma\xi\alpha}\delta_{km}\delta_{lm}\delta(\hbar\omega' - E_{p''}) \\ &\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p''}|m\lambda\downarrow\rangle\langle\phi_{p'}|n\beta\downarrow\rangle\langle n\beta\downarrow|\phi_{p''}\rangle}{\hbar\omega - E_{p'} + E_p} \\ &- \hbar\int d\omega' f(\omega') \sum_{\substack{mn\\ \alpha\beta}} \sum_{pp'p''} \sum_{\lambda} U^{n}_{\beta\gamma\xi\lambda}\delta_{kn}\delta_{ln}\delta(\hbar\omega' - E_{p''}) \\ &\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle\phi_{p''}|m\alpha\uparrow\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle n\lambda\uparrow|\phi_{p''}\rangle\langle\phi_{p'}|n\beta\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \ ,\end{aligned} \tag{3.131}$$

$$\begin{aligned}\sum_{\substack{mn\\ \alpha\beta}} \Gamma^{12\,\mu\nu\alpha\beta}_{ijmn}\overline{B}^{21\,\alpha\beta\gamma\xi}_{mnkl} =& \hbar\int d\omega' f(\omega') \sum_{\substack{mn\\ \alpha\beta}} \sum_{pp'p''} \sum_{\lambda} U^{m}_{\lambda\gamma\xi\alpha}\delta_{km}\delta_{lm}\delta(\hbar\omega' - E_{p''}) \\ &\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p''}|m\lambda\downarrow\rangle\langle\phi_{p'}|n\beta\uparrow\rangle\langle n\beta\uparrow|\phi_{p''}\rangle}{\hbar\omega - E_{p'} + E_p} \ , \text{ and}\end{aligned} \tag{3.132}$$

$$\begin{aligned}\sum_{\substack{mn\\ \alpha\beta}} \Gamma^{13\,\mu\nu\alpha\beta}_{ijmn}\overline{B}^{31\,\alpha\beta\gamma\xi}_{mnkl} =& -\hbar\int d\omega' f(\omega') \sum_{\substack{mn\\ \alpha\beta}} \sum_{pp'p''} \sum_{\lambda} U^{n}_{\beta\gamma\xi\lambda}\delta_{kn}\delta_{ln}\delta(\hbar\omega' - E_{p''}) \\ &\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle\phi_{p''}|m\alpha\downarrow\rangle\langle m\alpha\downarrow|\phi_p\rangle\langle n\lambda\uparrow|\phi_{p''}\rangle\langle\phi_{p'}|n\beta\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \ .\end{aligned} \tag{3.133}$$

Summing all these terms and using the completeness relation, we obtain

$$\begin{aligned}\Lambda^{11\,\mu\nu\gamma\xi}_{ijkl} =& \sum_{\substack{m\\ \alpha\lambda}} U^{m}_{\lambda\gamma\xi\alpha}\delta_{km}\delta_{lm}\ \hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega'-E_{p'}) \\ &\times\frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p'}|m\lambda\downarrow\rangle}{\hbar\omega-E_{p'}+E_p} \\ &-\sum_{\substack{n\\ \beta\lambda}} U^{n}_{\beta\gamma\xi\lambda}\delta_{kn}\delta_{ln}\ \hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega'-E_{p}) \\ &\times\frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle n\lambda\uparrow|\phi_p\rangle\langle\phi_{p'}|n\beta\downarrow\rangle}{\hbar\omega-E_{p'}+E_p}\ .\end{aligned} \tag{3.134}$$

If we make the changes of indices $n\to m$, $\lambda\to\alpha$ and $\beta\to\lambda$ on the second term, we end up with

$$\begin{aligned}\Lambda^{11\,\mu\nu\gamma\xi}_{ijkl} =& \sum_{\substack{m\\ \alpha\lambda}} U^{m}_{\lambda\gamma\xi\alpha}\delta_{km}\delta_{lm}\ \hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega'-E_{p'}) \\ &\times\frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p'}|m\lambda\downarrow\rangle}{\hbar\omega-E_{p'}+E_p} \\ &-\sum_{\substack{m\\ \alpha\lambda}} U^{m}_{\lambda\gamma\xi\alpha}\delta_{km}\delta_{lm}\ \hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega'-E_{p}) \\ &\times\frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p'}|m\lambda\downarrow\rangle}{\hbar\omega-E_{p'}+E_p} \\ =& \sum_{\substack{m\\ \alpha\lambda}} U^{m}_{\lambda\gamma\xi\alpha}\delta_{km}\delta_{lm}\ \Bigg\{\hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega'-E_{p'}) \\ &\times\frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle l\alpha\uparrow|\phi_p\rangle\langle\phi_{p'}|k\lambda\downarrow\rangle}{\hbar\omega-E_{p'}+E_p} \\ &-\hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega'-E_{p}) \\ &\times\frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle l\alpha\uparrow|\phi_p\rangle\langle\phi_{p'}|k\lambda\downarrow\rangle}{\hbar\omega-E_{p'}+E_p}\Bigg\}\ .\end{aligned} \tag{3.135}$$

Comparing this result with Eq. (3.72), we note that

$$\begin{aligned}\Lambda^{11\,\mu\nu\gamma\xi}_{ijkl} &= -\sum_{\substack{m\\ \alpha\lambda}} U^{m}_{\lambda\gamma\xi\alpha}\delta_{km}\delta_{lm}\chi^{+-\,\mu\nu\lambda\alpha}_{(0)\,ijkl}(\omega) \\ &= -\sum_{\substack{m\\ \alpha\beta}} \chi^{+-\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega)U^{m}_{\alpha\gamma\xi\beta}\delta_{km}\delta_{lm}\ .\end{aligned} \tag{3.136}$$

The elements Λ^{21}, Λ^{31} and Λ^{41} are calculated the same way, and it's easy to see that the only difference will be the two first indices of spins. So we have

$$\Lambda^{21}{}_{ijkl}^{\mu\nu\gamma\xi} = -\sum_{\substack{m \\ \alpha\beta}} \chi^{\uparrow - \,\mu\nu\alpha\beta}_{(0)ijmm}(\omega) U^m_{\alpha\gamma\xi\beta} \delta_{km}\delta_{lm} \ , \tag{3.137}$$

$$\Lambda^{31}{}_{ijkl}^{\mu\nu\gamma\xi} = -\sum_{\substack{m \\ \alpha\beta}} \chi^{\downarrow - \,\mu\nu\alpha\beta}_{(0)ijmm}(\omega) U^m_{\alpha\gamma\xi\beta} \delta_{km}\delta_{lm} \ , \tag{3.138}$$

and

$$\Lambda^{41}{}_{ijkl}^{\mu\nu\gamma\xi} = -\sum_{\substack{m \\ \alpha\beta}} \chi^{- - \,\mu\nu\alpha\beta}_{(0)ijmm}(\omega) U^m_{\alpha\gamma\xi\beta} \delta_{km}\delta_{lm} \ . \tag{3.139}$$

The first element of the second column is

$$\Lambda^{12}{}_{ijkl}^{\mu\nu\gamma\xi} = \left[\Gamma^{11}\overline{B}^{12} + \Gamma^{12}\overline{B}^{22} + \Gamma^{13}\overline{B}^{32} + \Gamma^{14}\overline{B}^{42}\right]^{\mu\nu\gamma\xi}_{ijkl} \ . \tag{3.140}$$

The first product is

$$\begin{aligned}
\sum_{\substack{mn \\ \alpha\beta}} \Gamma^{11}{}_{ijmn}^{\mu\nu\alpha\beta} \overline{B}^{12}{}_{mnkl}^{\alpha\beta\gamma\xi} =& \hbar \int d\omega' f(\omega') \sum_{\substack{mn \\ \alpha\beta}} \sum_{pp'p''} \sum_{\lambda} \left(U^m_{\lambda\gamma\xi\alpha} - U^m_{\gamma\lambda\xi\alpha}\right) \delta_{km}\delta_{lm} \delta(\hbar\omega' - E_{p''}) \\
& \times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p''}|m\lambda\uparrow\rangle\langle\phi_{p'}|n\beta\downarrow\rangle\langle n\beta\downarrow|\phi_{p''}\rangle}{\hbar\omega - E_{p'} + E_p} \\
& + \hbar \int d\omega' f(\omega') \sum_{\substack{mn \\ \alpha\beta}} \sum_{pp'p''} \sum_{\lambda} U^n_{\beta\gamma\lambda\xi} \delta_{kn}\delta_{ln} \delta(\hbar\omega' - E_{p''}) \\
& \times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle\phi_{p''}|m\alpha\uparrow\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle n\lambda\downarrow|\phi_{p''}\rangle\langle\phi_{p'}|n\beta\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \ .
\end{aligned} \tag{3.141}$$

The second is given by

$$\begin{aligned}
\sum_{\substack{mn \\ \alpha\beta}} \Gamma^{12}{}_{ijmn}^{\mu\nu\alpha\beta} \overline{B}^{22}{}_{mnkl}^{\alpha\beta\gamma\xi} =& \hbar \int d\omega' f(\omega') \sum_{\substack{mn \\ \alpha\beta}} \sum_{pp'p''} \sum_{\lambda} \left(U^m_{\lambda\gamma\xi\alpha} - U^m_{\gamma\lambda\xi\alpha}\right) \delta_{km}\delta_{lm} \delta(\hbar\omega' - E_{p''}) \\
& \times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p''}|m\lambda\uparrow\rangle\langle\phi_{p'}|n\beta\uparrow\rangle\langle n\beta\uparrow|\phi_{p''}\rangle}{\hbar\omega - E_{p'} + E_p} \\
& + \hbar \int d\omega' f(\omega') \sum_{\substack{mn \\ \alpha\beta}} \sum_{pp'p''} \sum_{\lambda} \left(U^n_{\beta\gamma\lambda\xi} - U^n_{\beta\gamma\xi\lambda}\right) \delta_{kn}\delta_{ln} \delta(\hbar\omega' - E_{p''}) \\
& \times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle\phi_{p''}|m\alpha\uparrow\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle n\lambda\uparrow|\phi_{p''}\rangle\langle\phi_{p'}|n\beta\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \ .
\end{aligned} \tag{3.142}$$

The third and fourth terms are

$$\begin{aligned}
\sum_{\substack{mn\\ \alpha\beta}} \Gamma^{13\,\mu\nu\alpha\beta}_{ijmn} \overline{B}^{32\,\alpha\beta\gamma\xi}_{mnkl} =& -\hbar \int d\omega' f(\omega') \sum_{\substack{mn\\ \alpha\beta}} \sum_{pp'p''} \sum_{\lambda} U^{m}_{\gamma\lambda\xi\alpha} \delta_{km} \delta_{lm} \delta(\hbar\omega' - E_{p''}) \\
&\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\downarrow|\phi_p\rangle\langle\phi_{p''}|m\lambda\downarrow\rangle\langle\phi_{p'}|n\beta\downarrow\rangle\langle n\beta\downarrow|\phi_{p''}\rangle}{\hbar\omega - E_{p'} + E_p} \\
&+ \hbar \int d\omega' f(\omega') \sum_{\substack{mn\\ \alpha\beta}} \sum_{pp'p''} \sum_{\lambda} U^{n}_{\beta\gamma\lambda\xi} \delta_{kn} \delta_{ln} \delta(\hbar\omega' - E_{p''}) \\
&\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle\phi_{p''}|m\alpha\downarrow\rangle\langle m\alpha\downarrow|\phi_p\rangle\langle n\lambda\downarrow|\phi_{p''}\rangle\langle\phi_{p'}|n\beta\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \ ,
\end{aligned} \tag{3.143}$$

$$\begin{aligned}
\sum_{\substack{mn\\ \alpha\beta}} \Gamma^{14\,\mu\nu\alpha\beta}_{ijmn} \overline{B}^{42\,\alpha\beta\gamma\xi}_{mnkl} =& -\hbar \int d\omega' f(\omega') \sum_{\substack{mn\\ \alpha\beta}} \sum_{pp'p''} \sum_{\lambda} U^{m}_{\gamma\lambda\xi\alpha} \delta_{km} \delta_{lm} \delta(\hbar\omega' - E_{p''}) \\
&\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\downarrow|\phi_p\rangle\langle\phi_{p''}|m\lambda\downarrow\rangle\langle\phi_{p'}|n\beta\uparrow\rangle\langle n\beta\uparrow|\phi_{p''}\rangle}{\hbar\omega - E_{p'} + E_p} \\
&+ \hbar \int d\omega' f(\omega') \sum_{\substack{mn\\ \alpha\beta}} \sum_{pp'p''} \sum_{\lambda} \left(U^{n}_{\beta\gamma\lambda\xi} - U^{n}_{\beta\gamma\xi\lambda}\right) \delta_{kn} \delta_{ln} \delta(\hbar\omega' - E_{p''}) \\
&\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle\phi_{p''}|m\alpha\downarrow\rangle\langle m\alpha\downarrow|\phi_p\rangle\langle n\lambda\uparrow|\phi_{p''}\rangle\langle\phi_{p'}|n\beta\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \ .
\end{aligned} \tag{3.144}$$

Summing all the terms and taking into account the completeness of the states $|m\alpha\sigma\rangle$, we obtain

$$
\begin{aligned}
\overset{12}{\Lambda}{}^{\mu\nu\gamma\xi}_{ijkl} =& \hbar \int d\omega' f(\omega') \sum_{\substack{m\\ \alpha}} \sum_{pp'} \sum_{\lambda} \left(U^m_{\lambda\gamma\xi\alpha} - U^m_{\gamma\lambda\xi\alpha} \right) \delta_{km}\delta_{lm}\delta(\hbar\omega' - E_{p'}) \\
& \times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p'}|m\lambda\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \\
& + \hbar \int d\omega' f(\omega') \sum_{\substack{n\\ \beta}} \sum_{pp'} \sum_{\lambda} \left(U^n_{\beta\gamma\lambda\xi} - U^n_{\beta\gamma\xi\lambda} \right) \delta_{kn}\delta_{ln}\delta(\hbar\omega' - E_p) \\
& \times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle n\lambda\uparrow|\phi_p\rangle\langle\phi_{p'}|n\beta\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \\
& - \hbar \int d\omega' f(\omega') \sum_{\substack{m\\ \alpha}} \sum_{pp'} \sum_{\lambda} U^m_{\gamma\lambda\xi\alpha} \delta_{km}\delta_{lm}\delta(\hbar\omega' - E_{p'}) \\
& \times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\downarrow|\phi_p\rangle\langle\phi_{p'}|m\lambda\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \\
& + \hbar \int d\omega' f(\omega') \sum_{\substack{n\\ \beta}} \sum_{pp'} \sum_{\lambda} U^n_{\beta\gamma\lambda\xi} \delta_{kn}\delta_{ln}\delta(\hbar\omega' - E_p) \\
& \times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle n\lambda\downarrow|\phi_p\rangle\langle\phi_{p'}|n\beta\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \, .
\end{aligned}
\tag{3.145}
$$

Changing the indices $n \to m$, $\lambda \to \alpha$ and $\beta \to \lambda$ on the second and fourth terms, we have

$$
\begin{aligned}
\overset{12}{\Lambda}{}^{\mu\nu\gamma\xi}_{ijkl} =& \hbar \int d\omega' f(\omega') \sum_{\substack{m\\ \alpha\lambda}} \sum_{pp'} \left(U^m_{\lambda\gamma\xi\alpha} - U^m_{\gamma\lambda\xi\alpha} \right) \delta_{km}\delta_{lm}\delta(\hbar\omega' - E_{p'}) \\
& \times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p'}|m\lambda\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \\
& + \hbar \int d\omega' f(\omega') \sum_{\substack{m\\ \alpha\lambda}} \sum_{pp'} \left(U^m_{\lambda\gamma\alpha\xi} - U^m_{\lambda\gamma\xi\alpha} \right) \delta_{km}\delta_{lm}\delta(\hbar\omega' - E_p) \\
& \times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p'}|m\lambda\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \\
& - \hbar \int d\omega' f(\omega') \sum_{\substack{m\\ \alpha\lambda}} \sum_{pp'} U^m_{\gamma\lambda\xi\alpha} \delta_{km}\delta_{lm}\delta(\hbar\omega' - E_{p'}) \\
& \times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\downarrow|\phi_p\rangle\langle\phi_{p'}|m\lambda\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \\
& + \hbar \int d\omega' f(\omega') \sum_{\substack{m\\ \alpha\lambda}} \sum_{pp'} U^m_{\lambda\gamma\alpha\xi} \delta_{km}\delta_{lm}\delta(\hbar\omega' - E_p) \\
& \times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\downarrow|\phi_p\rangle\langle\phi_{p'}|m\lambda\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \, .
\end{aligned}
\tag{3.146}
$$

Using the general symmetry of U, we can merge the two first terms, as well as the two last terms.

$$
\begin{aligned}
\Lambda^{12\,\mu\nu\gamma\xi}_{ijkl} = &\sum_{\substack{m\\ \alpha\lambda}} \Big(U^m_{\lambda\gamma\xi\alpha} - U^m_{\gamma\lambda\xi\alpha}\Big)\delta_{km}\delta_{lm}\Bigg\{\hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega' - E_{p'})\\
&\times\frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p'}|m\lambda\uparrow\rangle}{\hbar\omega - E_{p'} + E_p}\\
&-\hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega' - E_p)\\
&\times\frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p'}|m\lambda\uparrow\rangle}{\hbar\omega - E_{p'} + E_p}\Bigg\}\\
&-\sum_{\substack{m\\ \alpha\lambda}} U^m_{\gamma\lambda\xi\alpha}\delta_{km}\delta_{lm}\Bigg\{\hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega' - E_{p'})\\
&\times\frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\downarrow|\phi_p\rangle\langle\phi_{p'}|m\lambda\downarrow\rangle}{\hbar\omega - E_{p'} + E_p}\\
&-\hbar\int d\omega' f(\omega')\sum_{pp'}\delta(\hbar\omega' - E_p)\\
&\times\frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\downarrow|\phi_p\rangle\langle\phi_{p'}|m\lambda\downarrow\rangle}{\hbar\omega - E_{p'} + E_p}\Bigg\}\ .
\end{aligned}
\tag{3.147}
$$

Comparing the result inside curly brackets with the definitions of Hartree-Fock susceptibilities that we obtained in Eq. (3.72) and 3.79, for example, we can write the equation above as

$$
\begin{aligned}
\Lambda^{12\,\mu\nu\gamma\xi}_{ijkl} &= -\sum_{\substack{m\\ \alpha\lambda}}\Big(U^m_{\lambda\gamma\xi\alpha} - U^m_{\gamma\lambda\xi\alpha}\Big)\delta_{km}\delta_{lm}\chi^{+\uparrow\,\mu\nu\lambda\alpha}_{(0)\,ijmm}(\omega) + \sum_{\substack{m\\ \alpha\lambda}} U^m_{\gamma\lambda\xi\alpha}\delta_{km}\delta_{lm}\chi^{+\downarrow\,\mu\nu\lambda\alpha}_{(0)\,ijmm}(\omega)\\
&= \sum_{\substack{m\\ \alpha\beta}}\left\{\chi^{+\uparrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega)\Big(U^m_{\gamma\alpha\xi\beta} - U^m_{\alpha\gamma\xi\beta}\Big)\delta_{km}\delta_{lm} + \chi^{+\downarrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega)U^m_{\gamma\alpha\xi\beta}\delta_{km}\delta_{lm}\right\}\ .
\end{aligned}
\tag{3.148}
$$

As we have noticed before, the elements Λ^{22}, Λ^{32} and Λ^{42} can be obtained from the result above since only the spin indexes of the product $\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle$ changes. Thus, we have

$$
\Lambda^{22\,\mu\nu\gamma\xi}_{ijkl} = \sum_{\substack{m\\ \alpha\beta}}\left\{\chi^{\uparrow\uparrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega)\Big(U^m_{\gamma\alpha\xi\beta} - U^m_{\alpha\gamma\xi\beta}\Big)\delta_{km}\delta_{lm} + \chi^{\uparrow\downarrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega)U^m_{\gamma\alpha\xi\beta}\delta_{km}\delta_{lm}\right\}\ ,
\tag{3.149}
$$

$$\Lambda^{32\,\mu\nu\gamma\xi}_{ijkl} = \sum_{\substack{m\\ \alpha\beta}} \left\{ \chi^{\downarrow\uparrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega) \left(U^{m}_{\gamma\alpha\xi\beta} - U^{m}_{\alpha\gamma\xi\beta} \right) \delta_{km}\delta_{lm} + \chi^{\downarrow\downarrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega) U^{m}_{\gamma\alpha\xi\beta}\delta_{km}\delta_{lm} \right\} \ , \tag{3.150}$$

and

$$\Lambda^{42\,\mu\nu\gamma\xi}_{ijkl} = \sum_{\substack{m\\ \alpha\beta}} \left\{ \chi^{-\uparrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega) \left(U^{m}_{\gamma\alpha\xi\beta} - U^{m}_{\alpha\gamma\xi\beta} \right) \delta_{km}\delta_{lm} + \chi^{-\downarrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega) U^{m}_{\gamma\alpha\xi\beta}\delta_{km}\delta_{lm} \right\} \ . \tag{3.151}$$

Next, we proceed to calculate the elements of the third column of Λ:

$$\Lambda^{13\,\mu\nu\gamma\xi}_{ijkl} = \left[\Gamma^{11}\overline{B}^{13} + \Gamma^{12}\overline{B}^{23} + \Gamma^{13}\overline{B}^{33} + \Gamma^{14}\overline{B}^{43} \right]^{\mu\nu\gamma\xi}_{ijkl} \ . \tag{3.152}$$

In this case, we can simplify all the work by noting that the elements that we need are related to the ones used to calculate Λ^{12}. By inverting the spin indices, $\overline{B}^{13}$ changes to $\overline{B}^{42}$; $\overline{B}^{23}$ changes to $\overline{B}^{32}$; $\overline{B}^{33}$ changes to $\overline{B}^{22}$; and $\overline{B}^{43}$ changes to $\overline{B}^{12}$. If we call this elements with inverted spins $\tilde{\overline{B}}$, we can write

$$\begin{aligned}
\Lambda^{13\,\mu\nu\gamma\xi}_{ijkl} &= \left[\Gamma^{11}\tilde{\overline{B}}^{42} + \Gamma^{12}\tilde{\overline{B}}^{32} + \Gamma^{13}\tilde{\overline{B}}^{22} + \Gamma^{14}\tilde{\overline{B}}^{12} \right]^{\mu\nu\gamma\xi}_{ijkl} \\
&= \left[\Gamma^{14}\tilde{\overline{B}}^{12} + \Gamma^{13}\tilde{\overline{B}}^{22} + \Gamma^{12}\tilde{\overline{B}}^{32} + \Gamma^{11}\tilde{\overline{B}}^{42} \right]^{\mu\nu\gamma\xi}_{ijkl} \ .
\end{aligned} \tag{3.153}$$

Comparing to Eqs. 3.140 and 3.141, and noting that the difference between Γ^{14} and Γ^{11} and also between Γ^{12} and Γ^{13} are the spin indices of the last two expectation values, we can follow the same calculations as we did before, changing the last four spin indices of each term in Eq. (3.141) and so on. In this manner, it is straight forward to note that the result is

$$\Lambda^{13\,\mu\nu\gamma\xi}_{ijkl} = \sum_{\substack{m\\ \alpha\beta}} \left\{ \chi^{+\downarrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega) \left(U^{m}_{\gamma\alpha\xi\beta} - U^{m}_{\alpha\gamma\xi\beta} \right) \delta_{km}\delta_{lm} + \chi^{+\uparrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega) U^{m}_{\gamma\alpha\xi\beta}\delta_{km}\delta_{lm} \right\} \ . \tag{3.154}$$

The other elements are given by

$$\Lambda^{23\,\mu\nu\gamma\xi}_{ijkl} = \sum_{\substack{m\\ \alpha\beta}} \left\{ \chi^{\uparrow\downarrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega) \left(U^{m}_{\gamma\alpha\xi\beta} - U^{m}_{\alpha\gamma\xi\beta} \right) \delta_{km}\delta_{lm} + \chi^{\uparrow\uparrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega) U^{m}_{\gamma\alpha\xi\beta}\delta_{km}\delta_{lm} \right\} \ , \tag{3.155}$$

$$\Lambda^{33\,\mu\nu\gamma\xi}_{ijkl} = \sum_{\substack{m\\ \alpha\beta}} \left\{ \chi^{\downarrow\downarrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega) \left(U^{m}_{\gamma\alpha\xi\beta} - U^{m}_{\alpha\gamma\xi\beta} \right) \delta_{km}\delta_{lm} + \chi^{\downarrow\uparrow\,\mu\nu\alpha\beta}_{(0)\,ijmm}(\omega) U^{m}_{\gamma\alpha\xi\beta}\delta_{km}\delta_{lm} \right\} \ , \tag{3.156}$$

and

$$\Lambda_{ijkl}^{43\mu\nu\gamma\xi} = \sum_{\substack{m \\ \alpha\beta}} \left\{ \chi_{(0)ijmm}^{-\downarrow\mu\nu\alpha\beta}(\omega)\Big(U^m_{\gamma\alpha\xi\beta} - U^m_{\alpha\gamma\xi\beta}\Big)\delta_{km}\delta_{lm} + \chi_{(0)ijmm}^{-\uparrow\mu\nu\alpha\beta}(\omega)U^m_{\gamma\alpha\xi\beta}\delta_{km}\delta_{lm} \right\} \ . \tag{3.157}$$

Lastly, we can do the same consideration to obtain the last column of Λ through the values obtained for the first column. Therefore

$$\Lambda_{ijkl}^{14\mu\nu\gamma\xi} = -\sum_{\substack{m \\ \alpha\beta}} \chi_{(0)ijmm}^{++\mu\nu\alpha\beta}(\omega)U^m_{\alpha\gamma\xi\beta}\delta_{km}\delta_{lm} \ , \tag{3.158}$$

$$\Lambda_{ijkl}^{24\mu\nu\gamma\xi} = -\sum_{\substack{m \\ \alpha\beta}} \chi_{(0)ijmm}^{\uparrow+\mu\nu\alpha\beta}(\omega)U^m_{\alpha\gamma\xi\beta}\delta_{km}\delta_{lm} \ , \tag{3.159}$$

$$\Lambda_{ijkl}^{34\mu\nu\gamma\xi} = -\sum_{\substack{m \\ \alpha\beta}} \chi_{(0)ijmm}^{\downarrow+\mu\nu\alpha\beta}(\omega)U^m_{\alpha\gamma\xi\beta}\delta_{km}\delta_{lm} \ , \tag{3.160}$$

and

$$\Lambda_{ijkl}^{44\mu\nu\gamma\xi} = -\sum_{\substack{m \\ \alpha\beta}} \chi_{(0)ijmm}^{-+\mu\nu\alpha\beta}(\omega)U^m_{\alpha\gamma\xi\beta}\delta_{km}\delta_{lm} \ . \tag{3.161}$$

In summary, the elements of Λ matrix are

$$\Lambda_{ijkl}^{11\mu\nu\gamma\xi} = -\sum_{\substack{m \\ \alpha\beta}} \chi_{(0)ijmm}^{+-\mu\nu\alpha\beta}(\omega)U^m_{\alpha\gamma\xi\beta}\delta_{km}\delta_{lm} \ , \tag{3.162}$$

$$\Lambda_{ijkl}^{21\mu\nu\gamma\xi} = -\sum_{\substack{m \\ \alpha\beta}} \chi_{(0)ijmm}^{\uparrow-\mu\nu\alpha\beta}(\omega)U^m_{\alpha\gamma\xi\beta}\delta_{km}\delta_{lm} \ , \tag{3.163}$$

$$\Lambda_{ijkl}^{31\mu\nu\gamma\xi} = -\sum_{\substack{m \\ \alpha\beta}} \chi_{(0)ijmm}^{\downarrow-\mu\nu\alpha\beta}(\omega)U^m_{\alpha\gamma\xi\beta}\delta_{km}\delta_{lm} \ , \tag{3.164}$$

$$\Lambda_{ijkl}^{41\mu\nu\gamma\xi} = -\sum_{\substack{m \\ \alpha\beta}} \chi_{(0)ijmm}^{--\mu\nu\alpha\beta}(\omega)U^m_{\alpha\gamma\xi\beta}\delta_{km}\delta_{lm} \ , \tag{3.165}$$

$$\Lambda_{ijkl}^{12\mu\nu\gamma\xi} = \sum_{\substack{m \\ \alpha\beta}} \left\{ \chi_{(0)ijmm}^{+\uparrow\mu\nu\alpha\beta}(\omega)\Big(U^m_{\gamma\alpha\xi\beta} - U^m_{\alpha\gamma\xi\beta}\Big) + \chi_{(0)ijmm}^{+\downarrow\mu\nu\alpha\beta}(\omega)U^m_{\gamma\alpha\xi\beta} \right\} \delta_{km}\delta_{lm} \ , \tag{3.166}$$

$$\Lambda_{ijkl}^{22\,\mu\nu\gamma\xi} = \sum_{\substack{m\\ \alpha\beta}} \left\{ \chi_{(0)ijmm}^{\uparrow\uparrow\,\mu\nu\alpha\beta}(\omega) \Big(U_{\gamma\alpha\xi\beta}^{m} - U_{\alpha\gamma\xi\beta}^{m} \Big) + \chi_{(0)ijmm}^{\uparrow\downarrow\,\mu\nu\alpha\beta}(\omega) U_{\gamma\alpha\xi\beta}^{m} \right\} \delta_{km}\delta_{lm} \ , \tag{3.167}$$

$$\Lambda_{ijkl}^{32\,\mu\nu\gamma\xi} = \sum_{\substack{m\\ \alpha\beta}} \left\{ \chi_{(0)ijmm}^{\downarrow\uparrow\,\mu\nu\alpha\beta}(\omega) \Big(U_{\gamma\alpha\xi\beta}^{m} - U_{\alpha\gamma\xi\beta}^{m} \Big) + \chi_{(0)ijmm}^{\downarrow\downarrow\,\mu\nu\alpha\beta}(\omega) U_{\gamma\alpha\xi\beta}^{m} \right\} \delta_{km}\delta_{lm} \ , \tag{3.168}$$

$$\Lambda_{ijkl}^{42\,\mu\nu\gamma\xi} = \sum_{\substack{m\\ \alpha\beta}} \left\{ \chi_{(0)ijmm}^{-\uparrow\,\mu\nu\alpha\beta}(\omega) \Big(U_{\gamma\alpha\xi\beta}^{m} - U_{\alpha\gamma\xi\beta}^{m} \Big) + \chi_{(0)ijmm}^{-\downarrow\,\mu\nu\alpha\beta}(\omega) U_{\gamma\alpha\xi\beta}^{m} \right\} \delta_{km}\delta_{lm} \ , \tag{3.169}$$

$$\Lambda_{ijkl}^{13\,\mu\nu\gamma\xi} = \sum_{\substack{m\\ \alpha\beta}} \left\{ \chi_{(0)ijmm}^{+\downarrow\,\mu\nu\alpha\beta}(\omega) \Big(U_{\gamma\alpha\xi\beta}^{m} - U_{\alpha\gamma\xi\beta}^{m} \Big) + \chi_{(0)ijmm}^{+\uparrow\,\mu\nu\alpha\beta}(\omega) U_{\gamma\alpha\xi\beta}^{m} \right\} \delta_{km}\delta_{lm} \ , \tag{3.170}$$

$$\Lambda_{ijkl}^{23\,\mu\nu\gamma\xi} = \sum_{\substack{m\\ \alpha\beta}} \left\{ \chi_{(0)ijmm}^{\uparrow\downarrow\,\mu\nu\alpha\beta}(\omega) \Big(U_{\gamma\alpha\xi\beta}^{m} - U_{\alpha\gamma\xi\beta}^{m} \Big) + \chi_{(0)ijmm}^{\uparrow\uparrow\,\mu\nu\alpha\beta}(\omega) U_{\gamma\alpha\xi\beta}^{m} \right\} \delta_{km}\delta_{lm} \ , \tag{3.171}$$

$$\Lambda_{ijkl}^{33\,\mu\nu\gamma\xi} = \sum_{\substack{m\\ \alpha\beta}} \left\{ \chi_{(0)ijmm}^{\downarrow\downarrow\,\mu\nu\alpha\beta}(\omega) \Big(U_{\gamma\alpha\xi\beta}^{m} - U_{\alpha\gamma\xi\beta}^{m} \Big) + \chi_{(0)ijmm}^{\downarrow\uparrow\,\mu\nu\alpha\beta}(\omega) U_{\gamma\alpha\xi\beta}^{m} \right\} \delta_{km}\delta_{lm} \ , \tag{3.172}$$

$$\Lambda_{ijkl}^{43\,\mu\nu\gamma\xi} = \sum_{\substack{m\\ \alpha\beta}} \left\{ \chi_{(0)ijmm}^{-\downarrow\,\mu\nu\alpha\beta}(\omega) \Big(U_{\gamma\alpha\xi\beta}^{m} - U_{\alpha\gamma\xi\beta}^{m} \Big) + \chi_{(0)ijmm}^{-\uparrow\,\mu\nu\alpha\beta}(\omega) U_{\gamma\alpha\xi\beta}^{m} \right\} \delta_{km}\delta_{lm} \ , \tag{3.173}$$

$$\Lambda_{ijkl}^{14\,\mu\nu\gamma\xi} = - \sum_{\substack{m\\ \alpha\beta}} \chi_{(0)ijmm}^{++\,\mu\nu\alpha\beta}(\omega) U_{\alpha\gamma\xi\beta}^{m} \delta_{km}\delta_{lm} \ , \tag{3.174}$$

$$\Lambda_{ijkl}^{24\,\mu\nu\gamma\xi} = - \sum_{\substack{m\\ \alpha\beta}} \chi_{(0)ijmm}^{\uparrow+\,\mu\nu\alpha\beta}(\omega) U_{\alpha\gamma\xi\beta}^{m} \delta_{km}\delta_{lm} \ , \tag{3.175}$$

$$\Lambda_{ijkl}^{34\,\mu\nu\gamma\xi} = - \sum_{\substack{m\\ \alpha\beta}} \chi_{(0)ijmm}^{\downarrow+\,\mu\nu\alpha\beta}(\omega) U_{\alpha\gamma\xi\beta}^{m} \delta_{km}\delta_{lm} \ , \tag{3.176}$$

$$\Lambda_{ijkl}^{44\,\mu\nu\gamma\xi} = - \sum_{\substack{m\\ \alpha\beta}} \chi_{(0)ijmm}^{-+\,\mu\nu\alpha\beta}(\omega) U_{\alpha\gamma\xi\beta}^{m} \delta_{km}\delta_{lm} \ . \tag{3.177}$$

3.3 U matrix and RPA-HF relation

The result above can be summarized as

$$\Lambda_{ijkl}^{\mu\nu\gamma\xi} = -\sum_{\substack{mn\\\alpha\beta}} [\chi_{(0)}]_{ijmn}^{\mu\nu\alpha\beta}(\omega)[U]_{\alpha\beta\gamma\xi}^{mnkl} \ , \tag{3.178}$$

where

$$[\chi_{(0)}]_{ijmn}^{\mu\nu\alpha\beta}(\omega) = \begin{pmatrix} \chi_{(0)ijmn}^{+-\ \mu\nu\alpha\beta}(\omega) & \chi_{(0)ijmn}^{+\uparrow\ \mu\nu\alpha\beta}(\omega) & \chi_{(0)ijmn}^{+\downarrow\ \mu\nu\alpha\beta}(\omega) & \chi_{(0)ijmn}^{++\ \mu\nu\alpha\beta}(\omega) \\ \chi_{(0)ijmn}^{\uparrow-\ \mu\nu\alpha\beta}(\omega) & \chi_{(0)ijmn}^{\uparrow\uparrow\ \mu\nu\alpha\beta}(\omega) & \chi_{(0)ijmn}^{\uparrow\downarrow\ \mu\nu\alpha\beta}(\omega) & \chi_{(0)ijmn}^{\uparrow+\ \mu\nu\alpha\beta}(\omega) \\ \chi_{(0)ijmn}^{\downarrow-\ \mu\nu\alpha\beta}(\omega) & \chi_{(0)ijmn}^{\downarrow\uparrow\ \mu\nu\alpha\beta}(\omega) & \chi_{(0)ijmn}^{\downarrow\downarrow\ \mu\nu\alpha\beta}(\omega) & \chi_{(0)ijmn}^{\downarrow+\ \mu\nu\alpha\beta}(\omega) \\ \chi_{(0)ijmn}^{--\ \mu\nu\alpha\beta}(\omega) & \chi_{(0)ijmn}^{-\uparrow\ \mu\nu\alpha\beta}(\omega) & \chi_{(0)ijmn}^{-\downarrow\ \mu\nu\alpha\beta}(\omega) & \chi_{(0)ijmn}^{-+\ \mu\nu\alpha\beta}(\omega) \end{pmatrix} \tag{3.179}$$

$$[U]_{\alpha\beta\gamma\xi}^{mnkl} = \begin{pmatrix} U_{\alpha\gamma\xi\beta}^{m} & 0 & 0 & 0 \\ 0 & U_{\alpha\gamma\xi\beta}^{m} - U_{\gamma\alpha\xi\beta}^{m} & -U_{\gamma\alpha\xi\beta}^{m} & 0 \\ 0 & -U_{\gamma\alpha\xi\beta}^{m} & U_{\alpha\gamma\xi\beta}^{m} - U_{\gamma\alpha\xi\beta}^{m} & 0 \\ 0 & 0 & 0 & U_{\alpha\gamma\xi\beta}^{m} \end{pmatrix} \delta_{km}\delta_{lm}\delta_{mn} \ . \tag{3.180}$$

With this definition, we have

$$\begin{pmatrix} \delta S^{+} \\ \delta S^{\uparrow} \\ \delta S^{\downarrow} \\ \delta S^{-} \end{pmatrix} = [\chi] \begin{pmatrix} \delta B^{+} \\ \delta B^{\uparrow} \\ \delta B^{\downarrow} \\ \delta B^{-} \end{pmatrix} \ . \tag{3.181}$$

Using Lowde-Windsor parametrization for the matrix $[U]$, i.e., $U_{\gamma\beta\xi\alpha}^{m} = U^{m}\delta_{\gamma\alpha}\delta_{\beta\xi}$, we obtain

$$\begin{aligned} [U]_{\alpha\beta\gamma\xi}^{mnkl} &= \begin{pmatrix} \delta_{\alpha\beta}\delta_{\gamma\xi} & 0 & 0 & 0 \\ 0 & \delta_{\alpha\beta}\delta_{\gamma\xi} - \delta_{\gamma\beta}\delta_{\alpha\xi} & -\delta_{\gamma\beta}\delta_{\alpha\xi} & 0 \\ 0 & -\delta_{\gamma\beta}\delta_{\alpha\xi} & \delta_{\alpha\beta}\delta_{\gamma\xi} - \delta_{\gamma\beta}\delta_{\alpha\xi} & 0 \\ 0 & 0 & 0 & \delta_{\alpha\beta}\delta_{\gamma\xi} \end{pmatrix} U^{m}\delta_{km}\delta_{lm}\delta_{mn} \\ &= \left[\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \delta_{\alpha\beta}\delta_{\gamma\xi} + \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & -1 & -1 & 0 \\ 0 & -1 & -1 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \delta_{\gamma\beta}\delta_{\alpha\xi} \right] U^{m}\delta_{km}\delta_{lm}\delta_{mn} \end{aligned} \tag{3.182}$$

which can be written as

$$[U]_{\alpha\beta\gamma\xi}^{m} = \{[U_1^{m}]\delta_{\alpha\beta}\delta_{\gamma\xi} + [U_2^{m}]\delta_{\gamma\alpha}\delta_{\beta\xi}\} \ , \tag{3.183}$$

where

$$[U_1^m] = \begin{pmatrix} U^m & 0 & 0 & 0 \\ 0 & U^m & 0 & 0 \\ 0 & 0 & U^m & 0 \\ 0 & 0 & 0 & U^m \end{pmatrix} \quad \text{and} \tag{3.184}$$

$$[U_2^m] = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & -U^m & -U^m & 0 \\ 0 & -U^m & -U^m & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \; . \tag{3.185}$$

This way, Eq. (3.109) becomes

$$\chi_{ijkl}^{N\ \mu\nu\gamma\xi}(\omega) = \chi_{(0)ijkl}^{N\ \mu\nu\gamma\xi}(\omega) - [\chi_{(0)}]_{ijmm}^{NP\ \mu\nu\alpha\beta}(\omega)[U]_{\alpha\beta\lambda\rho}^{PL\ m}\chi_{mmkl}^{L\ \lambda\rho\gamma\xi}(\omega) \; , \tag{3.186}$$

where we have omitted all the sums for simplicity.

We can generalize even further noting from Eq. (3.4) that if we want to calculate the equation of motion for the susceptibilities involving $S_{kl}^{\uparrow\gamma\xi}$, $S_{kl}^{\downarrow\gamma\xi}$ and $S_{kl}^{+\gamma\xi}$ instead of $S_{kl}^{-\gamma\xi}$ on the second term inside the commutator, the only matrix in Eqs. 3.42 and 3.108 that will change is $\mathbf{A}$. But even in this case, the relation obtained in Eq. (3.109) doesn't change, since it does not depend on $\mathbf{A}$ directly. The only differences are in the expressions for each Hartree-Fock susceptibility.

In this case, the more general relation between RPA and HF susceptibilities is

$$[\chi]_{ijkl}^{MN\ \mu\nu\gamma\xi}(\omega) = [\chi_{(0)}]_{ijkl}^{MN\ \mu\nu\gamma\xi}(\omega) - [\chi_{(0)}]_{ijmm}^{MP\ \mu\nu\alpha\beta}(\omega)[U]_{\alpha\beta\lambda\rho}^{PL\ m}[\chi]_{mmkl}^{LN\ \lambda\rho\gamma\xi}(\omega) \; , \tag{3.187}$$

which is the generalization of Eq. (3.186).

Substituting Eq. (3.183) in 3.187, we can write

$$\begin{aligned}[\chi]_{ijkl}^{MN\ \mu\nu\gamma\xi}(\omega) =& [\chi_{(0)}]_{ijkl}^{MN\ \mu\nu\gamma\xi}(\omega) - [\chi_{(0)}]_{ijmm}^{MP\ \mu\nu\alpha\beta}(\omega)\left\{[U_1^m]^{PL}\delta_{\alpha\beta}\delta_{\lambda\rho} + [U_2^m]^{PL}\delta_{\lambda\alpha}\delta_{\beta\rho}\right\}[\chi]_{mmkl}^{LN\ \lambda\rho\gamma\xi}(\omega) \\ =& [\chi_{(0)}]_{ijkl}^{MN\ \mu\nu\gamma\xi}(\omega) - [\chi_{(0)}]_{ijmm}^{MP\ \mu\nu\alpha\alpha}(\omega)[U_1^m]^{PL}[\chi]_{mmkl}^{LN\ \lambda\lambda\gamma\xi}(\omega) - [\chi_{(0)}]_{ijmm}^{MP\ \mu\nu\alpha\beta}(\omega)[U_2^m]^{PL}[\chi]_{mmkl}^{LN\ \alpha\beta\gamma\xi}(\omega\end{aligned} \tag{3.188}$$

$$[\chi]_{iijj}^{MN\ \mu\nu\gamma\xi}(\omega) = [\chi]_{(0)iijj}^{MN\ \mu\nu\gamma\xi}(\omega) - [\chi]_{(0)iimm}^{MP\ \mu\nu\alpha\alpha}(\omega)[U_1^m]^{PL}[\chi]_{mmjj}^{LN\ \lambda\lambda\gamma\xi}(\omega) - [\chi]_{(0)iimm}^{MP\ \mu\nu\alpha\beta}(\omega)[U_2^m]^{PL}[\chi]_{mmjj}^{LN\ \alpha\beta\gamma\xi}(\omega) \; , \tag{3.189}$$

To calculate the additional Hartree-Fock susceptibilities, we need to obtain the complete matrix $[A]$. For $S_{kl}^{\uparrow\gamma\xi}$, the correspondent column can be obtained by

$$\begin{aligned}[c_{i\mu\sigma}^\dagger c_{j\nu\sigma'}, S_{kl}^{\uparrow\gamma\xi}] =& c_{i\mu\sigma}^\dagger \underbrace{c_{j\nu\sigma'} c_{k\gamma\uparrow}^\dagger} c_{l\xi\uparrow} - c_{k\gamma\uparrow}^\dagger c_{l\xi\uparrow} c_{i\mu\sigma}^\dagger c_{j\nu\sigma'} \\ =& c_{i\mu\sigma}^\dagger c_{l\xi\uparrow}\delta_{jk}\delta_{\gamma\nu}\delta_{\sigma'\uparrow} - \underbrace{c_{i\mu\sigma}^\dagger c_{k\gamma\uparrow}^\dagger}\underbrace{c_{j\nu\sigma'} c_{l\xi\uparrow}} - c_{k\gamma\uparrow}^\dagger c_{l\xi\uparrow} c_{i\mu\sigma}^\dagger c_{j\nu\sigma'} \\ =& c_{i\mu\sigma}^\dagger c_{l\xi\uparrow}\delta_{jk}\delta_{\gamma\nu}\delta_{\sigma'\uparrow} - c_{k\gamma\uparrow}^\dagger \underbrace{c_{i\mu\sigma}^\dagger c_{l\xi\uparrow}} c_{j\nu\sigma'} - c_{k\gamma\uparrow}^\dagger c_{l\xi\uparrow} c_{i\mu\sigma}^\dagger c_{j\nu\sigma'} \\ =& c_{i\mu\sigma}^\dagger c_{l\xi\uparrow}\delta_{jk}\delta_{\gamma\nu}\delta_{\sigma'\uparrow} - c_{k\gamma\uparrow}^\dagger c_{j\nu\sigma'}\delta_{il}\delta_{\mu\xi}\delta_{\sigma\uparrow} \; .\end{aligned} \tag{3.190}$$

So, each of its elements are

$$A^{12\,\mu\nu\gamma\xi}_{ijkl} = -\langle c^\dagger_{k\gamma\uparrow} c_{j\nu\downarrow}\rangle \delta_{il}\delta_{\mu\xi} \ , \tag{3.191}$$

$$A^{22\,\mu\nu\gamma\xi}_{ijkl} = \langle c^\dagger_{i\mu\uparrow} c_{l\xi\uparrow}\rangle \delta_{jk}\delta_{\gamma\nu} - \langle c^\dagger_{k\gamma\uparrow} c_{j\nu\uparrow}\rangle \delta_{il}\delta_{\mu\xi} \ , \tag{3.192}$$

$$A^{32\,\mu\nu\gamma\xi}_{ijkl} = 0 \ , \tag{3.193}$$

$$A^{42\,\mu\nu\gamma\xi}_{ijkl} = \langle c^\dagger_{i\mu\downarrow} c_{l\xi\uparrow}\rangle \delta_{jk}\delta_{\gamma\nu} \ , \tag{3.194}$$

For $S^{\downarrow\gamma\xi}_{kl}$, we have

$$\begin{aligned}
[c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, S^{\downarrow\gamma\xi}_{kl}] =& c^\dagger_{i\mu\sigma} \underbrace{c_{j\nu\sigma'} c^\dagger_{k\gamma\downarrow}} c_{l\xi\downarrow} - c^\dagger_{k\gamma\downarrow} c_{l\xi\downarrow} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{l\xi\downarrow}\delta_{jk}\delta_{\gamma\nu}\delta_{\sigma'\downarrow} - \underbrace{c^\dagger_{i\mu\sigma} c^\dagger_{k\gamma\downarrow}} \underbrace{c_{j\nu\sigma'} c_{l\xi\downarrow}} - c^\dagger_{k\gamma\downarrow} c_{l\xi\downarrow} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{l\xi\downarrow}\delta_{jk}\delta_{\gamma\nu}\delta_{\sigma'\downarrow} - c^\dagger_{k\gamma\downarrow} \underbrace{c^\dagger_{i\mu\sigma} c_{l\xi\downarrow}} c_{j\nu\sigma'} - c^\dagger_{k\gamma\downarrow} c_{l\xi\downarrow} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{l\xi\downarrow}\delta_{jk}\delta_{\gamma\nu}\delta_{\sigma'\downarrow} - c^\dagger_{k\gamma\downarrow} c_{j\nu\sigma'}\delta_{il}\delta_{\mu\xi}\delta_{\sigma\downarrow} \ .
\end{aligned} \tag{3.195}$$

So, each of its elements are

$$A^{13\,\mu\nu\gamma\xi}_{ijkl} = \langle c^\dagger_{i\mu\uparrow} c_{l\xi\downarrow}\rangle \delta_{jk}\delta_{\gamma\nu} \ , \tag{3.196}$$

$$A^{23\,\mu\nu\gamma\xi}_{ijkl} = 0 \ , \tag{3.197}$$

$$A^{33\,\mu\nu\gamma\xi}_{ijkl} = \langle c^\dagger_{i\mu\downarrow} c_{l\xi\downarrow}\rangle \delta_{jk}\delta_{\gamma\nu} - \langle c^\dagger_{k\gamma\downarrow} c_{j\nu\downarrow}\rangle \delta_{il}\delta_{\mu\xi} \ , \tag{3.198}$$

$$A^{43\,\mu\nu\gamma\xi}_{ijkl} = -\langle c^\dagger_{k\gamma\downarrow} c_{j\nu\uparrow}\rangle \delta_{il}\delta_{\mu\xi} \ , \tag{3.199}$$

And lastly,

$$\begin{aligned}
[c^\dagger_{i\mu\sigma} c_{j\nu\sigma'}, S^{+\gamma\xi}_{kl}] =& c^\dagger_{i\mu\sigma} \underbrace{c_{j\nu\sigma'} c^\dagger_{k\gamma\uparrow}} c_{l\xi\downarrow} - c^\dagger_{k\gamma\uparrow} c_{l\xi\downarrow} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{l\xi\downarrow}\delta_{jk}\delta_{\gamma\nu}\delta_{\sigma'\uparrow} - \underbrace{c^\dagger_{i\mu\sigma} c^\dagger_{k\gamma\uparrow}} \underbrace{c_{j\nu\sigma'} c_{l\xi\downarrow}} - c^\dagger_{k\gamma\uparrow} c_{l\xi\downarrow} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{l\xi\downarrow}\delta_{jk}\delta_{\gamma\nu}\delta_{\sigma'\uparrow} - c^\dagger_{k\gamma\uparrow} \underbrace{c^\dagger_{i\mu\sigma} c_{l\xi\downarrow}} c_{j\nu\sigma'} - c^\dagger_{k\gamma\uparrow} c_{l\xi\downarrow} c^\dagger_{i\mu\sigma} c_{j\nu\sigma'} \\
=& c^\dagger_{i\mu\sigma} c_{l\xi\downarrow}\delta_{jk}\delta_{\gamma\nu}\delta_{\sigma'\uparrow} - c^\dagger_{k\gamma\uparrow} c_{j\nu\sigma'}\delta_{il}\delta_{\mu\xi}\delta_{\sigma\downarrow} \ ,
\end{aligned} \tag{3.200}$$

which gives us the elements

$$A^{14\,\mu\nu\gamma\xi}_{ijkl} = 0 \ , \tag{3.201}$$

$$A^{24\,\mu\nu\gamma\xi}_{ijkl} = \langle c^\dagger_{i\mu\uparrow} c_{l\xi\downarrow}\rangle \delta_{jk}\delta_{\gamma\nu} \ , \tag{3.202}$$

$$A^{34\,\mu\nu\gamma\xi}_{ijkl} = -\langle c^\dagger_{k\gamma\uparrow} c_{j\nu\downarrow}\rangle \delta_{il}\delta_{\mu\xi} \ , \tag{3.203}$$

$$A^{44\,\mu\nu\gamma\xi}_{ijkl} = \langle c^\dagger_{i\mu\downarrow} c_{l\xi\downarrow}\rangle \delta_{jk}\delta_{\gamma\nu} - \langle c^\dagger_{k\gamma\uparrow} c_{j\nu\uparrow}\rangle \delta_{il}\delta_{\mu\xi} \ , \tag{3.204}$$

To obtain the Hartree-Fock susceptibilities, we need ΓA. So, for example, we have the susceptibility $\chi^{+\uparrow}_{(0)}$ given by

$$\chi^{+\uparrow\,\mu\nu\gamma\xi}_{(0)ijkl}(\omega) = \chi^{12\,\mu\nu\gamma\xi}_{(0)ijkl}(\omega) = \left[\Gamma^{11}A^{12} + \Gamma^{12}A^{22} + \Gamma^{14}A^{42}\right]^{\mu\nu\gamma\xi}_{ijkl} \ , \tag{3.205}$$

since $A^{32} = 0$.

Following the same steps as before, we have

$$\begin{aligned}\sum_{\substack{mn\\ \alpha\beta}} \Gamma^{11\,\mu\nu\alpha\beta}_{ijmn} A^{12\,\alpha\beta\gamma\xi}_{mnkl} = &-\hbar \sum_{\substack{n\\ \beta}} \int d\omega' f(\omega') \sum_{pp'p''} \langle \phi_{p''}|k\gamma\uparrow\rangle\langle n\beta\downarrow|\phi_{p''}\rangle \delta(\hbar\omega' - E_{p''}) \\ &\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle l\xi\uparrow|\phi_p\rangle\langle\phi_{p'}|n\beta\downarrow\rangle}{\hbar\omega - E_{p'} + E_p} \ ,\end{aligned} \tag{3.206}$$

$$\begin{aligned}\sum_{\substack{mn\\ \alpha\beta}} \Gamma^{12\,\mu\nu\alpha\beta}_{ijmn} A^{22\,\alpha\beta\gamma\xi}_{mnkl} = &\hbar \sum_{\substack{m\\ \alpha}} \int d\omega' f(\omega') \sum_{pp'p''} \langle \phi_{p''}|m\alpha\uparrow\rangle\langle l\xi\uparrow|\phi_{p''}\rangle \delta(\hbar\omega' - E_{p''}) \\ &\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\uparrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \\ &-\hbar \sum_{\substack{n\\ \beta}} \int d\omega' f(\omega') \sum_{pp'p''} \langle \phi_{p''}|k\gamma\uparrow\rangle\langle n\beta\uparrow|\phi_{p''}\rangle \delta(\hbar\omega' - E_{p''}) \\ &\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle l\xi\uparrow|\phi_p\rangle\langle\phi_{p'}|n\beta\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \ , \text{ and}\end{aligned} \tag{3.207}$$

$$\begin{aligned}\sum_{\substack{mn\\ \alpha\beta}} \Gamma^{14\,\mu\nu\alpha\beta}_{ijmn} A^{42\,\alpha\beta\gamma\xi}_{mnkl} = &\hbar \sum_{\substack{m\\ \alpha}} \int d\omega' f(\omega') \sum_{pp'p''} \langle \phi_{p''}|m\alpha\downarrow\rangle\langle l\xi\uparrow|\phi_{p''}\rangle \delta(\hbar\omega' - E_{p''}) \\ &\times \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle m\alpha\downarrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \ .\end{aligned} \tag{3.208}$$

Once again, using the orthogonality of the wave functions $\langle\phi_p|i\mu\sigma\rangle$, we have

$$\begin{aligned}\chi^{+\uparrow\,\mu\nu\gamma\xi}_{(0)ijkl}(\omega) = &\hbar \int d\omega' f(\omega') \sum_{pp'} \delta(\hbar\omega' - E_p) \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle l\xi\uparrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \\ &-\hbar \int d\omega' f(\omega') \sum_{pp'} \delta(\hbar\omega' - E_{p'}) \frac{\langle\phi_p|i\mu\uparrow\rangle\langle j\nu\downarrow|\phi_{p'}\rangle\langle l\xi\uparrow|\phi_p\rangle\langle\phi_{p'}|k\gamma\uparrow\rangle}{\hbar\omega - E_{p'} + E_p} \ .\end{aligned} \tag{3.209}$$

Moreover, we note again that an easy way to obtain the susceptibilities is to change the spin indices according to the indices M, N of the susceptibility $\chi_{ijkl}^{MN\,\mu\nu\gamma\xi}$ we are calculating. For example, in the equation above, we have $M = +$, which is related to the states $|i\mu\uparrow\rangle$ and $|j\nu\downarrow\rangle$, and $N =\uparrow$, which is related to $|l\xi\uparrow\rangle$ and $|k\gamma\uparrow\rangle$. In terms of the monoeletronic propagators,

$$\begin{aligned}\chi_{(0)ijkl}^{+\uparrow\,\mu\nu\gamma\xi}(\omega) = \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\Big(G_{jk}^{\downarrow\uparrow\,\nu\gamma}(\omega'+\omega)\Big\{G_{li}^{\uparrow\uparrow\,\xi\mu}(\omega') - \Big[G_{il}^{\uparrow\uparrow\,\mu\xi}(\omega')\Big]^*\Big\} \\ +[G_{il}^{\uparrow\uparrow\,\mu\xi}(\omega'-\omega)]^*\Big\{G_{jk}^{\downarrow\uparrow\,\nu\gamma}(\omega') - \Big[G_{kj}^{\uparrow\downarrow\,\gamma\nu}(\omega')\Big]^*\Big\}\Big)\ .\end{aligned} \tag{3.210}$$

Chapter 4

One electron propagators

To obtain the Hartree-Fock susceptibilities, we need to calculate the monoelectronic Green functions for a system where the spin orbit coupling is present. In order to do this, we can write

$$G(E) = \left(E - \hat{H}\right)^{-1} \ , \tag{4.1}$$

where $\hat{H}$ is a matrix in spin and orbital space. So for each site, $\hat{H}$ is a 2×2 matrix where its components are 9×9 matrices. Due to the poles of this function, it's convenient to define a Green function over the whole complex plane by doing $E \to z = E + i\eta$. By doing this, the Green function can be obtained by either doing a limit of $\eta \to 0$ from the upper complex plane (the poles are shifted to the lower complex half plane) or from the lower half plane (when the poles are shifted to the upper half-plane). They are called retarded and advanced Green functions, respectively, and can be written as

$$\begin{aligned} G(E+i\eta) &= \left(E - \hat{H} + i\eta\right)^{-1} \\ G(E-i\eta) &= \left(E - \hat{H} - i\eta\right)^{-1} \end{aligned} \ , \tag{4.2}$$

This operator can be written as

$$G(z) = \sum_\alpha |\alpha\rangle \left(z - \epsilon_\alpha\right)^{-1} \langle\alpha| \ , \tag{4.3}$$

where α denotes the eigenenergies ϵ_α and eigenvalues $|\alpha\rangle$ of the hamiltonian. Projecting on the real space basis that we use, which included site, orbital and spin indices, we obtain

$$\langle i\mu\sigma| G(z) \left|j\nu\sigma'\right\rangle = G_{ij}^{\sigma\sigma'\,\mu\nu}(z) = \sum_\alpha \langle i\mu\sigma\ |\alpha\rangle \left(z - \epsilon_\alpha\right)^{-1} \langle\alpha|\ j\nu\sigma'\rangle \ , \tag{4.4}$$

The retarded Green function can be written as

$$G_{ij}^{\sigma\sigma'\,\mu\nu}(E+i\eta) = \sum_\alpha \langle i\mu\sigma\ |\alpha\rangle \left(E - \epsilon_\alpha + i\eta\right)^{-1} \langle\alpha|\ j\nu\sigma'\rangle \ , \tag{4.5}$$

Chapter 5

Spin and Charge Perturbations

5.1 Current and torque operators in real space

Now we want to calculate spin currents, charge currents and orbital angular momentum currents in systems where the spin-orbit coupling is present.

5.1.1 Spin currents

To obtain the spin current operator with polarization m that flows out of a unit cell of volume V_i, we start on the continuity equation for the spin density and integrate over the cell volume

$$\begin{aligned}\int_{V_i} \hbar \frac{dS^m(\mathbf{r})}{dt} d\mathbf{r} + \int_{V_i} \nabla \cdot \mathbf{J}^m(\mathbf{r}) d\mathbf{r} = \int_{V_i} \tau^m(\mathbf{r})\mathbf{r} \\ \hbar \frac{dS_i^m}{dt} + \oint_S \mathbf{J}^m(\mathbf{r}) \cdot d\mathbf{A} = \tau_i^m \\ \hbar \frac{dS_i^m}{dt} + \sum_j I_{ij}^m = \tau_i^m \ .\end{aligned} \tag{5.1}$$

Here τ_i represents the local torque acting on cell i, and the current I_{ij}^m include non-local torques. Using Heinsenberg's equation,

$$\begin{aligned}\hbar \frac{dS_i^m}{dt} &= \frac{1}{i}[S_i^m, \hat{H}] \\ &= \frac{1}{i}[S_i^m, \hat{H}_0 + \hat{H}_{\text{int}} + \hat{H}_{\text{so}} + \hat{H}_{\text{ZS}} + \hat{H}_{\text{ZL}}] \\ &= -\sum_j I_{ij}^m + \tau_i^m \ .\end{aligned} \tag{5.2}$$

The terms $\frac{1}{i}[S_i^m, \hat{H}_{\text{so}} + \hat{H}_{\text{ZS}}] = \tau_i^m$ (since $[S_i^m, \hat{H}_{\text{ZL}}] = 0$) give the $m-$component of the local torques acting on the spin moment due to the spin-orbit coupling ($\propto \mathbf{S}_i \times \mathbf{L}_i$) and external magnetic fields ($\propto \mathbf{S}_i \times \mathbf{B}_i^{\mathrm{ext}}$). Therefore, we end up with

$$\begin{aligned}\sum_j I_{ij}^m =& i[S_i^m, \hat{H}_0 + \hat{H}_{\mathrm{int}}] \\ =& \frac{i}{2}\sum_\mu \sum_{\alpha\beta} [c^\dagger_{i\mu\alpha}\sigma^m_{\alpha\beta}c_{i\mu\beta}, \hat{H}_0 + \hat{H}_{\mathrm{int}}] \ .\end{aligned} \tag{5.3}$$

Using Eqs. 1.2 and 1.3, we can calculate the following commutators

$$\begin{aligned}\frac{i}{2}\sum_{\mu,\alpha\beta}\sigma^m_{\alpha\beta}[c^\dagger_{i\mu\alpha}c_{i\mu\beta}, \hat{H}_0] =& \frac{i}{2}\sum_{\alpha\beta}\sigma^m_{\alpha\beta}\sum_{\substack{mn\\ s}}\sum_{\mu\gamma\xi} t^{\gamma\xi}_{mn}[c^\dagger_{i\mu\alpha}c_{i\mu\beta}, c^\dagger_{m\gamma s}c_{n\xi s}] \\ =& \frac{i}{2}\sum_{\alpha\beta}\sigma^m_{\alpha\beta}\sum_{\substack{mn\\ s}}\sum_{\mu\gamma\xi} t^{\gamma\xi}_{mn}\left\{c^\dagger_{i\mu\alpha}c_{i\mu\beta}c^\dagger_{m\gamma s}c_{n\xi s} - c^\dagger_{m\gamma s}c_{n\xi s}c^\dagger_{i\mu\alpha}c_{i\mu\beta}\right\} \\ =& \frac{i}{2}\sum_{\alpha\beta}\sigma^m_{\alpha\beta}\sum_{\substack{mn\\ s}}\sum_{\mu\gamma\xi} t^{\gamma\xi}_{mn}\left\{c^\dagger_{i\mu\alpha}c_{n\xi s}\delta_{im}\delta_{\mu\gamma}\delta_{\beta s} - c^\dagger_{m\gamma s}c^\dagger_{i\mu\alpha}c_{n\xi s}c_{i\mu\beta} - c^\dagger_{m\gamma s}c_{n\xi s}c^\dagger_{i\mu\alpha}c_{i\mu\beta}\right\} \\ =& \frac{i}{2}\sum_{\alpha\beta}\sigma^m_{\alpha\beta}\sum_{\substack{mn\\ s}}\sum_{\mu\gamma\xi} t^{\gamma\xi}_{mn}\left\{c^\dagger_{i\mu\alpha}c_{n\xi s}\delta_{im}\delta_{\mu\gamma}\delta_{\beta s} - c^\dagger_{m\gamma s}c_{i\mu\beta}\delta_{in}\delta_{\mu\xi}\delta_{\alpha s}\right\} \\ =& \frac{i}{2}\sum_{\alpha\beta}\sum_{n,\mu\xi} t^{\mu\xi}_{in}c^\dagger_{i\mu\alpha}\sigma^m_{\alpha\beta}c_{n\xi\beta} - t^{\xi\mu}_{ni}c^\dagger_{n\xi\alpha}\sigma^m_{\alpha\beta}c_{i\mu\beta} \\ =& i\sum_n\sum_{\mu\xi} t^{\mu\xi}_{in}S^{m\,\mu\xi}_{in} - t^{\xi\mu}_{ni}S^{m\,\xi\mu}_{ni}\end{aligned} \tag{5.4}$$

where we have defined a generalized spin operator $S^{m\,\mu\nu}_{ij} = \frac{1}{2}\sum_{\alpha\beta} c^\dagger_{i\mu\alpha}\sigma^m_{\alpha\beta}c_{j\nu\beta}$, and

$$\begin{aligned}\sum_\mu [c^\dagger_{i\mu\sigma}c_{i\mu\sigma'}, \hat{H}_{\mathrm{int}}] =& \frac{1}{2}\sum_\mu\sum_{\substack{m\\ s,s'}}\sum_{\alpha\beta\gamma\xi} U^m_{\alpha\beta\gamma\xi}[c^\dagger_{i\mu\sigma}c_{i\mu\sigma'}, c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}] \\ =& \frac{1}{2}\sum_\mu\sum_{\substack{m\\ s,s'}}\sum_{\alpha\beta\gamma\xi} U^m_{\alpha\beta\gamma\xi}\Big\{c^\dagger_{i\mu\sigma}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}\delta_{im}\delta_{\mu\alpha}\delta_{\sigma' s} - c^\dagger_{i\mu\sigma}c^\dagger_{m\alpha s}c_{m\xi s'}c_{m\gamma s}\delta_{im}\delta_{\mu\beta}\delta_{\sigma' s'} \\ & + c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\gamma s}c_{i\mu\sigma'}\delta_{im}\delta_{\mu\xi}\delta_{\sigma s'} - c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{i\mu\sigma'}\delta_{im}\delta_{\mu\gamma}\delta_{\sigma s}\Big\} \\ =& \frac{1}{2}\sum_\mu\sum_s\sum_{\alpha\beta\gamma\xi} U^i_{\alpha\beta\gamma\xi}\Big\{c^\dagger_{i\mu\sigma}c^\dagger_{i\beta s}c_{i\xi s}c_{i\gamma\sigma'}\delta_{\mu\alpha} - c^\dagger_{i\mu\sigma}c^\dagger_{i\alpha s}c_{i\xi\sigma'}c_{i\gamma s}\delta_{\mu\beta} \\ & + c^\dagger_{i\alpha s}c^\dagger_{i\beta\sigma}c_{i\gamma s}c_{i\mu\sigma'}\delta_{\mu\xi} - c^\dagger_{i\alpha\sigma}c^\dagger_{i\beta s}c_{i\xi s}c_{i\mu\sigma'}\delta_{\mu\gamma}\Big\} \\ =& \frac{1}{2}\sum_s\sum_{\alpha\beta\gamma\xi} U^i_{\alpha\beta\gamma\xi}\Big\{c^\dagger_{i\alpha\sigma}c^\dagger_{i\beta s}c_{i\xi s}c_{i\gamma\sigma'} - c^\dagger_{i\beta\sigma}c^\dagger_{i\alpha s}c_{i\xi\sigma'}c_{i\gamma s} \\ & + c^\dagger_{i\alpha s}c^\dagger_{i\beta\sigma}c_{i\gamma s}c_{i\xi\sigma'} - c^\dagger_{i\alpha\sigma}c^\dagger_{i\beta s}c_{i\xi s}c_{i\gamma\sigma'}\Big\} = 0\end{aligned} \tag{5.5}$$

since we can change $\alpha \rightleftharpoons \beta$ and $\gamma \rightleftharpoons \xi$ on the second term and use the symmetry of the effective electronic interaction $U^i_{\beta\alpha\xi\gamma} = U^i_{\alpha\beta\gamma\xi}$.

The spin current can then be written as

$$\hat{I}^m_{ij} = i \sum_{\mu\nu} \left\{ t^{\mu\nu}_{ij} S^{m\,\mu\nu}_{ij} - t^{\nu\mu}_{ji} S^{m\,\nu\mu}_{ji} \right\} \ , \tag{5.6}$$

which gives the current that flows from site i to site j.

In equilibrium, $\frac{dS^m_i}{dt} = 0$ and we have

$$\begin{aligned} \sum_j I^m_{ij} =& \tau^m_i \\ [S^m_i, \hat{H}_0 + \hat{H}_{\text{int}}] =& [S^m_i, \hat{H}_{\text{so}} + \hat{H}_{\text{ZS}}] \end{aligned} \tag{5.7}$$

In the Hartree-Fock approximation, the Hamiltonian is given by Eq. (1.21), and the current can be obtained as

$$\sum_j I^m_{ij} - \tau^m_i = \frac{i}{2} \sum_{\mu} \sum_{\alpha\beta} [c^\dagger_{i\mu\alpha} \sigma^m_{\alpha\beta} c_{i\mu\beta}, \hat{H}'_0 + \hat{H}_{\text{sf}}] \ . \tag{5.8}$$

The first commutator is given by

$$\begin{aligned} [c^\dagger_{i\mu\alpha} \sigma^m_{\alpha\beta} c_{i\mu\beta}, \hat{H}'_0] =& \sum_{\substack{jk \\ s}} \sum_{\nu\gamma} t^{s\,\nu\gamma}_{jk} [c^\dagger_{i\mu\alpha} \sigma^m_{\alpha\beta} c_{i\mu\beta}, c^\dagger_{j\nu s} c_{k\gamma s}] \\ =& \sum_{\substack{jk \\ s}} \sum_{\nu\gamma} t^{s\,\nu\gamma}_{jk} \sigma^m_{\alpha\beta} \left\{ c^\dagger_{i\mu\alpha} c_{k\gamma s} \delta_{ij} \delta_{\mu\nu} \delta_{\beta s} - c^\dagger_{j\nu s} c_{i\mu\beta} \delta_{ik} \delta_{\mu\gamma} \delta_{\alpha s} \right\} \\ =& \sum_{\substack{j \\ \nu}} t^{\beta\,\mu\nu}_{ij} c^\dagger_{i\mu\alpha} \sigma^m_{\alpha\beta} c_{j\nu\beta} - t^{\alpha\,\nu\mu}_{ji} c^\dagger_{j\nu\alpha} \sigma^m_{\alpha\beta} c_{i\mu\beta} \ . \end{aligned} \tag{5.9}$$

So the contribution of this term to the current is

$$\sum_j I^m_{ij} - \tau^m_i = \frac{i}{2} \sum_{\alpha\beta} \sum_{\substack{j \\ \mu\nu}} t^{\beta\,\mu\nu}_{ij} c^\dagger_{i\mu\alpha} \sigma^m_{\alpha\beta} c_{j\nu\beta} - t^{\alpha\,\nu\mu}_{ji} c^\dagger_{j\nu\alpha} \sigma^m_{\alpha\beta} c_{i\mu\beta} \ . \tag{5.10}$$

The onsite terms—related to the torque—can be written as

$$\begin{aligned} & \frac{i}{2} \sum_{\alpha\beta} \sum_{\mu\nu} t^{\beta\,\mu\nu}_{ii} c^\dagger_{i\mu\alpha} \sigma^m_{\alpha\beta} c_{i\nu\beta} - t^{\alpha\,\nu\mu}_{ii} c^\dagger_{i\nu\alpha} \sigma^m_{\alpha\beta} c_{i\mu\beta} \\ =& \frac{i}{2} \sum_{\alpha\beta} \sum_{\mu\nu} t^{\beta\,\mu\nu}_{ii} c^\dagger_{i\mu\alpha} \sigma^m_{\alpha\beta} c_{i\nu\beta} - t^{\alpha\,\mu\nu}_{ii} c^\dagger_{i\mu\alpha} \sigma^m_{\alpha\beta} c_{i\nu\beta} \\ =& \frac{i}{2} \sum_{\alpha\beta} \sum_{\mu\nu} \left(t^{\beta\,\mu\nu}_{ii} - t^{\alpha\,\mu\nu}_{ii} \right) c^\dagger_{i\mu\alpha} \sigma^m_{\alpha\beta} c_{i\nu\beta} \ . \end{aligned} \tag{5.11}$$

Therefore, the spin-independent terms will cancel and we end up with
and the second commutator

$$
\begin{aligned}
[c^\dagger_{i\mu\alpha}\sigma^m_{\alpha\beta}c_{i\mu\beta}, \hat{H}_{\text{sf}}] &= \sum_j \sum_{\nu\gamma} [c^\dagger_{i\mu\alpha}\sigma^m_{\alpha\beta}c_{i\mu\beta}, a_{j,\nu\gamma}c^\dagger_{j\nu\uparrow}c_{j\gamma\downarrow} + a^*_{j,\gamma\nu}c^\dagger_{j\nu\downarrow}c_{j\gamma\uparrow}] \\
&= \sum_j \sum_{\nu\gamma} \sigma^m_{\alpha\beta} \Big\{ a_{j,\nu\gamma} \left[c^\dagger_{i\mu\alpha}c_{i\mu\beta}c^\dagger_{j\nu\uparrow}c_{j\gamma\downarrow} - c^\dagger_{j\nu\uparrow}c_{j\gamma\downarrow}c^\dagger_{i\mu\alpha}c_{i\mu\beta} \right] \\
&\qquad + a^*_{j,\gamma\nu} \left[c^\dagger_{i\mu\alpha}c_{i\mu\beta}c^\dagger_{j\nu\downarrow}c_{j\gamma\uparrow} - c^\dagger_{j\nu\downarrow}c_{j\gamma\uparrow}c^\dagger_{i\mu\alpha}c_{i\mu\beta} \right] \Big\} \ .
\end{aligned}
\tag{5.12}
$$

5.1.2 Local spin torque

As defined in the previous section, the operator for the local torque acting on the spin moment is given by

$$
\begin{aligned}
\tau^m_i =& \frac{1}{i}[S^m_i, \hat{H}_{\text{so}} + \hat{H}_{\text{ZS}}] \\
=& \frac{1}{2i} \sum_{\mu,\alpha\beta} [c^\dagger_{i\mu\alpha}\sigma^m_{\alpha\beta}c_{i\mu\beta}, \hat{H}_{\text{so}} + \hat{H}_{\text{ZS}}] \ .
\end{aligned}
\tag{5.13}
$$

Using Eq. (1.11) to calculate the spin-orbit coupling commutator

$$
\begin{aligned}
& \sum_{\mu,\alpha\beta} \sum_{\substack{jn\gamma\xi \\ ss'}} \frac{\lambda_j}{2} \sigma^m_{\alpha\beta} \left(L'^n_{\gamma\xi}\sigma^n_{ss'} \right) [c^\dagger_{i\mu\alpha}c_{i\mu\beta}, c^\dagger_{j\gamma s}c_{j\xi s'}] \\
=& \sum_{\mu,\alpha\beta} \sum_{\substack{jn\gamma\xi \\ ss'}} \frac{\lambda_j}{2} \sigma^m_{\alpha\beta} \left(L'^n_{\gamma\xi}\sigma^n_{ss'} \right) \left\{ c^\dagger_{i\mu\alpha}c_{j\xi s'}\delta_{ij}\delta_{\mu\gamma}\delta_{\beta s} - c^\dagger_{j\gamma s}c_{i\mu\beta}\delta_{ij}\delta_{\mu\xi}\delta_{\alpha s'} \right\} \\
=& \sum_{\alpha\beta} \sum_{\substack{n\gamma\xi \\ ss'}} \frac{\lambda_i}{2} \sigma^m_{\alpha\beta} \left(L'^n_{\gamma\xi}\sigma^n_{ss'} \right) \left\{ c^\dagger_{i\gamma\alpha}c_{i\xi s'}\delta_{\beta s} - c^\dagger_{i\gamma s}c_{i\xi\beta}\delta_{\alpha s'} \right\} \\
=& \sum_{\alpha\beta} \sum_{n\gamma\xi} \frac{\lambda_i}{2} L'^n_{\gamma\xi} \left\{ c^\dagger_{i\gamma\alpha} \sum_s \sigma^m_{\alpha s}\sigma^n_{s\beta}c_{i\xi\beta} - c^\dagger_{i\gamma\alpha} \sum_s \sigma^n_{\alpha s}\sigma^m_{s\beta}c_{i\xi\beta} \right\} \\
=& i\lambda_i \sum_{\alpha\beta} \sum_{nk\gamma\xi} \epsilon_{mnk} L'^n_{\gamma\xi} \sigma^k_{\alpha\beta} c^\dagger_{i\gamma\alpha}c_{i\xi\beta} \\
=& 2i\lambda_i \sum_{nk\gamma\xi} \epsilon_{mnk} L'^n_{\gamma\xi} S^{k\,\gamma\xi}_{ii} \ ,
\end{aligned}
\tag{5.14}
$$

where $m, n, k = x, y, z$. Finally, we can write the spin-orbit torque components as

$$
\begin{aligned}
\tau^m_i =& \lambda_i \sum_{\substack{nk \\ \mu\nu}} \epsilon_{mnk} L'^n_{\mu\nu} S^{k\,\mu\nu}_{ii} \\
=& \frac{\lambda_i}{2} \sum_{\substack{nk \\ \mu\nu}} \sum_{\alpha\beta} \epsilon_{mnk} L'^n_{\mu\nu} c^\dagger_{i\mu\alpha}\sigma^k_{\alpha\beta}c_{i\nu\beta} \ .
\end{aligned}
\tag{5.15}
$$

or

$$
\begin{aligned}
\tau_i^x =&\lambda_i \sum_{\mu\nu} \left(L'^y_{\mu\nu} S_{ii}^{z\,\mu\nu} - L'^z_{\mu\nu} S_{ii}^{y\,\mu\nu} \right) \\
\tau_i^y =&\lambda_i \sum_{\mu\nu} \left(L'^z_{\mu\nu} S_{ii}^{x\,\mu\nu} - L'^x_{\mu\nu} S_{ii}^{z\,\mu\nu} \right) \quad . \\
\tau_i^z =&\lambda_i \sum_{\mu\nu} \left(L'^x_{\mu\nu} S_{ii}^{y\,\mu\nu} - L'^y_{\mu\nu} S_{ii}^{x\,\mu\nu} \right)
\end{aligned}
\tag{5.16}
$$

In general, we can use the hamiltonian written in the form of Eq. (1.38) to calculate the torque. Since only the spin dependent part contributes with the torque, we have

$$
\begin{aligned}
\tau_i^m =&\frac{1}{2i} \sum_{\mu,\alpha\beta} \sum_{\substack{n,j\\ \gamma\xi}} \sum_{ss'} [c^\dagger_{i\mu\alpha} \sigma^m_{\alpha\beta} c_{i\mu\beta}, c^\dagger_{j\gamma s} \sigma^n_{ss'} B_j^{n\,\gamma\xi} c_{j\xi s'}] \\
=&\frac{1}{2i} \sum_{\mu,\alpha\beta} \sum_{\substack{n,j\\ \gamma\xi}} \sum_{ss'} \sigma^m_{\alpha\beta} \sigma^n_{ss'} B_j^{n\,\gamma\xi} \left\{ c^\dagger_{i\mu\alpha} c_{i\mu\beta} c^\dagger_{j\gamma s} c_{j\xi s'} - c^\dagger_{j\gamma s} c_{j\xi s'} c^\dagger_{i\mu\alpha} c_{i\mu\beta} \right\} \\
=&\frac{1}{2i} \sum_{\mu,\alpha\beta} \sum_{\substack{n,j\\ \gamma\xi}} \sum_{ss'} \sigma^m_{\alpha\beta} \sigma^n_{ss'} B_j^{n\,\gamma\xi} \left\{ c^\dagger_{i\mu\alpha} c_{j\xi s'} \delta_{ij} \delta_{\mu\gamma} \delta_{\beta s} - c^\dagger_{j\gamma s} c_{i\mu\beta} \delta_{ij} \delta_{\mu\xi} \delta_{\alpha s'} \right\} \\
=&\frac{1}{2i} \sum_{\alpha\beta} \sum_{\substack{n\\ \gamma\xi}} \sum_{ss'} \sigma^m_{\alpha\beta} \sigma^n_{ss'} B_i^{n\,\gamma\xi} \left\{ c^\dagger_{i\gamma\alpha} c_{i\xi s'} \delta_{\beta s} - c^\dagger_{i\gamma s} c_{i\xi\beta} \delta_{\alpha s'} \right\} \quad . \\
=&\frac{1}{2i} \sum_{\alpha\beta} \sum_{\substack{n\\ \gamma\xi}} B_i^{n\,\gamma\xi} c^\dagger_{i\gamma\alpha} \sum_{s} \left\{ \sigma^m_{\alpha s} \sigma^n_{s\beta} - \sigma^n_{\alpha s} \sigma^m_{s\beta} \right\} c_{i\xi\beta} \\
=&\sum_{\alpha\beta} \sum_{\substack{nk\\ \gamma\xi}} B_i^{n\,\gamma\xi} c^\dagger_{i\gamma\alpha} \epsilon_{mnk} \sigma^k_{\alpha\beta} c_{i\xi\beta} \\
=&2 \sum_{\substack{nk\\ \gamma\xi}} \epsilon_{mnk} B_i^{n\,\gamma\xi} S_{ii}^{k\,\gamma\xi}
\end{aligned}
\tag{5.17}
$$

This can also be written as

$$
\boldsymbol{\tau}_i = \hat{\mathbf{S}}_i \times \mathbf{B}_i \ . \tag{5.18}
$$

If we use the spin-orbit effective field $\mathbf{B}_i^{\text{soc}\,\mu\nu} = \frac{\lambda_i}{2} \mathbf{L}'_{\mu\nu}$ (in the spin frame of reference), we recover the result of Eq. (5.15). The exchange-correlation torque can be obtained using the xc-field $\mathbf{B}_i^{\text{xc}\,\mu\nu} = -\delta_{\mu\nu} \frac{U^i}{2} \langle \mathbf{m}_i \rangle$. This results in

$$
\begin{aligned}
\tau_i^m =& - U_i \sum_{\substack{nk\\ \mu}} \epsilon_{mnk} \langle m_i^n \rangle S_{ii}^{k\,\mu\mu} \\
=& - \sum_{\substack{nk\\ \mu}} \sum_{\alpha\beta} \epsilon_{mnk} \frac{U_i \langle m_i^n \rangle}{2} c^\dagger_{i\mu\alpha} \sigma^k_{\alpha\beta} c_{i\mu\beta}
\end{aligned} \quad .
\tag{5.19}
$$

Finally, the torque caused by the external static field is obtained by taking into account that $\mathbf{B}_i^{\text{ext}\mu\nu} = \frac{g_S\mu_B}{2}\delta_{\mu\nu}\mathbf{B}'$ (in the spin frame of reference). Therefore, the external torque operator may be written as

$$\begin{aligned}\tau_i^m =& g_S\mu_B \sum_{\substack{nk\\ \mu}} \epsilon_{mnk} B'^n S_{ii}^{k\mu\mu} \\ =& \sum_{\substack{nk\\ \mu}} \epsilon_{mnk} \hbar\omega_n S_{ii}^{k\mu\mu}\end{aligned} \quad . \tag{5.20}$$

where $\hbar\omega_\alpha = g_S\mu_B B'^\alpha$, and we assume a uniform magnetic field over all sites. Once again, it is important to remember that we are considering negative values of $\hbar\omega_\alpha$ to obtain peaks on positive energies in the transverse dynamic spin susceptibility.

5.1.3 Charge currents

The charge current operator in the tight-binding model may also be obtained from the continuity equation,

$$\frac{d\rho_i}{dt} + \sum_m I_{im}^{\mathrm{C}} = 0 \ . \tag{5.21}$$

We get this equation by integrating the continuous version over an unit cell

$$\begin{aligned}\int_{V_i} \frac{d\rho(\mathbf{r})}{dt} d\mathbf{r} + \int_{V_i} \nabla\cdot\mathbf{J}^{\mathrm{C}}(\mathbf{r}) d\mathbf{r} =& 0 \\ \frac{d\rho_i}{dt} + \oint_{S_i} \mathbf{J}^{\mathrm{C}}(\mathbf{r})\cdot d\mathbf{A} =& 0 \\ \frac{d\rho_i}{dt} + \sum_m I_{im}^{\mathrm{C}} =& 0 \ .\end{aligned} \tag{5.22}$$

Now, we take the time derivative of the charge density operator

$$\rho_i = e \sum_{\mu\sigma} c_{i\mu\sigma}^\dagger c_{i\mu\sigma} \tag{5.23}$$

using Heisenberg's equation of motion

$$\begin{aligned}\frac{d\rho_i}{dt} =& \frac{1}{i\hbar}[\rho_i, \hat{H}] \\ =& \frac{e}{i\hbar}\sum_{\mu\sigma}[c_{i\mu\sigma}^\dagger c_{i\mu\sigma}, \hat{H}] \\ =& \frac{e}{i\hbar}\sum_{\mu\sigma}[c_{i\mu\sigma}^\dagger c_{i\mu\sigma}, \hat{H}_0 + \hat{H}_{\text{int}} + \hat{H}_{\text{so}} + \hat{H}_{\text{ZS}} + \hat{H}_{\text{ZL}}] \ .\end{aligned} \tag{5.24}$$

The commutators are given by

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma}c_{i\mu\sigma},\hat{H}_0] &= \sum_{\substack{mn\\ \sigma'}}\sum_{\alpha\beta} t^{\alpha\beta}_{mn}[c^\dagger_{i\mu\sigma}c_{i\mu\sigma},c^\dagger_{m\alpha\sigma'}c_{n\beta\sigma'}] \\
&= \sum_{\substack{mn\\ \sigma'}}\sum_{\alpha\beta} t^{\alpha\beta}_{mn}\left\{c^\dagger_{i\mu\sigma}c_{n\beta\sigma'}\delta_{im}\delta_{\mu\alpha}\delta_{\sigma\sigma'} - c^\dagger_{m\alpha\sigma'}c_{i\mu\sigma}\delta_{in}\delta_{\mu\beta}\delta_{\sigma\sigma'}\right\} \\
&= \sum_{m\alpha}\left\{t^{\mu\alpha}_{im}c^\dagger_{i\mu\sigma}c_{m\alpha\sigma} - t^{\alpha\mu}_{mi}c^\dagger_{m\alpha\sigma}c_{i\mu\sigma}\right\} \;,
\end{aligned}
\tag{5.25}
$$

$$
\sum_{\mu\sigma}[c^\dagger_{i\mu\sigma}c_{i\mu\sigma},\hat{H}_{\mathrm{int}}] = 0 \;, \tag{5.26}
$$

$$
\sum_{\mu\sigma}[c^\dagger_{i\mu\sigma}c_{i\mu\sigma},\hat{H}_{\mathrm{so}}] = 0 \;. \tag{5.27}
$$

In general,

$$
[c^\dagger_{i\mu\sigma}c_{i\mu\sigma},c^\dagger_{m\alpha s}c_{m\beta s'}] = c^\dagger_{i\mu\sigma}c_{m\beta s'}\delta_{im}\delta_{\mu\alpha}\delta_{\sigma s} - c^\dagger_{m\alpha s}c_{i\mu\sigma}\delta_{im}\delta_{\mu\beta}\delta_{\sigma s'} \;. \tag{5.28}
$$

Substituting back in Eq. (5.24), we have

$$
\frac{d\rho_i}{dt} - \frac{e}{i\hbar}\sum_m\sum_\sigma\sum_{\mu\alpha}\left\{t^{\mu\alpha}_{im}c^\dagger_{i\mu\sigma}c_{m\alpha\sigma} - t^{\alpha\mu}_{mi}c^\dagger_{m\alpha\sigma}c_{i\mu\sigma}\right\} = 0 \;. \tag{5.29}
$$

Comparing to Eq. (5.21), it gives us the charge current operator

$$
\hat{I}^{\mathrm{C}}_{ij} = \frac{e}{\hbar}i\sum_\sigma\sum_{\mu\nu}\left\{t^{\mu\nu}_{ij}c^\dagger_{i\mu\sigma}c_{j\nu\sigma} - t^{\nu\mu}_{ji}c^\dagger_{j\nu\sigma}c_{i\mu\sigma}\right\} \;. \tag{5.30}
$$

This is the charge current operator that flows through the bond $i-m$. It is easy to see that it may be written in terms of the operator obtained in Eq. (5.6) as

$$
\hat{I}^{\mathrm{C}}_{ij} = \frac{e}{\hbar}\sum_\sigma \hat{I}^\sigma_{ij} = \frac{e}{\hbar}\left(\hat{I}^\uparrow_{ij} + \hat{I}^\downarrow_{ij}\right) \;. \tag{5.31}
$$

We can also obtain the charge current density using the expression

$$
\hat{J}^{\mathrm{C}}(\mathbf{r}) = \frac{e\hbar}{2mi}\sum_\sigma\left\{\hat{\psi}^\dagger_\sigma(\mathbf{r})\nabla\hat{\psi}_\sigma(\mathbf{r}) - \left[\nabla\hat{\psi}^\dagger_\sigma(\mathbf{r})\right]\hat{\psi}_\sigma(\mathbf{r})\right\} \;. \tag{5.32}
$$

Using the relation between the field operator and the creation and annihilation operators, i.e.,

$$
\begin{aligned}
\hat{\psi}^\dagger_\sigma(\mathbf{r}) &= \sum_{i\mu}\phi^*_\mu(\mathbf{r}-\mathbf{R}_i)c^\dagger_{i\mu\sigma} \;, \\
\hat{\psi}_\sigma(\mathbf{r}) &= \sum_{j\nu}\phi_\nu(\mathbf{r}-\mathbf{R}_j)c_{j\nu\sigma} \;,
\end{aligned}
\tag{5.33}
$$

we can write the charge current density as

$$\hat{\mathbf{J}}^{\mathrm{C}}(\mathbf{r}) = \frac{e}{2m}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}} \mathbf{w}_{ij}^{\mu\nu}(\mathbf{r}) c_{i\mu\sigma}^{\dagger} c_{j\nu\sigma} \ , \tag{5.34}$$

where

$$\mathbf{w}_{ij}^{\mu\nu}(\mathbf{r}) = \left\{ \phi_{\mu}^{*}(\mathbf{r}-\mathbf{R}_i)\frac{\hbar}{i}\nabla\phi_{\nu}(\mathbf{r}-\mathbf{R}_j) - \left[\frac{\hbar}{i}\nabla\phi_{\mu}^{*}(\mathbf{r}-\mathbf{R}_i)\right]\phi_{\nu}(\mathbf{r}-\mathbf{R}_j)\right\} \ . \tag{5.35}$$

5.1.4 Diamagnetic current

When we apply an external field given by a potential vector, the current operator will be modified. To obtain this new term, we take into account a new term in the Heisenberg equation of motion which involves the external perturbation (this hamiltonian is obtained in Eq. (5.211))

$$\hat{H}_{\mathrm{ext}}(t) = -\int d\mathbf{r} \hat{\mathbf{J}}^{\mathrm{C}}(\mathbf{r},t)\cdot\mathbf{A}(\mathbf{r},t) \ . \tag{5.36}$$

The term that should be added to the Eq. of motion 5.24 involves the commutator

$$\frac{e}{i\hbar}\sum_{\mu\sigma}[c_{i\mu\sigma}^{\dagger}c_{i\mu\sigma}, \hat{H}_{\mathrm{ext}}] = -\frac{e}{i\hbar}\sum_{\mu\sigma}\int d\mathbf{r}[c_{i\mu\sigma}^{\dagger}c_{i\mu\sigma}, \hat{\mathbf{J}}^{\mathrm{C}}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r})] \ , \tag{5.37}$$

where $\hat{\mathbf{J}}^{\mathrm{C}}(\mathbf{r})$ is given by Eq. (5.34). Substituting in the expression above, we have

$$\begin{aligned}
[c_{i\mu\sigma}^{\dagger}c_{i\mu\sigma}, \hat{\mathbf{J}}^{\mathrm{C}}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r})] =& \frac{e}{2m}\sum_{\sigma'}\sum_{\substack{mn\\ \alpha\beta}}[c_{i\mu\sigma}^{\dagger}c_{i\mu\sigma}, c_{m\alpha\sigma'}^{\dagger}c_{n\beta\sigma'}]\mathbf{w}_{mn}^{\alpha\beta}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r}) \\
=& \frac{e}{2m}\sum_{\sigma'}\sum_{\substack{mn\\ \alpha\beta}}\left\{c_{i\mu\sigma}^{\dagger}c_{n\beta\sigma'}\delta_{im}\delta_{\mu\alpha}\delta_{\sigma\sigma'} - c_{m\alpha\sigma'}^{\dagger}c_{i\mu\sigma}\delta_{in}\delta_{\mu\beta}\delta_{\sigma\sigma'}\right\}\mathbf{w}_{mn}^{\alpha\beta}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r}) \\
=& \frac{e}{2m}\sum_{m\alpha}\left\{c_{i\mu\sigma}^{\dagger}c_{m\alpha\sigma}\mathbf{w}_{im}^{\mu\alpha}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r}) - c_{m\alpha\sigma}^{\dagger}c_{i\mu\sigma}\mathbf{w}_{mi}^{\alpha\mu}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r})\right\} \ .
\end{aligned} \tag{5.38}$$

Substituting back in Eq. (5.37),

$$\begin{aligned}
\frac{e}{i\hbar}\sum_{\mu\sigma}[c_{i\mu\sigma}^{\dagger}c_{i\mu\sigma}, \hat{H}_{\mathrm{ext}}] =& -\frac{e^2}{2im\hbar}\int d\mathbf{r}\sum_{\substack{m\mu\alpha\\ \sigma}}\left\{c_{i\mu\sigma}^{\dagger}c_{m\alpha\sigma}\mathbf{w}_{im}^{\mu\alpha}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r}) - c_{m\alpha\sigma}^{\dagger}c_{i\mu\sigma}\mathbf{w}_{mi}^{\alpha\mu}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r})\right\} \\
=& \frac{e}{i\hbar}\sum_{m}\sum_{\sigma}\sum_{\mu\alpha}\left\{D_{im}^{\mu\alpha}c_{i\mu\sigma}^{\dagger}c_{m\alpha\sigma} - D_{mi}^{\alpha\mu}c_{m\alpha\sigma}^{\dagger}c_{i\mu\sigma}\right\} \ ,
\end{aligned} \tag{5.39}$$

where we have defined

$$D_{im}^{\mu\alpha} = -\frac{e}{2m}\int d\mathbf{r}\mathbf{w}_{im}^{\mu\alpha}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r}) \ . \tag{5.40}$$

This term should be added to the RHS of Eq. (5.29). So, comparing both equations, we can see that the diamagnetic current can be written as

$$\hat{I}_{ij}^{\text{dia}} = \frac{e}{\hbar} i \sum_{\sigma} \sum_{\mu\nu} \left\{ D_{ij}^{\mu\nu} c_{i\mu\sigma}^{\dagger} c_{j\nu\sigma} - D_{ji}^{\nu\mu} c_{j\nu\sigma}^{\dagger} c_{i\mu\sigma} \right\} . \tag{5.41}$$

For the potential vector we consider, obtained in Eq. (5.215), the D components are given by

$$\begin{aligned} D_{ij}^{\mu\nu} =& - \frac{e}{2m} \int d\mathbf{r} \mathbf{w}_{ij}^{\mu\nu}(\mathbf{r}) \cdot \left\{ \frac{1}{2i\omega} E_0 \hat{\mathbf{u}}_E \left[e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} - e^{-i(\mathbf{q}\cdot\mathbf{r}-\omega t)} \right] \right\} \\ =& - \frac{eE_0}{4im\omega} \int d\mathbf{r} \mathbf{w}_{ij}^{\mu\nu}(\mathbf{r}) \cdot \hat{\mathbf{u}}_E \left[e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} - e^{-i(\mathbf{q}\cdot\mathbf{r}-\omega t)} \right] \\ =& - \frac{eE_0}{4im\omega} \left[\mathbf{w}_{ij}^{\mu\nu}(\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{-i\omega t} - \mathbf{w}_{ij}^{\mu\nu}(-\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{i\omega t} \right] , \end{aligned} \tag{5.42}$$

where $\mathbf{w}_{ij}^{\mu\alpha}(\mathbf{q})$ is the Fourier transform of $\mathbf{w}_{ij}^{\mu\alpha}(\mathbf{r})$, and is given by Eq. (5.219). For $\mathbf{q} = 0$ (uniform electric field), $\mathbf{w}(\mathbf{q} = 0)$ becomes the momentum operator (see Eq. (5.220)), and we have

$$\begin{aligned} D_{ij}^{\mu\nu} =& - \frac{eE_0}{2im\omega} \mathbf{p}_{ij}^{\mu\nu} \cdot \hat{\mathbf{u}}_E \left(e^{-i\omega t} - e^{i\omega t} \right) \\ =& \frac{eE_0}{m\omega} \mathbf{p}_{ij}^{\mu\nu} \cdot \hat{\mathbf{u}}_E \sin(\omega t) . \end{aligned} \tag{5.43}$$

5.1.5 Diamagnetic spin current

The spin current operator will also be modified by the external perturbation. Including the new term in the Heisenberg equation of motion, The term that should be added to the Eq. of motion 5.2 involves the commutator

$$\frac{1}{2i} \sum_{\mu,ss'} \sigma_{ss'}^{m} [c_{i\mu s}^{\dagger} c_{i\mu s'}, \hat{H}_{\text{ext}}] = - \frac{1}{2i} \sum_{\mu,ss'} \sigma_{ss'}^{m} [c_{i\mu s}^{\dagger} c_{i\mu s'}, \hat{\mathbf{J}}^{\text{C}}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r})] , \tag{5.44}$$

where $\hat{\mathbf{J}}^{\text{C}}(\mathbf{r})$ is given by Eq. (5.34). Substituting in the expression above, we have

$$\begin{aligned} [c_{i\mu s}^{\dagger} c_{i\mu s'}, \hat{\mathbf{J}}^{\text{C}}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r})] =& \frac{e}{2m} \sum_{\sigma'} \sum_{\substack{mn \\ \alpha\beta}} [c_{i\mu s}^{\dagger} c_{i\mu s'}, c_{m\alpha\sigma'}^{\dagger} c_{n\beta\sigma'}] \mathbf{w}_{mn}^{\alpha\beta}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) \\ =& \frac{e}{2m} \sum_{\sigma'} \sum_{\substack{mn \\ \alpha\beta}} \left\{ c_{i\mu s}^{\dagger} c_{n\beta\sigma'} \delta_{im} \delta_{\mu\alpha} \delta_{s'\sigma'} - c_{m\alpha\sigma'}^{\dagger} c_{i\mu s'} \delta_{in} \delta_{\mu\beta} \delta_{s\sigma'} \right\} \mathbf{w}_{mn}^{\alpha\beta}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) \\ =& \frac{e}{2m} \sum_{m\alpha} \left\{ c_{i\mu s}^{\dagger} c_{m\alpha s'} \mathbf{w}_{im}^{\mu\alpha}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) - c_{m\alpha s}^{\dagger} c_{i\mu s'} \mathbf{w}_{mi}^{\alpha\mu}(\mathbf{r}) \cdot \mathbf{A}(\mathbf{r}) \right\} . \end{aligned} \tag{5.45}$$

Substituting back in Eq. (5.44),

$$\begin{aligned}\frac{1}{2i}\sum_{\mu,ss'}\sigma^m_{ss'}[c^\dagger_{i\mu s}c_{i\mu s'},\hat{H}_{\text{ext}}] =& -\frac{e}{4im}\int d\mathbf{r}\sum_{j\alpha}\sum_{\mu,ss'}\left\{c^\dagger_{i\mu s}\sigma^m_{ss'}c_{j\alpha s'}\mathbf{w}^{\mu\alpha}_{ij}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r}) - c^\dagger_{j\alpha s}\sigma^m_{ss'}c_{i\mu s'}\mathbf{w}^{\alpha\mu}_{ji}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r})\right\} \\ =&\frac{i}{2}\sum_j\sum_{\mu\alpha}\sum_{ss'}\left\{D^{\mu\alpha}_{ij}c^\dagger_{i\mu s}\sigma^m_{ss'}c_{j\alpha s'} - D^{\alpha\mu}_{ji}c^\dagger_{j\alpha s}\sigma^m_{ss'}c_{i\mu s'}\right\} \ ,\end{aligned} \tag{5.46}$$

where we have defined

$$D^{\mu\alpha}_{im} = -\frac{e}{2m}\int d\mathbf{r}\mathbf{w}^{\mu\alpha}_{im}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r}) \ . \tag{5.47}$$

This term should be added to the RHS of Eq. (5.3). So, comparing both equations, we can see that the diamagnetic spin current is

$$\hat{I}^{m,\text{dia}}_{ij} = \frac{i}{2}\sum_\sigma\sum_{\mu\nu}\sum_{ss'}\left\{D^{\mu\nu}_{ij}c^\dagger_{i\mu s}\sigma^m_{ss'}c_{j\nu s'} - D^{\nu\mu}_{ji}c^\dagger_{j\nu s}\sigma^m_{ss'}c_{i\mu s'}\right\} \ . \tag{5.48}$$

For the potential vector we consider, D components are given by Eq. (5.43).

5.1.6 Orbital Angular Momentum currents

Following the same steps as before, we can write a discrete continuity equation for the orbital angular momentum as

$$\frac{d\mathbf{L}_i}{dt} + \sum_n \mathbf{I}^L_{in} = \boldsymbol{\tau}_i \ . \tag{5.49}$$

Here, τ_i represents the torque exerted by external sources (magnetic fields and effective fields such as spin-orbit coupling). The m-component of the orbital angular momentum operator is given by

$$\hat{L}^m_i = \sum_\sigma\sum_{\mu\nu}L^m_{\mu\nu}c^\dagger_{i\mu\sigma}c_{i\nu\sigma} \ , \tag{5.50}$$

where the matrices $L^m_{\mu\nu}$ were obtained for the real spherical harmonics in appendix D. Once again,

$$\begin{aligned}\frac{d\hat{L}^m_i}{dt} =& \frac{1}{i\hbar}[\hat{L}^m_i,\hat{H}] \\ =& \frac{1}{i\hbar}\sum_{\mu\nu\sigma}L^m_{\mu\nu}[c^\dagger_{i\mu\sigma}c_{i\nu\sigma},\hat{H}] \\ =& \frac{1}{i\hbar}\sum_{\mu\nu\sigma}L^m_{\mu\nu}[c^\dagger_{i\mu\sigma}c_{i\nu\sigma},\hat{H}_0+\hat{H}_{\text{int}}+\hat{H}_{\text{so}}+\hat{H}_{\text{ZS}}+\hat{H}_{\text{ZL}}] \ .\end{aligned} \tag{5.51}$$

The first term on the right hand side is given by

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma}c_{i\nu\sigma},\hat{H}_0] &= \sum_{\substack{mn\\ \sigma'}}\sum_{\alpha\beta} t^{\alpha\beta}_{mn}[c^\dagger_{i\mu\sigma}c_{i\nu\sigma}, c^\dagger_{m\alpha\sigma'}c_{n\beta\sigma'}] \\
&= \sum_{\substack{mn\\ \sigma'}}\sum_{\alpha\beta} t^{\alpha\beta}_{mn}\left\{c^\dagger_{i\mu\sigma}c_{n\beta\sigma'}\delta_{im}\delta_{\nu\alpha}\delta_{\sigma\sigma'} - c^\dagger_{m\alpha\sigma'}c_{i\nu\sigma}\delta_{in}\delta_{\mu\beta}\delta_{\sigma\sigma'}\right\} \\
&= \sum_{m\alpha}\left\{t^{\nu\alpha}_{im}c^\dagger_{i\mu\sigma}c_{m\alpha\sigma} - t^{\alpha\mu}_{mi}c^\dagger_{m\alpha\sigma}c_{i\nu\sigma}\right\} \ .
\end{aligned} \tag{5.52}
$$

So,

$$
\frac{1}{i\hbar}\sum_{\mu\nu\sigma} L^m_{\mu\nu}[c^\dagger_{i\mu\sigma}c_{i\nu\sigma},\hat{H}_0] = \frac{1}{i\hbar}\sum_{n\sigma}\sum_{\alpha\mu\nu} L^m_{\mu\nu}\left\{t^{\nu\alpha}_{in}c^\dagger_{i\mu\sigma}c_{n\alpha\sigma} - t^{\alpha\mu}_{ni}c^\dagger_{n\alpha\sigma}c_{i\nu\sigma}\right\} \ . \tag{5.53}
$$

The second one, related to the commutator between the orbital angular momentum with the electron-electron interaction part of the hamiltonian, can be written as

$$
[c^\dagger_{i\mu\sigma}c_{i\nu\sigma},\hat{H}_{\text{int}}] = \frac{1}{2}\sum_{\substack{m\\ s,s'}}\sum_{\alpha\beta\gamma\xi} U^m_{\alpha\beta\gamma\xi}[c^\dagger_{i\mu\sigma}c_{i\nu\sigma}, c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}] \ . \tag{5.54}
$$

The commutator above is given by

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma}c_{i\nu\sigma}, c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}] &= c^\dagger_{i\mu\sigma}c_{i\nu\sigma}c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s} - c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}c^\dagger_{i\mu\sigma}c_{i\nu\sigma} \\
&= c^\dagger_{i\mu\sigma}c^\dagger_{m\beta s'}c_{m\xi s'}c_{m\gamma s}\delta_{im}\delta_{\nu\alpha}\delta_{\sigma s} - c^\dagger_{i\mu\sigma}c^\dagger_{m\alpha s}c_{m\xi s'}c_{m\gamma s}\delta_{im}\delta_{\nu\beta}\delta_{\sigma s'} \\
&\quad + c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\gamma s}c_{i\nu\sigma}\delta_{im}\delta_{\mu\xi}\delta_{\sigma s'} - c^\dagger_{m\alpha s}c^\dagger_{m\beta s'}c_{m\xi s'}c_{i\nu\sigma}\delta_{im}\delta_{\mu\gamma}\delta_{\sigma s} \ .
\end{aligned} \tag{5.55}
$$

Substituting it back, we have

$$
\begin{aligned}
[c^\dagger_{i\mu\sigma}c_{i\nu\sigma},\hat{H}_{\text{int}}] = \frac{1}{2}\sum_{s,s'}\sum_{\alpha\beta\gamma\xi} U^i_{\alpha\beta\gamma\xi}\Big\{ & c^\dagger_{i\mu\sigma}c^\dagger_{i\beta s'}c_{i\xi s'}c_{i\gamma s}\delta_{\nu\alpha}\delta_{\sigma s} - c^\dagger_{i\mu\sigma}c^\dagger_{i\alpha s}c_{i\xi s'}c_{i\gamma s}\delta_{\nu\beta}\delta_{\sigma s'} \\
& + c^\dagger_{i\alpha s}c^\dagger_{i\beta s'}c_{i\gamma s}c_{i\nu\sigma}\delta_{\mu\xi}\delta_{\sigma s'} - c^\dagger_{i\alpha s}c^\dagger_{i\beta s'}c_{i\xi s'}c_{i\nu\sigma}\delta_{\mu\gamma}\delta_{\sigma s}\Big\} \ .
\end{aligned} \tag{5.56}
$$

Using Lowde-Windsor approximation, i.e., $U^i_{\alpha\beta\gamma\xi} = U^i\delta_{\alpha\xi}\delta_{\beta\gamma}$, it's possible to show, using the equation above, that

$$
\sum_{\mu\nu\sigma} L^m_{\mu\nu}[c^\dagger_{i\mu\sigma}c_{i\nu\sigma},\hat{H}_{\text{int}}] = 0 \ . \tag{5.57}
$$

All the other terms correspond to "external" (with respect to orbital angular momentum) torques - due to external magnetic fields or effective magnetic field from the spin-orbit coupling -, so they cancel with the RHS of the continuity equation.

Finally, we can rewrite the continuity equation for the orbital angular momentum as

$$\frac{d\hat{L}_i^m}{dt} - \frac{1}{i\hbar}\sum_{n\sigma}\sum_{\alpha\mu\nu} L_{\mu\nu}^m \left\{ t_{in}^{\nu\alpha} c_{i\mu\sigma}^\dagger c_{n\alpha\sigma} - t_{ni}^{\alpha\mu} c_{n\alpha\sigma}^\dagger c_{i\nu\sigma} \right\} = 0 \ . \tag{5.58}$$

Comparing to Eq. (5.49), we see that the orbital angular momentum current vector between sites i and n is given by

$$\begin{aligned} \mathbf{I}_{ij}^L &= \frac{i}{\hbar}\sum_{\sigma}\sum_{\alpha\mu\nu} \mathbf{L}_{\mu\nu} \left\{ t_{ij}^{\nu\alpha} c_{i\mu\sigma}^\dagger c_{j\alpha\sigma} - t_{ji}^{\alpha\mu} c_{j\alpha\sigma}^\dagger c_{i\nu\sigma} \right\} \\ &= \frac{i}{\hbar}\sum_{\sigma}\sum_{\alpha\mu\nu} \left\{ \mathbf{L}_{\alpha\mu} t_{ij}^{\mu\nu} c_{i\alpha\sigma}^\dagger c_{j\nu\sigma} - t_{ji}^{\nu\mu} \mathbf{L}_{\mu\alpha} c_{j\nu\sigma}^\dagger c_{i\alpha\sigma} \right\} \ , \end{aligned} \tag{5.59}$$

or, the $m-$component,

$$\begin{aligned} I_{ij}^{Lm} &= \frac{i}{\hbar}\sum_{\sigma}\sum_{\alpha\mu\nu} L_{\mu\nu}^m \left\{ t_{ij}^{\nu\alpha} c_{i\mu\sigma}^\dagger c_{j\alpha\sigma} - t_{ji}^{\alpha\mu} c_{j\alpha\sigma}^\dagger c_{i\nu\sigma} \right\} \\ &= \frac{i}{\hbar}\sum_{\sigma}\sum_{\alpha\mu\nu} \left\{ L_{\alpha\mu}^m t_{ij}^{\mu\nu} c_{i\alpha\sigma}^\dagger c_{j\nu\sigma} - t_{ji}^{\nu\mu} L_{\mu\alpha}^m c_{j\nu\sigma}^\dagger c_{i\alpha\sigma} \right\} \ , \end{aligned} \tag{5.60}$$

5.2 Currents and torque operators in multilayers

5.2.1 Charge current

The charge current operator is given by Eq. (5.30), which can be rewritten as

$$\hat{I}_{ij}^{\mathrm{C}}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel) = \frac{e}{\hbar} i \sum_{\sigma}\sum_{\mu\nu} \left\{ t_{ij}^{\mu\nu}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel) c_{i\mu\sigma}^\dagger(\mathbf{R}_\parallel) c_{j\nu\sigma}(\mathbf{R}'_\parallel) - t_{ji}^{\nu\mu}(\mathbf{R}'_\parallel, \mathbf{R}_\parallel) c_{j\nu\sigma}^\dagger(\mathbf{R}'_\parallel) c_{i\mu\sigma}(\mathbf{R}_\parallel) \right\} \ . \tag{5.61}$$

where i and j now denotes layers and $\mathbf{R}_\parallel, \mathbf{R}'_\parallel$ are vectors inside these layers. Using the Fourier transformation of the creation operator,

$$c_{i\mu\sigma}^\dagger(\mathbf{R}_\parallel) = \frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel\cdot\mathbf{R}_\parallel} c_{i\mu\sigma}^\dagger(\mathbf{k}_\parallel) \ , \tag{5.62}$$

and its conjugate

$$c_{j\nu\sigma}(\mathbf{R}'_\parallel) = \frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel} e^{i\mathbf{k}_\parallel\cdot\mathbf{R}'_\parallel} c_{j\nu\sigma}(\mathbf{k}_\parallel) \ , \tag{5.63}$$

we can write

$$\hat{I}^{\rm C}_{ij}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel) = \frac{e}{\hbar}\frac{i}{N_\parallel^2}\sum_\sigma\sum_{\mu\nu}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel}\Big\{t^{\mu\nu}_{ij}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)c^\dagger_{i\mu\sigma}(\mathbf{k}_\parallel)c_{j\nu\sigma}(\mathbf{k}'_\parallel)e^{-i\mathbf{k}_\parallel\cdot\mathbf{R}_\parallel}e^{i\mathbf{k}'_\parallel\cdot\mathbf{R}'_\parallel}$$
$$-t^{\nu\mu}_{ji}(\mathbf{R}'_\parallel,\mathbf{R}_\parallel)c^\dagger_{j\nu\sigma}(\mathbf{k}_\parallel)c_{i\mu\sigma}(\mathbf{k}'_\parallel)e^{-i\mathbf{k}_\parallel\cdot\mathbf{R}'_\parallel}e^{i\mathbf{k}'_\parallel\cdot\mathbf{R}_\parallel}\Big\}$$
$$= \frac{e}{\hbar}\frac{i}{N_\parallel^2}\sum_\sigma\sum_{\mu\nu}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel}\Big\{t^{\mu\nu}_{ij}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)c^\dagger_{i\mu\sigma}(\mathbf{k}_\parallel)c_{j\nu\sigma}(\mathbf{k}'_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}e^{i(\mathbf{k}'_\parallel-\mathbf{k}_\parallel)\cdot\mathbf{R}'_\parallel}$$
$$-t^{\nu\mu}_{ji}(\mathbf{R}'_\parallel,\mathbf{R}_\parallel)c^\dagger_{j\nu\sigma}(\mathbf{k}_\parallel)c_{i\mu\sigma}(\mathbf{k}'_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}e^{i(\mathbf{k}'_\parallel-\mathbf{k}_\parallel)\cdot\mathbf{R}_\parallel}\Big\}$$
$$= \frac{e}{\hbar}\frac{i}{N_\parallel^2}\sum_\sigma\sum_{\mu\nu}\sum_{\mathbf{k}_\parallel,\mathbf{q}_\parallel}\Big\{t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)c^\dagger_{i\mu\sigma}(\mathbf{k}_\parallel)c_{j\nu\sigma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}e^{i\mathbf{q}_\parallel\cdot\mathbf{R}'_\parallel}$$
$$-t^{\nu\mu}_{ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)c^\dagger_{j\nu\sigma}(\mathbf{k}_\parallel)c_{i\mu\sigma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}\Big\} \ . \tag{5.64}$$

We have used the fact that, in multilayers, $t^{\mu\nu}_{ij}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel) = t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)$. Finally, we can write this equation as

$$\hat{I}^{\rm C}_{ij}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel) = \frac{1}{N_\parallel}\sum_{\mathbf{q}_\parallel}\Big\{\hat{\mathcal{I}}^{\rm C}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{q}_\parallel)e^{i\mathbf{q}_\parallel\cdot\mathbf{R}'_\parallel} - \hat{\mathcal{I}}^{\rm C}_{ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel,\mathbf{q}_\parallel)e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}\Big\} \ , \tag{5.65}$$

where

$$\hat{\mathcal{I}}^{\rm C}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{q}_\parallel) = \frac{ie}{\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_\sigma\sum_{\mu\nu}t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)c^\dagger_{i\mu\sigma}(\mathbf{k}_\parallel)c_{j\nu\sigma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)} \ . \tag{5.66}$$

5.2.2 Diamagnetic current

For multilayers, the diamagnetic current follows the same derivation as above and Eq. (5.41) becomes,

$$\hat{I}^{\rm dia}_{\ell\ell'}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel) = \frac{1}{N_\parallel}\sum_{\mathbf{q}_\parallel}\Big\{\hat{\mathcal{I}}^{\rm dia}_{\ell\ell'}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{q}_\parallel)e^{i\mathbf{q}_\parallel\cdot\mathbf{R}'_\parallel} - \hat{\mathcal{I}}^{\rm dia}_{\ell'\ell}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel,\mathbf{q}_\parallel)e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}\Big\} \ , \tag{5.67}$$

where

$$\hat{\mathcal{I}}^{\rm dia}_{\ell\ell'}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{q}_\parallel) = \frac{ie}{\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_\sigma\sum_{\mu\nu}D^{\mu\nu}_{\ell\ell'}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel)c_{\ell'\nu\sigma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)} \ . \tag{5.68}$$

where

$$
\begin{aligned}
D^{\mu\nu}_{\ell\ell'}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) =& \frac{eE_0}{m\omega}\mathbf{p}^{\mu\nu}_{\ell\ell'}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t) \\
=& \frac{eE_0}{m\omega}\sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\mathbf{p}^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t)\ .
\end{aligned}
\tag{5.69}
$$

for uniform electric fields. Using the relation

$$
\frac{\hbar}{m}\hat{p}^\alpha = \sum_{\substack{\ell\ell' \\ \mu\nu}}\sum_{\mathbf{k}_\parallel\sigma}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{e}}^\alpha c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel)c_{\ell'\nu\sigma}(\mathbf{k}_\parallel)\ .
\tag{5.70}
$$

obtained in Appendix A, we can write

$$
\frac{\hbar}{m}\mathbf{p}^{\mu\nu}_{\ell\ell'}(\mathbf{R}_j - \mathbf{R}_i)\cdot\hat{\mathbf{u}}_E = \sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j - \mathbf{R}_i)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E
\tag{5.71}
$$

and

$$
D^{\mu\nu}_{\ell\ell'}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) = \frac{eE_0}{\hbar\omega}\sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t)\ .
\tag{5.72}
$$

With this definition, we can see that

$$
\begin{aligned}
\Big[D^{\mu\nu}_{\ell\ell'}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)\Big]^* =& \frac{eE_0}{\hbar\omega}\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\mu\nu}_{\ell\ell'}(-\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t) \\
=& -\frac{eE_0}{\hbar\omega}\sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t) \\
=& -D^{\mu\nu}_{\ell\ell'}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)
\end{aligned}
\tag{5.73}
$$

which shows that $D^{\mu\nu}_{\ell\ell'}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)$ is purely imaginary, and also

$$
\begin{aligned}
\Big[D^{\mu\nu}_{\ell\ell'}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)\Big]^* =& \frac{eE_0}{\hbar\omega}\sum_{\mathbf{k}} e^{i\mathbf{k}\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\nu\mu}_{\ell'\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t) \\
=& D^{\nu\mu}_{\ell'\ell}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)\ .
\end{aligned}
\tag{5.74}
$$

We can also obtain the following relation

$$
\begin{aligned}
D^{\mu\nu}_{\ell'\ell}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) =& \frac{eE_0}{\hbar\omega}\sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\mu\nu}_{\ell'\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t) \\
=& -\frac{eE_0}{\hbar\omega}\sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\mu\nu}_{\ell'\ell}(-\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t) \\
=& -\frac{eE_0}{\hbar\omega}\sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\nu\mu}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t) \\
=& -D^{\nu\mu}_{\ell\ell'}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)\ .
\end{aligned}
\tag{5.75}
$$

5.2.3 Spin current

The spin current operators with polarization m can be obtained comparing Eqs. 5.30 and 5.6 - instead of the generalized charge operator $\rho_{ij}^{\mu\nu} = e\sum_\sigma c_{i\mu\sigma}^\dagger c_{j\nu\sigma}$, we have the generalized spin operator $S_{ij}^{m\,\mu\nu} = \frac{\hbar}{2}\sum_{\alpha\beta} c_{i\mu\alpha}^\dagger \sigma_{\alpha\beta}^m c_{j\nu\beta}$. Thus,

$$\hat{I}_{ij}^m(\mathbf{R}_\parallel, \mathbf{R}'_\parallel) = \frac{1}{N_\parallel}\sum_{\mathbf{q}_\parallel}\left\{\hat{\mathcal{I}}_{ij}^m(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{q}_\parallel)e^{i\mathbf{q}_\parallel\cdot\mathbf{R}'_\parallel} - \hat{\mathcal{I}}_{ji}^m(\mathbf{R}_\parallel - \mathbf{R}'_\parallel, \mathbf{q}_\parallel)e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}\right\} , \tag{5.76}$$

where

$$\hat{\mathcal{I}}_{ij}^m(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{q}_\parallel) = \frac{i}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\mu\nu} t_{ij}^{\mu\nu}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) S_{ij}^{m\,\mu\nu}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{q}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \tag{5.77}$$

and the generalized spin operator in mixed representation is given by

$$S_{ij}^{m\,\mu\nu}(\mathbf{k}_\parallel, \mathbf{k}'_\parallel) = \frac{1}{2}\sum_{\alpha\beta} c_{i\mu\alpha}^\dagger(\mathbf{k}_\parallel)\sigma_{\alpha\beta}^m c_{j\nu\beta}(\mathbf{k}'_\parallel) . \tag{5.78}$$

The total spin current operator between layers i and j can be obtained summing Eq. (5.76) in all sites $\mathbf{R}_\parallel$ of plane i and all sites $\mathbf{R}'_\parallel$ of plane j, i.e.,

$$I_{ij}^m = \sum_{\mathbf{R}_\parallel, \mathbf{R}'_\parallel} \hat{I}_{ij}^m(\mathbf{R}_\parallel, \mathbf{R}'_\parallel) . \tag{5.79}$$

Since

$$\begin{aligned} t_{ij}^{\mu\nu}(\mathbf{k}_\parallel) &= \sum_{\mathbf{R}''_\parallel} e^{i\mathbf{k}_\parallel\cdot\mathbf{R}''_\parallel} t_{ij}^{\mu\nu}(\mathbf{R}''_\parallel) \\ &= \sum_{\mathbf{R}_\parallel} e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} t_{ij}^{\mu\nu}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) \end{aligned} \tag{5.80}$$

and

$$\sum_{\mathbf{R}'_\parallel} e^{i\mathbf{q}_\parallel\cdot\mathbf{R}'_\parallel} = N_\parallel \delta_{\mathbf{q}_\parallel,0} , \tag{5.81}$$

we end up with

$$I_{ij}^m = \frac{i}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\mu\nu}\left[t_{ij}^{\mu\nu}(\mathbf{k}_\parallel)\; S_{ij}^{m\,\mu\nu}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) - t_{ji}^{\nu\mu}(\mathbf{k}_\parallel)\; S_{ji}^{m\,\nu\mu}(\mathbf{k}_\parallel, \mathbf{k}_\parallel)\right] . \tag{5.82}$$

5.2.4 Orbital Angular Momentum Current

We can now write the orbital angular momentum current operator, given by Eq. (5.60), as

$$\hat{I}^{Lm}_{ij}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)=\frac{i}{\hbar}\sum_{\sigma}\sum_{\alpha\mu\nu}\Big\{L^m_{\alpha\mu}t^{\mu\nu}_{ij}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)c^\dagger_{i\alpha\sigma}(\mathbf{R}_\parallel)c_{j\nu\sigma}(\mathbf{R}'_\parallel)-t^{\nu\mu}_{ji}(\mathbf{R}'_\parallel,\mathbf{R}_\parallel)L^m_{\mu\alpha}c^\dagger_{j\nu\sigma}(\mathbf{R}'_\parallel)c_{i\alpha\sigma}(\mathbf{R}_\parallel)\Big\}\ . \tag{5.83}$$

Comparing with Eq. (5.61) and following the same steps, we obtain

$$\begin{aligned}\hat{I}^{Lm}_{ij}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)=\frac{e}{\hbar}\frac{i}{N_\parallel^2}\sum_{\sigma}\sum_{\alpha\mu\nu}\sum_{\mathbf{k}_\parallel,\mathbf{q}_\parallel}&\Big\{L^m_{\alpha\mu}t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)c^\dagger_{i\alpha\sigma}(\mathbf{k}_\parallel)c_{j\nu\sigma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}e^{i\mathbf{q}_\parallel\cdot\mathbf{R}'_\parallel}\\&-t^{\nu\mu}_{ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)L^m_{\mu\alpha}c^\dagger_{j\nu\sigma}(\mathbf{k}_\parallel)c_{i\alpha\sigma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}\Big\}\ .\end{aligned} \tag{5.84}$$

To simplify the analogies with other cases, we are going to define each term on the right hand side as

$$\hat{I}^{Lm}_{ij}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)=\frac{1}{N_\parallel}\sum_{\mathbf{q}_\parallel}\Big\{\hat{\mathcal{I}}^{Lm}_{(1)ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{q}_\parallel)e^{i\mathbf{q}_\parallel\cdot\mathbf{R}'_\parallel}-\hat{\mathcal{I}}^{Lm}_{(2)ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel,\mathbf{q}_\parallel)e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}\Big\}\ . \tag{5.85}$$

where

$$\hat{\mathcal{I}}^{Lm}_{(1)ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{q}_\parallel)=\frac{ie}{\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\sigma}\sum_{\alpha\mu\nu}L^m_{\alpha\mu}t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)c^\dagger_{i\alpha\sigma}(\mathbf{k}_\parallel)c_{j\nu\sigma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\ . \tag{5.86}$$

$$\hat{\mathcal{I}}^{Lm}_{(2)ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel,\mathbf{q}_\parallel)=\frac{ie}{\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\sigma}\sum_{\alpha\mu\nu}t^{\nu\mu}_{ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)L^m_{\mu\alpha}c^\dagger_{j\nu\sigma}(\mathbf{k}_\parallel)c_{i\alpha\sigma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}\ . \tag{5.87}$$

For $\mathbf{q}_\parallel=0$ and $m=x,y,z$ we have

$$\begin{aligned}\Big[\hat{\mathcal{I}}^{Lm}_{(1)ji}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,0)\Big]^\dagger&=-\frac{ie}{\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\sigma}\sum_{\alpha\mu\nu}L^m_{\mu\alpha}t^{\mu\nu}_{ji}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)c^\dagger_{j\nu\sigma}(\mathbf{k}_\parallel)c_{i\alpha\sigma}(\mathbf{k}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}\\&=-\frac{ie}{\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\sigma}\sum_{\alpha\mu\nu}t^{\nu\mu}_{ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)L^m_{\mu\alpha}c^\dagger_{j\nu\sigma}(\mathbf{k}_\parallel)c_{i\alpha\sigma}(\mathbf{k}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}\\&=-\hat{\mathcal{I}}^{Lm}_{(2)ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel,0)\ .\end{aligned} \tag{5.88}$$

5.2.5 Local spin torque

In multilayers, the torque operator given in Eq. (5.15) can be written as

$$\begin{aligned}
\tau_\ell^m(\mathbf{q}_\parallel) &= \sum_{\mathbf{R}_\parallel} e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}\tau_\ell^m(\mathbf{R}_\parallel) \\
&= \frac{\lambda_\ell}{2}\sum_{\mathbf{R}_\parallel}\sum_{\substack{nk\\ \mu\nu}}\sum_{\alpha\beta} e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}\epsilon_{mnk}L'^n_{\mu\nu}c^\dagger_{\ell\mu\alpha}(\mathbf{R}_\parallel)\sigma^k_{\alpha\beta}c_{\ell\nu\beta}(\mathbf{R}_\parallel) \\
&= \frac{\lambda_\ell}{2N^2}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_{\mathbf{R}_\parallel}\sum_{\substack{nk\\ \mu\nu}}\sum_{\alpha\beta} e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}e^{-i\mathbf{k}'_\parallel\cdot\mathbf{R}_\parallel}\epsilon_{mnk}L'^n_{\mu\nu}c^\dagger_{\ell\mu\alpha}(\mathbf{k}'_\parallel)\sigma^k_{\alpha\beta}\; e^{i\mathbf{k}_\parallel\cdot\mathbf{R}_\parallel}c_{\ell\nu\beta}(\mathbf{k}_\parallel) \\
&= \frac{\lambda_\ell}{2N}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_{\substack{nk\\ \mu\nu}}\sum_{\alpha\beta}\delta_{\mathbf{k}'_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel}\epsilon_{mnk}L'^n_{\mu\nu}c^\dagger_{\ell\mu\alpha}(\mathbf{k}'_\parallel)\sigma^k_{\alpha\beta}c_{\ell\nu\beta}(\mathbf{k}_\parallel) \\
&= \frac{\lambda_\ell}{2N}\sum_{\mathbf{k}_\parallel}\sum_{\substack{nk\\ \mu\nu}}\sum_{\alpha\beta}\epsilon_{mnk}L'^n_{\mu\nu}c^\dagger_{\ell\mu\alpha}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)\sigma^k_{\alpha\beta}c_{\ell\nu\beta}(\mathbf{k}_\parallel) \\
&= \frac{\lambda_\ell}{N}\sum_{\mathbf{k}_\parallel}\sum_{\substack{nk\\ \mu\nu}}\epsilon_{mnk}L'^n_{\mu\nu}S^{k\,\mu\nu}_{\ell\ell}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\mathbf{k}_\parallel)\ .
\end{aligned} \tag{5.89}$$

Analogously, assuming that the effective fields are uniform inside the layers, the torque in Eq. (5.17) are given by

$$\tau_\ell^m(\mathbf{q}_\parallel) = \frac{2}{N}\sum_{\mathbf{k}_\parallel}\sum_{\substack{nk\\ \mu\nu}}\epsilon_{mnk}B_\ell^{n\,\mu\nu}S^{k\,\mu\nu}_{\ell\ell}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\mathbf{k}_\parallel)\ . \tag{5.90}$$

The xc-torque, given by Eq. (5.19) in real space, is given by

$$\tau_\ell^m(\mathbf{q}_\parallel) = -\frac{U_\ell}{N}\sum_{\mathbf{k}_\parallel}\sum_{\substack{nk\\ \mu}}\epsilon_{mnk}\langle m_\ell^n\rangle S^{k\,\mu\mu}_{\ell\ell}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\mathbf{k}_\parallel)\ . \tag{5.91}$$

This is basically the spin density operator, renormalized by the xc-field.

5.2.6 Torque-torque correlation

The most general form for the susceptibility matrix is given by Eq.(3.1), that we can write as

$$\chi^{mm'\,\mu\nu\gamma\xi}_{ijkl}(t) = -\frac{i}{4\hbar}\Theta(t)\langle[c^\dagger_{i\mu\alpha}(t)\sigma^m_{\alpha\alpha'}c_{j\nu\alpha'}(t), c^\dagger_{k\gamma\beta}\sigma^{m'}_{\beta\beta'}c_{\ell\xi\beta'}]\rangle\ . \tag{5.92}$$

where m, n are the cartesian components x, y, z and the density. It can also be written in the $+, \uparrow, \downarrow, -$ basis.

The local torque-torque correlation function can then be written in terms of this generalized susceptibility as

$$\chi_{\tau_i\tau_i}^{mm'}(t) = -\frac{i}{\hbar}\Theta(t)\langle[\tau_i^m(t), \tau_i^{m'}]\rangle \ . \tag{5.93}$$

where the torque operator in real space is given by 5.15. Hence,

$$\begin{aligned}
\chi_{\tau_i\tau_i}^{mm'}(t) =& -\frac{i}{\hbar}\Theta(t)\langle[\lambda_i \sum_{\substack{nk\\ \mu\nu}} \epsilon_{mnk} L'^{n}_{\mu\nu} S_{ii}^{k\,\mu\nu}(t), \lambda_i \sum_{\substack{n'k'\\ \gamma\xi}} \epsilon_{m'n'k'} L'^{n'}_{\gamma\xi} S_{ii}^{k'\,\gamma\xi}]\rangle \\
=& -(\lambda_i)^2 \sum_{\substack{nk\\ \mu\nu}} \epsilon_{mnk} L'^{n}_{\mu\nu} \sum_{\substack{n'k'\\ \gamma\xi}} \epsilon_{m'n'k'} L'^{n'}_{\gamma\xi} \frac{i}{\hbar}\Theta(t)\langle[S_{ii}^{k\,\mu\nu}(t), S_{ii}^{k'\,\gamma\xi}]\rangle \\
=& \lambda_i^2 \sum_{\substack{nk\\ \mu\nu}} \epsilon_{mnk} L'^{n}_{\mu\nu} \sum_{\substack{n'k'\\ \gamma\xi}} \epsilon_{m'n'k'} L'^{n'}_{\gamma\xi} \left\{-\frac{i}{4\hbar}\Theta(t)\langle[c^\dagger_{i\mu\alpha}(t)\sigma^k_{\alpha\alpha'}c_{i\nu\alpha'}(t), c^\dagger_{i\gamma\beta}(t)\sigma^{k'}_{\beta\beta'}c_{i\xi\beta'}]\rangle\right\} \\
=& \lambda_i^2 \sum_{\substack{nk\\ \mu\nu}} \epsilon_{mnk} L'^{n}_{\mu\nu} \sum_{\substack{n'k'\\ \gamma\xi}} \epsilon_{m'n'k'} L'^{n'}_{\gamma\xi} \chi_{iiii}^{kk'\,\mu\nu\gamma\xi}(t) \ .
\end{aligned} \tag{5.94}$$

5.2.7 Torque-spin correlation

The local torque-spin correlation function can also be written in terms of the generalized susceptibility as

$$\chi_{\tau_i S_i}^{mm'}(t) = -\frac{i}{\hbar}\Theta(t)\langle[\tau_i^m(t), S_i^{m'}]\rangle \ . \tag{5.95}$$

where the torque operator in real space is given by 5.15. Hence,

$$\begin{aligned}
\chi_{\tau_i S_i}^{mm'}(t) =& -\frac{i}{\hbar}\Theta(t)\langle[\lambda_i \sum_{\substack{nk\\ \mu\nu}} \epsilon_{mnk} L'^{n}_{\mu\nu} S_{ii}^{k\,\mu\nu}(t), \sum_{\gamma} S_{ii}^{m'\,\gamma\gamma}]\rangle \\
=& -\lambda_i \sum_{\substack{nk\\ \mu\nu}} \epsilon_{mnk} L'^{n}_{\mu\nu} \sum_{\gamma} \frac{i}{\hbar}\Theta(t)\langle[S_{ii}^{k\,\mu\nu}(t), S_{ii}^{m'\,\gamma\gamma}]\rangle \\
=& \lambda_i \sum_{\substack{nk\\ \mu\nu}} \epsilon_{mnk} L'^{n}_{\mu\nu} \sum_{\gamma} \left\{-\frac{i}{4\hbar}\Theta(t)\langle[c^\dagger_{i\mu\alpha}(t)\sigma^k_{\alpha\alpha'}c_{i\nu\alpha'}(t), c^\dagger_{i\gamma\beta}(t)\sigma^{m'}_{\beta\beta'}c_{i\gamma\beta'}]\rangle\right\} \\
=& \lambda_i \sum_{\substack{nk\\ \mu\nu}} \epsilon_{mnk} L'^{n}_{\mu\nu} \sum_{\gamma} \chi_{iiii}^{km'\,\mu\nu\gamma\gamma}(t) \ .
\end{aligned} \tag{5.96}$$

5.3 Magnetic Perturbations

First, let us calculate the spin currents generated by an oscillatory transverse magnetic field. After that, we will obtain the charge currents generated by the same magnetic field (Inverse Spin Hall Effect) and the spin currents generated by an electric field (Spin Hall Effect).

Besides a static magnetic field $\mathbf{B}' = B_0'\hat{z}$, we are going to consider an oscillatory transverse magnetic field written in the spin coordinates as

$$\mathbf{b}'_\perp(t) = b_0 \left[\cos(\omega t)\hat{\mathbf{x}}' - \sin(\omega t)\hat{\mathbf{y}}'\right] \tag{5.97}$$

which couples to the spin density and to the orbital angular momentum at sites ℓ. The first contribution is described by the interaction hamiltonian

$$\begin{aligned}\hat{H}_{S\perp} =& g_S\mu_B\mathbf{b}'_\perp(t)\cdot\sum_\ell \hat{\mathbf{S}}_\ell(t)\\ =& g_S\mu_B b_0\sum_\ell\left[\cos(\omega t)\hat{S}^x_\ell - \sin(\omega t)\hat{S}^y_\ell\right]\\ =& \frac{g_S\mu_B b_0}{2}\sum_\ell\left[e^{i\omega t}\hat{S}^+_\ell + e^{-i\omega t}\hat{S}^-_\ell\right]\ .\end{aligned} \tag{5.98}$$

To calculate the perturbation due to the orbital angular momentum, we need to consider the orbital angular momentum in the spin coordinates (obtained in appendix D). So, the perturbation hamiltonian due to the coupling with the angular momentum is

$$\begin{aligned}\hat{H}_{L\perp} =& g_L\mu_B\sum_\ell \mathbf{b}'_\perp(t)\cdot\hat{\mathbf{L}}'_\ell(t)\\ =& \frac{g_L\mu_B\sum_\ell}{2}\left(b'^+_\perp\hat{L}'^-_\ell + b'^-_\perp\hat{L}'^+_\ell + 2b'^z_\perp\hat{L}'^z_\ell\right)\\ =& \frac{\mu_B b_0}{2}\sum_\ell e^{-i\omega t}\left\{\hat{L}^x_\ell\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] + \hat{L}^y_\ell\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] - \hat{L}^z_\ell\sin(\theta)\right\}\\ &+e^{i\omega t}\left\{\hat{L}^x_\ell\left[\cos(\theta)\cos(\phi)-i\sin(\phi)\right] + \hat{L}^y_\ell\left[\cos(\theta)\sin(\phi)+i\cos(\phi)\right] - \hat{L}^z_\ell\sin(\theta)\right\}\ .\end{aligned} \tag{5.99}$$

In second quantization,

$$\hat{H}_{L\perp} = \frac{\mu_B b_0}{2}\sum_{\substack{\ell,\gamma\xi\\ s}}\left\{e^{-i\omega t}\hat{L}'^-_{\gamma\xi} + e^{i\omega t}\hat{L}'^+_{\gamma\xi}\right\}c^\dagger_{\ell\gamma s}c_{\ell\xi s}\ , \tag{5.100}$$

where the matrices

$$\begin{aligned}\hat{L}'^-_{\gamma\xi} =& \hat{L}^x_{\gamma\xi}\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] + \hat{L}^y_{\gamma\xi}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] - \hat{L}^z_{\gamma\xi}\sin(\theta) \text{ and}\\ \hat{L}'^+_{\gamma\xi} =& \hat{L}^x_{\gamma\xi}\left[\cos(\theta)\cos(\phi)-i\sin(\phi)\right] + \hat{L}^y_{\gamma\xi}\left[\cos(\theta)\sin(\phi)+i\cos(\phi)\right] - \hat{L}^z_{\gamma\xi}\sin(\theta)\end{aligned} \tag{5.101}$$

are calculated in appendix D.

For multilayers, the external magnetic field in the spin coordinate system can be written as

$$\mathbf{b}'_\perp(\mathbf{R}_\parallel, t) = b_0\left[\cos(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel)\hat{\mathbf{x}}' - \sin(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel)\hat{\mathbf{y}}'\right] \tag{5.102}$$

and the spin contribution to the interaction hamiltonian is given by

$$\hat{H}_{S\perp} = \frac{g_S\mu_B b_0}{2}\sum_\ell \left\{e^{i\omega t}S_\ell^+(-\mathbf{Q}_\parallel,t) + e^{-i\omega t}S_\ell^-(\mathbf{Q}_\parallel,t)\right\} \ , \tag{5.103}$$

where

$$\begin{aligned} S_\ell^+(-\mathbf{Q}_\parallel) &= \frac{1}{N_\parallel}\sum_\gamma\sum_{\mathbf{k}_\parallel} c^\dagger_{\ell\gamma\uparrow}(\mathbf{k}_\parallel - \mathbf{Q}_\parallel)c_{\ell\gamma\downarrow}(\mathbf{k}_\parallel) \\ &= \frac{1}{N_\parallel}\sum_\gamma\sum_{\mathbf{k}_\parallel} S_{\ell\ell}^{+\gamma\gamma}(\mathbf{k}_\parallel - \mathbf{Q}_\parallel, \mathbf{k}_\parallel) \ , \text{ and} \end{aligned} \tag{5.104}$$

$$\begin{aligned} S_\ell^-(\mathbf{Q}_\parallel) &= \frac{1}{N_\parallel}\sum_\gamma\sum_{\mathbf{k}_\parallel} c^\dagger_{\ell\gamma\downarrow}(\mathbf{k}_\parallel + \mathbf{Q}_\parallel)c_{\ell\gamma\uparrow}(\mathbf{k}_\parallel) \\ &= \frac{1}{N_\parallel}\sum_\gamma\sum_{\mathbf{k}_\parallel} S_{\ell\ell}^{-\gamma\gamma}(\mathbf{k}_\parallel + \mathbf{Q}_\parallel, \mathbf{k}_\parallel) \end{aligned} \tag{5.105}$$

are the $\mathbf{Q}_\parallel$ and $-\mathbf{Q}_\parallel$ components of the Fourier transform of $S_\ell^-(\mathbf{R}_\parallel)$ and $S_\ell^+(\mathbf{R}_\parallel)$, respectively. Note that $[S_\ell^-(\mathbf{Q}_\parallel)]^\dagger = S_\ell^+(-\mathbf{Q}_\parallel)$.

The orbital angular momentum contribution in this case can be found following the same steps as before, and we get

$$\hat{H}_{L\perp} = \frac{\mu_B b_0}{2}\sum_{\substack{\ell,\gamma\xi \\ s}}\sum_{\mathbf{R}_\parallel}\left\{e^{i\left(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel\right)}\hat{L}'^+_{\gamma\xi} + e^{-i\left(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel\right)}\hat{L}'^-_{\gamma\xi}\right\} c^\dagger_{\ell\gamma s}(\mathbf{R}_\parallel)c_{\ell\xi s}(\mathbf{R}_\parallel) \ . \tag{5.106}$$

Using the Fourier transform of the creation and annihilation operators given by Eqs. 5.62 and 5.63,

$$\begin{aligned} \hat{H}_{L\perp} &= \frac{\mu_B b_0}{2}\sum_{\substack{\ell,\gamma\xi \\ s}}\frac{1}{N_\parallel^2}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_{\mathbf{R}_\parallel}\left\{e^{i\left(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel\right)}\hat{L}'^+_{\gamma\xi} + e^{-i\left(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel\right)}\hat{L}'^-_{\gamma\xi}\right\} \ e^{-i\left(\mathbf{k}_\parallel - \mathbf{k}'_\parallel\right)\cdot\mathbf{R}_\parallel} \\ &\quad \times c^\dagger_{\ell\gamma s}(\mathbf{k}_\parallel)c_{\ell\xi s}(\mathbf{k}'_\parallel) \\ &= \frac{\mu_B b_0}{2}\sum_{\substack{\ell,\gamma\xi \\ s}}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\left\{e^{i\omega t}\hat{L}'^+_{\gamma\xi}c^\dagger_{\ell\gamma s}(\mathbf{k}_\parallel - \mathbf{Q}_\parallel)c_{\ell\xi s}(\mathbf{k}_\parallel) + e^{-i\omega t}\hat{L}'^-_{\gamma\xi}c^\dagger_{\ell\gamma s}(\mathbf{k}_\parallel + \mathbf{Q}_\parallel)c_{\ell\xi s}(\mathbf{k}_\parallel)\right\} \ . \end{aligned} \tag{5.107}$$

Finally, we can write this result as

$$\hat{H}_{L\perp} = \frac{\mu_B b_0}{2}\sum_{\substack{\ell,\gamma\xi \\ s}}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\left\{e^{i\omega t}\hat{L}'^+_{\gamma\xi}S_{\ell\ell}^{s\gamma\xi}(\mathbf{k}_\parallel - \mathbf{Q}_\parallel, \mathbf{k}_\parallel) + e^{-i\omega t}\hat{L}'^-_{\gamma\xi}S_{\ell\ell}^{s\gamma\xi}(\mathbf{k}_\parallel + \mathbf{Q}_\parallel, \mathbf{k}_\parallel)\right\} \ , \tag{5.108}$$

where $s = \uparrow$ or $\downarrow$ and

$$\sigma^\uparrow = \begin{pmatrix} 2 & 0 \\ 0 & 0 \end{pmatrix} \quad , \quad \sigma^\downarrow = \begin{pmatrix} 0 & 0 \\ 0 & 2 \end{pmatrix} \ . \tag{5.109}$$

5.3.1 Spin currents in real space

Assuming that the intensity of the transverse field is much smaller than the static field, i.e., $b_0 \ll H_0$, we can use the linear response theory to calculate the spin current through the bond $i-j$ (with polarization m) $\langle \hat{I}^m_{ij} \rangle$ generated by this perturbation. In this case,

$$\langle \hat{I}^m_{ij} \rangle(t) = -\frac{i}{\hbar} \int dt' \Theta(t-t') \left\langle \left[\hat{I}^m_{ij}(t), \hat{H}_\perp(t') \right] \right\rangle \ . \tag{5.110}$$

Substituting Eqs. 5.6 and 5.98, we get

$$\begin{aligned}
\langle \hat{I}^m_{ij} \rangle(t) &= \frac{g_S \mu_B b_0}{2\hbar} \sum_{\substack{\ell \\ \mu\nu}} \int dt' \Theta(t-t') \left\langle \left[\left\{ t^{\mu\nu}_{ij} \overset{m}{S}{}^{\mu\nu}_{ij}(t) - t^{\nu\mu}_{ji} \overset{m}{S}{}^{\nu\mu}_{ji}(t) \right\}, e^{i\omega t'} \hat{S}^+_\ell(t') + e^{-i\omega t'} \hat{S}^-_\ell(t') \right] \right\rangle \\
&= \frac{g_S \mu_B b_0}{2\hbar} \sum_{\substack{\ell \\ \mu\nu}} \int dt' \Theta(t-t') \left\{ t^{\mu\nu}_{ij} e^{i\omega t'} \left\langle \left[\overset{m}{S}{}^{\mu\nu}_{ij}(t), \hat{S}^+_\ell(t') \right] \right\rangle + t^{\mu\nu}_{ij} e^{-i\omega t'} \left\langle \left[\overset{m}{S}{}^{\mu\nu}_{ij}(t), \hat{S}^-_\ell(t') \right] \right\rangle \right. \\
&\qquad \left. - t^{\nu\mu}_{ji} e^{i\omega t'} \left\langle \left[\overset{m}{S}{}^{\nu\mu}_{ji}(t), \hat{S}^+_\ell(t') \right] \right\rangle - t^{\nu\mu}_{ji} e^{-i\omega t'} \left\langle \left[\overset{m}{S}{}^{\nu\mu}_{ji}(t), \hat{S}^-_\ell(t') \right] \right\rangle \right\} \\
&= \frac{i g_S \mu_B b_0}{2} \sum_{\substack{\ell \\ \mu\nu\gamma}} \int dt' \left\{ t^{\mu\nu}_{ij} e^{i\omega t'} \overset{m+}{\chi}{}^{\mu\nu\gamma\gamma}_{ij\ell\ell}(t-t') + t^{\mu\nu}_{ij} e^{-i\omega t'} \overset{m-}{\chi}{}^{\mu\nu\gamma\gamma}_{ij\ell\ell}(t-t') \right. \\
&\qquad \left. - t^{\nu\mu}_{ji} e^{i\omega t'} \overset{m+}{\chi}{}^{\nu\mu\gamma\gamma}_{ji\ell\ell}(t-t') - t^{\nu\mu}_{ji} e^{-i\omega t'} \overset{m-}{\chi}{}^{\nu\mu\gamma\gamma}_{ji\ell\ell}(t-t') \right\} \ ,
\end{aligned} \tag{5.111}$$

where we have used that

$$\overset{m\pm}{\chi}{}^{\mu\nu\gamma\xi}_{ijkl}(t-t') = -\frac{i}{\hbar} \Theta(t-t') \langle [\overset{m}{S}{}^{\mu\nu}_{ij}(t), S^{\pm\gamma\xi}_{kl}(t')] \rangle \ . \tag{5.112}$$

Using the Fourier transform of the susceptibilities given by

$$\overset{m\pm}{\chi}{}^{\mu\nu\gamma\xi}_{ijkl}(\omega) = \int dt' e^{i\omega(t-t')} \overset{m\pm}{\chi}{}^{\mu\nu\gamma\xi}_{ijkl}(t-t') \ , \tag{5.113}$$

we can obtain

$$\begin{aligned}
\langle \hat{I}^m_{ij} \rangle(t) = \frac{i g_S \mu_B b_0}{2} \sum_{\substack{\ell \\ \mu\nu\gamma}} & \left\{ t^{\mu\nu}_{ij} e^{i\omega t} \overset{m+}{\chi}{}^{\mu\nu\gamma\gamma}_{ij\ell\ell}(-\omega) + t^{\mu\nu}_{ij} e^{-i\omega t} \overset{m-}{\chi}{}^{\mu\nu\gamma\gamma}_{ij\ell\ell}(\omega) \right. \\
& \left. - t^{\nu\mu}_{ji} e^{i\omega t} \overset{m+}{\chi}{}^{\nu\mu\gamma\gamma}_{ji\ell\ell}(-\omega) - t^{\nu\mu}_{ji} e^{-i\omega t} \overset{m-}{\chi}{}^{\nu\mu\gamma\gamma}_{ji\ell\ell}(\omega) \right\} \ .
\end{aligned} \tag{5.114}$$

If $m = x, y, z$, we can write

$$
\begin{aligned}
\langle \hat{I}_{ij}^{m} \rangle(t) &= \frac{g_S \mu_B b_0}{2} \sum_{\substack{\ell \\ \mu\nu\gamma}} \Big\{ - \Big[i t_{ji}^{\nu\mu} e^{-i\omega t} \overset{m-}{\chi}{}_{ji\ell\ell}^{\nu\mu\gamma\gamma}(\omega) \Big]^{*} + i t_{ij}^{\mu\nu} e^{-i\omega t} \overset{m-}{\chi}{}_{ij\ell\ell}^{\mu\nu\gamma\gamma}(\omega) \\
&\qquad\qquad + \Big[i t_{ij}^{\mu\nu} e^{-i\omega t} \overset{m-}{\chi}{}_{ij\ell\ell}^{\mu\nu\gamma\gamma}(\omega) \Big]^{*} - i t_{ji}^{\nu\mu} e^{-i\omega t} \overset{m-}{\chi}{}_{ji\ell\ell}^{\nu\mu\gamma\gamma}(\omega) \Big\} \\
&= g_S \mu_B b_0 \sum_{\substack{\ell \\ \mu\nu\gamma}} \mathrm{Re} \Big\{ i t_{ij}^{\mu\nu} e^{-i\omega t} \overset{m-}{\chi}{}_{ij\ell\ell}^{\mu\nu\gamma\gamma}(\omega) - i t_{ji}^{\nu\mu} e^{-i\omega t} \overset{m-}{\chi}{}_{ji\ell\ell}^{\nu\mu\gamma\gamma}(\omega) \Big\} \\
&= -g_S \mu_B b_0 \sum_{\substack{\ell \\ \mu\nu\gamma}} \mathrm{Im} \Big\{ t_{ij}^{\mu\nu} e^{-i\omega t} \overset{m-}{\chi}{}_{ij\ell\ell}^{\mu\nu\gamma\gamma}(\omega) - t_{ji}^{\nu\mu} e^{-i\omega t} \overset{m-}{\chi}{}_{ji\ell\ell}^{\nu\mu\gamma\gamma}(\omega) \Big\} \\
&= -g_S \mu_B b_0 \left| \mathcal{J}_{ij}^{m} \right| \sin(\omega t - \phi^{m}) \ .
\end{aligned}
\tag{5.115}
$$

where

$$
\mathcal{J}_{ij}^{m} = \sum_{\substack{\ell \\ \mu\nu\gamma}} t_{ij}^{\mu\nu} \overset{m-}{\chi}{}_{ij\ell\ell}^{\mu\nu\gamma\gamma}(\omega) - t_{ji}^{\nu\mu} \overset{m-}{\chi}{}_{ji\ell\ell}^{\nu\mu\gamma\gamma}(\omega) \ . \tag{5.116}
$$

The RPA susceptibility can be calculated as

$$
\chi_{ijll}^{\mu\nu\gamma\gamma} = \overset{0}{\chi}{}_{ijll}^{\mu\nu\gamma\gamma} - \sum_{\substack{m \\ \lambda\alpha}} \overset{0}{\chi}{}_{ijmm}^{\mu\nu\lambda\lambda} U_{m}^{\lambda\alpha} \chi_{ml}^{\alpha\gamma} \ , \tag{5.117}
$$

where the susceptibilities are matrices in spin space. In matrix form, this can be calculated simply as $\chi = \overset{0}{\chi}(1 - U\chi^{+-})$, where χ and χ^{0} are the non-local RPA and HF susceptibility and χ^{+-} is the usual transverse magnetic susceptibility matrix.

We can obtain the spin currents with polarization x and y calculating the I^{+} or I^{-} current and taking the real and imaginary part, respectively. For example,

$$
\begin{aligned}
\langle \hat{I}_{ij}^{-} \rangle(t) = \frac{i g_S \mu_B b_0}{2} \sum_{\substack{\ell \\ \mu\nu\gamma}} \Big\{ & t_{ij}^{\mu\nu} e^{i\omega t} \overset{-+}{\chi}{}_{ij\ell\ell}^{\mu\nu\gamma\gamma}(-\omega) + t_{ij}^{\mu\nu} e^{-i\omega t} \overset{--}{\chi}{}_{ij\ell\ell}^{\mu\nu\gamma\gamma}(\omega) \\
& - t_{ji}^{\nu\mu} e^{i\omega t} \overset{-+}{\chi}{}_{ji\ell\ell}^{\nu\mu\gamma\gamma}(-\omega) - t_{ji}^{\nu\mu} e^{-i\omega t} \overset{--}{\chi}{}_{ji\ell\ell}^{\nu\mu\gamma\gamma}(\omega) \Big\} \ ,
\end{aligned}
\tag{5.118}
$$

Using the relation $\Big[\overset{+-}{\chi}{}_{(0)ijkl}^{\mu\nu\gamma\xi}(\omega) \Big]^{*} = \overset{-+}{\chi}{}_{(0)jilk}^{\nu\mu\xi\gamma}(-\omega)$,

$$
\begin{aligned}
\langle \hat{I}_{ij}^{-} \rangle(t) = \frac{i g_S \mu_B b_0}{2} \sum_{\substack{\ell \\ \mu\nu\gamma}} \Big\{ & \Big[t_{ji}^{\nu\mu} e^{-i\omega t} \overset{+-}{\chi}{}_{ji\ell\ell}^{\nu\mu\gamma\gamma}(\omega) \Big]^{*} + t_{ij}^{\mu\nu} e^{-i\omega t} \overset{--}{\chi}{}_{ij\ell\ell}^{\mu\nu\gamma\gamma}(\omega) \\
& - \Big[t_{ij}^{\mu\nu} e^{-i\omega t} \overset{+-}{\chi}{}_{ij\ell\ell}^{\mu\nu\gamma\gamma}(\omega) \Big]^{*} - t_{ji}^{\nu\mu} e^{-i\omega t} \overset{--}{\chi}{}_{ji\ell\ell}^{\nu\mu\gamma\gamma}(\omega) \Big\} \ .
\end{aligned}
\tag{5.119}
$$

$$
\begin{aligned}
\langle \hat{I}^{+}_{ij}\rangle(t) &= \frac{ig_S\mu_B b_0}{2}\sum_{\substack{\ell\\ \mu\nu\gamma}}\Big\{t^{\mu\nu}_{ij}e^{i\omega t}\overset{++}{\chi}{}^{\mu\nu\gamma\gamma}_{ij\ell\ell}(-\omega)+t^{\mu\nu}_{ij}e^{-i\omega t}\overset{+-}{\chi}{}^{\mu\nu\gamma\gamma}_{ij\ell\ell}(\omega)\\
&\qquad -t^{\nu\mu}_{ji}e^{i\omega t}\overset{++}{\chi}{}^{\nu\mu\gamma\gamma}_{ji\ell\ell}(-\omega)-t^{\nu\mu}_{ji}e^{-i\omega t}\overset{+-}{\chi}{}^{\nu\mu\gamma\gamma}_{ji\ell\ell}(\omega)\Big\}\\
&= \frac{ig_S\mu_B b_0}{2}\sum_{\substack{\ell\\ \mu\nu\gamma}}\Big\{\Big[t^{\nu\mu}_{ji}e^{-i\omega t}\overset{--}{\chi}{}^{\nu\mu\gamma\gamma}_{ji\ell\ell}(\omega)-t^{\mu\nu}_{ij}e^{-i\omega t}\overset{--}{\chi}{}^{\mu\nu\gamma\gamma}_{ij\ell\ell}(\omega)\Big]^{*}\\
&\qquad +t^{\mu\nu}_{ij}e^{-i\omega t}\overset{+-}{\chi}{}^{\mu\nu\gamma\gamma}_{ij\ell\ell}(\omega)-t^{\nu\mu}_{ji}e^{-i\omega t}\overset{+-}{\chi}{}^{\nu\mu\gamma\gamma}_{ji\ell\ell}(\omega)\Big\}\ ,
\end{aligned}
\tag{5.120}
$$

5.3.2 Total perpendicular spin currents in multilayers

For multilayers, the total perpendicular spin current operator between layers ℓ_i and ℓ_j is given by Eq. (5.82). So, substituting it together with the perturbation hamiltonian given by Eq. (5.103) in the spin current calculated by linear response, Eq. (5.110), we obtain the contribution of the coupling of the external field with the spin as

$$
\begin{aligned}
\langle \hat{I}^{m}_{\ell_i\ell_j}\rangle(t) = \frac{g_S\mu_B b_0}{2\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell\\ \mu\nu}}\int dt'\Theta(t-t')\Big\langle\Big[\Big\{t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\ \overset{m}{S}{}^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)-t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\ \overset{m}{S}{}^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)\Big\}\ ,\\
\Big\{e^{i\omega t'}S^{+}_{\ell}(-\mathbf{Q}_\parallel,t')+e^{-i\omega t'}S^{-}_{\ell}(\mathbf{Q}_\parallel,t')\Big\}\Big]\Big\rangle\ ,
\end{aligned}
\tag{5.121}
$$

where the spin operators $S^{+}_{\ell}(-\mathbf{Q},t')$ and $S^{-}_{\ell}(\mathbf{Q},t')$ are given by Eqs. 5.104 and 5.105, respectively.

We can rewrite Eq. (5.121) in terms of generalized susceptibilities as

$$
\begin{aligned}
\langle \hat{I}^{m}_{\ell_i\ell_j}\rangle(t) = \frac{ig_S\mu_B b_0}{2}\frac{1}{N^2_\parallel}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel}\sum_{\substack{\ell\\ \mu\nu\gamma}}\int dt'\ \Big\{&e^{i\omega t'}t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\overset{m+}{\chi}{}^{\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,-\mathbf{k}_\parallel,\mathbf{k}'_\parallel-\mathbf{Q}_\parallel,-\mathbf{k}'_\parallel;t-t')\\
&+e^{-i\omega t'}t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\overset{m-}{\chi}{}^{\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,-\mathbf{k}_\parallel,\mathbf{k}'_\parallel+\mathbf{Q}_\parallel,-\mathbf{k}'_\parallel;t-t')\\
&-e^{i\omega t'}t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\overset{m+}{\chi}{}^{\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,-\mathbf{k}_\parallel,\mathbf{k}'_\parallel-\mathbf{Q}_\parallel,-\mathbf{k}'_\parallel;t-t')\\
&-e^{-i\omega t'}t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\overset{m-}{\chi}{}^{\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,-\mathbf{k}_\parallel,\mathbf{k}'_\parallel+\mathbf{Q}_\parallel,-\mathbf{k}'_\parallel;t-t')\Big\}\ ,
\end{aligned}
\tag{5.122}
$$

where

$$
\overset{m\sigma}{\chi}{}^{\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_1,-\mathbf{k}_2,\mathbf{k}_3,-\mathbf{k}_4;t-t') = -\frac{i}{\hbar}\Theta(t-t')\Big\langle\Big[\overset{m}{S}{}^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_1,\mathbf{k}_2;t),\overset{\sigma}{S}{}^{\gamma\gamma}_{\ell\ell}(\mathbf{k}_3,\mathbf{k}_4;t')\Big]\Big\rangle\ ,\ \text{and} \tag{5.123}
$$

Taking into account the symmetry of the multilayer, we obtain the relation

$$
\overset{m\sigma}{\chi}{}^{\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_1,-\mathbf{k}_2,\mathbf{k}_3,-\mathbf{k}_4) = N^2_\parallel\overset{m\sigma}{\chi}{}^{\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_1,\mathbf{k}_3)\delta_{\mathbf{k}_3\mathbf{k}_2}\delta_{\mathbf{k}_1\mathbf{k}_4}\ . \tag{5.124}
$$

Using the Fourier transform of the susceptibilities given by Eq. (5.113), the spin current per bond is

$$
\begin{aligned}
\langle \hat{I}^m_{\ell_i\ell_j}\rangle(t) = \frac{ig_S\mu_B b_0}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell\\ \mu\nu\gamma}}\Big\{ & e^{i\omega t} t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{m+\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;-\omega) \\
& + e^{-i\omega t} t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{m-\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) \\
& - e^{i\omega t} t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{m+\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;-\omega) \\
& - e^{-i\omega t} t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{m-\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\Big\}\,\delta_{\mathbf{Q}_\parallel 0}\ ,
\end{aligned}
\tag{5.125}
$$

where the RPA susceptibilities can be calculated from the HF susceptibilities as (see Eq. (I.41)),

$$
\chi_{ij\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \chi^0_{ij\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) - \sum_m \chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U_m\chi_{m\ell}(\mathbf{k}'_\parallel-\mathbf{k}_\parallel)\ . \tag{5.126}
$$

The total perpendicular spin current with transverse polarization is given by

$$
\begin{aligned}
\langle \hat{I}^+_{\ell_i\ell_j}\rangle(t) = \frac{ig_S\mu_B b_0}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell\\ \mu\nu\gamma}}\Big\{ & e^{i\omega t} t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{++\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;-\omega) \\
& + e^{-i\omega t} t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{+-\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) \\
& - e^{i\omega t} t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{++\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;-\omega) \\
& - e^{-i\omega t} t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{+-\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\Big\}\,\delta_{\mathbf{Q}_\parallel 0}\ ,
\end{aligned}
\tag{5.127}
$$

or

$$
\begin{aligned}
\langle \hat{I}^+_{\ell_i\ell_j}\rangle(t) = \frac{ig_S\mu_B b_0}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell\\ \mu\nu\gamma}}\Big\{ & e^{i\omega t}\left[t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{--\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{--\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\right]^* \\
& + e^{-i\omega t}\left[t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{+-\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{+-\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\right]\Big\}\,\delta_{\mathbf{Q}_\parallel 0}\ .
\end{aligned}
\tag{5.128}
$$

To simplify the calculation, we can define

$$
I_+(\omega) = \frac{i}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell\\ \mu\nu\gamma}}\left[t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{--\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{--\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\right]\ , \tag{5.129}
$$

$$
I_-(\omega) = \frac{i}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell\\ \mu\nu\gamma}}\left[t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{+-\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{+-\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\right]\ . \tag{5.130}
$$

Then, the spin current of the x-component polarization is

$$
\begin{aligned}
\langle \hat{I}^{x}_{\ell_i\ell_j}\rangle(t) =& \mathrm{Re}\left[\langle \hat{I}^{+}_{\ell_i\ell_j}\rangle(t)\right] \\
=& g_S\mu_B b_0 \ \mathrm{Re}\left\{(\cos\omega t + i\sin\omega t)\left[I_+(\omega)\right]^* + (\cos\omega t - i\sin\omega t)\left[I_-(\omega)\right]\right\}\delta_{\mathbf{Q}_\parallel 0} \\
=& g_S\mu_B b_0 \ \left\{\cos\omega t \ \mathrm{Re}\left[I_+(\omega) + I_-(\omega)\right] + \sin\omega t \ \mathrm{Im}\left[I_+(\omega) + I_-(\omega)\right]\right\}\delta_{\mathbf{Q}_\parallel 0} \\
=& g_S\mu_B b_0 \ |I_+(\omega) + I_-(\omega)|\left\{\cos\omega t\cos\phi^x + \sin\omega t\sin\phi^x\right\}\delta_{\mathbf{Q}_\parallel 0} \\
=& g_S\mu_B b_0 \ |I_+(\omega) + I_-(\omega)|\cos(\omega t - \phi^x)\delta_{\mathbf{Q}_\parallel 0} \ ,
\end{aligned}
\tag{5.131}
$$

where the amplitude is given by the absolute value of

$$
\begin{aligned}
I_+(\omega) + I_-(\omega) = \frac{i}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell \\ \mu\nu\gamma}} \Big[& t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{--\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{--\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) \\
& + t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{+-\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{+-\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\Big] \ ,
\end{aligned}
\tag{5.132}
$$

and the phase is given by

$$
\cos\phi^x = \frac{\mathrm{Re}\left[I_+(\omega) + I_-(\omega)\right]}{|I_+(\omega) + I_-(\omega)|} \ , \tag{5.133}
$$

$$
\sin\phi^x = \frac{\mathrm{Im}\left[I_+(\omega) + I_-(\omega)\right]}{|I_+(\omega) + I_-(\omega)|} \ . \tag{5.134}
$$

Analogously, the spin current of the y-component polarization is

$$
\begin{aligned}
\langle \hat{I}^{y}_{\ell_i\ell_j}\rangle(t) =& \mathrm{Im}\left[\langle \hat{I}^{+}_{\ell_i\ell_j}\rangle(t)\right] \\
=& g_S\mu_B b_0 \ \mathrm{Im}\left\{(\cos\omega t + i\sin\omega t)\left[I_+(\omega)\right]^* + (\cos\omega t - i\sin\omega t)\left[I_-(\omega)\right]\right\}\delta_{\mathbf{Q}_\parallel 0} \\
=& g_S\mu_B b_0 \ \left\{\sin\omega t \ \mathrm{Re}\left[I_+(\omega) - I_-(\omega)\right] - \cos\omega t \ \mathrm{Im}\left[I_+(\omega) - I_-(\omega)\right]\right\}\delta_{\mathbf{Q}_\parallel 0} \\
=& g_S\mu_B b_0 \ |I_+(\omega) - I_-(\omega)|\left\{\sin\omega t\cos\phi^y - \cos\omega t\sin\phi^y\right\}\delta_{\mathbf{Q}_\parallel 0} \\
=& g_S\mu_B b_0 \ |I_+(\omega) - I_-(\omega)|\sin(\omega t - \phi^y)\delta_{\mathbf{Q}_\parallel 0} \ ,
\end{aligned}
\tag{5.135}
$$

where the amplitude is now given by the absolute value of

$$
\begin{aligned}
I_+(\omega) - I_-(\omega) = \frac{i}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell \\ \mu\nu\gamma}} \Big[& t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{--\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{--\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) \\
& - t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{+-\mu\nu\gamma\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) + t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{+-\nu\mu\gamma\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\Big] \ ,
\end{aligned}
\tag{5.136}
$$

and the phase by

$$\cos\phi^y = \frac{\mathrm{Re}\left[I_+(\omega) - I_-(\omega)\right]}{|I_+(\omega) - I_-(\omega)|} \ , \tag{5.137}$$

$$\sin\phi^y = \frac{\mathrm{Im}\left[I_+(\omega) - I_-(\omega)\right]}{|I_+(\omega) - I_-(\omega)|} \ . \tag{5.138}$$

The spin current due to the coupling of the external field with the orbital angular momentum is obtained with analogous calculations, and we find

$$\begin{aligned}\langle \hat{I}^+_{\ell_i\ell_j}\rangle^L(t) = \frac{i\mu_B b_0}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell\\ \mu\nu\gamma\xi}}\sum_{\sigma}\Big\{ & e^{i\omega t}\hat{L}'^{+}_{\gamma\xi} t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{+\sigma\,\mu\nu\gamma\xi}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;-\omega) \\ & + e^{-i\omega t}\hat{L}'^{-}_{\gamma\xi} t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{+\sigma\,\mu\nu\gamma\xi}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) \\ & - e^{i\omega t}\hat{L}'^{+}_{\gamma\xi} t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{+\sigma\,\nu\mu\gamma\xi}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;-\omega) \\ & - e^{-i\omega t}\hat{L}'^{-}_{\gamma\xi} t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{+\sigma\,\nu\mu\gamma\xi}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\Big\}\,\delta_{\mathbf{Q}_\parallel 0} \ .\end{aligned} \tag{5.139}$$

In terms of the conjugates

$$\begin{aligned}\langle \hat{I}^+_{\ell_i\ell_j}\rangle^L(t) = & \frac{i\mu_B b_0}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell\\ \mu\nu\gamma\xi}}\sum_{\sigma}\delta_{\mathbf{Q}_\parallel 0} \\ & \Big\{e^{i\omega t}\Big[\hat{L}'^{-}_{\xi\gamma}\Big(t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{-\sigma\,\nu\mu\xi\gamma}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{-\sigma\,\mu\nu\xi\gamma}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\Big)\Big]^* \\ & + e^{-i\omega t}\Big[\hat{L}'^{-}_{\gamma\xi}\Big(t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{+\sigma\,\mu\nu\gamma\xi}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{+\sigma\,\nu\mu\gamma\xi}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\Big)\Big]\Big\} \ .\end{aligned} \tag{5.140}$$

Changing $\gamma \rightleftharpoons \xi$ on the first two terms

$$\begin{aligned}\langle \hat{I}^+_{\ell_i\ell_j}(t)\rangle^L = & \frac{i\mu_B b_0}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell\\ \mu\nu\gamma\xi}}\sum_{\sigma}\delta_{\mathbf{Q}_\parallel 0} \\ & \Big\{e^{i\omega t}\Big[\hat{L}'^{-}_{\gamma\xi}\Big(t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{-\sigma\,\nu\mu\gamma\xi}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{-\sigma\,\mu\nu\gamma\xi}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\Big)\Big]^* \\ & + e^{-i\omega t}\Big[\hat{L}'^{-}_{\gamma\xi}\Big(t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{+\sigma\,\mu\nu\gamma\xi}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{+\sigma\,\nu\mu\gamma\xi}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\Big)\Big]\Big\} \ .\end{aligned} \tag{5.141}$$

To simplify the calculation, we can define

$$I^L_+(\omega) = \frac{i}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell\\ \mu\nu\gamma\xi}}\sum_{\sigma}\hat{L}'^{-}_{\gamma\xi}\Big[t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{-\sigma\,\nu\mu\gamma\xi}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{-\sigma\,\mu\nu\gamma\xi}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\Big] \ , \tag{5.142}$$

$$I_-^L(\omega) = \frac{i}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell \\ \mu\nu\gamma\xi}}\sum_{\sigma}\hat{L}'^{-}_{\gamma\xi}\left[t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{+\sigma\,\mu\nu\gamma\xi}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{+\sigma\,\nu\mu\gamma\xi}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\right] . \tag{5.143}$$

Then, following the same steps as before, the orbital contribution to the spin current of the x-component polarization can be written as

$$\begin{aligned}\langle\hat{I}^x_{\ell_i\ell_j}(t)\rangle^L =&\mathrm{Re}\left[\langle\hat{I}^+_{\ell_i\ell_j}(t)\rangle^L\right] \\ =&\mu_B b_0\;|I_+^L(\omega)+I_-^L(\omega)|\cos(\omega t-\phi^x_L)\delta_{\mathbf{Q}_\parallel 0}\ ,\end{aligned} \tag{5.144}$$

where the amplitude is given by the absolute value of

$$\begin{aligned}I_+^L(\omega)+I_-^L(\omega) = \frac{i}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell \\ \mu\nu\gamma\xi}}\sum_{\sigma}\hat{L}'^{-}_{\gamma\xi}&\left[t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{-\sigma\,\nu\mu\gamma\xi}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{-\sigma\,\mu\nu\gamma\xi}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\right.\\ &\left.+t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{+\sigma\,\mu\nu\gamma\xi}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{+\sigma\,\nu\mu\gamma\xi}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\right] ,\end{aligned} \tag{5.145}$$

and the phase is given by

$$\cos\phi^x_L = \frac{\mathrm{Re}\left[I_+^L(\omega)+I_-^L(\omega)\right]}{|I_+^L(\omega)+I_-^L(\omega)|}\ , \tag{5.146}$$

$$\sin\phi^x_L = \frac{\mathrm{Im}\left[I_+^L(\omega)+I_-^L(\omega)\right]}{|I_+^L(\omega)+I_-^L(\omega)|}\ . \tag{5.147}$$

Analogously, the spin current of the y-component polarization is

$$\begin{aligned}\langle\hat{I}^y_{\ell_i\ell_j}(t)\rangle^L =&\mathrm{Im}\left[\langle\hat{I}^+_{\ell_i\ell_j}(t)\rangle^L\right] \\ =&\mu_B b_0\;|I_+^L(\omega)-I_-^L(\omega)|\sin(\omega t-\phi^y_L)\delta_{\mathbf{Q}_\parallel 0}\ ,\end{aligned} \tag{5.148}$$

where the amplitude is now given by the absolute value of

$$\begin{aligned}I_+^L(\omega)-I_-^L(\omega) = \frac{i}{2}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\ell \\ \mu\nu\gamma\xi}}\sum_{\sigma}\hat{L}'^{-}_{\gamma\xi}&\left[t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{-\sigma\,\nu\mu\gamma\xi}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) - t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{-\sigma\,\mu\nu\gamma\xi}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\right.\\ &\left.-t^{\mu\nu}_{\ell_i\ell_j}(\mathbf{k}_\parallel)\chi^{+\sigma\,\mu\nu\gamma\xi}_{\ell_i\ell_j\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega) + t^{\nu\mu}_{\ell_j\ell_i}(\mathbf{k}_\parallel)\chi^{+\sigma\,\nu\mu\gamma\xi}_{\ell_j\ell_i\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel;\omega)\right] ,\end{aligned} \tag{5.149}$$

and the phase by

$$\cos\phi^y_L = \frac{\mathrm{Re}\left[I_+^L(\omega)-I_-^L(\omega)\right]}{|I_+^L(\omega)-I_-^L(\omega)|}\ , \tag{5.150}$$

$$\sin\phi^y_L = \frac{\mathrm{Im}\left[I_+^L(\omega)-I_-^L(\omega)\right]}{|I_+^L(\omega)-I_-^L(\omega)|}\ . \tag{5.151}$$

5.3.3 Charge Current in real space

We can also calculate the charge current I^{C} generated on this system. This current is created only in systems where the spin-orbit coupling is present, and this is known as the Inverse Spin Hall Effect. The charge current operator is given by Eqs. 5.30 and 5.31. So, we need

$$\begin{aligned}
\langle \hat{I}_{ij}^{\uparrow} \rangle(t) &= \frac{ig_S\mu_B b_0}{2} \sum_{\substack{\ell \\ \mu\nu\gamma}} \Big\{ t_{ij}^{\mu\nu} e^{i\omega t} \left[\chi_{ji\ell\ell}^{\uparrow-\nu\mu\gamma\gamma}(\omega) \right]^* + t_{ij}^{\mu\nu} e^{-i\omega t} \chi_{ij\ell\ell}^{\uparrow-\mu\nu\gamma\gamma}(\omega) \\
&\qquad\qquad - t_{ji}^{\nu\mu} e^{i\omega t} \left[\chi_{ij\ell\ell}^{\uparrow-\mu\nu\gamma\gamma}(\omega) \right]^* - t_{ji}^{\nu\mu} e^{-i\omega t} \chi_{ji\ell\ell}^{\uparrow-\nu\mu\gamma\gamma}(\omega) \Big\} \\
&= \frac{ig_S\mu_B b_0}{2} \sum_{\substack{\ell \\ \mu\nu\gamma}} \Big\{ \left[t_{ji}^{\nu\mu} e^{-i\omega t} \chi_{ji\ell\ell}^{\uparrow-\nu\mu\gamma\gamma}(\omega) \right]^* + t_{ij}^{\mu\nu} e^{-i\omega t} \chi_{ij\ell\ell}^{\uparrow-\mu\nu\gamma\gamma}(\omega) \\
&\qquad\qquad - \left[t_{ij}^{\mu\nu} e^{-i\omega t} \chi_{ij\ell\ell}^{\uparrow-\mu\nu\gamma\gamma}(\omega) \right]^* - t_{ji}^{\nu\mu} e^{-i\omega t} \chi_{ji\ell\ell}^{\uparrow-\nu\mu\gamma\gamma}(\omega) \Big\} \\
&= -g_S\mu_B b_0 \sum_{\substack{\ell \\ \mu\nu\gamma}} \mathrm{Im} \Big\{ t_{ij}^{\mu\nu} e^{-i\omega t} \chi_{ij\ell\ell}^{\uparrow-\mu\nu\gamma\gamma}(\omega) - t_{ji}^{\nu\mu} e^{-i\omega t} \chi_{ji\ell\ell}^{\uparrow-\nu\mu\gamma\gamma}(\omega) \Big\} \ .
\end{aligned} \tag{5.152}$$

Analogously,

$$\langle \hat{I}_{ij}^{\downarrow} \rangle(t) = -g_S\mu_B b_0 \sum_{\substack{\ell \\ \mu\nu\gamma}} \mathrm{Im} \Big\{ t_{ij}^{\mu\nu} e^{-i\omega t} \chi_{ij\ell\ell}^{\downarrow-\mu\nu\gamma\gamma}(\omega) - t_{ji}^{\nu\mu} e^{-i\omega t} \chi_{ji\ell\ell}^{\downarrow-\nu\mu\gamma\gamma}(\omega) \Big\} \ . \tag{5.153}$$

5.3.4 Parallel charge current in multilayers

Now we can obtain the charge current which is created in multilayer systems due to the Inverse Spin Hall Effect. In this case, the spin current is pumped perpendicularly to the layers, and the charge current flows parallel to them. So, we need to obtain the expectation value of Eq. (5.65), i.e.,

$$\langle \hat{I}_{ij}^{\mathrm{C}}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel; t) \rangle = \frac{1}{N_\parallel} \sum_{\mathbf{q}_\parallel} \Big\{ \langle \hat{\mathcal{I}}_{ij}^{\mathrm{C}}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{q}_\parallel; t) \rangle e^{i\mathbf{q}_\parallel \cdot \mathbf{R}'_\parallel} - \langle \hat{\mathcal{I}}_{ji}^{\mathrm{C}}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel, \mathbf{q}_\parallel; t) \rangle e^{i\mathbf{q}_\parallel \cdot \mathbf{R}_\parallel} \Big\} \ , \tag{5.154}$$

Here,

$$\begin{aligned}
\hat{\mathcal{I}}_{ij}^{\mathrm{C}}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{q}_\parallel) &= \frac{ie}{\hbar} \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\sigma} \sum_{\mu\nu} t_{ij}^{\mu\nu}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) c_{i\mu\sigma}^{\dagger}(\mathbf{k}_\parallel) c_{j\nu\sigma}(\mathbf{k}_\parallel + \mathbf{q}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \\
&= \frac{ie}{\hbar} \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\sigma} \sum_{\mu\nu} t_{ij}^{\mu\nu}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) S_{ij}^{\sigma\,\mu\nu}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{q}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \ ,
\end{aligned} \tag{5.155}$$

where we have used the generalized form of the spin operator given by Eq. (5.78).

Using the interaction hamiltonian given by Eq. (5.103) we obtain the contribution of the perturbation on the spin as

$$\begin{aligned}
\langle \hat{\mathcal{I}}^{\mathrm{C}}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel; t)\rangle &= -\frac{ig_S\mu_B b_0}{2\hbar}\sum_{\ell}\int dt'\Theta(t-t')\Big\langle\Big[\hat{\mathcal{I}}^{\mathrm{C}}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{q}_\parallel; t), \\
&\qquad \Big\{e^{i\omega t'}S^{+}_{\ell}(-\mathbf{Q}_\parallel, t') + e^{-i\omega t'}S^{-}_{\ell}(\mathbf{Q}_\parallel, t')\Big\}\Big]\Big\rangle \\
&= \frac{ig_S\mu_B b_0 e}{2\hbar}\frac{-i}{\hbar}\frac{1}{N_\parallel^2}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_{\substack{\mu\nu\gamma\\ \sigma}}\sum_{\ell} t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\int dt'\Theta(t-t')\Big\langle\Big[S^{\sigma\,\mu\nu}_{ij}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{q}_\parallel; t), \\
&\qquad \Big\{e^{i\omega t'}S^{+\gamma\gamma}_{\ell\ell}(\mathbf{k}'_\parallel - \mathbf{Q}_\parallel, \mathbf{k}'_\parallel; t') + e^{-i\omega t'}S^{-\gamma\gamma}_{\ell\ell}(\mathbf{k}'_\parallel + \mathbf{Q}_\parallel, \mathbf{k}'_\parallel; t')\Big\}\Big]\Big\rangle \\
&= \frac{ig_S\mu_B b_0 e}{2\hbar}\frac{1}{N_\parallel^2}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_{\substack{\mu\nu\gamma\\ \sigma}}\sum_{\ell} t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \\
&\qquad \times\int dt'\Big\{e^{i\omega t'}\chi^{\sigma+\,\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel, -\mathbf{k}_\parallel - \mathbf{q}_\parallel, \mathbf{k}'_\parallel - \mathbf{Q}_\parallel, -\mathbf{k}'_\parallel; t-t') \\
&\qquad + e^{-i\omega t'}\chi^{\sigma-\,\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel, -\mathbf{k}_\parallel - \mathbf{q}_\parallel, \mathbf{k}'_\parallel + \mathbf{Q}_\parallel, -\mathbf{k}'_\parallel; t-t')\Big\}\ ,
\end{aligned} \tag{5.156}$$

To obtain this result, we have used the spin operators given by Eqs. 5.105 and 5.104, and the generalized susceptibilities defined in Eq. (5.123). When we take into account the layered symmetry, the susceptibilities are simplified as shown in Eq. (5.124). In this case,

$$\begin{aligned}
\langle \hat{\mathcal{I}}^{\mathrm{C}}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel; t)\rangle &= \frac{ig_S\mu_B b_0 e}{2\hbar}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\mu\nu\gamma\\ \sigma}}\sum_{\ell} t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \\
&\quad \times\int dt'\Big\{e^{i\omega t'}\chi^{\sigma+\,\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel - \mathbf{Q}_\parallel; t-t')\delta_{-\mathbf{q}_\parallel\mathbf{Q}_\parallel} \\
&\qquad + e^{-i\omega t'}\chi^{\sigma-\,\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; t-t')\delta_{\mathbf{q}_\parallel\mathbf{Q}_\parallel}\Big\}\ .
\end{aligned} \tag{5.157}$$

Using the Fourier transform given by Eq. (5.113), we obtain

$$\begin{aligned}
\langle \hat{\mathcal{I}}^{\mathrm{C}}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel; t)\rangle &= \frac{ig_S\mu_B b_0 e}{2\hbar}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\mu\nu\gamma\\ \sigma}}\sum_{\ell} t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \\
&\quad \times\Big\{e^{i\omega t}\chi^{\sigma+\,\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel - \mathbf{Q}_\parallel; -\omega)\delta_{-\mathbf{q}_\parallel\mathbf{Q}_\parallel} \\
&\qquad + e^{-i\omega t}\chi^{\sigma-\,\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega)\delta_{\mathbf{q}_\parallel\mathbf{Q}_\parallel}\Big\}\ .
\end{aligned} \tag{5.158}$$

Substituting this result in Eq. (5.154),

$$
\begin{aligned}
\langle \hat{I}^{\mathrm{C}}_{ij}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel;t)\rangle &= \frac{ig_S\mu_B b_0 e}{2\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\mu\nu\gamma\\ \sigma}}\sum_{\ell}\Big\{t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\\
&\times\Big[e^{i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)}\chi^{\sigma+\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel-\mathbf{Q}_\parallel;-\omega)+e^{-i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)}\chi^{\sigma-\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{Q}_\parallel;\omega)\Big]\\
&-t^{\nu\mu}_{ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}\\
&\times\Big[e^{i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel)}\chi^{\sigma+\nu\mu\gamma\gamma}_{ji\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel-\mathbf{Q}_\parallel;-\omega)+e^{-i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel)}\chi^{\sigma-\nu\mu\gamma\gamma}_{ji\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{Q}_\parallel;\omega)\Big]\Big\}\ ,
\end{aligned}
\tag{5.159}
$$

or, multiplying all the terms,

$$
\begin{aligned}
\langle \hat{I}^{\mathrm{C}}_{ij}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel;t)\rangle &= \frac{ig_S\mu_B b_0 e}{2\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\mu\nu\gamma\\ \sigma}}\sum_{\ell}\\
&\times\Big\{t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}e^{i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)}\chi^{\sigma+\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel-\mathbf{Q}_\parallel;-\omega)\\
&+t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}e^{-i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)}\chi^{\sigma-\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{Q}_\parallel;\omega)\\
&-t^{\nu\mu}_{ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}e^{i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel)}\chi^{\sigma+\nu\mu\gamma\gamma}_{ji\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel-\mathbf{Q}_\parallel;-\omega)\\
&-t^{\nu\mu}_{ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}e^{-i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel)}\chi^{\sigma-\nu\mu\gamma\gamma}_{ji\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{Q}_\parallel;\omega)\Big\}\\
&=\frac{ig_S\mu_B b_0 e}{2\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\mu\nu\gamma\\ \sigma}}\sum_{\ell}\\
&\times\Big\{t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}e^{i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)}\chi^{\sigma+\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel-\mathbf{Q}_\parallel;-\omega)\\
&+t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}e^{-i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)}\chi^{\sigma-\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{Q}_\parallel;\omega)\\
&-t^{\nu\mu}_{ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)e^{i(\mathbf{k}_\parallel-\mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}e^{i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)}\chi^{\sigma+\nu\mu\gamma\gamma}_{ji\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel-\mathbf{Q}_\parallel;-\omega)\\
&-t^{\nu\mu}_{ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)e^{i(\mathbf{k}_\parallel+\mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}e^{-i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)}\chi^{\sigma-\nu\mu\gamma\gamma}_{ji\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{Q}_\parallel;\omega)\Big\}\ .
\end{aligned}
\tag{5.160}
$$

Changing the position of the second and third terms,

$$
\begin{aligned}
\langle \hat{I}^{\mathrm{C}}_{ij}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel;t)\rangle &= \frac{ig_S\mu_B b_0 e}{2\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\mu\nu\gamma\\ \sigma}}\sum_{\ell}\\
&\times\Big\{\Big[t^{\nu\mu}_{ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}e^{-i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)}\chi^{\sigma-\nu\mu\gamma\gamma}_{ji\ell\ell}(\mathbf{k}_\parallel-\mathbf{Q}_\parallel,\mathbf{k}_\parallel;\omega)\Big]^*\\
&-\Big[t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{-i(\mathbf{k}_\parallel-\mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}e^{-i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)}\chi^{\sigma-\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel-\mathbf{Q}_\parallel,\mathbf{k}_\parallel;\omega)\Big]^*\\
&+t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}e^{-i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)}\chi^{\sigma-\mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{Q}_\parallel;\omega)\\
&-t^{\nu\mu}_{ji}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)e^{i(\mathbf{k}_\parallel+\mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}e^{-i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)}\chi^{\sigma-\nu\mu\gamma\gamma}_{ji\ell\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{Q}_\parallel;\omega)\Big\}\ ,
\end{aligned}
\tag{5.161}
$$

where we have already written the first two terms in respect to their conjugates. Changing the variable $\mathbf{k}_\parallel \to \mathbf{k}_\parallel + \mathbf{Q}_\parallel$ in this two terms,

$$
\begin{aligned}
&\langle \hat{I}^{\mathrm{C}}_{ij}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel; t)\rangle = \frac{i g_S \mu_B b_0 e}{2\hbar} \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\substack{\mu\nu\gamma \\ \sigma}} \sum_{\ell} \\
&\left\{ \left[t^{\nu\mu}_{ji}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i(\mathbf{k}_\parallel + \mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel \cdot \mathbf{R}'_\parallel)} \chi^{\sigma - \nu\mu\gamma\gamma}_{ji\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \right]^* \right. \\
&- \left[t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel \cdot \mathbf{R}'_\parallel)} \chi^{\sigma - \mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \right]^* \\
&+ t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel \cdot \mathbf{R}'_\parallel)} \chi^{\sigma - \mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \\
&\left. - t^{\nu\mu}_{ji}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i(\mathbf{k}_\parallel + \mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel \cdot \mathbf{R}'_\parallel)} \chi^{\sigma - \nu\mu\gamma\gamma}_{ji\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \right\} \ .
\end{aligned} \tag{5.162}
$$

Now we can see that the first two terms are the complex conjugate of the last two. So, the charge current can be written as

$$
\begin{aligned}
&\langle \hat{I}^{\mathrm{C}}_{ij}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel; t)\rangle = -\frac{g_S \mu_B b_0 e}{\hbar} \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\substack{\mu\nu\gamma \\ \sigma}} \sum_{\ell} \\
&\mathrm{Im} \left\{ t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel \cdot \mathbf{R}'_\parallel)} \chi^{\sigma - \mu\nu\gamma\gamma}_{ij\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \right. \\
&\left. - t^{\nu\mu}_{ji}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i(\mathbf{k}_\parallel + \mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel \cdot \mathbf{R}'_\parallel)} \chi^{\sigma - \nu\mu\gamma\gamma}_{ji\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \right\} \ ,
\end{aligned} \tag{5.163}
$$

which shows that the charge current is real, as expected.

For sites in the same plane (i.e., $i = j$), we can change the indices $\mu \rightleftharpoons \nu$ in the second term and we end up with

$$
\begin{aligned}
\langle \hat{I}^{\mathrm{C}}_{ii}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel; t)\rangle = &- \frac{g_S \mu_B b_0 e}{\hbar} \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\substack{\mu\nu\gamma \\ \sigma}} \sum_{\ell} \mathrm{Im} \left\{ e^{-i(\omega t - \mathbf{Q}_\parallel \cdot \mathbf{R}'_\parallel)} \chi^{\sigma - \mu\nu\gamma\gamma}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \right. \\
&\left. \times \left[t^{\mu\nu}_{ii}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} - t^{\mu\nu}_{ii}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i(\mathbf{k}_\parallel + \mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} \right] \right\} \ ,
\end{aligned} \tag{5.164}
$$

If we call

$$
\begin{aligned}
I^{\mathrm{C}}_i(\mathbf{R}'_\parallel - \mathbf{R}_\parallel; \mathbf{Q}_\parallel; \omega) = &\frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\substack{\mu\nu\gamma \\ \sigma}} \sum_{\ell} \chi^{\sigma - \mu\nu\gamma\gamma}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \\
&\times \left[t^{\mu\nu}_{ii}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} - t^{\mu\nu}_{ii}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i(\mathbf{k}_\parallel + \mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} \right] \\
= &|I^{\mathrm{C}}_i(\mathbf{R}'_\parallel - \mathbf{R}_\parallel; \mathbf{Q}_\parallel; \omega)| e^{i\phi_C} \ ,
\end{aligned} \tag{5.165}
$$

where

$$
\begin{aligned}
\cos\phi_C =& \frac{\mathrm{Re}\left[I_i^{\mathrm{C}}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel;\mathbf{Q}_\parallel;\omega)\right]}{|I_i^{\mathrm{C}}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel;\mathbf{Q}_\parallel;\omega)|}\ \text{, and} \\
\sin\phi_C =& \frac{\mathrm{Im}\left[I_i^{\mathrm{C}}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel;\mathbf{Q}_\parallel;\omega)\right]}{|I_i^{\mathrm{C}}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel;\mathbf{Q}_\parallel;\omega)|}\ ,
\end{aligned}
\tag{5.166}
$$

we finally end up with

$$
\begin{aligned}
\langle \hat{I}_{ii}^{\mathrm{C}}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel;\mathbf{Q}_\parallel;t)\rangle =& -\frac{g_S\mu_B b_0 e}{\hbar}\mathrm{Im}\left\{e^{-i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)}I^{\mathrm{C}}(\omega)\right\} \\
=& -\frac{g_S\mu_B b_0 e}{\hbar}|I_i^{\mathrm{C}}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel;\mathbf{Q}_\parallel;\omega)|\mathrm{Im}\left\{e^{-i(\omega t-\mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel-\phi_C)}\right\} \\
=& \frac{g_S\mu_B b_0 e}{\hbar}|I_i^{\mathrm{C}}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel;\mathbf{Q}_\parallel;\omega)|\sin(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel - \phi_C)\ .
\end{aligned}
\tag{5.167}
$$

For $\omega = 0$, $\mathbf{Q}_\parallel = 0$ and $i = j$,

$$
\begin{aligned}
\langle \hat{I}^{\mathrm{C}}_{ii}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel; t)\rangle &= \frac{i g_S \mu_B b_0 e}{2\hbar} \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\substack{\mu\nu\gamma \\ \sigma}} \sum_{\ell} \\
&\times \Big\{ t^{\mu\nu}_{ii}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \chi^{\sigma+\mu\nu\gamma\gamma}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; 0) \\
&+ t^{\mu\nu}_{ii}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \chi^{\sigma-\mu\nu\gamma\gamma}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; 0) \\
&- t^{\nu\mu}_{ii}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} \chi^{\sigma+\nu\mu\gamma\gamma}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; 0) \\
&- t^{\nu\mu}_{ii}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} \chi^{\sigma-\nu\mu\gamma\gamma}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; 0) \Big\} \ .
\end{aligned}
\tag{5.168}
$$

Changing $\mu \rightleftharpoons \nu$ in the last 2 terms,

$$
\begin{aligned}
\langle \hat{I}^{\mathrm{C}}_{ii}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel; t)\rangle &= \frac{i g_S \mu_B b_0 e}{2\hbar} \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\substack{\mu\nu\gamma \\ \sigma}} \sum_{\ell} \\
&\times \Big\{ \Big[t^{\mu\nu}_{ii}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} - t^{\mu\nu}_{ii}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} \Big] \chi^{\sigma+\mu\nu\gamma\gamma}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; 0) \\
&+ \Big[t^{\mu\nu}_{ii}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} - t^{\mu\nu}_{ii}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} \Big] \chi^{\sigma-\mu\nu\gamma\gamma}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; 0) \Big\} \\
&= \frac{i g_S \mu_B b_0 e}{2\hbar} \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\substack{\mu\nu\gamma \\ \sigma}} \sum_{\ell} \Big[\chi^{\sigma+\mu\nu\gamma\gamma}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; 0) + \chi^{\sigma-\mu\nu\gamma\gamma}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; 0) \Big] \\
&\times \Big[t^{\mu\nu}_{ii}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} - t^{\mu\nu}_{ii}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} \Big] \ .
\end{aligned}
\tag{5.169}
$$

$$
\begin{aligned}
\langle \hat{I}^{\mathrm{C}}_{ii}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel; t)\rangle &= \frac{i g_S \mu_B b_0 e}{2\hbar} \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\substack{\mu\nu\gamma \\ \sigma}} \sum_{\ell} \Big[\chi^{\sigma+\mu\nu\gamma\gamma}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; 0) + \chi^{\sigma-\mu\nu\gamma\gamma}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; 0) \Big] \\
&\times \Big[t^{\mu\nu}_{ii}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} - t^{\mu\nu}_{ii}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} \Big] \ .
\end{aligned}
\tag{5.170}
$$

Writing the susceptibilities within mean field approximation in terms of the monoeletronic propagators,

$$
\chi^{\sigma+\mu\nu\gamma\gamma}_{(0)ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; 0) = \frac{i\hbar}{2\pi} \int_{-\infty}^{\omega_F} d\omega' \Big\{ G^{\sigma\uparrow\nu\gamma}_{i\ell}(\omega') G^{\downarrow\sigma\gamma\mu}_{\ell i}(\omega') - \Big[G^{\sigma\downarrow\mu\gamma}_{i\ell}(\omega') G^{\uparrow\sigma\gamma\nu}_{\ell i}(\omega') \Big]^* \Big\} \ .
\tag{5.171}
$$

$$
\chi^{\sigma-\mu\nu\gamma\gamma}_{(0)ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; 0) = \frac{i\hbar}{2\pi} \int_{-\infty}^{\omega_F} d\omega' \Big\{ G^{\sigma\downarrow\nu\gamma}_{i\ell}(\omega') G^{\uparrow\sigma\gamma\mu}_{\ell i}(\omega') - \Big[G^{\sigma\uparrow\mu\gamma}_{i\ell}(\omega') G^{\downarrow\sigma\gamma\nu}_{\ell i}(\omega') \Big]^* \Big\} \ .
\tag{5.172}
$$

The orbital contribution may be found following the same steps using the hamiltonian given by Eq. (5.108). Therefore, the analogous of Eq. (5.158) is

$$
\begin{aligned}
\langle \hat{\mathcal{I}}_{ij}^{\mathrm{C}}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel; t)\rangle^L = \frac{i\mu_B b_0 e}{2\hbar} \sum_{\mathbf{k}_\parallel} \sum_{\substack{\mu\nu\gamma\xi \\ \sigma\sigma'}} \sum_{\ell} t_{ij}^{\mu\nu}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel' - \mathbf{R}_\parallel)} \\
\times \Big\{ e^{i\omega t} \hat{L}'^{+}_{\gamma\xi} \chi_{ij\ell\ell}^{\sigma\sigma'\,\mu\nu\gamma\xi}(\mathbf{k}_\parallel, \mathbf{k}_\parallel - \mathbf{Q}_\parallel; -\omega)\delta_{-\mathbf{q}_\parallel\mathbf{Q}_\parallel} \\
+ e^{-i\omega t} \hat{L}'^{-}_{\gamma\xi} \chi_{ij\ell\ell}^{\sigma\sigma'\,\mu\nu\gamma\xi}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega)\delta_{\mathbf{q}_\parallel\mathbf{Q}_\parallel} \Big\} .
\end{aligned} \tag{5.173}
$$

Substituting in Eq. (5.154) and multiplying all the terms we can write

$$
\begin{aligned}
\langle \hat{I}_{ij}^{\mathrm{C}}(\mathbf{R}_\parallel, \mathbf{R}_\parallel'; t)\rangle^L = \frac{i\mu_B b_0 e}{2\hbar} \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\substack{\mu\nu\gamma\xi \\ \sigma\sigma'}} \sum_{\ell} \\
\times \Big\{ t_{ij}^{\mu\nu}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel' - \mathbf{R}_\parallel)} e^{i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel')} \hat{L}'^{+}_{\gamma\xi} \chi_{ij\ell\ell}^{\sigma\sigma'\,\mu\nu\gamma\xi}(\mathbf{k}_\parallel, \mathbf{k}_\parallel - \mathbf{Q}_\parallel; -\omega) \\
+ t_{ij}^{\mu\nu}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel' - \mathbf{R}_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel')} \hat{L}'^{-}_{\gamma\xi} \chi_{ij\ell\ell}^{\sigma\sigma'\,\mu\nu\gamma\xi}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \\
- t_{ji}^{\nu\mu}(\mathbf{R}_\parallel - \mathbf{R}_\parallel') e^{i(\mathbf{k}_\parallel - \mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel - \mathbf{R}_\parallel')} e^{i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel')} \hat{L}'^{+}_{\gamma\xi} \chi_{ji\ell\ell}^{\sigma\sigma'\,\nu\mu\gamma\xi}(\mathbf{k}_\parallel, \mathbf{k}_\parallel - \mathbf{Q}_\parallel; -\omega) \\
- t_{ji}^{\nu\mu}(\mathbf{R}_\parallel - \mathbf{R}_\parallel') e^{i(\mathbf{k}_\parallel + \mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel - \mathbf{R}_\parallel')} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel')} \hat{L}'^{-}_{\gamma\xi} \chi_{ji\ell\ell}^{\sigma\sigma'\,\nu\mu\gamma\xi}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \Big\} .
\end{aligned} \tag{5.174}
$$

Changing the position of the second and third terms and writing the first two in terms of the conjugate,

$$
\begin{aligned}
\langle \hat{I}_{ij}^{\mathrm{C}}(\mathbf{R}_\parallel, \mathbf{R}_\parallel'; t)\rangle^L = \frac{i\mu_B b_0 e}{2\hbar} \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\substack{\mu\nu\gamma\xi \\ \sigma\sigma'}} \sum_{\ell} \\
\times \Big\{ \Big[t_{ji}^{\nu\mu}(\mathbf{R}_\parallel - \mathbf{R}_\parallel') e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel' - \mathbf{R}_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel')} \hat{L}'^{-}_{\xi\gamma} \chi_{ji\ell\ell}^{\sigma\sigma'\,\nu\mu\xi\gamma}(\mathbf{k}_\parallel - \mathbf{Q}_\parallel, \mathbf{k}_\parallel; \omega) \Big]^* \\
- \Big[t_{ij}^{\mu\nu}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel) e^{-i(\mathbf{k}_\parallel - \mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel - \mathbf{R}_\parallel')} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel')} \hat{L}'^{-}_{\xi\gamma} \chi_{ij\ell\ell}^{\sigma\sigma'\,\mu\nu\xi\gamma}(\mathbf{k}_\parallel - \mathbf{Q}_\parallel, \mathbf{k}_\parallel; \omega) \Big]^* \\
+ t_{ij}^{\mu\nu}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel' - \mathbf{R}_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel')} \hat{L}'^{-}_{\gamma\xi} \chi_{ij\ell\ell}^{\sigma\sigma'\,\mu\nu\gamma\xi}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \\
- t_{ji}^{\nu\mu}(\mathbf{R}_\parallel - \mathbf{R}_\parallel') e^{i(\mathbf{k}_\parallel + \mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel - \mathbf{R}_\parallel')} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}_\parallel')} \hat{L}'^{-}_{\gamma\xi} \chi_{ji\ell\ell}^{\sigma\sigma'\,\nu\mu\gamma\xi}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \Big\} .
\end{aligned} \tag{5.175}
$$

Changing the variable $\mathbf{k}_\parallel \to \mathbf{k}_\parallel + \mathbf{Q}_\parallel$ and also $\gamma \rightleftharpoons \xi$ in the first two terms,

$$\begin{aligned}
\langle \hat{I}^{\mathrm{C}}_{ij}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel; t)\rangle^L = &\frac{i\mu_B b_0 e}{2\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\mu\nu\gamma\xi\\ \sigma\sigma'}}\sum_{\ell} \\
&\times \left\{ \left[t^{\nu\mu}_{ji}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{-i\left(\mathbf{k}_\parallel + \mathbf{Q}_\parallel\right)\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)} \hat{L}'^{-}_{\gamma\xi} \chi^{\sigma\sigma'\,\nu\mu\gamma\xi}_{ji\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega)\right]^* \right. \\
&- \left[t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)} \hat{L}'^{-}_{\gamma\xi} \chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{ij\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega)\right]^* \\
&+ t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)} \hat{L}'^{-}_{\gamma\xi} \chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{ij\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \\
&\left. - t^{\nu\mu}_{ji}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i(\mathbf{k}_\parallel + \mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)} \hat{L}'^{-}_{\gamma\xi} \chi^{\sigma\sigma'\,\nu\mu\gamma\xi}_{ji\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \right\} ,
\end{aligned} \tag{5.176}$$

or

$$\begin{aligned}
\langle \hat{I}^{\mathrm{C}}_{ij}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel; t)\rangle^L = &-\frac{\mu_B b_0 e}{\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\mu\nu\gamma\xi\\ \sigma\sigma'}}\sum_{\ell} \\
&\times \mathrm{Im} \left\{ t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)} \hat{L}'^{-}_{\gamma\xi} \chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{ij\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \right. \\
&\left. - t^{\nu\mu}_{ji}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i(\mathbf{k}_\parallel + \mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)} \hat{L}'^{-}_{\gamma\xi} \chi^{\sigma\sigma'\,\nu\mu\gamma\xi}_{ji\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \right\} .
\end{aligned} \tag{5.177}$$

For sites in the same plane, using the relation $t^{\mu\nu}_{ii}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) = t^{\nu\mu}_{ii}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)$, we can write

$$\begin{aligned}
\langle \hat{I}^{\mathrm{C}}_{ii}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel; t)\rangle^L = &-\frac{\mu_B b_0 e}{\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\mu\nu\gamma\xi\\ \sigma\sigma'}}\sum_{\ell} \\
&\times \mathrm{Im} \left\{ t^{\mu\nu}_{ii}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)} \hat{L}'^{-}_{\gamma\xi} \chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \right. \\
&\left. - t^{\nu\mu}_{ii}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i(\mathbf{k}_\parallel + \mathbf{Q}_\parallel)\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} e^{-i(\omega t - \mathbf{Q}_\parallel\cdot\mathbf{R}'_\parallel)} \hat{L}'^{-}_{\gamma\xi} \chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega) \right\} ,
\end{aligned} \tag{5.178}$$

Defining

$$\begin{aligned}
I^{CL}_i(\mathbf{R}'_\parallel - \mathbf{R}_\parallel; \mathbf{Q}_\parallel; \omega) = &\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\substack{\mu\nu\gamma\xi\\ \sigma\sigma'}}\sum_{\ell} \chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{ii\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{Q}_\parallel; \omega)\hat{L}'^{-}_{\gamma\xi} \\
&\times \left[t^{\mu\nu}_{ii}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} - t^{\nu\mu}_{ii}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{-i(\mathbf{k}_\parallel + \mathbf{Q}_\parallel)\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \right] \\
= &|I^{CL}_i(\mathbf{R}'_\parallel - \mathbf{R}_\parallel; \mathbf{Q}_\parallel; \omega)| e^{i\phi^L_C} ,
\end{aligned} \tag{5.179}$$

where

$$\begin{aligned}
\cos\phi_C^L =& \frac{\mathrm{Re}\left[I_i^{CL}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel; \mathbf{Q}_\parallel; \omega)\right]}{|I_i^{CL}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel; \mathbf{Q}_\parallel; \omega)|}\ , \text{ and} \\
\sin\phi_C^L =& \frac{\mathrm{Im}\left[I_i^{CL}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel; \mathbf{Q}_\parallel; \omega)\right]}{|I_i^{CL}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel; \mathbf{Q}_\parallel; \omega)|}\ ,
\end{aligned} \tag{5.180}$$

we finally end up with

$$\langle \hat{I}_{ii}^{\mathrm{C}}(\mathbf{R}_\parallel, \mathbf{R}_\parallel'; \mathbf{Q}_\parallel; t)\rangle^L = \frac{\mu_B b_0 e}{\hbar} |I_i^{CL}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel; \mathbf{Q}_\parallel; \omega)| \sin(\omega t - \mathbf{Q}_\parallel \cdot \mathbf{R}_\parallel' - \phi_C^L)\ . \tag{5.181}$$

For uniform excitations, i.e., $\mathbf{Q}_\parallel = 0$, the spin contribution to the charge current is given by

$$\langle \hat{I}_{ii}^{\mathrm{C}}(\mathbf{R}_\parallel, \mathbf{R}_\parallel'; 0; t)\rangle = \frac{g_S \mu_B b_0 e}{\hbar} |I_i^{\mathrm{C}}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel; 0; \omega)| \sin(\omega t - \phi_C)\ , \tag{5.182}$$

where

$$\begin{aligned}
I_i^{\mathrm{C}}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel; 0; \omega) =& \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\substack{\mu\nu\gamma \\ \sigma}} \sum_{\ell} \chi_{ii\ell\ell}^{\sigma-\ \mu\nu\gamma\gamma}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; \omega) \\
& \times \left[t_{ii}^{\mu\nu}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}_\parallel' - \mathbf{R}_\parallel)} - t_{ii}^{\nu\mu}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel) e^{-i\mathbf{k}_\parallel \cdot (\mathbf{R}_\parallel' - \mathbf{R}_\parallel)}\right]\ .
\end{aligned} \tag{5.183}$$

The orbital angular momentum part is

$$\langle \hat{I}_{ii}^{\mathrm{C}}(\mathbf{R}_\parallel, \mathbf{R}_\parallel'; 0; t)\rangle^L = \frac{g_S \mu_B b_0 e}{\hbar} |I_i^{CL}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel; 0; \omega)| \sin(\omega t - \phi_C^L)\ , \tag{5.184}$$

with

$$\begin{aligned}
I_i^{CL}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel; 0; \omega) =& \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\substack{\mu\nu\gamma\xi \\ \sigma\sigma'}} \sum_{\ell} \chi_{ii\ell\ell}^{\sigma\sigma'\ \mu\nu\gamma\xi}(\mathbf{k}_\parallel, \mathbf{k}_\parallel; \omega) \hat{L}_{\gamma\xi}'^{-} \\
& \times \left[t_{ii}^{\mu\nu}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel \cdot (\mathbf{R}_\parallel' - \mathbf{R}_\parallel)} - t_{ii}^{\nu\mu}(\mathbf{R}_\parallel' - \mathbf{R}_\parallel) e^{-i\mathbf{k}_\parallel \cdot (\mathbf{R}_\parallel' - \mathbf{R}_\parallel)}\right]\ .
\end{aligned} \tag{5.185}$$

5.3.5 Spin Disturbance

The spin disturbance caused by the transverse magnetic field is given by

$$\begin{aligned}
\delta\langle\hat{S}_i^m\rangle(t) =& -\frac{i}{\hbar}\int dt'\Theta(t-t')\left\langle\left[\hat{S}_i^m(t),\hat{H}_{\rm int}(t')\right]\right\rangle \\
=& -\frac{ig_S\mu_B b_0}{2\hbar}\sum_\ell\int dt'\Theta(t-t')\left\langle\left[\hat{S}_i^m(t),e^{i\omega t'}\hat{S}_\ell^+(t')+e^{-i\omega t'}\hat{S}_\ell^-(t')\right]\right\rangle \\
=&\frac{g_S\mu_B b_0}{2}\sum_{\mu\nu}\sum_\ell\int dt'\left\{e^{i\omega t'}\chi_{i\ell}^{m+\mu\nu}(t-t')+e^{-i\omega t'}\chi_{i\ell}^{m-\mu\nu}(t-t')\right\} \\
=&\frac{g_S\mu_B b_0}{2}\sum_{\mu\nu}\sum_\ell\int dt'\left\{e^{i\omega t}e^{-i\omega(t-t')}\chi_{i\ell}^{m+\mu\nu}(t-t')+e^{-i\omega t}e^{i\omega(t-t')}\chi_{i\ell}^{m-\mu\nu}(t-t')\right\} \\
=&\frac{g_S\mu_B b_0}{2}\sum_{\mu\nu}\sum_\ell\left\{e^{i\omega t}\chi_{i\ell}^{m+\mu\nu}(-\omega)+e^{-i\omega t}\chi_{i\ell}^{m-\mu\nu}(\omega)\right\} \ .
\end{aligned} \tag{5.186}$$

The transverse components can be obtained through

$$\begin{aligned}
\delta\langle\hat{S}_i^+\rangle(t) =&\frac{g_S\mu_B b_0}{2}\sum_{\mu\nu}\sum_\ell\left\{e^{i\omega t}\chi_{i\ell}^{++\mu\nu}(-\omega)+e^{-i\omega t}\chi_{i\ell}^{+-\mu\nu}(\omega)\right\} \\
=&\frac{g_S\mu_B b_0}{2}\sum_{\mu\nu}\sum_\ell\left\{e^{i\omega t}\left[\chi_{i\ell}^{--\mu\nu}(\omega)\right]^*+e^{-i\omega t}\chi_{i\ell}^{+-\mu\nu}(\omega)\right\} \ .
\end{aligned} \tag{5.187}$$

So, the expectation value of S^x and S^y are

$$\begin{aligned}
\delta\langle\hat{S}_i^x\rangle(t) =&\frac{g_S\mu_B b_0}{2}\sum_{\mu\nu}\sum_\ell{\rm Re}\left\{e^{i\omega t}\left[\chi_{i\ell}^{--\mu\nu}(\omega)\right]^*+e^{-i\omega t}\chi_{i\ell}^{+-\mu\nu}(\omega)\right\} \\
=&g_S\mu_B b_0\ {\rm Re}\left\{e^{i\omega t}\left[S_+(\omega)\right]^*+e^{-i\omega t}S_-(\omega)\right\} \ .
\end{aligned} \tag{5.188}$$

$$\begin{aligned}
\delta\langle\hat{S}_i^y\rangle(t) =&\frac{g_S\mu_B b_0}{2}\sum_{\mu\nu}\sum_\ell{\rm Im}\left\{e^{i\omega t}\left[\chi_{i\ell}^{--\mu\nu}(\omega)\right]^*+e^{-i\omega t}\chi_{i\ell}^{+-\mu\nu}(\omega)\right\} \\
=&g_S\mu_B b_0\ {\rm Im}\left\{e^{i\omega t}\left[S_+(\omega)\right]^*+e^{-i\omega t}S_-(\omega)\right\} \ ,
\end{aligned} \tag{5.189}$$

where

$$S_+(\omega) = \frac{1}{2}\sum_{\mu\nu}\sum_\ell\chi_{i\ell}^{--\mu\nu}(\omega) \ , \tag{5.190}$$

$$S_-(\omega) = \frac{1}{2}\sum_{\mu\nu}\sum_\ell\chi_{i\ell}^{+-\mu\nu}(\omega) \ . \tag{5.191}$$

To obtain the precession amplitude and the phase delay to the applied field, we can rewrite the above equations as

$$\begin{aligned}\delta\langle\hat{S}_i^x\rangle(t) &= g_S\mu_B b_0\ \mathrm{Re}\left\{[\cos(\omega t)+i\sin(\omega t)]\,[S_+(\omega)]^* + [\cos(\omega t)-i\sin(\omega t)]\,S_-(\omega)\right\}\\ &= g_S\mu_B b_0\ \left\{\cos(\omega t)\mathrm{Re}\,[S_+(\omega)] + \sin(\omega t)\mathrm{Im}\,[S_+(\omega)]\right.\\ &\qquad\left. + \cos(\omega t)\mathrm{Re}\,[S_-(\omega)] + \sin(\omega t)\mathrm{Im}\,[S_-(\omega)]\right\}\\ &= g_S\mu_B b_0\ \left\{\cos(\omega t)\mathrm{Re}\,[S_+(\omega)+S_-(\omega)] + \sin(\omega t)\mathrm{Im}\,[S_+(\omega)+S_-(\omega)]\right\}\ .\end{aligned}\tag{5.192}$$

and

$$\begin{aligned}\delta\langle\hat{S}_i^y\rangle(t) &= g_S\mu_B b_0\ \mathrm{Im}\left\{[\cos(\omega t)+i\sin(\omega t)]\,[S_+(\omega)]^* + [\cos(\omega t)-i\sin(\omega t)]\,S_-(\omega)\right\}\\ &= g_S\mu_B b_0\ \left\{\sin(\omega t)\mathrm{Re}\,[S_+(\omega)] - \cos(\omega t)\mathrm{Im}\,[S_+(\omega)]\right.\\ &\qquad\left. - \sin(\omega t)\mathrm{Re}\,[S_-(\omega)] + \cos(\omega t)\mathrm{Im}\,[S_-(\omega)]\right\}\\ &= g_S\mu_B b_0\ \left\{\sin(\omega t)\mathrm{Re}\,[S_+(\omega)-S_-(\omega)] - \cos(\omega t)\mathrm{Im}\,[S_+(\omega)-S_-(\omega)]\right\}\ .\end{aligned}\tag{5.193}$$

When spin-orbit coupling is not present, $S_+(\omega) = \frac{1}{2}\sum_{\mu\nu}\sum_\ell \chi_{i\ell}^{--} = 0$ and we can see that

$$\begin{aligned}\delta\langle\hat{S}_i^x\rangle(t) =& g_S\mu_B b_0\ \left\{\cos(\omega t)\mathrm{Re}\,[S_-(\omega)] + \sin(\omega t)\mathrm{Im}\,[S_-(\omega)]\right\}\\ =& g_S\mu_B b_0\ \left\{\cos(\omega t)\mathrm{Re}\,[S_-(\omega)] + \sin(\omega t)\mathrm{Re}\,[iS_-(\omega)]\right\}\\ =& g_S\mu_B b_0\ \mathrm{Re}\left\{\cos(\omega t)S_-(\omega) - i\sin(\omega t)S_-(\omega)\right\}\\ =& g_S\mu_B b_0\ \mathrm{Re}\left\{e^{-i\omega t}S_-(\omega)\right\}\\ =& \frac{g_S\mu_B b_0}{2}\sum_{\mu\nu}\sum_\ell \mathrm{Re}\left\{e^{-i\omega t}\chi_{i\ell}^{+-\,\mu\nu}(\omega)\right\}\\ =& \mathrm{Re}\left[\delta\langle\hat{S}_i^+\rangle(t)\right]\ ,\end{aligned}\tag{5.194}$$

and, analogously,

$$\begin{aligned}\delta\langle\hat{S}_i^y\rangle(t) =& g_S\mu_B b_0\ \left\{\sin(\omega t)\mathrm{Re}\,[-S_-(\omega)] - \cos(\omega t)\mathrm{Im}\,[-S_-(\omega)]\right\}\\ =& g_S\mu_B b_0\ \left\{\sin(\omega t)\mathrm{Im}\,[-iS_-(\omega)] + \cos(\omega t)\mathrm{Im}\,[S_-(\omega)]\right\}\\ =& g_S\mu_B b_0\ \mathrm{Im}\left\{-i\sin(\omega t)S_-(\omega) + \cos(\omega t)S_-(\omega)\right\}\\ =& g_S\mu_B b_0\ \mathrm{Im}\left\{e^{-i\omega t}S_-(\omega)\right\}\\ =& \frac{g_S\mu_B b_0}{2}\sum_{\mu\nu}\sum_\ell \mathrm{Im}\left\{e^{-i\omega t}\chi_{i\ell}^{+-\,\mu\nu}(\omega)\right\}\\ =& \mathrm{Im}\left[\delta\langle\hat{S}_i^+\rangle(t)\right]\ ,\end{aligned}\tag{5.195}$$

as expected from previous calculations.

It is convenient to write the change in S^x and S^y as

$$\begin{aligned}
\delta\langle\hat{S}_i^x\rangle(t) &= g_S\mu_B b_0\ \ |S_+(\omega)+S_-(\omega)|\left\{\cos(\omega t)\frac{\mathrm{Re}\left[S_+(\omega)+S_-(\omega)\right]}{|S_+(\omega)+S_-(\omega)|}\right.\\
&\qquad\qquad\left.+\sin(\omega t)\frac{\mathrm{Im}\left[S_+(\omega)+S_-(\omega)\right]}{|S_+(\omega)+S_-(\omega)|}\right\} \\
&= g_S\mu_B b_0\ \ |S_+(\omega)+S_-(\omega)|\left\{\cos(\omega t)\cos\phi^x+\sin(\omega t)\sin\phi^x\right\}\\
&= g_S\mu_B b_0\ \ |S_+(\omega)+S_-(\omega)|\cos(\omega t-\phi^x)\ .
\end{aligned} \tag{5.196}$$

where

$$|S_+(\omega)+S_-(\omega)| = \left|\frac{1}{2}\sum_{\mu\nu}\sum_{\ell}\chi_{i\ell}^{--\,\mu\nu}(\omega)+\chi_{i\ell}^{+-\,\mu\nu}(\omega)\right| \tag{5.197}$$

$$\cos\phi^x = \frac{\mathrm{Re}\left[S_+(\omega)+S_-(\omega)\right]}{|S_+(\omega)+S_-(\omega)|} \tag{5.198}$$

$$\sin\phi^x = \frac{\mathrm{Im}\left[S_+(\omega)+S_-(\omega)\right]}{|S_+(\omega)+S_-(\omega)|} \tag{5.199}$$

Similarly,

$$\begin{aligned}
\delta\langle\hat{S}_i^y\rangle(t) &= g_S\mu_B b_0\ \ |S_+(\omega)-S_-(\omega)|\left\{\sin(\omega t)\frac{\mathrm{Re}\left[S_+(\omega)-S_-(\omega)\right]}{|S_+(\omega)-S_-(\omega)|}\right.\\
&\qquad\qquad\left.-\cos(\omega t)\frac{\mathrm{Im}\left[S_+(\omega)-S_-(\omega)\right]}{|S_+(\omega)-S_-(\omega)|}\right\} \\
&= g_S\mu_B b_0\ \ |S_+(\omega)-S_-(\omega)|\left\{\sin(\omega t)\cos(\phi^y)-\cos(\omega t)\sin(\phi^y)\right\}\\
&= g_S\mu_B b_0\ \ |S_+(\omega)-S_-(\omega)|\sin(\omega t-\phi^y)\ ,
\end{aligned} \tag{5.200}$$

where

$$|S_+(\omega)-S_-(\omega)| = \left|\frac{1}{2}\sum_{\mu\nu}\sum_{\ell}\chi_{i\ell}^{--\,\mu\nu}(\omega)-\chi_{i\ell}^{+-\,\mu\nu}(\omega)\right| \tag{5.201}$$

$$\cos\phi^y = \frac{\mathrm{Re}\left[S_+(\omega)-S_-(\omega)\right]}{|S_+(\omega)-S_-(\omega)|} \tag{5.202}$$

$$\sin\phi^y = \frac{\mathrm{Im}\left[S_+(\omega)-S_-(\omega)\right]}{|S_+(\omega)-S_-(\omega)|} \tag{5.203}$$

5.3.6 Charge Disturbance

The charge disturbance can be obtained calculating the expectation value of the operator

$$
\begin{aligned}
\hat{\rho}_i =& e\sum_{\sigma} \hat{n}_{i\sigma} \\
=& e\sum_{\mu\sigma} c^{\dagger}_{i\mu\sigma} c_{i\mu\sigma}
\end{aligned}
\tag{5.204}
$$

$$
\begin{aligned}
\delta\langle\hat{\rho}_i\rangle(t) =& -\frac{i}{\hbar}\int dt'\Theta(t-t')\left\langle\left[\hat{\rho}_i(t), \hat{H}_{\mathrm{int}}(t')\right]\right\rangle \\
=& -\frac{ig_S\mu_B b_0}{2\hbar}\sum_{\ell}\int dt'\Theta(t-t')\left\langle\left[\hat{\rho}_i(t), e^{i\omega t'}\hat{S}^{+}_{\ell}(t') + e^{-i\omega t'}\hat{S}^{-}_{\ell}(t')\right]\right\rangle \\
=& -\frac{ieg_S\mu_B b_0}{2\hbar}\sum_{\ell\mu\sigma}\int dt'\Theta(t-t')\left\langle\left[c^{\dagger}_{i\mu\sigma}c_{i\mu\sigma}(t), e^{i\omega t'}\hat{S}^{+}_{\ell}(t') + e^{-i\omega t'}\hat{S}^{-}_{\ell}(t')\right]\right\rangle \\
=& \frac{eg_S\mu_B b_0}{2}\sum_{\ell\mu\sigma}\int dt'\left\{e^{i\omega t'}\chi^{\sigma+\mu\nu}_{i\ell}(t-t') + e^{-i\omega t'}\chi^{\sigma-\mu\nu}_{i\ell}(t-t')\right\} \\
=& \frac{eg_S\mu_B b_0}{2}\sum_{\ell\mu\sigma}\left\{e^{i\omega t}\chi^{\sigma+\mu\nu}_{i\ell}(-\omega) + e^{-i\omega t}\chi^{\sigma-\mu\nu}_{i\ell}(\omega)\right\} \\
=& \frac{eg_S\mu_B b_0}{2}\sum_{\ell\mu\sigma}\left\{\left[e^{-i\omega t}\chi^{\sigma-\mu\nu}_{i\ell}(\omega)\right]^{*} + e^{-i\omega t}\chi^{\sigma-\mu\nu}_{i\ell}(\omega)\right\} \\
=& eg_S\mu_B b_0\sum_{\ell\mu\sigma}\mathrm{Re}\left\{e^{-i\omega t}\chi^{\sigma-\mu\nu}_{i\ell}(\omega)\right\} \ .
\end{aligned}
\tag{5.205}
$$

5.3.7 Orbital Angular Momentum Disturbance

The orbital disturbance caused by the transverse magnetic field can be obtained using the operator given in Eq. (5.50). In linear response theory, we have

$$\hat{L}_i^m = \sum_\sigma \sum_{\mu\nu} L_{\mu\nu}^m c_{i\mu\sigma}^\dagger c_{i\nu\sigma} \ , \tag{5.206}$$

$$\begin{aligned}
\delta\langle\hat{L}_i^m\rangle(t) =& -\frac{i}{\hbar}\int dt'\Theta(t-t')\left\langle\left[\hat{L}_i^m(t),\hat{H}_{\text{int}}(t')\right]\right\rangle \\
=& -\frac{ig_S\mu_B b_0}{2\hbar}\sum_\ell\int dt'\Theta(t-t')\left\langle\left[\hat{L}_i^m(t),e^{i\omega t'}\hat{S}_\ell^+(t')+e^{-i\omega t'}\hat{S}_\ell^-(t')\right]\right\rangle \\
=&\frac{g_S\mu_B b_0}{2}\sum_{\mu\nu\gamma}\sum_\ell\sum_\sigma\int dt' L_{\mu\nu}^m\left\{e^{i\omega t'}\chi_{ii\ell\ell}^{\sigma+\mu\nu\gamma\gamma}(t-t')+e^{-i\omega t'}\chi_{ii\ell\ell}^{\sigma-\mu\nu\gamma\gamma}(t-t')\right\} \\
=&\frac{g_S\mu_B b_0}{2}\sum_{\mu\nu\gamma}\sum_\ell\sum_\sigma\int dt' L_{\mu\nu}^m\left\{e^{i\omega t}e^{-i\omega(t-t')}\chi_{ii\ell\ell}^{\sigma+\mu\nu\gamma\gamma}(t-t')+e^{-i\omega t}e^{i\omega(t-t')}\chi_{ii\ell\ell}^{\sigma-\mu\nu\gamma\gamma}(t-t')\right\} \\
=&\frac{g_S\mu_B b_0}{2}\sum_{\mu\nu\gamma}\sum_\ell\sum_\sigma L_{\mu\nu}^m\left\{e^{i\omega t}\chi_{ii\ell\ell}^{\sigma+\mu\nu\gamma\gamma}(-\omega)+e^{-i\omega t}\chi_{ii\ell\ell}^{\sigma-\mu\nu\gamma\gamma}(\omega)\right\} \ .
\end{aligned} \tag{5.207}$$

For $m = x, y, z$, we can write

$$\begin{aligned}
\delta\langle\hat{L}_i^m\rangle(t) =&\frac{g_S\mu_B b_0}{2}\sum_{\mu\nu\gamma}\sum_\ell\sum_\sigma\left\{\left[L_{\nu\mu}^m e^{-i\omega t}\chi_{ii\ell\ell}^{\sigma-\nu\mu\gamma\gamma}(\omega)\right]^*+L_{\mu\nu}^m e^{-i\omega t}\chi_{ii\ell\ell}^{\sigma-\mu\nu\gamma\gamma}(\omega)\right\} \\
=&\frac{g_S\mu_B b_0}{2}\sum_{\mu\nu\gamma}\sum_\ell\sum_\sigma\left\{\left[L_{\mu\nu}^m e^{-i\omega t}\chi_{ii\ell\ell}^{\sigma-\mu\nu\gamma\gamma}(\omega)\right]^*+L_{\mu\nu}^m e^{-i\omega t}\chi_{ii\ell\ell}^{\sigma-\mu\nu\gamma\gamma}(\omega)\right\} \\
=&g_S\mu_B b_0\sum_{\mu\nu\gamma}\sum_\ell\sum_\sigma \mathrm{Re}\left\{L_{\mu\nu}^m e^{-i\omega t}\chi_{ii\ell\ell}^{\sigma-\mu\nu\gamma\gamma}(\omega)\right\} \ .
\end{aligned} \tag{5.208}$$

5.4 Electric Field Perturbation (Through Vector Potential)

Another way to describe an electric field is using a vector potential $\mathbf{A}(\mathbf{r}, t)$. The perturbation hamiltonian can be obtained from the unperturbed one using the transformation in momentum $\mathbf{p} \to \mathbf{p} - e\mathbf{A}$. So, the total hamiltonian have the form

$$\hat{H} = \sum_\ell \frac{(\mathbf{p}_\ell - e\mathbf{A}(\mathbf{r}_\ell, t))^2}{2m} + U(\mathbf{r}_\ell) \ . \tag{5.209}$$

where $\mathbf{r}_\ell$ and $\mathbf{p}_\ell$ are the position and momentum of particle ℓ, respectively.

5.4.1 Linear term

For relatively small electric fields, the linear perturbation term of the hamiltonian is given by

$$\begin{aligned}\hat{H}_{\text{int}}(t) =& - e \sum_{\ell} \frac{\mathbf{p}_\ell(t) \cdot \mathbf{A}(\mathbf{r}_\ell, t)}{m} \\ =& - e \sum_{\ell} \dot{\mathbf{r}}_\ell(t) \cdot \mathbf{A}(\mathbf{r}_\ell, t) \ .\end{aligned} \tag{5.210}$$

If we consider a distribution of charges $\rho(\mathbf{r})$, this expression can be modified to

$$\begin{aligned}\hat{H}_{\text{int}}(t) =& - \int d\mathbf{r} \rho(\mathbf{r}) \dot{\mathbf{r}}(t) \cdot \mathbf{A}(\mathbf{r}, t) \\ =& - \int d\mathbf{r} \hat{\mathbf{J}}^{\text{C}}(\mathbf{r}, t) \cdot \mathbf{A}(\mathbf{r}, t) \ .\end{aligned} \tag{5.211}$$

$\hat{\mathbf{J}}^{\text{C}}(\mathbf{r}, t) = \rho(\mathbf{r})\dot{\mathbf{r}}(t)$ is the charge current density operator. Note that if we use the distribution of a set of discrete particles $\rho(\mathbf{r}) = \sum_\ell e\delta(\mathbf{r} - \mathbf{r}_\ell)$ in Eq. (5.211), we recover Eq. (5.210).

Let's consider an harmonic electric field with frequency ω and wave vector $\mathbf{q}$ applied in direction of the unit vector $\hat{\mathbf{u}}_E$

$$\mathbf{E}(\mathbf{r}, t) = E_0 \cos(\mathbf{q} \cdot \mathbf{r} - \omega t) \hat{\mathbf{u}}_E \ . \tag{5.212}$$

The relation between the electric field and the vector potential is given by

$$\mathbf{E}(\mathbf{r}, t) = -\nabla \phi - \frac{\partial \mathbf{A}(\mathbf{r}, t)}{\partial t} \ . \tag{5.213}$$

For a neutral distribution of charges (e.g., a background of positively charged ions where the electrons move through), $\nabla \cdot \mathbf{E} = 0$. Within Coulomb's gauge ($\nabla \cdot \mathbf{A} = 0$), this means that the scalar potential ϕ vanishes, or $\nabla \phi = 0$. Then, we have

$$\mathbf{E}(\mathbf{r}, t) = -\frac{\partial \mathbf{A}(\mathbf{r}, t)}{\partial t} \ , \tag{5.214}$$

and

$$\begin{aligned}\mathbf{A}(\mathbf{r}, t) =& - \int^t \mathbf{E}(\mathbf{r}, t') dt' \\ =& - E_0 \hat{\mathbf{u}}_E \int^t \cos(\mathbf{q} \cdot \mathbf{r} - \omega t') dt' \\ =& \frac{1}{\omega} E_0 \hat{\mathbf{u}}_E \sin(\mathbf{q} \cdot \mathbf{r} - \omega t) \\ =& \frac{1}{2i\omega} E_0 \hat{\mathbf{u}}_E \left[e^{i(\mathbf{q} \cdot \mathbf{r} - \omega t)} - e^{-i(\mathbf{q} \cdot \mathbf{r} - \omega t)} \right] \ .\end{aligned} \tag{5.215}$$

In general, we have

$$\begin{aligned}\mathbf{A}(\mathbf{r},t) =& -\int^t dt'\mathbf{E}(\mathbf{r},t') \\ =& -\frac{1}{2\pi}\int^t dt'\int d\omega e^{-i\omega t'}\mathbf{E}(\mathbf{r},\omega) \\ =& \frac{1}{2\pi}\int d\omega\frac{1}{i\omega}e^{-i\omega t}\mathbf{E}(\mathbf{r},\omega)\ .\end{aligned} \tag{5.216}$$

Finally, the perturbation hamiltonian is

$$\hat{H}_{\text{int}}(t) = -\frac{E_0}{2i\omega}\int d\mathbf{r}\hat{\mathbf{J}}^{\mathrm{C}}(\mathbf{r},t)\cdot\hat{\mathbf{u}}_E\left[e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} - e^{-i(\mathbf{q}\cdot\mathbf{r}-\omega t)}\right]\ . \tag{5.217}$$

Using $\hat{\mathbf{J}}^{\mathrm{C}}(\mathbf{r},t)$ given by Eq. (5.34), we obtain

$$\begin{aligned}\hat{H}_{\text{int}}(t) =& -\frac{eE_0}{4im\omega}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}}\int d\mathbf{r}\ \mathbf{w}^{\mu\nu}_{ij}(\mathbf{r})\cdot\hat{\mathbf{u}}_E\left[e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} - e^{-i(\mathbf{q}\cdot\mathbf{r}-\omega t)}\right]c^\dagger_{i\mu\sigma}(t)c_{j\nu\sigma}(t) \\ =& -\frac{eE_0}{4im\omega}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}}\left[\mathbf{w}^{\mu\nu}_{ij}(\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{-i\omega t} - \mathbf{w}^{\mu\nu}_{ij}(-\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{i\omega t}\right]c^\dagger_{i\mu\sigma}(t)c_{j\nu\sigma}(t)\ .\end{aligned} \tag{5.218}$$

where

$$\begin{aligned}\mathbf{w}^{\mu\nu}_{ij}(\mathbf{q}) =& \int d\mathbf{r}e^{i\mathbf{q}\cdot\mathbf{r}}\mathbf{w}^{\mu\nu}_{ij}(\mathbf{r}) \\ =& \int d\mathbf{r}e^{i\mathbf{q}\cdot\mathbf{r}}\left\{\phi^*_\mu(\mathbf{r}-\mathbf{R}_i)\left(\frac{\hbar}{i}\nabla\right)\phi_\nu(\mathbf{r}-\mathbf{R}_j) - \left[\frac{\hbar}{i}\nabla\phi^*_\mu(\mathbf{r}-\mathbf{R}_i)\right]\phi_\nu(\mathbf{r}-\mathbf{R}_j)\right\}\ .\end{aligned} \tag{5.219}$$

Note that

$$\begin{aligned}\mathbf{w}^{\mu\nu}_{ij}(0) =& \int d\mathbf{r}\left\{\phi^*_\mu(\mathbf{r}-\mathbf{R}_i)\left(\frac{\hbar}{i}\nabla\right)\phi_\nu(\mathbf{r}-\mathbf{R}_j) - \left[\frac{\hbar}{i}\nabla\phi^*_\mu(\mathbf{r}-\mathbf{R}_i)\right]\phi_\nu(\mathbf{r}-\mathbf{R}_j)\right\} \\ =& \int d\mathbf{r}\left\{\phi^*_\mu(\mathbf{r}-\mathbf{R}_i)\left(\frac{\hbar}{i}\nabla\right)\phi_\nu(\mathbf{r}-\mathbf{R}_j) + \phi^*_\mu(\mathbf{r}-\mathbf{R}_i)\left(\frac{\hbar}{i}\nabla\right)\phi_\nu(\mathbf{r}-\mathbf{R}_j)\right\} \\ =& 2\langle i\mu|\hat{\mathbf{p}}|j\nu\rangle\ .\end{aligned} \tag{5.220}$$

We have integrated by parts from the first to the second line, assuming that the wave functions are zero on the infinite surface.

In the case of $\mathbf{q} = 0$, we can also write

$$\begin{aligned}\hat{H}_{\text{int}}(t) =& -\frac{eE_0}{2im\omega}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}}\mathbf{p}^{\mu\nu}_{ij}\cdot\hat{\mathbf{u}}_E\left[e^{-i\omega t} - e^{i\omega t}\right]c^\dagger_{i\mu\sigma}(t)c_{j\nu\sigma}(t) \\ =& \frac{eE_0}{m\omega}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}}\mathbf{p}^{\mu\nu}_{ij}\cdot\hat{\mathbf{u}}_E\sin(\omega t)c^\dagger_{i\mu\sigma}(t)c_{j\nu\sigma}(t)\ .\end{aligned} \tag{5.221}$$

For a multilayer system, $i \to \{\ell, \mathbf{R}_\parallel\}$ and $j \to \{\ell', \mathbf{R}'_\parallel\}$. So,

$$\begin{aligned}
\hat{H}_{\rm int}(t) =& \frac{eE_0}{m\omega} \sum_\sigma \sum_{\substack{\ell\ell' \\ \mu\nu}} \sum_{\mathbf{R}_\parallel, \mathbf{R}'_\parallel} \mathbf{p}^{\mu\nu}_{\ell\ell'}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) \cdot \hat{\mathbf{u}}_E \sin(\omega t) c^\dagger_{\ell\mu\sigma}(\mathbf{R}_\parallel, t) c_{\ell'\nu\sigma}(\mathbf{R}'_\parallel, t) \\
=& \frac{eE_0}{m\omega} \sum_\sigma \sum_{\substack{\ell\ell' \\ \mu\nu}} \sum_{\mathbf{R}_\parallel, \mathbf{R}'_\parallel} \frac{1}{N^2} \sum_{\mathbf{k}_\parallel, \mathbf{k}'_\parallel} \mathbf{p}^{\mu\nu}_{\ell\ell'}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) \cdot \hat{\mathbf{u}}_E \sin(\omega t) e^{-i\mathbf{k}_\parallel \cdot \mathbf{R}_\parallel} c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel, t) e^{i\mathbf{k}'_\parallel \cdot \mathbf{R}'_\parallel} c_{\ell'\nu\sigma}(\mathbf{k}'_\parallel, t) \\
=& \frac{eE_0}{m\omega} \sum_\sigma \sum_{\substack{\ell\ell' \\ \mu\nu}} \sum_{\mathbf{R}_\parallel, \mathbf{R}'_\parallel} \frac{1}{N^2} \sum_{\mathbf{k}_\parallel, \mathbf{k}'_\parallel} e^{i\mathbf{k}'_\parallel \cdot (\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \mathbf{p}^{\mu\nu}_{\ell\ell'}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) \cdot \hat{\mathbf{u}}_E \sin(\omega t) e^{i(\mathbf{k}'_\parallel - \mathbf{k}_\parallel)\cdot \mathbf{R}_\parallel} c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel, t) c_{\ell'\nu\sigma}(\mathbf{k}'_\parallel, t) \\
=& \frac{eE_0}{m\omega} \sum_\sigma \sum_{\substack{\ell\ell' \\ \mu\nu}} \frac{1}{N} \sum_{\mathbf{k}_\parallel, \mathbf{k}'_\parallel} \mathbf{p}^{\mu\nu}_{\ell\ell'}(\mathbf{k}'_\parallel) \cdot \hat{\mathbf{u}}_E \sin(\omega t) \delta_{\mathbf{k}'_\parallel, \mathbf{k}_\parallel} c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel, t) c_{\ell'\nu\sigma}(\mathbf{k}'_\parallel, t) \\
=& \frac{eE_0}{m\omega} \frac{1}{N} \sum_{\mathbf{k}_\parallel} \sum_\sigma \sum_{\substack{\ell\ell' \\ \mu\nu}} \mathbf{p}^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \sin(\omega t) c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel, t) c_{\ell'\nu\sigma}(\mathbf{k}_\parallel, t) \; .
\end{aligned} \tag{5.222}$$

Using the relation

$$\frac{\hbar}{m} \hat{p}^\alpha = \sum_{\substack{\ell\ell' \\ \mu\nu}} \sum_{\mathbf{k}_\parallel \sigma} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{e}}^\alpha c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel) c_{\ell'\nu\sigma}(\mathbf{k}_\parallel) \; . \tag{5.223}$$

obtained in Appendix A, we end up with

$$\hat{H}_{\rm int}(t) = \frac{eE_0}{\hbar\omega} \frac{1}{N} \sum_{\mathbf{k}_\parallel, \sigma} \sum_{\substack{\ell\ell' \\ \mu\nu}} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \;\; \sin(\omega t) c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel, t) c_{\ell'\nu\sigma}(\mathbf{k}_\parallel, t) \; . \tag{5.224}$$

If instead we use an unphysical complex field (as a tool to obtain the response of the system),

$$\mathbf{E}(\mathbf{r}, t) = E_0 e^{i(\mathbf{q}\cdot\mathbf{r} - \omega t)} \hat{\mathbf{u}}_E \; , \tag{5.225}$$

the vector potential is given by

$$\begin{aligned}
\mathbf{A}(\mathbf{r}, t) =& -\int^t \mathbf{E}(\mathbf{r}, t') dt' \\
=& -E_0 \hat{\mathbf{u}}_E \int^t e^{i(\mathbf{q}\cdot\mathbf{r} - \omega t')} dt' \\
=& \frac{1}{i\omega} E_0 e^{i(\mathbf{q}\cdot\mathbf{r} - \omega t)} \hat{\mathbf{u}}_E \\
=& \frac{\mathbf{E}(\mathbf{r}, t)}{i\omega} \; .
\end{aligned} \tag{5.226}$$

The perturbation hamiltonian is then given by

$$
\begin{aligned}
\hat{H}_{\text{int}}(t) &= -\frac{E_0}{i\omega}\int d\mathbf{r}\hat{\mathbf{J}}^{\mathrm{C}}(\mathbf{r},t)\cdot\hat{\mathbf{u}}_E e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} \\
&= -\frac{eE_0}{2im\omega}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}}\int d\mathbf{r}\ \mathbf{w}^{\mu\nu}_{ij}(\mathbf{r})\cdot\hat{\mathbf{u}}_E e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)}c^{\dagger}_{i\mu\sigma}(t)c_{j\nu\sigma}(t) \\
&= -\frac{eE_0}{2im\omega}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}}\mathbf{w}^{\mu\nu}_{ij}(\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{-i\omega t}c^{\dagger}_{i\mu\sigma}(t)c_{j\nu\sigma}(t)\ .
\end{aligned}
\tag{5.227}
$$

For $\mathbf{q}=0$,

$$
\begin{aligned}
\hat{H}_{\text{int}}(t) &= -\frac{eE_0}{im\omega}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}}\mathbf{p}^{\mu\nu}_{ij}\cdot\hat{\mathbf{u}}_E e^{-i\omega t}c^{\dagger}_{i\mu\sigma}(t)c_{j\nu\sigma}(t) \\
&= -\frac{e}{im\omega}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}}\mathbf{p}^{\mu\nu}_{ij}\cdot\mathbf{E}(t)c^{\dagger}_{i\mu\sigma}(t)c_{j\nu\sigma}(t)\ .
\end{aligned}
\tag{5.228}
$$

For multilayers, we can follow the same calculations done in Eq. (5.222) to obtain

$$
\begin{aligned}
\hat{H}_{\text{int}}(t) &= -\frac{eE_0}{i\hbar\omega}\frac{1}{N}\sum_{\mathbf{k}_{\parallel},\sigma}\sum_{\substack{\ell\ell'\\ \mu\nu}}\boldsymbol{\nabla}_{\mathbf{k}_{\parallel}}t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_{\parallel})\cdot\hat{\mathbf{u}}_E\ e^{-i\omega t}c^{\dagger}_{\ell\mu\sigma}(\mathbf{k}_{\parallel},t)c_{\ell'\nu\sigma}(\mathbf{k}_{\parallel},t) \\
&= -\frac{e}{i\hbar\omega}\frac{1}{N}\sum_{\mathbf{k}_{\parallel},\sigma}\sum_{\substack{\ell\ell'\\ \mu\nu}}\boldsymbol{\nabla}_{\mathbf{k}_{\parallel}}t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_{\parallel})\cdot\mathbf{E}(t)c^{\dagger}_{\ell\mu\sigma}(\mathbf{k}_{\parallel},t)c_{\ell'\nu\sigma}(\mathbf{k}_{\parallel},t)\ .
\end{aligned}
\tag{5.229}
$$

We can also keep $\mathbf{A}(\mathbf{r},t)$ directly in Eq.(5.211), and substitute J_C via Eq. (5.34) to obtain

$$
\hat{H}_{\text{int}}(t) = -\frac{e}{2m}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}}\int d\mathbf{r}\mathbf{w}^{\mu\nu}_{ij}(\mathbf{r})\cdot\mathbf{A}(\mathbf{r},t)c^{\dagger}_{i\mu\sigma}(t)c_{j\nu\sigma}(t)\ .
\tag{5.230}
$$

When the vector potential is uniform, i.e. $\mathbf{A}(\mathbf{r},t)=\mathbf{A}(t)$

$$
\begin{aligned}
\hat{H}_{\text{int}}(t) &= -\frac{e}{2m}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}}\int d\mathbf{r}\mathbf{w}^{\mu\nu}_{ij}(\mathbf{r})\cdot\mathbf{A}(t)c^{\dagger}_{i\mu\sigma}(t)c_{j\nu\sigma}(t) \\
&= -\frac{e}{2m}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}}\mathbf{w}^{\mu\nu}_{ij}(\mathbf{q}=0)\cdot\mathbf{A}(t)c^{\dagger}_{i\mu\sigma}(t)c_{j\nu\sigma}(t) \\
&= -\frac{e}{m}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}}\mathbf{p}^{\mu\nu}_{ij}\cdot\mathbf{A}(t)c^{\dagger}_{i\mu\sigma}(t)c_{j\nu\sigma}(t)\ .
\end{aligned}
\tag{5.231}
$$

For multilayers, we follow again the steps in Eq. (5.222) and Eq. (5.224) to obtain

$$\begin{aligned}
\hat{H}_{\text{int}}(t) &= -\frac{e}{m}\sum_{\sigma}\sum_{\substack{ij\\ \mu\nu}} \mathbf{p}_{ij}^{\mu\nu}\cdot\mathbf{A}(t)c_{i\mu\sigma}^{\dagger}(t)c_{j\nu\sigma}(t) \\
&= -\frac{e}{m}\frac{1}{N}\sum_{\mathbf{k}_{\parallel},\sigma}\sum_{\substack{\ell\ell'\\ \mu\nu}} \mathbf{p}_{\ell\ell'}^{\mu\nu}(\mathbf{k}_{\parallel})\cdot\mathbf{A}(t)c_{\ell\mu\sigma}^{\dagger}(\mathbf{k}_{\parallel},t)c_{\ell'\nu\sigma}(\mathbf{k}_{\parallel},t) \\
&= -\frac{e}{\hbar}\frac{1}{N}\sum_{\mathbf{k}_{\parallel},\sigma}\sum_{\substack{\ell\ell'\\ \mu\nu}} \boldsymbol{\nabla}_{\mathbf{k}_{\parallel}} t_{\ell\ell'}^{\mu\nu}(\mathbf{k}_{\parallel})\cdot\mathbf{A}(t)c_{\ell\mu\sigma}^{\dagger}(\mathbf{k}_{\parallel},t)c_{\ell'\nu\sigma}(\mathbf{k}_{\parallel},t)\ .
\end{aligned} \tag{5.232}$$

5.4.2 Second order term

The second order term gives rise to a diamagnetic current that is important for small frequencies. The hamiltonian is given by

$$\hat{H}_{(2)} = e^2\sum_{\ell}\frac{\mathbf{A}^2(\mathbf{r}_\ell,t)}{2m}\ . \tag{5.233}$$

Analogously as we have done before, we can write

$$\begin{aligned}
\hat{H}_{(2)} =& e^2\int d\mathbf{r}\sum_{\ell}\delta(\mathbf{r}-\mathbf{r}_\ell)\frac{\mathbf{A}^2(\mathbf{r},t)}{2m} \\
=& e\int d\mathbf{r}\hat{\rho}(\mathbf{r})\frac{\mathbf{A}^2(\mathbf{r},t)}{2m}\ ,
\end{aligned} \tag{5.234}$$

where $\hat{\rho}$ is the charge density operator. In the tight-binding basis, we can write

$$\hat{H}_{(2)} = e^2\sum_{\mu\sigma} c_{i\mu\sigma}^{\dagger}c_{i\mu\sigma}\frac{\mathbf{A}_i^2(t)}{2m}\ . \tag{5.235}$$

We can also write this term as

$$\hat{H}_{(2)}(t) = -\int d\mathbf{r}\hat{\mathbf{J}}^{\text{dia}}(\mathbf{r},t)\cdot\mathbf{A}(\mathbf{r},t)\ , \tag{5.236}$$

if we define

$$\hat{\mathbf{J}}^{\text{dia}}(\mathbf{r},t) = -\frac{e\hat{\rho}(\mathbf{r})}{2m}\mathbf{A}(\mathbf{r},t)\ . \tag{5.237}$$

$$\hat{\mathbf{J}}^{\text{dia}}(\mathbf{r},t) = -\frac{1}{2i\omega}\frac{e\hat{\rho}(\mathbf{r})}{2m}\mathbf{E}_0\left[e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} - e^{-i(\mathbf{q}\cdot\mathbf{r}-\omega t)}\right]\ . \tag{5.238}$$

Using Eq. (5.215),

$$\hat{H}_{(2)}(t) = -\frac{1}{2i\omega}\int d\mathbf{r}\hat{\mathbf{J}}^{\text{dia}}(\mathbf{r},t)\cdot\mathbf{E}_0\left[e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} - e^{-i(\mathbf{q}\cdot\mathbf{r}-\omega t)}\right]\ . \tag{5.239}$$

5.4.3 Charge Current

Now are able to obtain the charge current that flows from site i to j due to the hamiltonian 5.218.

In multilayers, the charge current operator from site $\mathbf{R}'_\parallel$ in layer ℓ_2 to site $\mathbf{R}_\parallel$ in layer ℓ_1 is given by Eqs. 5.65 and 5.66, which we repeat here for completeness:

$$\hat{I}^{\mathrm{C}}_{\ell_1\ell_2}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel) = \frac{1}{N_\parallel}\sum_{\mathbf{q}_\parallel}\left\{\hat{\mathcal{I}}^{\mathrm{C}}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{q}_\parallel)e^{i\mathbf{q}_\parallel\cdot\mathbf{R}'_\parallel} - \hat{\mathcal{I}}^{\mathrm{C}}_{\ell_2\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel, \mathbf{q}_\parallel)e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}\right\} \ , \tag{5.240}$$

where

$$\hat{\mathcal{I}}^{\mathrm{C}}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{q}_\parallel) = \frac{ie}{\hbar}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\sigma}\sum_{\mu\nu} t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)c^\dagger_{\ell_1\mu\sigma}(\mathbf{k}_\parallel)c_{\ell_2\nu\sigma}(\mathbf{k}_\parallel + \mathbf{q}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \ . \tag{5.241}$$

Its expectation value in linear response is

$$\langle\hat{I}^{\mathrm{C}}_{\ell_1\ell_2}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel; t)\rangle = \frac{1}{N_\parallel}\sum_{\mathbf{q}_\parallel}\left\{\langle\hat{\mathcal{I}}^{\mathrm{C}}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{q}_\parallel; t)\rangle e^{i\mathbf{q}_\parallel\cdot\mathbf{R}'_\parallel} - \langle\hat{\mathcal{I}}^{\mathrm{C}}_{\ell_2\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel, \mathbf{q}_\parallel; t)\rangle e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}\right\} \ , \tag{5.242}$$

where

$$\begin{aligned}
\langle\hat{\mathcal{I}}^{\mathrm{C}}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{q}_\parallel; t)\rangle &= -\frac{i}{\hbar}\int dt'\Theta(t-t')\left\langle\left[\hat{\mathcal{I}}^{\mathrm{C}}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{q}_\parallel; t), \hat{H}_{\mathrm{int}}(t')\right]\right\rangle \\
=& -\frac{i}{\hbar}\frac{eE_0}{\hbar\omega}\frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma'}}\sum_{\substack{\ell\ell'\\ \gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ \sin(\omega t')\left\langle\left[\hat{\mathcal{I}}^{m}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{q}_\parallel; t), c^\dagger_{\ell\gamma\sigma'}(\mathbf{k}_\parallel, t')c_{\ell'\xi\sigma'}(\mathbf{k}_\parallel, t'\right.\right. \\
=& \frac{1}{\hbar}\frac{e^2E_0}{\hbar^2\omega}\frac{1}{N^2_\parallel}\sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ \sin(\omega t')t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)e^{i\mathbf{k}'_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \\
&\times\left\langle\left[c^\dagger_{\ell_1\mu\sigma}(\mathbf{k}'_\parallel, t)c_{\ell_2\nu\sigma}(\mathbf{k}'_\parallel + \mathbf{q}'_\parallel, t), c^\dagger_{\ell\gamma\sigma'}(\mathbf{k}_\parallel, t')c_{\ell'\xi\sigma'}(\mathbf{k}_\parallel, t')\right]\right\rangle \ .
\end{aligned} \tag{5.243}$$

Identifying once again the susceptibility

$$\begin{aligned}
\chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}'_\parallel, -\mathbf{k}'_\parallel - \mathbf{q}_\parallel, \mathbf{k}_\parallel, -\mathbf{k}_\parallel, t-t') =& -\frac{i}{\hbar}\Theta(t-t')\left\langle\left[c^\dagger_{\ell_1\mu\sigma}(\mathbf{k}'_\parallel, t)c_{\ell_2\nu\sigma}(\mathbf{k}'_\parallel + \mathbf{q}'_\parallel, t), c^\dagger_{\ell\gamma\sigma'}(\mathbf{k}_\parallel, t')c_{\ell'\xi\sigma'}(\mathbf{k}_\parallel, t')\right.\right. \\
=& N^2_\parallel\chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}'_\parallel, \mathbf{k}_\parallel, t-t')\delta_{\mathbf{k}'_\parallel + \mathbf{q}_\parallel, \mathbf{k}_\parallel}\delta_{\mathbf{k}'_\parallel, \mathbf{k}_\parallel} \ ,
\end{aligned} \tag{5.244}$$

we get

$$
\begin{aligned}
\langle \hat{\mathcal{I}}^{\mathrm{C}}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{q}_\parallel)\rangle =& \frac{e^2 E_0}{2\hbar^2\omega} \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \int dt' \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \ \left[e^{i\omega t'} - e^{-i\omega t'} \right] t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \\
& \times \chi^{\sigma\sigma' \ \mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, t - t') \delta_{\mathbf{q}_\parallel, 0} \\
=& - \frac{e^2 E_0}{2\hbar^2\omega} \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \ t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \\
& \times \left[e^{-i\omega t} \chi^{\sigma\sigma' \ \mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega) - e^{i\omega t} \chi^{\sigma\sigma' \ \mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, -\omega) \right] \delta_{\mathbf{q}_\parallel, 0} \ .
\end{aligned}
\tag{5.245}
$$

where we have used the Fourier transform given by Eq. (5.113). Taking two sites in the same plane, i.e., $\ell_2 = \ell_1$, the parallel current becomes

$$
\langle \hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel)\rangle = \frac{1}{N_\parallel} \left\{ \langle \hat{\mathcal{I}}^{\mathrm{C}}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)\rangle - \langle \hat{\mathcal{I}}^{\mathrm{C}}_{\ell_1\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)\rangle \right\} \ , \tag{5.246}
$$

where we have already taken into account the $\delta_{\mathbf{q}_\parallel,0}$. Here,

$$
\begin{aligned}
\langle \hat{\mathcal{I}}^{\mathrm{C}}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)\rangle =& - \frac{e^2 E_0}{2\hbar^2\omega} \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \ t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \\
& \times \left[e^{-i\omega t} \chi^{\sigma\sigma' \ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega) - e^{i\omega t} \chi^{\sigma\sigma' \ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, -\omega) \right] \\
=& - \frac{e^2 E_0}{2\hbar^2\omega} \left[e^{-i\omega t} \mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega) - e^{i\omega t} \mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, -\omega) \right] \ .
\end{aligned}
\tag{5.247}
$$

where we have defined

$$
\mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega) = \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \ t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \chi^{\sigma\sigma' \ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega) \ .
\tag{5.248}
$$

Now we can see that

$$
\begin{aligned}
[\mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega)]^* =& \sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}} \sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}} \left[\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot \hat{\mathbf{u}}_E\ t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega) \right]^* \\
=& \sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}} \sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\xi\gamma}_{\ell'\ell}(\mathbf{k}_\parallel)\cdot \hat{\mathbf{u}}_E\ t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \chi^{\sigma\sigma'\ \nu\mu\xi\gamma}_{\ell_1\ell_1\ell'\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega) \\
=& \sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}} \sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot \hat{\mathbf{u}}_E\ t^{\nu\mu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega) \\
=& \sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}} \sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot \hat{\mathbf{u}}_E\ t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} \chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega) \\
=& \mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel,-\omega)\ .
\end{aligned}
\tag{5.249}
$$

Using the results above, we can write Eq. (5.246) as

$$
\begin{aligned}
\langle \hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)\rangle =& -\frac{e^2E_0}{2\hbar^2\omega}\frac{1}{N_\parallel}\Big\{ e^{-i\omega t}\left[\mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega) - \mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel,\omega)\right] \\
&\qquad - e^{i\omega t}\left[\mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,-\omega) - \mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel,-\omega)\right]\Big\} \\
=& -\frac{e^2E_0}{2\hbar^2\omega}\frac{1}{N_\parallel}\Big\{ e^{-i\omega t}\left[\mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega) - \mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel,\omega)\right] \\
&\qquad + e^{i\omega t}\left[\mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega) - \mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel,\omega)\right]^*\Big\} \\
=& -\frac{e^2E_0}{\hbar}\frac{1}{N_\parallel}\,\mathrm{Re}\left\{\frac{e^{-i\omega t}}{\hbar\omega}\left[\mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega) - \mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel,\omega)\right]\right\} \\
=& -\frac{e^2E_0}{\hbar}\,\mathrm{Re}\Big\{\frac{e^{-i\omega t}}{\hbar\omega}\frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}} \sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot \hat{\mathbf{u}}_E\ \chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega) \\
&\qquad \times\left[t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} - t^{\nu\mu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\right]\Big\}\ .
\end{aligned}
\tag{5.250}
$$

Finally, this may be expressed as

$$
\begin{aligned}
\langle \hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)\rangle =& -\frac{e^2E_0}{\hbar^2\omega}\,\mathrm{Re}\left\{e^{-i\omega t}\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega)\right\} \\
=& -\frac{e^2E_0}{\hbar^2\omega}|\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega)|\cos[\omega t - \phi^{\mathrm{C}}(\omega)]\ .
\end{aligned}
\tag{5.251}
$$

where the absolute value of $\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega)$ and its phase $\phi^{\mathrm{C}}(\omega)$ are given by the complex number

$$
\begin{aligned}
\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega) =& \frac{1}{N_\parallel} \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \Big[t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} - t^{\nu\mu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \Big] \\
& \times \chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega) \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \ .
\end{aligned}
\tag{5.252}
$$

Note that the dc limit is obtained by

$$
\langle \hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel) \rangle = -\frac{e^2}{\hbar} \lim_{\omega\to 0} \mathrm{Re} \left\{ \frac{1}{\hbar\omega} \mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega) \right\} E_0 \ . \tag{5.253}
$$

This is different than what is usually done (that is, to take the imaginary part) because we have used the imaginary unit to change to the real part. This way, the phase ϕ^{C} can be directly related to the external electric field given by Eq. (5.212).

We can also obtain the response function using the complex external field field given in Eq. (5.225) through by the perturbation hamiltonian obtained in Eq. (5.229). In this case,

$$
\begin{aligned}
\langle \hat{\mathcal{I}}^{\mathrm{C}}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{q}_\parallel) \rangle =& \frac{i}{\hbar} \frac{e^2 E_0}{\hbar^2\omega} \frac{1}{N^2_\parallel} \sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \int dt' \Theta(t-t') \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \ e^{-i\omega t'} t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}'_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \\
& \times \Big\langle \Big[c^\dagger_{\ell_1\mu\sigma}(\mathbf{k}'_\parallel, t) c_{\ell_2\nu\sigma}(\mathbf{k}'_\parallel + \mathbf{q}'_\parallel, t), c^\dagger_{\ell\gamma\sigma'}(\mathbf{k}_\parallel, t') c_{\ell'\xi\sigma'}(\mathbf{k}_\parallel, t') \Big] \Big\rangle \\
=& -\frac{e^2 E_0}{\hbar^2\omega} \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \int dt' \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \ e^{-i\omega t'} t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \\
& \times \chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, t-t') \delta_{\mathbf{q}_\parallel,0} \\
=& -\frac{e^2 E_0}{\hbar^2\omega} \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \ t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} e^{-i\omega t} \chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega) \delta \\
=& -\frac{e^2}{\hbar^2\omega} \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \mathbf{E}(t) \ t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega) \delta_{\mathbf{q}_\parallel,0}
\end{aligned}
\tag{5.254}
$$

The total current will then be

$$
\begin{aligned}
\langle \hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel)\rangle &= -\frac{e^2 E_0}{\hbar^2\omega}\frac{1}{N_\parallel} e^{-i\omega t}\left[\mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega) - \mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel, \omega)\right] \\
&= -\frac{e^2 E_0}{\hbar}\frac{1}{N_\parallel}\frac{e^{-i\omega t}}{\hbar\omega}\left[\mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega) - \mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel, \omega)\right] \\
&= -\frac{e^2 E_0}{\hbar}\frac{e^{-i\omega t}}{\hbar\omega}\frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}}\sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ \chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega) \\
&\quad\times\left[t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} - t^{\nu\mu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\right] \\
&= -\frac{e^2}{\hbar}\frac{1}{\hbar\omega}\frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}}\sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}}\left[t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} - t^{\nu\mu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\right] \\
&\quad\times\chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\mathbf{E}(t)\ .
\end{aligned}
\tag{5.255}
$$

where the complex number $\mathcal{C}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega)$ is given by Eq. (5.248). To obtain the real currents (given by Eq. (5.251)) from this result, we just need to take the real part.

5.4.4 Diamagnetic current

The diamagnetic current comes from the change in the current operator due to the external potential vector. The linear response term is given by the expectation value of this operator with the unperturbed stated. (The Kubo formalism for the response of this operator gives a second order term.) A similar calculation was done in appendix H. Using the result of Eq. (H.12) but taking into account the diamagnetic current operator obtained in Eq. (5.41), we can write the in-plane diamagnetic current as

$$
\begin{aligned}
\langle \hat{I}^{\mathrm{dia}}_{\ell\ell}(\mathbf{R}_i, \mathbf{R}_j)\rangle = -\frac{e}{2\pi}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F} d\omega'\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left\{G^{\sigma\sigma\ \nu\mu}_{\ell\ell}(\mathbf{k}_\parallel;\omega') - \left[G^{\sigma\sigma\ \mu\nu}_{\ell\ell}(\mathbf{k}_\parallel;\omega')\right]^*\right\} \\
\times\left\{D^{\mu\nu}_{\ell\ell}(\mathbf{R}_j - \mathbf{R}_i;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i - \mathbf{R}_j)} - D^{\mu\nu}_{\ell\ell}(\mathbf{R}_i - \mathbf{R}_j;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j - \mathbf{R}_i)}\right\}\ .
\end{aligned}
\tag{5.256}
$$

Here, we have explicitly written the dependence of D on the external frequency. For the in-plane case, Eq. (5.72) becomes

$$
D^{\mu\nu}_{\ell\ell}(\mathbf{R}_j - \mathbf{R}_i;\omega) = \frac{eE_0}{\hbar\omega}\sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j - \mathbf{R}_i)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\mu\nu}_{\ell\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t)\ . \tag{5.257}
$$

Therefore,

$$
\begin{aligned}
\langle \hat{I}^{\text{dia}}_{\ell\ell}(\mathbf{R}_i,\mathbf{R}_j)\rangle =& -\frac{e}{2\pi}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F} d\omega' \frac{1}{N}\sum_{\mathbf{k}_\parallel} \\
&\Big\{ G^{\sigma\sigma}_{\ell\ell}{}^{\nu\mu}(\mathbf{k}_\parallel;\omega')\left[D^{\mu\nu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)} - D^{\mu\nu}_{\ell\ell}(\mathbf{R}_i-\mathbf{R}_j;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\right] \\
&-\left[G^{\sigma\sigma}_{\ell\ell}{}^{\mu\nu}(\mathbf{k}_\parallel;\omega')\right]^*\left[D^{\mu\nu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)} - D^{\mu\nu}_{\ell\ell}(\mathbf{R}_i-\mathbf{R}_j;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\right]\Big\} \\
=& -\frac{e}{2\pi}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F} d\omega' \frac{1}{N}\sum_{\mathbf{k}_\parallel} \\
&\Big\{ G^{\sigma\sigma}_{\ell\ell}{}^{\nu\mu}(\mathbf{k}_\parallel;\omega')\left[D^{\mu\nu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)} - D^{\mu\nu}_{\ell\ell}(\mathbf{R}_i-\mathbf{R}_j;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\right] \\
&+\left[G^{\sigma\sigma}_{\ell\ell}{}^{\mu\nu}(\mathbf{k}_\parallel;\omega')\left[D^{\mu\nu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)} - D^{\mu\nu}_{\ell\ell}(\mathbf{R}_i-\mathbf{R}_j;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)}\right]\right]^*\Big\}
\end{aligned} \tag{5.258}
$$

where we have used the fact that $D^{\mu\nu}_{\ell\ell}(\mathbf{R}_i-\mathbf{R}_j;\omega)$ is purely imaginary, obtained in Eq. (5.73). Using now Eq. (5.75) to change its position indices adding another minus sign and changing orbital indices on the last line, we get

$$
\begin{aligned}
\langle \hat{I}^{\text{dia}}_{\ell\ell}(\mathbf{R}_i,\mathbf{R}_j)\rangle =& -\frac{e}{2\pi}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F} d\omega' \frac{1}{N}\sum_{\mathbf{k}_\parallel} \\
&\Big\{ G^{\sigma\sigma}_{\ell\ell}{}^{\nu\mu}(\mathbf{k}_\parallel;\omega')\left[D^{\mu\nu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)} - D^{\mu\nu}_{\ell\ell}(\mathbf{R}_i-\mathbf{R}_j;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\right] \\
&+\left[G^{\sigma\sigma}_{\ell\ell}{}^{\mu\nu}(\mathbf{k}_\parallel;\omega')\left[D^{\nu\mu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)} - D^{\nu\mu}_{\ell\ell}(\mathbf{R}_i-\mathbf{R}_j;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\right]\right]^*\Big\}
\end{aligned} \tag{5.259}
$$

This equation can be rewritten as

$$
\begin{aligned}
\langle \hat{I}^{\text{dia}}_{\ell\ell}(\mathbf{R}_i,\mathbf{R}_j)\rangle =& -\frac{e}{\pi}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F} d\omega' \frac{1}{N}\sum_{\mathbf{k}_\parallel} \\
&\mathrm{Re}\Big\{ G^{\sigma\sigma}_{\ell\ell}{}^{\nu\mu}(\mathbf{k}_\parallel;\omega')\left[D^{\mu\nu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)} - D^{\mu\nu}_{\ell\ell}(\mathbf{R}_i-\mathbf{R}_j;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\right]\Big\}
\end{aligned} \tag{5.260}
$$

Using again Eq. (5.75) to change the position indices of the second term, we obtain, changing $\mu \rightleftharpoons \nu$,

$$
\begin{aligned}
\langle \hat{I}^{\text{dia}}_{\ell\ell}(\mathbf{R}_i,\mathbf{R}_j)\rangle =& -\frac{e}{\pi}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F} d\omega' \frac{1}{N}\sum_{\mathbf{k}_\parallel} \\
&\mathrm{Re}\Big\{ G^{\sigma\sigma}_{\ell\ell}{}^{\mu\nu}(\mathbf{k}_\parallel;\omega')\left[D^{\nu\mu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i;\omega)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)} + D^{\mu\nu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i;\omega)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\right]\Big\}
\end{aligned} \tag{5.261}
$$

Substituting Eq. (5.257),

$$\langle \hat{I}^{\mathrm{dia}}_{\ell\ell}(\mathbf{R}_i,\mathbf{R}_j)\rangle = -\frac{e^2E_0}{\pi m\omega}\sin(\omega t)\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F} d\omega' \frac{1}{N}\sum_{\mathbf{k}_\parallel} \mathrm{Re}\left\{ G^{\sigma\sigma}_{\ell\ell}{}^{\mu\nu}(\mathbf{k}_\parallel;\omega')\left[\mathbf{p}^{\nu\mu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i)\cdot\hat{\mathbf{u}}_E e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)} + \mathbf{p}^{\mu\nu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i)\cdot\hat{\mathbf{u}}_E e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\right]\right\} . \tag{5.262}$$

where $\mathbf{p}^{\mu\nu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i)\cdot\hat{\mathbf{u}}_E = \frac{m}{\hbar}\sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\mu\nu}_{\ell\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E$.

We can also identify the Fourier transform of the Green function $G^{\sigma\sigma}_{ij}{}^{\mu\nu}(\omega') = \frac{1}{N}\sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)} G^{\sigma\sigma}_{\ell\ell}{}^{\mu\nu}(\mathbf{k}_\parallel;\omega')$, where i and j are sites in plane ℓ, to obtain an equation in real space

$$\begin{aligned}\langle \hat{I}^{\mathrm{dia}}_{\ell\ell}(\mathbf{R}_i,\mathbf{R}_j)\rangle &= -\frac{e^2E_0}{\pi m\omega}\sin(\omega t)\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F} d\omega'\,\mathrm{Re}\left\{ G^{\sigma\sigma}_{ji}{}^{\mu\nu}(\omega')\mathbf{p}^{\nu\mu}_{ij} + G^{\sigma\sigma}_{ij}{}^{\mu\nu}(\omega')\mathbf{p}^{\mu\nu}_{ij}\cdot\hat{\mathbf{u}}_E\right\} \\ &= -\frac{e^2E_0}{\pi m\omega}\sin(\omega t)\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F} d\omega'\,\mathrm{Re}\left\{\left[G^{\sigma\sigma}_{ji}{}^{\nu\mu}(\omega') + G^{\sigma\sigma}_{ij}{}^{\mu\nu}(\omega')\right]\mathbf{p}^{\mu\nu}_{ij}\cdot\hat{\mathbf{u}}_E\right\} .\end{aligned} \tag{5.263}$$

Here we have changed the orbital indices on the first term and used the notation $\mathbf{p}^{\mu\nu}_{ij}\cdot\hat{\mathbf{u}}_E = \mathbf{p}^{\mu\nu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i)\cdot\hat{\mathbf{u}}_E$.

To compare this current with the one obtained in Eq. (5.251), we can write this equation as

$$\langle \hat{I}^{\mathrm{dia}}_{\ell\ell}(\mathbf{R}_i,\mathbf{R}_j)\rangle = -\frac{e^2E_0}{\hbar^2\omega}\mathrm{Re}\left\{e^{-i\omega t}\mathcal{J}^{\mathrm{dia}}_{\ell}(\mathbf{R}_j-\mathbf{R}_i)\right\} . \tag{5.264}$$

where

$$\begin{aligned}\mathcal{J}^{\mathrm{dia}}_{\ell}(\mathbf{R}_j-\mathbf{R}_i) &= \frac{i\hbar}{\pi m}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{E_{\mathrm{F}}} d\epsilon'\,\mathrm{Re}\left\{\left[G^{\sigma\sigma}_{ji}{}^{\nu\mu}(\epsilon') + G^{\sigma\sigma}_{ij}{}^{\mu\nu}(\epsilon')\right]\mathbf{p}^{\mu\nu}_{ij}\cdot\hat{\mathbf{u}}_E\right\} \\ &= \frac{i}{\pi}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{E_{\mathrm{F}}} d\epsilon'\sum_{\mathbf{k}_\parallel}\mathrm{Re}\left\{\left[G^{\sigma\sigma}_{ji}{}^{\nu\mu}(\epsilon') + G^{\sigma\sigma}_{ij}{}^{\mu\nu}(\epsilon')\right] e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\mu\nu}_{\ell\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\right\}\end{aligned} \tag{5.265}$$

Note that $\mathcal{J}^{\mathrm{dia}}_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)$ is independent of ω, so the frequency dependence of the diamagnetic current follows a simple $1/\omega$ behavior. Nevertheless, this contribution to the current may depend on the external static magnetic fields.

5.4.5 Spin Current

We can also calculate the linear response of the current of polarization m that flows from site i to j, $\langle \hat{I}^m_{ij} \rangle$, generated by the perturbation hamiltonian written in Eq. (5.218),

$$
\begin{aligned}
\langle \hat{I}^m_{ij} \rangle(t) =& -\frac{i}{\hbar} \int dt' \Theta(t-t') \left\langle \left[\hat{I}^m_{ij}(t), \hat{H}_{\text{int}}(t') \right] \right\rangle \\
=& \frac{eE_0}{4m\hbar\omega} \sum_\sigma \sum_{\substack{k\ell \\ \gamma\xi}} \int dt' \Theta(t-t') \left[\mathbf{w}^{\gamma\xi}_{k\ell}(\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{-i\omega t'} - \mathbf{w}^{\gamma\xi}_{k\ell}(-\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{i\omega t'} \right] \left\langle \left[\hat{I}^m_{ij}(t), c^\dagger_{k\gamma\sigma}(t') c_{\ell\xi\sigma}(t') \right] \right\rangle \; .
\end{aligned}
\tag{5.266}
$$

Substituting Eq. (5.6)

$$
\begin{aligned}
\langle \hat{I}^m_{ij} \rangle(t) =& \frac{ieE_0}{4m\hbar\omega} \sum_\sigma \sum_{\substack{k\ell \\ \mu\nu\gamma\xi}} \int dt' \Theta(t-t') \left[\mathbf{w}^{\gamma\xi}_{k\ell}(\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{-i\omega t'} - \mathbf{w}^{\gamma\xi}_{k\ell}(-\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{i\omega t'} \right] \\
& \times \left\langle \left[\left\{ t^{\mu\nu}_{ij} S^{m\,\mu\nu}_{ij}(t) - t^{\nu\mu}_{ji} S^{m\,\nu\mu}_{ji}(t) \right\}, c^\dagger_{k\gamma\sigma}(t') c_{\ell\xi\sigma}(t') \right] \right\rangle \\
=& -\frac{eE_0}{4m\omega} \sum_\sigma \sum_{\substack{k\ell \\ \mu\nu\gamma\xi}} \int dt' \left[\mathbf{w}^{\gamma\xi}_{k\ell}(\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{-i\omega t'} - \mathbf{w}^{\gamma\xi}_{k\ell}(-\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{i\omega t'} \right] \\
& \times \left\{ t^{\mu\nu}_{ij} \chi^{m\sigma\,\mu\nu\gamma\xi}_{ijkl}(t-t') - t^{\nu\mu}_{ji} \chi^{m\sigma\,\nu\mu\gamma\xi}_{jikl}(t-t') \right\} \; .
\end{aligned}
\tag{5.267}
$$

And we finally end up with

$$
\langle \hat{I}^m_{ij} \rangle(t) = -\frac{eE_0}{4m\omega} \sum_\sigma \sum_{\substack{k\ell \\ \mu\nu\gamma\xi}} \left[\mathbf{w}^{\gamma\xi}_{k\ell}(\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{-i\omega t} - \mathbf{w}^{\gamma\xi}_{k\ell}(-\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{i\omega t} \right] \left\{ t^{\mu\nu}_{ij} \chi^{m\sigma\,\mu\nu\gamma\xi}_{ijkl}(\omega) - t^{\nu\mu}_{ji} \chi^{m\sigma\,\nu\mu\gamma\xi}_{jikl}(\omega) \right\} \; .
\tag{5.268}
$$

We have used

$$
\chi^{m\sigma\,\mu\nu\gamma\xi}_{ijkl}(t-t') = -\frac{i}{\hbar} \Theta(t-t') \left\langle \left[S^{m\,\mu\nu}_{ij}(t), c^\dagger_{k\gamma\sigma}(t') c_{\ell\xi\sigma}(t') \right] \right\rangle
\tag{5.269}
$$

and Eq. (5.113) for the Fourier transform.

For multilayers, the operator for the total perpendicular spin current flowing from layer ℓ_1 to ℓ_2 is given by

$$
\hat{I}^m_{\ell_1\ell_2} = \frac{i}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\mu\nu} \left[t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{k}_\parallel) \; S^{m\,\mu\nu}_{\ell_1\ell_2}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) - t^{\nu\mu}_{\ell_2\ell_1}(\mathbf{k}_\parallel) \; S^{m\,\nu\mu}_{\ell_2\ell_1}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) \right] \; ,
\tag{5.270}
$$

where

$$
S^{m\,\mu\nu}_{\ell_1\ell_2}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) = \frac{1}{2} \sum_{\alpha\beta} c^\dagger_{\ell_1\mu\alpha}(\mathbf{k}_\parallel) \sigma^m_{\alpha\beta} c_{\ell_2\nu\beta}(\mathbf{k}_\parallel) \; .
\tag{5.271}
$$

Its expectation value in linear response theory can be calculated using the Hamiltonian 5.224. Thus, for a uniform field,

$$\begin{aligned}\langle \hat{I}^m_{\ell_1\ell_2}(t)\rangle =& -\frac{i}{\hbar}\int dt'\Theta(t-t')\left\langle\left[\hat{I}^m_{\ell_1\ell_2}(t),\hat{H}_{\rm int}(t')\right]\right\rangle\\ =& -\frac{i}{\hbar}\frac{eE_0}{\hbar\omega}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel,\sigma}\sum_{\substack{\ell\ell'\\ \gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ \sin(\omega t')\left\langle\left[\hat{I}^m_{\ell_1\ell_2}(t),c^\dagger_{\ell\gamma\sigma}(\mathbf{k}_\parallel,t')c_{\ell'\xi\sigma}(\mathbf{k}_\parallel,t')\right]\right\rangle\ .\end{aligned} \tag{5.272}$$

Using Eq. (5.270),

$$\begin{aligned}\langle \hat{I}^m_{\ell_1\ell_2}(t)\rangle =&\frac{1}{\hbar}\frac{eE_0}{\hbar\omega}\frac{1}{N_\parallel^2}\sum_{\substack{\mathbf{k}_\parallel,\mathbf{k}'_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ \sin(\omega t')\\ &\times\left\langle\left[t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{k}'_\parallel)\ \overset{m}{S}{}^{\mu\nu}_{\ell_1\ell_2}(\mathbf{k}'_\parallel,\mathbf{k}'_\parallel,t)-t^{\nu\mu}_{\ell_2\ell_1}(\mathbf{k}'_\parallel)\ \overset{m}{S}{}^{\nu\mu}_{\ell_2\ell_1}(\mathbf{k}'_\parallel,\mathbf{k}'_\parallel,t),c^\dagger_{\ell\gamma\sigma}(\mathbf{k}_\parallel,t')c_{\ell'\xi\sigma}(\mathbf{k}_\parallel,t')\right]\right\rangle\\ =&\frac{eE_0}{2\hbar\omega}\frac{1}{N_\parallel^2}\sum_{\substack{\mathbf{k}_\parallel,\mathbf{k}'_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\int dt'\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ \left(e^{i\omega t'}-e^{-i\omega t'}\right)\\ &\times\left\{t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{k}'_\parallel)\ \overset{m\sigma}{\chi}{}^{\mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}'_\parallel,-\mathbf{k}'_\parallel,\mathbf{k}_\parallel,-\mathbf{k}_\parallel,t-t')-t^{\nu\mu}_{\ell_2\ell_1}(\mathbf{k}'_\parallel)\ \overset{m\sigma}{\chi}{}^{\nu\mu\gamma\xi}_{\ell_2\ell_1\ell\ell'}(\mathbf{k}'_\parallel,-\mathbf{k}'_\parallel,\mathbf{k}_\parallel,-\mathbf{k}_\parallel,t-t')\right\}\ ,\end{aligned} \tag{5.273}$$

where

$$\begin{aligned}\overset{m\sigma}{\chi}{}^{\mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}'_\parallel,-\mathbf{k}'_\parallel,\mathbf{k}_\parallel,-\mathbf{k}_\parallel,t-t') =& -\frac{i}{\hbar}\Theta(t-t')\left\langle\left[\overset{m}{S}{}^{\mu\nu}_{\ell_1\ell_2}(\mathbf{k}'_\parallel,\mathbf{k}'_\parallel),c^\dagger_{\ell\gamma\sigma}(\mathbf{k}_\parallel,t')c_{\ell'\xi\sigma}(\mathbf{k}_\parallel,t')\right]\right\rangle\\ =&N_\parallel^2\overset{m\sigma}{\chi}{}^{\mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}'_\parallel,\mathbf{k}_\parallel,t-t')\delta_{\mathbf{k}'_\parallel,\mathbf{k}_\parallel}\delta_{\mathbf{k}'_\parallel,\mathbf{k}_\parallel}\ .\end{aligned} \tag{5.274}$$

Finally, identifying $\overset{m\sigma}{\chi}{}^{\mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,t-t')=\overset{m\sigma}{\chi}{}^{\mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,t-t')$, we get

$$\langle \hat{I}^m_{\ell_1\ell_2}(t)\rangle =\frac{eE_0}{\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\left\{t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{k}_\parallel)\ e^{i\omega t}\overset{m\sigma}{\chi}{}^{\mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,-\omega)-t^{\nu\mu}_{\ell_2\ell_1}(\mathbf{k}_\parallel)\ e^{-i\omega t}\overset{m\sigma}{\chi}{}^{\nu\mu\gamma\xi}_{\ell_2\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\right\}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ . \tag{5.275}$$

where we have used the Fourier transform given by Eq. (5.113).

We can also calculate the spin current that flows parallel to the layers. The parallel spin current operator is given by Eq. (5.76), and the expectation value in linear response is

$$\langle \hat{I}^m_{\ell_1\ell_2}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)\rangle = \frac{1}{N_\parallel}\sum_{\mathbf{q}_\parallel}\left\{\langle\hat{\mathcal{I}}^m_{\ell_1\ell_2}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{q}_\parallel)\rangle e^{i\mathbf{q}_\parallel\cdot\mathbf{R}'_\parallel}-\langle\hat{\mathcal{I}}^m_{\ell_2\ell_1}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel,\mathbf{q}_\parallel)\rangle e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}\right\}\ , \tag{5.276}$$

where

$$
\begin{aligned}
&\langle \hat{\mathcal{I}}^{m}_{\ell_1\ell_2}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{q}_\parallel)\rangle = -\frac{i}{\hbar}\int dt'\Theta(t-t')\left\langle\left[\hat{\mathcal{I}}^{m}_{\ell_1\ell_2}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{q}_\parallel),\hat{H}_{\text{int}}(t')\right]\right\rangle\\
&=-\frac{i}{\hbar}\frac{eE_0}{\hbar\omega}\frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ \sin(\omega t')\left\langle\left[\hat{\mathcal{I}}^{m}_{\ell_1\ell_2}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{q}_\parallel),c^\dagger_{\ell\gamma\sigma}(\mathbf{k}_\parallel,t')c_{\ell'\xi\sigma}(\mathbf{k}_\parallel,t')\right]\right\rangle\\
&=\frac{1}{\hbar}\frac{eE_0}{\hbar\omega}\frac{1}{N_\parallel^2}\sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ \sin(\omega t')t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}'_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\\
&\quad\times\left\langle\left[S^{m\ \mu\nu}_{\ell_1\ell_2}(\mathbf{k}'_\parallel,\mathbf{k}'_\parallel+\mathbf{q}_\parallel,t),c^\dagger_{\ell\gamma\sigma}(\mathbf{k}_\parallel,t')c_{\ell'\xi\sigma}(\mathbf{k}_\parallel,t')\right]\right\rangle\ .
\end{aligned}
\tag{5.277}
$$

Identifying once again the susceptibility

$$
\begin{aligned}
\chi^{m\sigma\ \mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}'_\parallel,-\mathbf{k}'_\parallel-\mathbf{q}_\parallel,\mathbf{k}_\parallel,-\mathbf{k}_\parallel,t-t') &= -\frac{i}{\hbar}\Theta(t-t')\left\langle\left[S^{m\ \mu\nu}_{\ell_1\ell_2}(\mathbf{k}'_\parallel,\mathbf{k}'_\parallel+\mathbf{q}_\parallel,t),c^\dagger_{\ell\gamma\sigma}(\mathbf{k}_\parallel,t')c_{\ell'\xi\sigma}(\mathbf{k}_\parallel,t')\right]\right\rangle\\
&=N_\parallel^2\chi^{m\sigma\ \mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}'_\parallel,\mathbf{k}_\parallel,t-t')\delta_{\mathbf{k}'_\parallel+\mathbf{q}_\parallel,\mathbf{k}_\parallel}\delta_{\mathbf{k}'_\parallel,\mathbf{k}_\parallel}\ ,
\end{aligned}
\tag{5.278}
$$

we get

$$
\begin{aligned}
\langle \hat{\mathcal{I}}^{m}_{\ell_1\ell_2}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{q}_\parallel)\rangle =&\frac{eE_0}{2\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\int dt'\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ \left[e^{i\omega t'}-e^{-i\omega t'}\right]t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\\
&\times\chi^{m\sigma\ \mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,t-t')\delta_{\mathbf{q}_\parallel,0}\\
=&\frac{eE_0}{2\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\\
&\times\left[e^{i\omega t}\chi^{m\sigma\ \mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)-e^{-i\omega t}\chi^{m\sigma\ \mu\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)\right]\delta_{\mathbf{q}_\parallel,0}\ .
\end{aligned}
\tag{5.279}
$$

where we have used the Fourier transform given by Eq. (5.113). Taking two sites in the same plane, i.e., $\ell_2=\ell_1$, the parallel current becomes

$$
\langle \hat{I}^{m}_{\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)\rangle =\langle \hat{\mathcal{I}}^{m}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)\rangle-\langle \hat{\mathcal{I}}^{m}_{\ell_1\ell_1}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)\rangle\ ,
\tag{5.280}
$$

where we have already taken into account the $\delta_{\mathbf{q}_\parallel,0}$. Here,

$$
\begin{aligned}
\langle \hat{\mathcal{I}}^{m}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)\rangle = -\frac{eE_0}{2\hbar\omega}\frac{1}{N_\parallel}&\sum_{\mathbf{k}_\parallel,\sigma}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\\
&\times\left[e^{-i\omega t}\chi^{m\sigma\ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)-e^{i\omega t}\chi^{m\sigma\ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\right]\ .
\end{aligned}
\tag{5.281}
$$

Therefore, the spin current may be written as

$$
\begin{aligned}
\langle \hat{I}^m_{\ell_1\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)\rangle =& -\frac{eE_0}{2\hbar\omega}\frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel,\sigma}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\left[t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}-t^{\nu\mu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\right]\\
&\times\left[e^{-i\omega t}\chi^{m\sigma\;\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)-e^{i\omega t}\chi^{m\sigma\;\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\right]\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\\
=& -\frac{eE_0}{2\hbar\omega}\left[e^{-i\omega t}\mathcal{J}^m_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)-e^{i\omega t}\mathcal{J}^m_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,-\omega)\right]\ .
\end{aligned}
\tag{5.282}
$$

where we have defined

$$
\begin{aligned}
\mathcal{J}^m_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega) =& \frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel,\sigma}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\left[t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}-t^{\nu\mu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\right]\\
&\times\chi^{m\sigma\;\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ .
\end{aligned}
\tag{5.283}
$$

This expression is general, and can also be used to define the charge current obtained in Eqs. 5.251-5.252 as $\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)=\mathcal{J}^{\uparrow}_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)+\mathcal{J}^{\downarrow}_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)$.

Note that

$$
\begin{aligned}
\left[\mathcal{J}^m_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)\right]^* =& \frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel,\sigma}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\left[t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}-t^{\nu\mu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\right]\\
&\times\chi^{\overline{m}\sigma\;\nu\mu\xi\gamma}_{\ell_1\ell_1\ell'\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\xi\gamma}_{\ell'\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\\
=& \frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel,\sigma}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\left[t^{\nu\mu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}-t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\right]\\
&\times\chi^{\overline{m}\sigma\;\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\\
=& -\mathcal{J}^{\overline{m}}_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,-\omega)\ .
\end{aligned}
\tag{5.284}
$$

Therefore, for $m=x,y,z$, Eq. (5.282) becomes

$$
\begin{aligned}
\langle \hat{I}^m_{\ell_1\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)\rangle =& -\frac{eE_0}{2\hbar\omega}\left\{e^{-i\omega t}\mathcal{J}^m_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)+\left[e^{-i\omega t}\mathcal{J}^m_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)\right]^*\right\}\\
=& -\frac{eE_0}{\hbar\omega}\,\mathrm{Re}\left\{e^{-i\omega t}\mathcal{J}^m_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)\right\}\\
=& -\frac{eE_0}{\hbar\omega}\left|\mathcal{J}^m_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)\right|\cos\left(\omega t-\phi^m\right)\ .
\end{aligned}
\tag{5.285}
$$

where ϕ^m is the phase of the complex number $\mathcal{J}^m_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)$ - so it also depends on ℓ_1, $\mathbf{R}'_\parallel-\mathbf{R}_\parallel$ and ω.

With this result, we can can obtain the spin current with $x-$polarization as

$$\begin{aligned}\langle \hat{I}^x_{\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)\rangle =& -\frac{eE_0}{\hbar\omega}\left|\mathcal{J}^x_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)\right|\cos\left(\omega t-\phi^x\right)\\ =& -\frac{eE_0}{\hbar\omega}\left|\frac{\mathcal{J}^+_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)+\mathcal{J}^-_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)}{2}\right|\cos(\omega t-\phi^x)\ ,\end{aligned} \tag{5.286}$$

The $y-$polarization component may be obtained following the same steps:

$$\begin{aligned}\langle \hat{I}^y_{\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)\rangle =& -\frac{eE_0}{\hbar\omega}\left|\mathcal{J}^y_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)\right|\cos\left(\omega t-\phi^y\right)\\ =& -\frac{eE_0}{\hbar\omega}\left|\frac{\mathcal{J}^+_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)-\mathcal{J}^-_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)}{2i}\right|\cos(\omega t-\phi^y)\ ,\end{aligned} \tag{5.287}$$

And finally, the $z-$component is given by

$$\begin{aligned}\langle \hat{I}^z_{\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)\rangle =& -\frac{eE_0}{\hbar\omega}\left|\mathcal{J}^z_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)\right|\cos\left(\omega t-\phi^z\right)\\ =& -\frac{eE_0}{\hbar\omega}\left|\frac{\mathcal{J}^\uparrow_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)-\mathcal{J}^\downarrow_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)}{2}\right|\cos(\omega t-\phi^z)\ ,\end{aligned} \tag{5.288}$$

5.4.6 Orbital Angular Momentum Current

Following the same steps as we did with the charge current, but now using the operator given by Eq. (5.85), we obtain

$$\begin{aligned}\langle \hat{\mathcal{I}}^{L_m}_{(1)\ell_1\ell_2}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{q}_\parallel)\rangle =& -\delta_{\mathbf{q}_\parallel,0}\frac{e^2E_0}{2\hbar^2\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}} L^m_{\alpha\mu}t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\\ &\times\left[e^{-i\omega t}\chi^{\sigma\sigma'\ \alpha\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)-e^{i\omega t}\chi^{\sigma\sigma'\ \alpha\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\right]\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\\ =& -\delta_{\mathbf{q}_\parallel,0}\frac{e^2E_0}{2\hbar^2\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}} t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\\ &\times L^m_{\alpha\mu}\left[e^{-i\omega t}\chi^{\sigma\sigma'\ \alpha\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)-e^{i\omega t}\chi^{\sigma\sigma'\ \alpha\nu\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\right]\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ .\end{aligned} \tag{5.289}$$

The in-plane orbital angular momentum current is given by

$$
\begin{aligned}
\langle \hat{\mathcal{I}}^{L_m}_{(1)\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)\rangle = & -\frac{e^2 E_0}{2\hbar^2\omega}\frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}} L^m_{\mu\alpha} t^{\alpha\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \\
& \times \left[e^{-i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega) - e^{i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\right] \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E \\
= & -\frac{e^2 E_0}{2\hbar^2\omega}\left[e^{-i\omega t}\mathcal{C}^{L_m}_{(1)\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega) - e^{i\omega t}\mathcal{C}^{L_m}_{(1)\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,-\omega)\right] \ .
\end{aligned} \tag{5.290}
$$

where we have already taken into account $\mathbf{q}_\parallel = 0$ and included the factor $1/N_\parallel$. We have also defined

$$
\mathcal{C}^{L_m}_{(1)\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega) = \frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}} L^m_{\mu\alpha} t^{\alpha\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E \ . \tag{5.291}
$$

With this definition,

$$
\begin{aligned}
\mathcal{C}^{L_m}_{(1)\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,-\omega) = & \frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}} L^m_{\mu\alpha} t^{\alpha\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E \\
= & \frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}} L^m_{\mu\alpha} t^{\alpha\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\left[\chi^{\sigma\sigma'\,\nu\mu\xi\gamma}_{\ell_1\ell_1\ell'\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)\right]^*\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E \\
= & \frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}} \left[L^{\overline{m}}_{\alpha\mu} t^{\alpha\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)}\chi^{\sigma\sigma'\,\nu\mu\xi\gamma}_{\ell_1\ell_1\ell'\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\xi\gamma}_{\ell'\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\right]^* \\
= & \frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}} \left[t^{\nu\alpha}_{\ell_1\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) L^{\overline{m}}_{\alpha\mu} e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)}\chi^{\sigma\sigma'\,\nu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\right]^* \ .
\end{aligned} \tag{5.292}
$$

The second term is given by

$$
\begin{aligned}
\langle \hat{\mathcal{I}}^{L_m}_{(2)\ell_1\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel,\mathbf{q}_\parallel)\rangle = & -\frac{e^2 E_0}{2\hbar^2\omega}\frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}} t^{\nu\alpha}_{\ell_1\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) L^m_{\alpha\mu} e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} \\
& \times \left[e^{-i\omega t}\chi^{\sigma\sigma'\,\nu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega) - e^{i\omega t}\chi^{\sigma\sigma'\,\nu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\right] \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E \\
= & -\frac{e^2 E_0}{2\hbar^2\omega}\left[e^{-i\omega t}\mathcal{C}^{L_m}_{(2)\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel,\omega) - e^{i\omega t}\mathcal{C}^{L_m}_{(2)\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel,-\omega)\right] \ ,
\end{aligned} \tag{5.293}
$$

where

$$\mathcal{C}^{Lm}_{(2)\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel, \omega) = \frac{1}{N_\parallel} \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \alpha\mu\nu\gamma\xi}} t^{\nu\alpha}_{\ell_1\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) L^m_{\alpha\mu} e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} \chi^{\sigma\sigma'\,\nu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega) \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E$$

$$= \left[\mathcal{C}^{L\overline{m}}_{(1)\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, -\omega)\right]^* \ , \tag{5.294}$$

and

$$\begin{aligned}
\mathcal{C}^{Lm}_{(2)\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel, -\omega) =& \frac{1}{N_\parallel} \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \alpha\mu\nu\gamma\xi}} t^{\nu\alpha}_{\ell_1\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) L^m_{\alpha\mu} e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} \chi^{\sigma\sigma'\,\nu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, -\omega) \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \\
=& \frac{1}{N_\parallel} \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \alpha\mu\nu\gamma\xi}} t^{\nu\alpha}_{\ell_1\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel) L^m_{\alpha\mu} e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)} \left[\chi^{\sigma\sigma'\,\mu\nu\xi\gamma}_{\ell_1\ell_1\ell'\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega)\right]^* \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \\
=& \frac{1}{N_\parallel} \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \alpha\mu\nu\gamma\xi}} \left[L^{\overline{m}}_{\mu\alpha} t^{\alpha\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \chi^{\sigma\sigma'\,\mu\nu\xi\gamma}_{\ell_1\ell_1\ell'\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega) \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\xi\gamma}_{\ell'\ell}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E\right]^* \\
=& \frac{1}{N_\parallel} \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \alpha\mu\nu\gamma\xi}} \left[L^{\overline{m}}_{\mu\alpha} t^{\alpha\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} \chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega) \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E\right]^* \\
=& \left[\mathcal{C}^{L\overline{m}}_{(1)\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega)\right]^* \ ,
\end{aligned} \tag{5.295}$$

So, we can write the expectation value of the current operator as

$$\begin{aligned}
\langle \hat{I}^{Lm}_{\ell_1\ell_1}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel) &= \left\{\hat{\mathcal{I}}^{Lm}_{(1)\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) - \hat{\mathcal{I}}^{Lm}_{(2)\ell_1\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel)\right\} \\
&= -\frac{e^2 E_0}{2\hbar^2\omega} \Big[e^{-i\omega t} \mathcal{C}^{Lm}_{(1)\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega) - e^{i\omega t} \mathcal{C}^{Lm}_{(1)\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, -\omega) \\
&\qquad - e^{-i\omega t} \mathcal{C}^{Lm}_{(2)\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel, \omega) + e^{i\omega t} \mathcal{C}^{Lm}_{(2)\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel, -\omega)\Big] \\
&= -\frac{e^2 E_0}{2\hbar^2\omega} \Big\{e^{-i\omega t} \mathcal{C}^{Lm}_{(1)\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega) - \left[e^{-i\omega t} \mathcal{C}^{L\overline{m}}_{(2)\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel, \omega)\right]^* \\
&\qquad - e^{-i\omega t} \mathcal{C}^{Lm}_{(2)\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel, \omega) + \left[e^{-i\omega t} \mathcal{C}^{L\overline{m}}_{(1)\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega)\right]^*\Big\} \ .
\end{aligned} \tag{5.296}$$

For $m = x, y, z$, $\overline{m} = m$ and we can write

$$\langle \hat{I}^{Lm}_{\ell_1\ell_1}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel) = -\frac{e^2 E_0}{\hbar^2\omega} \operatorname{Re}\left\{e^{-i\omega t}\left[\mathcal{C}^{Lm}_{(1)\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega) - \mathcal{C}^{Lm}_{(2)\ell_1}(\mathbf{R}_\parallel - \mathbf{R}'_\parallel, \omega)\right]\right\} \ . \tag{5.297}$$

Finally, we can summarize this result as

$$\begin{aligned}\langle \hat{I}^{Lm}_{\ell_1\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel\rangle &= -\frac{e^2E_0}{\hbar^2\omega}\,\mathrm{Re}\left\{e^{-i\omega t}\mathcal{J}^{Lm}_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)\right\} \\ &= -\frac{e^2E_0}{\hbar^2\omega}|\mathcal{J}^{Lm}_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega)|\cos(\omega t-\phi^m_L)\ .\end{aligned} \tag{5.298}$$

where

$$\begin{aligned}\mathcal{J}^{Lm}_{\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\omega) = \frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}} &\left[L^m_{\mu\alpha}t^{\alpha\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)} - t^{\alpha\mu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)L^m_{\alpha\nu}e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\right] \\ &\times \chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\end{aligned} \tag{5.299}$$

Note that if we change L^m to the identity (i.e., $L^m_{\mu\alpha}=\delta_{\mu\alpha}$), we recover the result for charge current

$$
\begin{aligned}
\langle\hat{\mathcal{I}}^{L\,m}_{(2)\ell_2\ell_1}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel,\mathbf{q}_\parallel)\rangle =&\delta_{\mathbf{q}_\parallel,0}\frac{e^2E_0}{2\hbar^2\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}} t^{\nu\mu}_{\ell_2\ell_1}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)L^m_{\mu\alpha}e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}\\
&\times\left[e^{-i\omega t}\chi^{\sigma\sigma'\,\nu\alpha\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)-e^{i\omega t}\chi^{\sigma\sigma'\,\nu\alpha\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\right]\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\\
=&\delta_{\mathbf{q}_\parallel,0}\frac{e^2E_0}{2\hbar^2\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}} t^{\mu\nu}_{\ell_1\ell_2}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)}\\
&\times L^m_{\mu\alpha}\left[e^{-i\omega t}\chi^{\sigma\sigma'\,\nu\alpha\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)-e^{i\omega t}\chi^{\sigma\sigma'\,\nu\alpha\gamma\xi}_{\ell_1\ell_2\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\right]\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ .
\end{aligned}
\tag{5.300}
$$

So, the in-plane orbital angular momentum current is given by

$$
\langle\hat{I}^{L\,m}_{\ell_1\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)=\frac{1}{N_\parallel}\left\{\hat{\mathcal{I}}^{L\,m}_{(1)\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)-\hat{\mathcal{I}}^{L\,m}_{(2)\ell_1\ell_1}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)\right\}\ . \tag{5.301}
$$

Using the equations above, we obtain

$$
\begin{aligned}
\langle\hat{I}^{L\,m}_{\ell_1\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)\rangle =&\frac{e^2E_0}{2\hbar^2\omega}\frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}}\left[L^m_{\alpha\mu}t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}-t^{\alpha\mu}_{\ell_1\ell_1}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)L^m_{\mu\nu}e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\right]\\
&\times\left[e^{-i\omega t}\chi^{\sigma\sigma'\,\alpha\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)-e^{i\omega t}\chi^{\sigma\sigma'\,\alpha\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\right]\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\\
=&\frac{e^2E_0}{2\hbar^2\omega}\frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}}\left[L^m_{\mu\alpha}t^{\alpha\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}-t^{\mu\alpha}_{\ell_1\ell_1}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)L^m_{\alpha\nu}e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\right]\\
&\times\left[e^{-i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)-e^{i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\right]\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ .
\end{aligned}
\tag{5.302}
$$

Let us calculate the $x-$component onde again using

$$
\begin{aligned}
\langle\hat{I}^{L\,x}_{\ell_1\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)\rangle =&\mathrm{Re}\{\langle\hat{I}^{L\,+}_{\ell_1\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel)\rangle\}\\
=&\frac{e^2E_0}{2\hbar^2\omega}\frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}}\left[L^+_{\mu\alpha}t^{\alpha\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}-t^{\mu\alpha}_{\ell_1\ell_1}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)L^+_{\alpha\nu}e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\right]\\
&\times\left[e^{-i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)-e^{i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\right]\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ .
\end{aligned}
\tag{5.303}
$$

We can separate this into two terms, each containing one gerenalized susceptibility. The first one is given by

$$\frac{e^2 E_0}{2\hbar^2\omega}\frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}}\left[L^+_{\mu\alpha}t^{\alpha\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}-t^{\mu\alpha}_{\ell_1\ell_1}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)L^+_{\alpha\nu}e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\right]$$
$$\times\left[e^{-i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)\right]\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ . \tag{5.304}$$

and the second one

$$-\frac{e^2 E_0}{2\hbar^2\omega}\frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}}\mathrm{Re}\left\{\left[L^+_{\mu\alpha}t^{\alpha\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}-t^{\mu\alpha}_{\ell_1\ell_1}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)L^+_{\alpha\nu}e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\right]\right.$$
$$\left.\times\left[e^{i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\right]\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\right\}$$
$$-\frac{e^2 E_0}{2\hbar^2\omega}\frac{1}{N_\parallel}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \alpha\mu\nu\gamma\xi}}\mathrm{Re}\left\{\left[L^-_{\mu\alpha}t^{\alpha\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}-t^{\mu\alpha}_{\ell_1\ell_1}(\mathbf{R}_\parallel-\mathbf{R}'_\parallel)L^+_{\alpha\nu}e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\right]\right.$$
$$\left.\times\left[e^{-i\omega t}\chi^{\sigma\sigma'\,\nu\mu\xi\gamma}_{\ell_1\ell_1\ell'\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)\right]^*\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\right\}\ . \tag{5.305}$$

5.4.7 Charge Disturbance

The charge disturbance in multilayers is given by

$$\begin{aligned}\delta\langle\hat{\rho}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle &= -\frac{i}{\hbar}\int dt'\Theta(t-t')\left\langle\left[\hat{\rho}_{\ell_1}(\mathbf{q}_\parallel,t),\hat{H}_{\mathrm{int}}(t')\right]\right\rangle\\ &= -\frac{i}{\hbar}\frac{eE_0}{\hbar\omega}\frac{1}{N}\sum_{\mathbf{k}_\parallel,\sigma}\sum_{\substack{\ell\ell'\\ \gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t')\left\langle\left[\hat{\rho}_{\ell_1}(\mathbf{q}_\parallel,t),c^\dagger_{\ell\gamma\sigma}(\mathbf{k}_\parallel,t')c_{\ell'\xi\sigma}(\mathbf{k}_\parallel,t')\right]\right\rangle\\ &= -\frac{i}{\hbar}\frac{eE_0}{2i\hbar\omega}\frac{1}{N^2}\sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right]\\ &\quad\times\left\langle\left[c^\dagger_{\ell_1\mu\sigma'}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel,t)c_{\ell_1\mu\sigma'}(\mathbf{k}'_\parallel,t),c^\dagger_{\ell\gamma\sigma}(\mathbf{k}_\parallel,t')c_{\ell'\xi\sigma}(\mathbf{k}_\parallel,t')\right]\right\rangle\ ,\end{aligned} \tag{5.306}$$

where we have used

$$
\begin{aligned}
\rho_\ell(\mathbf{q}_\parallel) &= \sum_{\mathbf{R}_\parallel} e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}\rho_\ell(\mathbf{R}_\parallel) \\
&= \sum_{\mathbf{R}_\parallel}\sum_{\gamma}\sum_{\sigma'} e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel} c^\dagger_{\ell\gamma\sigma'}(\mathbf{R}_\parallel)c_{\ell\gamma\sigma'}(\mathbf{R}_\parallel) \\
&= \frac{1}{N^2}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_{\mathbf{R}_\parallel}\sum_{\gamma}\sum_{\sigma'} e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel} e^{-i\mathbf{k}'_\parallel\cdot\mathbf{R}_\parallel} c^\dagger_{\ell\gamma\sigma'}(\mathbf{k}'_\parallel) e^{i\mathbf{k}_\parallel\cdot\mathbf{R}_\parallel} c_{\ell\gamma\sigma'}(\mathbf{k}_\parallel) \\
&= \frac{1}{N}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_{\gamma}\sum_{\sigma'} \delta_{\mathbf{k}'_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel} c^\dagger_{\ell\gamma\sigma'}(\mathbf{k}'_\parallel)c_{\ell\gamma\sigma'}(\mathbf{k}_\parallel) \\
&= \frac{1}{N}\sum_{\mathbf{k}_\parallel}\sum_{\gamma}\sum_{\sigma'} c^\dagger_{\ell\gamma\sigma'}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)c_{\ell\gamma\sigma'}(\mathbf{k}_\parallel) \ .
\end{aligned}
\tag{5.307}
$$

Identifying the susceptibility analogous to Eq. (5.123),

$$
\begin{aligned}
\delta\langle\hat{\rho}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle =& \frac{eE_0}{2i\hbar\omega}\frac{1}{N^2}\sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\int dt'\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right]\chi^{\sigma'\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel,-\mathbf{k}'_\parallel,\mathbf{k}_\parallel,-\mathbf{k}_\parallel,t-t') \\
=& \frac{eE_0}{2i\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\int dt'\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right]\chi^{\sigma'\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel,\mathbf{k}_\parallel,t-t')\delta_{\mathbf{k}'_\parallel,\mathbf{k}_\parallel}\delta_{\mathbf{k}'_\parallel+\mathbf{q}_\parallel,\mathbf{k}_\parallel} \\
=& -\frac{ieE_0}{2\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\int dt'\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right]\chi^{\sigma'\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,t-t')\delta_{\mathbf{q}_\parallel 0} \\
=& -\frac{ieE_0}{2\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t}\chi^{\sigma'\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,-\omega)-e^{-i\omega t}\chi^{\sigma'\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)\right]\delta_{\mathbf{q}_\parallel 0} \\
=& \frac{ieE_0}{2\hbar\omega}\left[e^{-i\omega t}\mathcal{D}_{\ell_1}(\omega)-e^{i\omega t}\mathcal{D}_{\ell_1}(-\omega)\right]\delta_{\mathbf{q}_\parallel 0} \ .
\end{aligned}
\tag{5.308}
$$

where the complex number $\mathcal{D}_{\ell_1}$ is given by

$$
\mathcal{D}_{\ell_1}(\omega) = \sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\chi^{\sigma'\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega) \ . \tag{5.309}
$$

With this definition

$$
\begin{aligned}
\mathcal{D}_{\ell_1}(-\omega) =& \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\gamma\xi}} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \chi^{\sigma'\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, -\omega) \\
=& \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\gamma\xi}} \left[\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\xi\gamma}_{\ell'\ell}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \chi^{\sigma'\sigma\,\mu\mu\xi\gamma}_{\ell_1\ell_1\ell'\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega) \right]^* \\
=& \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\gamma\xi}} \left[\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \chi^{\sigma'\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel, \omega) \right]^* \\
=& [\mathcal{D}_{\ell_1}(\omega)]^*
\end{aligned}
\tag{5.310}
$$

So, we have

$$
\begin{aligned}
\delta\langle \hat{\rho}_{\ell_1}(\mathbf{q}_\parallel, t)\rangle =& \frac{ieE_0}{2\hbar\omega} \left\{ e^{-i\omega t}\mathcal{D}_{\ell_1}(\omega) - e^{i\omega t}[\mathcal{D}_{\ell_1}(\omega)]^* \right\} \delta_{\mathbf{q}_\parallel 0} \\
=& \frac{ieE_0}{2\hbar\omega} 2i \,\mathrm{Im} \left\{ e^{-i\omega t}\mathcal{D}_{\ell_1}(\omega) \right\} \delta_{\mathbf{q}_\parallel 0} \\
=& \frac{eE_0}{\hbar\omega} |\mathcal{D}_{\ell_1}(\omega)| \sin[\omega t - \phi^{\mathrm{C}}(\omega)] \delta_{\mathbf{q}_\parallel 0}
\end{aligned}
\tag{5.311}
$$

where $\phi^{\mathrm{C}}(\omega)$ is the phase of $\mathcal{D}_{\ell_1}(\omega)$.

We can also obtain the excitation energies from the Fluctuation-Dissipation theorem by calculating the susceptibility

$$
\begin{aligned}
\langle\langle \hat{\rho}_\ell(\mathbf{q}_\parallel), \hat{\rho}_\ell(-\mathbf{q}_\parallel)\rangle\rangle_\omega =& \sum_{\substack{\mathbf{k}_\parallel \mathbf{k}'_\parallel \\ \sigma\sigma'}} \sum_{\mu\nu} \chi^{\sigma\sigma'\,\mu\mu\nu\nu}_{\ell\ell\ell\ell}(\mathbf{k}+\mathbf{q}, \mathbf{k}, \mathbf{k}'-\mathbf{q}, \mathbf{k}'; \omega) \\
=& \sum_{\substack{\mathbf{k}_\parallel \mathbf{k}'_\parallel \\ \sigma\sigma'}} \sum_{\mu\nu} \chi^{\sigma\sigma'\,\mu\mu\nu\nu}_{\ell\ell\ell\ell}(\mathbf{k}+\mathbf{q}, \mathbf{k}'-\mathbf{q}; \omega) \delta_{\mathbf{k}'-\mathbf{q},\mathbf{k}} \delta_{\mathbf{k}+\mathbf{q},\mathbf{k}'} \\
=& \sum_{\substack{\mathbf{k}_\parallel \\ \sigma\sigma'}} \sum_{\mu\nu} \chi^{\sigma\sigma'\,\mu\mu\nu\nu}_{\ell\ell\ell\ell}(\mathbf{k}+\mathbf{q}, \mathbf{k}; \omega) \\
=& \sum_{\sigma\sigma'} \sum_{\mu\nu} \chi^{\sigma\sigma'\,\mu\nu}_{\ell\ell}(\mathbf{q}; \omega) \ ,
\end{aligned}
\tag{5.312}
$$

which involves the components σ, σ' of the usual susceptibility.

5.4.8 Spin Disturbance

We can also calculate the spin disturbance generated by an electric field. It is given by

$$\begin{aligned}
\delta\langle\hat{S}_i^m\rangle(t) =& -\frac{i}{\hbar}\int dt'\Theta(t-t')\left\langle\left[\hat{S}_i^m(t),\hat{H}_{\rm int}(t')\right]\right\rangle \\
=&\frac{ieE_0}{4im\hbar\omega}\sum_\sigma\sum_{\substack{k\ell\\ \gamma\xi}}\int dt'\Theta(t-t')\left[\mathbf{w}_{k\ell}^{\gamma\xi}(\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{-i\omega t'}-\mathbf{w}_{k\ell}^{\gamma\xi}(-\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{i\omega t'}\right]\left\langle\left[\hat{S}_i^m(t),c_{k\gamma\sigma}^\dagger(t')c_{\ell\xi\sigma}(t')\right]\right\rangle \\
=&-\frac{eE_0}{4im\omega}\sum_\sigma\sum_{\substack{k\ell\\ \mu\gamma\xi}}\int dt'\left[\mathbf{w}_{k\ell}^{\gamma\xi}(\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{-i\omega t'}-\mathbf{w}_{k\ell}^{\gamma\xi}(-\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{i\omega t'}\right]\chi_{iik\ell}^{m\sigma\mu\mu\gamma\xi}(t-t') \\
=&-\frac{eE_0}{4im\omega}\sum_\sigma\sum_{\substack{k\ell\\ \mu\gamma\xi}}\left[\mathbf{w}_{k\ell}^{\gamma\xi}(\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{-i\omega t}\chi_{iik\ell}^{m\sigma\mu\mu\gamma\xi}(\omega)-\mathbf{w}_{k\ell}^{\gamma\xi}(-\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{i\omega t}\chi_{iik\ell}^{m\sigma\mu\mu\gamma\xi}(-\omega)\right]\ .
\end{aligned}\tag{5.313}$$

For uniform electric fields ($\mathbf{q}=0$) we have

$$\begin{aligned}
\delta\langle\hat{S}_i^m\rangle(t) =& -\frac{eE_0}{2im\omega}\sum_\sigma\sum_{\substack{k\ell\\ \mu\gamma\xi}}\mathbf{p}_{k\ell}^{\gamma\xi}\cdot\hat{\mathbf{u}}_E\left[e^{-i\omega t}\chi_{iik\ell}^{m\sigma\mu\mu\gamma\xi}(\omega)-e^{i\omega t}\chi_{iik\ell}^{m\sigma\mu\mu\gamma\xi}(-\omega)\right] \\
=&-\frac{eE_0}{2im\omega}\sum_\sigma\sum_{\substack{k\ell\\ \mu\gamma\xi}}\left\{e^{-i\omega t}\mathbf{p}_{k\ell}^{\gamma\xi}\chi_{iik\ell}^{m\sigma\mu\mu\gamma\xi}(\omega)-e^{i\omega t}\left[\mathbf{p}_{\ell k}^{\xi\gamma}\chi_{ii\ell k}^{\overline{m}\sigma\mu\mu\xi\gamma}(\omega)\right]^*\right\}\cdot\hat{\mathbf{u}}_E \\
=&-\frac{eE_0}{2im\omega}\sum_\sigma\sum_{\substack{k\ell\\ \mu\gamma\xi}}\left\{e^{-i\omega t}\mathbf{p}_{k\ell}^{\gamma\xi}\chi_{iik\ell}^{m\sigma\mu\mu\gamma\xi}(\omega)-e^{i\omega t}\left[\mathbf{p}_{k\ell}^{\gamma\xi}\chi_{iik\ell}^{\overline{m}\sigma\mu\mu\gamma\xi}(\omega)\right]^*\right\}\cdot\hat{\mathbf{u}}_E\ ,
\end{aligned}\tag{5.314}$$

where $\overline{m}$ denotes the swap between the two spin indices (i.e., $\overline{m}=\mp$ if $m=\pm$ and $\overline{m}=\uparrow,\downarrow$ if $m=\uparrow,\downarrow$) and we have used the fact that

$$\begin{aligned}
\left[\mathbf{p}_{k\ell}^{\gamma\xi}\right]^* =& \int d\mathbf{r}\left[\phi_\gamma^*(\mathbf{r}-\mathbf{R}_k)\left(\frac{\hbar}{i}\nabla\right)\phi_\xi(\mathbf{r}-\mathbf{R}_\ell)\right]^* \\
=&\int d\mathbf{r}\phi_\gamma(\mathbf{r}-\mathbf{R}_k)\left(-\frac{\hbar}{i}\nabla\right)\phi_\xi^*(\mathbf{r}-\mathbf{R}_\ell) \\
=&\int d\mathbf{r}\phi_\xi^*(\mathbf{r}-\mathbf{R}_\ell)\left(\frac{\hbar}{i}\nabla\right)\phi_\gamma(\mathbf{r}-\mathbf{R}_k) \\
=&\mathbf{p}_{\ell k}^{\xi\gamma}\ .
\end{aligned}\tag{5.315}$$

In multilayers,

$$
\begin{aligned}
\delta\langle \hat{S}^m_{\ell_1}(\mathbf{q}_\parallel, t)\rangle &= -\frac{i}{\hbar}\int dt'\Theta(t-t')\left\langle\left[\hat{S}^m_{\ell_1}(\mathbf{q}_\parallel, t), \hat{H}_{\text{int}}(t')\right]\right\rangle \\
&= -\frac{i}{\hbar}\frac{eE_0}{\hbar\omega}\frac{1}{N}\sum_{\mathbf{k}_\parallel,\sigma}\sum_{\substack{\ell\ell' \\ \gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t')\Big\langle\Big[\hat{S}^m_{\ell_1}(\mathbf{q}_\parallel, t), c^\dagger_{\ell\gamma\sigma}(\mathbf{k}_\parallel, t')c_{\ell'\xi\sigma}(\mathbf{k}_\parallel, t') \\
&= -\frac{i}{\hbar}\frac{eE_0}{2i\hbar\omega}\frac{1}{N^2}\sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel \\ \sigma}}\sum_{\substack{\ell\ell' \\ \mu\gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'} - e^{-i\omega t'}\right] \\
&\quad\times\left\langle\left[S^{m\ \mu\mu}_{\ell_1\ell_1}(\mathbf{k}'_\parallel + \mathbf{q}_\parallel, \mathbf{k}'_\parallel, t), c^\dagger_{\ell\gamma\sigma}(\mathbf{k}_\parallel, t')c_{\ell'\xi\sigma}(\mathbf{k}_\parallel, t')\right]\right\rangle\ ,
\end{aligned}
\tag{5.316}
$$

where we have used

$$
\begin{aligned}
S^m_\ell(\mathbf{q}_\parallel) &= \sum_{\mathbf{R}_\parallel} e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel} S^m_\ell(\mathbf{R}_\parallel) \\
&= \frac{1}{2}\sum_{\mathbf{R}_\parallel}\sum_\gamma\sum_{\alpha\beta} e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel} c^\dagger_{\ell\gamma\alpha}(\mathbf{R}_\parallel)\sigma^m_{\alpha\beta}c_{\ell\gamma\beta}(\mathbf{R}_\parallel) \\
&= \frac{1}{2N^2}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_{\mathbf{R}_\parallel}\sum_\gamma\sum_{\alpha\beta} e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel}e^{-i\mathbf{k}'_\parallel\cdot\mathbf{R}_\parallel}c^\dagger_{\ell\gamma\alpha}(\mathbf{k}'_\parallel)\sigma^m_{\alpha\beta}\ e^{i\mathbf{k}_\parallel\cdot\mathbf{R}_\parallel}c_{\ell\gamma\beta}(\mathbf{k}_\parallel) \\
&= \frac{1}{2N}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_\gamma\sum_{\alpha\beta}\delta_{\mathbf{k}'_\parallel, \mathbf{k}_\parallel + \mathbf{q}_\parallel}c^\dagger_{\ell\gamma\alpha}(\mathbf{k}'_\parallel)\sigma^m_{\alpha\beta}c_{\ell\gamma\beta}(\mathbf{k}_\parallel) \\
&= \frac{1}{2N}\sum_{\mathbf{k}_\parallel}\sum_\gamma\sum_{\alpha\beta}c^\dagger_{\ell\gamma\alpha}(\mathbf{k}_\parallel + \mathbf{q}_\parallel)\sigma^m_{\alpha\beta}c_{\ell\gamma\beta}(\mathbf{k}_\parallel) \\
&= \frac{1}{N}\sum_{\mathbf{k}_\parallel}\sum_\gamma S^{m\ \gamma\gamma}_{\ell\ell}(\mathbf{k}_\parallel + \mathbf{q}_\parallel, \mathbf{k}_\parallel)\ .
\end{aligned}
\tag{5.317}
$$

Identifying the susceptibility given by Eq. (5.123),

$$
\begin{aligned}
\delta\langle \hat{S}^m_{\ell_1}(\mathbf{q}_\parallel,t)\rangle =&\frac{eE_0}{2i\hbar\omega}\frac{1}{N^2}\sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\int dt'\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right]\chi^{m\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel,-\mathbf{k}'_\parallel,\mathbf{k}_\parallel,-\mathbf{k}_\parallel,t-t')\\
=&\frac{eE_0}{2i\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\int dt'\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right]\chi^{m\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel,\mathbf{k}_\parallel,t-t')\delta_{\mathbf{k}'_\parallel,\mathbf{k}_\parallel}\delta_{\mathbf{k}'_\parallel+\mathbf{q}_\parallel,\mathbf{k}_\parallel}\\
=&-\frac{ieE_0}{2\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\int dt'\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right]\chi^{m\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,t-t')\delta_{\mathbf{q}_\parallel 0}\\
=&-\frac{ieE_0}{2\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t}\chi^{m\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,-\omega)-e^{-i\omega t}\chi^{m\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\right]\delta_{\mathbf{q}_\parallel 0}\\
=&\frac{ieE_0}{2\hbar\omega}\left[e^{-i\omega t}\mathcal{D}^m_{\ell_1}(\omega)-e^{i\omega t}\mathcal{D}^m_{\ell_1}(-\omega)\right]\delta_{\mathbf{q}_\parallel 0}\ .
\end{aligned}
\tag{5.318}
$$

where the complex number $\mathcal{D}^m_{\ell_1}$ is given by

$$
\mathcal{D}^m_{\ell_1}(\omega)=\sum_{\substack{\mathbf{k}_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\chi^{m\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\ . \tag{5.319}
$$

With this definition

$$
\begin{aligned}
\mathcal{D}^m_{\ell_1}(-\omega)=&\sum_{\substack{\mathbf{k}_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\chi^{m\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,-\omega)\\
=&\sum_{\substack{\mathbf{k}_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\left[\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\xi\gamma}_{\ell'\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\chi^{\overline{m}\sigma\,\mu\mu\xi\gamma}_{\ell_1\ell_1\ell'\ell}(\mathbf{k}_\parallel,\omega)\right]^*\\
=&\sum_{\substack{\mathbf{k}_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\left[\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\chi^{\overline{m}\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\right]^*\\
=&[\mathcal{D}^{\overline{m}}_{\ell_1}(\omega)]^*
\end{aligned}
\tag{5.320}
$$

So, for $m=x,y,z$ we have

$$
\begin{aligned}
\delta\langle \hat{S}^m_{\ell_1}(\mathbf{q}_\parallel,t)\rangle =&\frac{ieE_0}{2\hbar\omega}\left\{e^{-i\omega t}\mathcal{D}^m_{\ell_1}(\omega)-[e^{-i\omega t}\mathcal{D}^m_{\ell_1}(\omega)]^*\right\}\delta_{\mathbf{q}_\parallel 0}\\
=&\frac{ieE_0}{2\hbar\omega}2i\,\mathrm{Im}\left\{e^{-i\omega t}\mathcal{D}^m_{\ell_1}(\omega)\right\}\delta_{\mathbf{q}_\parallel 0}\\
=&\frac{eE_0}{\hbar\omega}|\mathcal{D}^m_{\ell_1}(\omega)|\sin[\omega t-\phi^m(\omega)]\delta_{\mathbf{q}_\parallel 0}
\end{aligned}
\tag{5.321}
$$

where

$$
\mathcal{D}^m_{\ell_1}(\omega)=\sum_{\substack{\mathbf{k}_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\chi^{m\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ . \tag{5.322}
$$

Therefore, the non-vanishing ($\mathbf{q}_\parallel = 0$) x-component spin disturbance is given by

$$\delta\langle\hat{S}^x_{\ell_1}(t)\rangle = \frac{eE_0}{\hbar\omega}|\mathcal{D}^x_{\ell_1}(\omega)|\sin(\omega t - \phi^x) \ , \tag{5.323}$$

where $\mathcal{D}^x_{\ell_1}(\omega)$ can be written as

$$\begin{aligned}
\mathcal{D}^x_{\ell_1}(\omega) =& \sum_{\substack{\mathbf{k}_\parallel \\ \sigma}} \sum_{\substack{\ell\ell' \\ \mu\gamma\xi}} \chi^{x\sigma\ \mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E \\
=& \frac{1}{2}\sum_{\substack{\mathbf{k}_\parallel \\ \sigma}} \sum_{\substack{\ell\ell' \\ \mu\gamma\xi}} \left[\chi^{+\sigma\ \mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega) + \chi^{-\sigma\ \mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\right]\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E \\
=& \frac{1}{2}\left[\mathcal{D}^+_{\ell_1}(\omega) + \mathcal{D}^-_{\ell_1}(\omega)\right] \ .
\end{aligned} \tag{5.324}$$

and

$$\begin{aligned}
\cos\phi^x =& \frac{\mathrm{Re}[\mathcal{D}^x_{\ell_1}(\omega)]}{|\mathcal{D}^x_{\ell_1}(\omega)|} \ , \\
\sin\phi^x =& \frac{\mathrm{Im}[\mathcal{D}^x_{\ell_1}(\omega)]}{|\mathcal{D}^x_{\ell_1}(\omega)|}.
\end{aligned} \tag{5.325}$$

For y-component, we have

$$\delta\langle\hat{S}^y_{\ell_1}(t)\rangle = \frac{eE_0}{\hbar\omega}|\mathcal{D}^y_{\ell_1}(\omega)|\sin(\omega t - \phi^y) \ , \tag{5.326}$$

where $\mathcal{D}^y_{\ell_1}(\omega)$ is

$$\begin{aligned}
\mathcal{D}^y_{\ell_1}(\omega) =& \sum_{\substack{\mathbf{k}_\parallel \\ \sigma}} \sum_{\substack{\ell\ell' \\ \mu\gamma\xi}} \chi^{y\sigma\ \mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E \\
=& \frac{1}{2i}\sum_{\substack{\mathbf{k}_\parallel \\ \sigma}} \sum_{\substack{\ell\ell' \\ \mu\gamma\xi}} \left[\chi^{+\sigma\ \mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega) - \chi^{-\sigma\ \mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\right]\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E \\
=& \frac{1}{2i}\left[\mathcal{D}^+_{\ell_1}(\omega) - \mathcal{D}^-_{\ell_1}(\omega)\right] \ .
\end{aligned} \tag{5.327}$$

$$\begin{aligned}
\cos\phi^y =& \frac{\mathrm{Re}[\mathcal{D}^y_{\ell_1}(\omega)]}{|\mathcal{D}^y_{\ell_1}(\omega)|} \ , \\
\sin\phi^y =& \frac{\mathrm{Im}[\mathcal{D}^y_{\ell_1}(\omega)]}{|\mathcal{D}^y_{\ell_1}(\omega)|}.
\end{aligned} \tag{5.328}$$

Finally, the uniform z-component is

$$\delta\langle\hat{S}^z_{\ell_1}(t)\rangle = \frac{eE_0}{\hbar\omega}|\mathcal{D}^z_{\ell_1}(\omega)|\sin(\omega t - \phi^z) \ , \tag{5.329}$$

where $\mathcal{D}^z_{\ell_1}(\omega)$ is

$$
\begin{aligned}
\mathcal{D}^z_{\ell_1}(\omega) =& \sum_{\substack{\mathbf{k}_\parallel \\ \sigma}} \sum_{\substack{\ell\ell' \\ \mu\gamma\xi}} \chi^{z\sigma\ \mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \\
=& \frac{1}{2} \sum_{\substack{\mathbf{k}_\parallel \\ \sigma}} \sum_{\substack{\ell\ell' \\ \mu\gamma\xi}} \left[\chi^{\uparrow\sigma\ \mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) - \chi^{\downarrow\sigma\ \mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) \right] \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \\
=& \frac{1}{2} \left[\mathcal{D}^\uparrow_{\ell_1}(\omega) - \mathcal{D}^\downarrow_{\ell_1}(\omega) \right] \ .
\end{aligned}
\tag{5.330}
$$

where

$$
\begin{aligned}
\cos\phi^z &= \frac{\mathrm{Re}[\mathcal{D}^z_{\ell_1}(\omega)]}{|\mathcal{D}^z_{\ell_1}(\omega)|} \ , \\
\sin\phi^z &= \frac{\mathrm{Im}[\mathcal{D}^z_{\ell_1}(\omega)]}{|\mathcal{D}^z_{\ell_1}(\omega)|} .
\end{aligned}
\tag{5.331}
$$

In frequency domain, we can write the Fourier transform of Eq. (5.318) as

$$
\begin{aligned}
\delta\langle \hat{S}^m_{\ell_1}(\mathbf{q}_\parallel, \omega') \rangle =& \frac{ieE_0}{2\hbar\omega} \int dt \left[e^{i(\omega'-\omega)t} \mathcal{D}^m_{\ell_1}(\omega) - e^{i(\omega'+\omega)t} \mathcal{D}^m_{\ell_1}(-\omega) \right] \delta_{\mathbf{q}_\parallel 0} \\
=& \frac{ieE_0}{2\hbar\omega} \left[\delta(\omega'-\omega) \mathcal{D}^m_{\ell_1}(\omega) - \delta(\omega'+\omega) \mathcal{D}^m_{\ell_1}(-\omega) \right] \delta_{\mathbf{q}_\parallel 0} \\
=& \frac{ieE_0}{2\hbar\omega} \left\{ \delta(\omega'-\omega) \mathcal{D}^m_{\ell_1}(\omega) - \delta(\omega'+\omega) \left[\mathcal{D}^m_{\ell_1}(\omega) \right]^* \right\} \delta_{\mathbf{q}_\parallel 0} \ ,
\end{aligned}
\tag{5.332}
$$

where ω is the frequency of the applied field.

5.4.9 Orbital Angular Momentum Disturbance

The charge current will also perturb the orbital angular momentum through the system. This current can be obtained using the m-component of the orbital angular momentum operator written in Eq. (5.50). It is given by

$$
\begin{aligned}
\delta\langle \hat{L}^m_i \rangle(t) =& -\frac{i}{\hbar} \int dt' \Theta(t-t') \left\langle \left[\hat{L}^m_i(t), \hat{H}_{\mathrm{int}}(t') \right] \right\rangle \\
=& \frac{ieE_0}{4im\hbar\omega} \sum_{\sigma'} \sum_{\substack{k\ell \\ \gamma\xi}} \int dt' \Theta(t-t') \left[\mathbf{w}^{\gamma\xi}_{k\ell}(\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{-i\omega t'} - \mathbf{w}^{\gamma\xi}_{k\ell}(-\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{i\omega t'} \right] \left\langle \left[\hat{L}^m_i(t), c^\dagger_{k\gamma\sigma'}(t') c_{\ell\xi\sigma'}(t') \right] \right\rangle \\
=& -\frac{eE_0}{4im\omega} \sum_{\sigma\sigma'} \sum_{\substack{k\ell \\ \mu\nu\gamma\xi}} \int dt' \left[\mathbf{w}^{\gamma\xi}_{k\ell}(\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{-i\omega t'} - \mathbf{w}^{\gamma\xi}_{k\ell}(-\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{i\omega t'} \right] L^m_{\mu\nu} \chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{iik\ell}(t-t') \\
=& -\frac{eE_0}{4im\omega} \sum_{\sigma\sigma'} \sum_{\substack{k\ell \\ \mu\nu\gamma\xi}} L^m_{\mu\nu} \left[\mathbf{w}^{\gamma\xi}_{k\ell}(\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{-i\omega t} \chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{iik\ell}(\omega) - \mathbf{w}^{\gamma\xi}_{k\ell}(-\mathbf{q}) \cdot \hat{\mathbf{u}}_E e^{i\omega t} \chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{iik\ell}(-\omega) \right] \ .
\end{aligned}
\tag{5.333}
$$

For uniform electric fields ($\mathbf{q}=0$) we have

$$
\begin{aligned}
\delta\langle \hat{L}_i^m\rangle(t) =& -\frac{eE_0}{2im\omega}\sum_{\sigma\sigma'}\sum_{\substack{k\ell\\ \mu\nu\gamma\xi}} L^m_{\mu\nu}\mathbf{p}^{\gamma\xi}_{k\ell}\cdot\hat{\mathbf{u}}_E\left[e^{-i\omega t}\chi^{\sigma\sigma'}_{iik\ell}{}^{\mu\nu\gamma\xi}(\omega) - e^{i\omega t}\chi^{\sigma\sigma'}_{iik\ell}{}^{\mu\nu\gamma\xi}(-\omega)\right]\\
=& -\frac{eE_0}{2im\omega}\sum_{\sigma\sigma'}\sum_{\substack{k\ell\\ \mu\nu\gamma\xi}}\left\{e^{-i\omega t}L^m_{\mu\nu}\mathbf{p}^{\gamma\xi}_{k\ell}\chi^{\sigma\sigma'}_{iik\ell}{}^{\mu\nu\gamma\xi}(\omega) - \left[e^{-i\omega t}L^m_{\nu\mu}\mathbf{p}^{\xi\gamma}_{\ell k}\chi^{\sigma\sigma'}_{ii\ell k}{}^{\nu\mu\xi\gamma}(\omega)\right]^*\right\}\cdot\hat{\mathbf{u}}_E\\
=& -\frac{eE_0}{2im\omega}\sum_{\sigma\sigma'}\sum_{\substack{k\ell\\ \mu\nu\gamma\xi}}\left\{e^{-i\omega t}L^m_{\mu\nu}\mathbf{p}^{\gamma\xi}_{k\ell}\chi^{\sigma\sigma'}_{iik\ell}{}^{\mu\nu\gamma\xi}(\omega) - \left[e^{-i\omega t}L^m_{\mu\nu}\mathbf{p}^{\gamma\xi}_{k\ell}\chi^{\sigma\sigma'}_{iik\ell}{}^{\mu\nu\gamma\xi}(\omega)\right]^*\right\}\cdot\hat{\mathbf{u}}_E\\
=& -\frac{eE_0}{m\omega}\sum_{\sigma\sigma'}\sum_{\substack{k\ell\\ \mu\nu\gamma\xi}}\mathrm{Im}\left\{e^{-i\omega t}L^m_{\mu\nu}\mathbf{p}^{\gamma\xi}_{k\ell}\chi^{\sigma\sigma'}_{iik\ell}{}^{\mu\nu\gamma\xi}(\omega)\right\}\cdot\hat{\mathbf{u}}_E\ .
\end{aligned}
\tag{5.334}
$$

where $m=x,y,z$. In multilayers,

$$
\begin{aligned}
\delta\langle \hat{L}^m_{\ell_1}(\mathbf{q}_\parallel,t)\rangle =& -\frac{i}{\hbar}\int dt'\Theta(t-t')\left\langle\left[\hat{L}^m_{\ell_1}(\mathbf{q}_\parallel,t),\hat{H}_{\mathrm{int}}(t')\right]\right\rangle\\
=& -\frac{i}{\hbar}\frac{eE_0}{\hbar\omega}\frac{1}{N}\sum_{\mathbf{k}_\parallel,\sigma'}\sum_{\substack{\ell\ell'\\ \gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t')\left\langle\left[\hat{L}^m_{\ell_1}(\mathbf{q}_\parallel,t),c^\dagger_{\ell\gamma\sigma'}(\mathbf{k}_\parallel,t')c_{\ell'\xi\sigma'}(\mathbf{k}_\parallel,t\right.\right.\\
=& -\frac{i}{\hbar}\frac{eE_0}{2i\hbar\omega}\frac{1}{N^2}\sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right]\\
&\times L^m_{\mu\nu}\left\langle\left[S^{\sigma}_{\ell_1\ell_1}{}^{\mu\nu}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel,\mathbf{k}'_\parallel,t),c^\dagger_{\ell\gamma\sigma'}(\mathbf{k}_\parallel,t')c_{\ell'\xi\sigma'}(\mathbf{k}_\parallel,t')\right]\right\rangle\ ,
\end{aligned}
\tag{5.335}
$$

where we have used

$$
\begin{aligned}
L_\ell^m(\mathbf{q}_\parallel) &= \sum_{\mathbf{R}_\parallel} e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel} L_\ell^m(\mathbf{R}_\parallel) \\
&= \sum_{\mathbf{R}_\parallel}\sum_{\mu\nu}\sum_{\sigma} e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel} L_{\mu\nu}^m c_{\ell\mu\sigma}^\dagger(\mathbf{R}_\parallel) c_{\ell\mu\sigma}(\mathbf{R}_\parallel) \\
&= \frac{1}{N^2}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_{\mathbf{R}_\parallel}\sum_{\mu\nu}\sum_{\sigma} e^{i\mathbf{q}_\parallel\cdot\mathbf{R}_\parallel} L_{\mu\nu}^m e^{-i\mathbf{k}'_\parallel\cdot\mathbf{R}_\parallel} c_{\ell\mu\sigma}^\dagger(\mathbf{k}'_\parallel)\ e^{i\mathbf{k}_\parallel\cdot\mathbf{R}_\parallel} c_{\ell\nu\sigma}(\mathbf{k}_\parallel) \\
&= \frac{1}{N}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_{\mu\nu}\sum_{\sigma} \delta_{\mathbf{k}'_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel} L_{\mu\nu}^m c_{\ell\mu\sigma}^\dagger(\mathbf{k}'_\parallel) c_{\ell\nu\sigma}(\mathbf{k}_\parallel) \\
&= \frac{1}{N}\sum_{\mathbf{k}_\parallel}\sum_{\mu\nu}\sum_{\sigma} L_{\mu\nu}^m c_{\ell\mu\sigma}^\dagger(\mathbf{k}_\parallel+\mathbf{q}_\parallel) c_{\ell\nu\sigma}(\mathbf{k}_\parallel) \\
&= \frac{1}{N}\sum_{\mathbf{k}_\parallel}\sum_{\mu\nu}\sum_{\sigma} L_{\mu\nu}^m S_{\ell\ell}^{\sigma\,\mu\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\mathbf{k}_\parallel)\ .
\end{aligned}
\tag{5.336}
$$

Identifying the susceptibility given by Eq. (5.123),

$$
\begin{aligned}
\delta\langle \hat{L}^m_{\ell_1}(\mathbf{q}_\parallel,t)\rangle =& \frac{eE_0}{2i\hbar\omega}\frac{1}{N^2}\sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\int dt' \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right] L^m_{\mu\nu}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel,-\mathbf{k}'_\parallel,\mathbf{k}_\parallel,-\mathbf{k}_\parallel,t-\\
=& \frac{eE_0}{2i\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\int dt' \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right] L^m_{\mu\nu}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel,\mathbf{k}_\parallel,t-t')\delta_{\mathbf{k}'_\parallel,\mathbf{k}_\parallel}\delta_{\mathbf{k}'_\parallel+\\
=& -\frac{ieE_0}{2\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\int dt' \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right] L^m_{\mu\nu}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,t-t')\delta_{\mathbf{q}_\parallel 0}\\
=& -\frac{ieE_0}{2\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E L^m_{\mu\nu}\left[e^{i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,-\omega)-e^{-i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\right]\delta_{\mathbf{q}_\parallel 0}\\
=& -\frac{ieE_0}{2\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\left\{\left[e^{-i\omega t}\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\xi\gamma}_{\ell'\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E L^m_{\nu\mu}\chi^{\sigma\sigma'\,\nu\mu\xi\gamma}_{\ell_1\ell_1\ell'\ell}(\mathbf{k}_\parallel,\omega)\right]^*\right.\\
&\qquad\qquad\qquad\left. - e^{-i\omega t}\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell'\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E L^m_{\mu\nu}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\right\}\delta_{\mathbf{q}_\parallel 0}\\
=& -\frac{ieE_0}{2\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\left\{\left[e^{-i\omega t}\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E L^m_{\mu\nu}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\right]^*\right.\\
&\qquad\qquad\qquad\left. - e^{-i\omega t}\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell'\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E L^m_{\mu\nu}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\right\}\delta_{\mathbf{q}_\parallel 0}\\
=& -\frac{eE_0}{\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\mathrm{Im}\left\{e^{-i\omega t}\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell'\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E L^m_{\mu\nu}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\right\}\delta_{\mathbf{q}_\parallel 0}\ .
\end{aligned}
\tag{5.337}
$$

Defining the complex number $\mathcal{D}^{L\,m}_{\ell_1}(\omega)$ as

$$
\mathcal{D}^{L\,m}_{\ell_1}(\omega) = \sum_{\substack{\mathbf{k}_\parallel\\ \sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}} L^m_{\mu\nu}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell'\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ , \tag{5.338}
$$

we can write the orbital angular momentum disturbance as

$$
\begin{aligned}
\delta\langle \hat{L}^m_{\ell_1}(\mathbf{q}_\parallel,t)\rangle =& -\frac{eE_0}{\hbar\omega}\,\mathrm{Im}\left\{e^{-i\omega t}\mathcal{D}^{L\,m}_{\ell_1}(\omega)\right\}\delta_{\mathbf{q}_\parallel 0}\\
=& \frac{eE_0}{\hbar\omega}|\mathcal{D}^{L\,m}_{\ell_1}(\omega)|\sin\left[\omega t-\phi^m_L(\omega)\right]\delta_{\mathbf{q}_\parallel 0}\ ,
\end{aligned}
\tag{5.339}
$$

where $\phi^m_L(\omega)$ is the phase of the complex number $\mathcal{D}^{L\,m}_{\ell_1}(\omega)$.

5.4.10 Effective magnetic field

The electric field create an induced spin accumulation given by Eq. (5.321) and an orbital angular momentum accumulation given by Eq. (5.339), and we can calculate what would be the magnetic field that would create these disturbances.

To obtain the spin contribution, we take into account that the interaction of an external magnetic field with the spins is given by

$$\begin{aligned}\hat{H}_{S\perp} =& g_S\mu_B \mathbf{B}_{\text{ext}}(t)\cdot\sum_{\ell'}\hat{\mathbf{S}}_{\ell'}(t)\\ =& g_S\mu_B\sum_{\ell'}\sum_{\beta}B^{\beta\text{ext}}_{\ell'}(t)\cdot\hat{S}^{\beta}_{\ell'}(t)\ ,\end{aligned}\tag{5.340}$$

where $\beta = x, y, z$ (it can also be written in terms of $+, -, \uparrow, \downarrow$, taking into account that B^+ couples to S^- and vice-versa). This will induce a spin disturbance given by

$$\delta\langle\hat{S}^{\alpha}_{\ell}\rangle(t) = g_S\mu_B\int dt'\sum_{\ell'}\sum_{\beta}\chi^{\alpha\beta}_{\ell\ell'}(t-t')B^{\beta\text{ext}}_{\ell'}(t')\ .\tag{5.341}$$

where $\chi^{\alpha\beta}_{\ell\ell'}(t-t') = \sum_{\mu\nu}\chi^{\alpha\beta\,\mu\nu}_{\ell\ell'}(t-t')$. Multiplying by the inverse from the left,

$$\begin{aligned}\int dt\sum_{\ell}\sum_{\alpha}[\chi^{-1}]^{\gamma\alpha}_{\ell''\ell}(t''-t)\delta\langle\hat{S}^{\alpha}_{\ell}\rangle(t) =& g_S\mu_B\int dt'dt''\sum_{\ell\ell'}\sum_{\alpha\beta}[\chi^{-1}]^{\gamma\alpha}_{\ell''\ell}(t''-t)\chi^{\alpha\beta}_{\ell\ell'}(t-t')B^{\beta\text{ext}}_{\ell'}(t')\\ =& g_S\mu_B\int dt'\sum_{\ell'}\sum_{\beta}\delta_{\ell''\ell'}\delta_{\gamma\beta}\delta(t''-t')B^{\beta\text{ext}}_{\ell'}(t')\\ =& g_S\mu_B B^{\alpha\text{ext}}_{\ell''}(t'')\ .\end{aligned}\tag{5.342}$$

Therefore,

$$B^{\alpha\text{ext}}_{\ell_2}(t) = \frac{1}{g_S\mu_B}\int dt'\sum_{\ell_1}\sum_{\beta}[\chi^{-1}]^{\alpha\beta}_{\ell_2\ell_1}(t-t')\delta\langle\hat{S}^{\beta}_{\ell_1}\rangle(t')\ .\tag{5.343}$$

Using now that the spin disturbance is caused by an external electric field (see Eq. (5.321)),

$$\delta\langle\hat{S}^{\beta}_{\ell_1}(t)\rangle = \frac{ieE_0}{2\hbar\omega}\left\{e^{-i\omega t}\mathcal{D}^{\beta}_{\ell_1}(\omega) - e^{i\omega t}[\mathcal{D}^{\beta}_{\ell_1}(\omega)]^*\right\}\tag{5.344}$$

where

$$\mathcal{D}^{\beta}_{\ell_1}(\omega) = \sum_{\substack{\mathbf{k}_\parallel\\ \sigma}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\chi^{\beta\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ ,\tag{5.345}$$

we can calculate the effective field as

$$
\begin{aligned}
B_{\ell_2}^{\alpha\,\mathrm{eff}}(t) =& \frac{1}{g_S\mu_B}\frac{ieE_0}{2\hbar\omega}\int dt' \sum_{\ell_1}\sum_{\beta}[\chi^{-1}]^{\alpha\beta}_{\ell_2\ell_1}(t-t')\left\{e^{-i\omega t'}\mathcal{D}^{\beta}_{\ell_1}(\omega) - e^{i\omega t'}[\mathcal{D}^{\beta}_{\ell_1}(\omega)]^*\right\} \\
=& \frac{1}{g_S\mu_B}\frac{ieE_0}{2\hbar\omega}\int dt' \sum_{\ell_1}\sum_{\beta}[\chi^{-1}]^{\alpha\beta}_{\ell_2\ell_1}(t-t')\left\{e^{-i\omega t}e^{i\omega(t-t')}\mathcal{D}^{\beta}_{\ell_1}(\omega) - e^{i\omega t}e^{-i\omega(t-t')}[\mathcal{D}^{\beta}_{\ell_1}(\omega)]^*\right\} \\
=& \frac{1}{g_S\mu_B}\frac{ieE_0}{2\hbar\omega}\sum_{\ell_1}\sum_{\beta}\left\{e^{-i\omega t}[\chi^{-1}]^{\alpha\beta}_{\ell_2\ell_1}(\omega)\mathcal{D}^{\beta}_{\ell_1}(\omega) - e^{i\omega t}\left[[\chi^{-1}]^{\alpha\beta}_{\ell_2\ell_1}(\omega)\mathcal{D}^{\beta}_{\ell_1}(\omega)\right]^*\right\} \\
=& -\frac{1}{g_S\mu_B}\frac{eE_0}{\hbar\omega}\sum_{\ell_1}\sum_{\beta}\mathrm{Im}\left\{e^{-i\omega t}[\chi^{-1}]^{\alpha\beta}_{\ell_2\ell_1}(\omega)\mathcal{D}^{\beta}_{\ell_1}(\omega)\right\} \ .
\end{aligned}
\tag{5.346}
$$

Defining

$$
\mathcal{B}^{\alpha}_{\ell_2}(\omega) = \sum_{\ell_1}\sum_{\beta}[\chi^{-1}]^{\alpha\beta}_{\ell_2\ell_1}(\omega)\mathcal{D}^{\beta}_{\ell_1}(\omega) \ , \tag{5.347}
$$

(note that the orbital indices were already summed up in χ^{-1} and $\mathcal{D}$) we can write the effective field as

$$
\begin{aligned}
B_{\ell_2}^{\alpha\,\mathrm{eff}}(t) =& -\frac{1}{g_S\mu_B}\frac{eE_0}{\hbar\omega}\,\mathrm{Im}\left\{e^{-i\omega t}\mathcal{B}^{\alpha}_{\ell_2}(\omega)\right\} \\
=& \frac{1}{g_S\mu_B}\frac{eE_0}{\hbar\omega}|\mathcal{B}^{\alpha}_{\ell_2}(\omega)|\sin[\omega t - \varphi^{\alpha}_{\ell_2}(\omega)] \ .
\end{aligned}
\tag{5.348}
$$

where $\varphi^{\alpha}_{\ell_2}$ is the phase of the complex number written in Eq. (5.347).

5.4.11 Spin-orbit torques

The torques applied by the electric field can be obtained using the operators given in Eq. (5.16)

$$
\begin{aligned}
\delta\langle\hat{\tau}^m_i\rangle(t) =& -\frac{i}{\hbar}\int dt'\Theta(t-t')\left\langle\left[\hat{\tau}^m_i(t), \hat{H}_{\mathrm{int}}(t')\right]\right\rangle \\
=& \frac{ieE_0}{4im\hbar\omega}\sum_{\sigma'}\sum_{\substack{k\ell\\ \gamma\xi}}\int dt'\Theta(t-t')\left[\mathbf{w}^{\gamma\xi}_{k\ell}(\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{-i\omega t'} - \mathbf{w}^{\gamma\xi}_{k\ell}(-\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{i\omega t'}\right]\left\langle\left[\hat{\tau}^m_i(t), c^{\dagger}_{k\gamma\sigma'}(t')c_{\ell\xi\sigma'}(t')\right]\right\rangle \\
=& -\frac{eE_0}{4im\omega}\lambda_i\sum_{n\sigma\sigma'}\sum_{\substack{k\ell\\ \mu\nu\gamma\xi}}\int dt'\left[\mathbf{w}^{\gamma\xi}_{k\ell}(\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{-i\omega t'} - \mathbf{w}^{\gamma\xi}_{k\ell}(-\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{i\omega t'}\right]\epsilon_{mn\sigma}L'^{n}_{\mu\nu}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{iik\ell}(t-t') \\
=& -\frac{eE_0}{4im\omega}\lambda_i\sum_{n\sigma\sigma'}\sum_{\substack{k\ell\\ \mu\nu\gamma\xi}}\epsilon_{mn\sigma}L'^{n}_{\mu\nu}\left[\mathbf{w}^{\gamma\xi}_{k\ell}(\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{-i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{iik\ell}(\omega) - \mathbf{w}^{\gamma\xi}_{k\ell}(-\mathbf{q})\cdot\hat{\mathbf{u}}_E e^{i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{iik\ell}(-\omega)\right] \ .
\end{aligned}
\tag{5.349}
$$

In multilayers,

$$
\begin{aligned}
\delta\langle\hat{\tau}^m_{\ell_1}(\mathbf{q}_\parallel,t)\rangle =& -\frac{i}{\hbar}\int dt'\Theta(t-t')\left\langle\left[\hat{\tau}^m_{\ell_1}(\mathbf{q}_\parallel,t),\hat{H}_{\text{int}}(t')\right]\right\rangle \\
=& -\frac{i}{\hbar}\frac{eE_0\lambda_{\ell_1}}{\hbar\omega}\frac{1}{N^2}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_{\substack{n\sigma\sigma'\\ \mu\nu}}\sum_{\substack{\ell\ell'\\ \gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\sin(\omega t')\epsilon_{mn\sigma}L'^n_{\mu\nu} \\
&\times\left\langle\left[S^{\sigma\,\mu\nu}_{\ell_1\ell_1}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel,\mathbf{k}'_\parallel,t),c^\dagger_{\ell\gamma\sigma'}(\mathbf{k}_\parallel,t')c_{\ell'\xi\sigma'}(\mathbf{k}_\parallel,t')\right]\right\rangle \\
=& -\frac{i}{\hbar}\frac{eE_0\lambda_{\ell_1}}{2i\hbar\omega}\frac{1}{N^2}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_{\substack{n\sigma\sigma'\\ \mu\nu}}\sum_{\substack{\ell\ell'\\ \gamma\xi}}\int dt'\Theta(t-t')\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right]\epsilon_{mn\sigma}L'^n_{\mu\nu} \\
&\times\left\langle\left[S^{\sigma\,\mu\nu}_{\ell_1\ell_1}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel,\mathbf{k}'_\parallel,t),c^\dagger_{\ell\gamma\sigma'}(\mathbf{k}_\parallel,t')c_{\ell'\xi\sigma'}(\mathbf{k}_\parallel,t')\right]\right\rangle\ ,
\end{aligned}
\tag{5.350}
$$

where we have used Eq. (5.89). Identifying the susceptibility given by Eq. (5.123),

$$
\begin{aligned}
\delta\langle\hat{\tau}^m_{\ell_1}(\mathbf{q}_\parallel,t)\rangle =& \frac{eE_0\lambda_{\ell_1}}{2i\hbar\omega}\frac{1}{N^2}\sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel\\ n\sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\int dt'\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right]\epsilon_{mn\sigma}L'^n_{\mu\nu}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel,-\mathbf{k}'_\parallel,\mathbf{k}_\parallel,- \\
=& \frac{eE_0\lambda_{\ell_1}}{2i\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\mathbf{k}'_\parallel\\ n\sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\int dt'\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right]\epsilon_{mn\sigma}L'^n_{\mu\nu}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel,\mathbf{k}_\parallel,t-t')\delta_{\mathbf{k}'_\parallel,} \\
=& -\frac{ieE_0\lambda_{\ell_1}}{2\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ n\sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\int dt'\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\left[e^{i\omega t'}-e^{-i\omega t'}\right]\epsilon_{mn\sigma}L'^n_{\mu\nu}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,t-t')\delta_{\mathbf{q}_\parallel 0} \\
=& -\frac{ieE_0\lambda_{\ell_1}}{2\hbar\omega}\sum_{\substack{\mathbf{k}_\parallel\\ n\sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\epsilon_{mn\sigma}L'^n_{\mu\nu}\left[e^{i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,-\omega)-e^{-i\omega t}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\right]\delta_{\mathbf{q}} \\
=& \frac{ieE_0\lambda_{\ell_1}}{2\hbar\omega}\left[e^{-i\omega t}\mathcal{T}^m_{\ell_1}(\omega)-e^{i\omega t}\mathcal{T}^m_{\ell_1}(-\omega)\right]\delta_{\mathbf{q}_\parallel 0}\ .
\end{aligned}
\tag{5.351}
$$

where the complex number $\mathcal{T}^m_{\ell_1}$ is given by

$$
\mathcal{T}^m_{\ell_1}(\omega)=\sum_{\substack{\mathbf{k}_\parallel\\ n\sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\nu\gamma\xi}}\epsilon_{mn\sigma}L'^n_{\mu\nu}\chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ . \tag{5.352}
$$

With this definition and considering $m, \sigma, \sigma' = x, y, z$,

$$
\begin{aligned}
\mathcal{T}^m_{\ell_1}(-\omega) &= \sum_{\substack{\mathbf{k}_\parallel \\ n\sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \epsilon_{mn\sigma} L'^n_{\mu\nu} \chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, -\omega) \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \\
&= \sum_{\substack{\mathbf{k}_\parallel \\ n\sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \left[\epsilon_{mn\sigma} L'^n_{\nu\mu} \chi^{\sigma\sigma'\,\nu\mu\xi\gamma}_{\ell_1\ell_1\ell'\ell}(\mathbf{k}_\parallel, \omega) \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\xi\gamma}_{\ell'\ell}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \right]^* \\
&= \sum_{\substack{\mathbf{k}_\parallel \\ n\sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \left[\epsilon_{mn\sigma} L'^n_{\mu\nu} \chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \right]^* \\
&= [\mathcal{T}^m_{\ell_1}(\omega)]^*
\end{aligned}
\tag{5.353}
$$

Finally, we end up with

$$
\begin{aligned}
\delta\langle \hat{\tau}^m_{\ell_1}(\mathbf{q}_\parallel, t)\rangle &= \frac{ieE_0\lambda_{\ell_1}}{2\hbar\omega} \left\{ e^{-i\omega t}\mathcal{T}^m_{\ell_1}(\omega) - [e^{-i\omega t}\mathcal{T}^m_{\ell_1}(\omega)]^* \right\} \delta_{\mathbf{q}_\parallel 0} \\
&= -\frac{eE_0\lambda_{\ell_1}}{\hbar\omega} \operatorname{Im}\left\{ e^{-i\omega t}\mathcal{T}^m_{\ell_1}(\omega) \right\} \delta_{\mathbf{q}_\parallel 0} \qquad , \\
&= \frac{eE_0\lambda_{\ell_1}}{\hbar\omega} \left| \mathcal{T}^m_{\ell_1}(\omega) \right| \sin[\omega t - \phi^m_T(\omega)] \delta_{\mathbf{q}_\parallel 0}
\end{aligned}
\tag{5.354}
$$

where

$$
\mathcal{T}^m_{\ell_1}(\omega) = \sum_{\substack{\mathbf{k}_\parallel \\ n\sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \epsilon_{mn\sigma} L'^n_{\mu\nu} \chi^{\sigma\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \quad . \tag{5.355}
$$

Each of the components can be written as

$$
\begin{aligned}
\mathcal{T}^x_{\ell_1}(\omega) &= \sum_{\substack{\mathbf{k}_\parallel \\ \sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \left\{ L'^y_{\mu\nu} \chi^{z\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) - L'^z_{\mu\nu} \chi^{y\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) \right\} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \\
&= \sum_{\substack{\mathbf{k}_\parallel \\ \sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \left\{ \frac{L'^y_{\mu\nu}}{2} \left[\chi^{\uparrow\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) - \chi^{\downarrow\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) \right] - \frac{L'^z_{\mu\nu}}{2i} \left[\chi^{+\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) - \chi^{-\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) \right] \right\} \boldsymbol{\nabla}_{\mathbf{k}_\parallel}
\end{aligned}
\tag{5.356}
$$

$$
\begin{aligned}
\mathcal{T}^y_{\ell_1}(\omega) &= \sum_{\substack{\mathbf{k}_\parallel \\ \sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \left\{ L'^z_{\mu\nu} \chi^{x\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) - L'^x_{\mu\nu} \chi^{z\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) \right\} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \\
&= \sum_{\substack{\mathbf{k}_\parallel \\ \sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \left\{ \frac{L'^z_{\mu\nu}}{2} \left[\chi^{+\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) + \chi^{-\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) \right] - \frac{L'^x_{\mu\nu}}{2} \left[\chi^{\uparrow\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) - \chi^{\downarrow\sigma'\,\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \omega) \right] \right\} \boldsymbol{\nabla}_{\mathbf{k}_\parallel}
\end{aligned}
\tag{5.357}
$$

$$
\begin{aligned}
\mathcal{T}^{z}_{\ell_1}(\omega) &= \sum_{\substack{\mathbf{k}_\parallel \\ \sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \left\{ L'^{x}_{\mu\nu} \chi^{y\sigma'\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega) - L'^{y}_{\mu\nu} \chi^{x\sigma'\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega) \right\} \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \\
&= \sum_{\substack{\mathbf{k}_\parallel \\ \sigma'}} \sum_{\substack{\ell\ell' \\ \mu\nu\gamma\xi}} \left\{ \frac{L'^{x}_{\mu\nu}}{2i} \left[\chi^{+\sigma'\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega) - \chi^{-\sigma'\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega) \right] - \frac{L'^{y}_{\mu\nu}}{2} \left[\chi^{+\sigma'\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega) + \chi^{-\sigma'\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega) \right] \right\} \boldsymbol{\nabla}_{\mathbf{k}}
\end{aligned}
\tag{5.358}
$$

5.4.12 Exchange-correlation torques

The xc-torque is obtained using the operator of Eq. (5.91):

$$
\delta\langle \hat{\tau}^{m}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle = -\frac{eE_0 U_{\ell_1}}{\hbar\omega} \left|\mathcal{T}^{m}_{\ell_1}(\omega)\right| \sin[\omega t - \phi^{m}_{T}(\omega)] \delta_{\mathbf{q}_\parallel 0} \ , \tag{5.359}
$$

where

$$
\mathcal{T}^{m}_{\ell_1}(\omega) = \sum_{\substack{\mathbf{k}_\parallel \\ n\sigma\sigma'}} \sum_{\substack{\ell\ell' \\ \mu\gamma\xi}} \epsilon_{mn\sigma} \langle m^{n}_{\ell_1}\rangle \chi^{\sigma\sigma'\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega) \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E \ . \tag{5.360}
$$

Since this is basically the spin density operator, the linear response of the xc-torque can be written as

$$
\delta\langle \hat{\tau}^{m}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle = -\frac{eE_0 U_{\ell_1}}{\hbar\omega} \sum_{n\sigma} \epsilon_{mn\sigma} \langle m^{n}_{\ell_1}\rangle \left|\mathcal{T}^{\sigma}_{\ell_1}(\omega)\right| \sin[\omega t - \phi^{m}_{T}(\omega)] \delta_{\mathbf{q}_\parallel 0} \ , \tag{5.361}
$$

where

$$
\mathcal{T}^{\sigma}_{\ell_1}(\omega) = \sum_{\substack{\mathbf{k}_\parallel \\ \sigma'}} \sum_{\substack{\ell\ell' \\ \mu\gamma\xi}} \chi^{\sigma\sigma'\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega) \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E = \mathcal{D}^{\sigma}_{\ell_1}(\omega) \ . \tag{5.362}
$$

$\mathcal{D}^{\sigma}_{\ell_1}(\omega)$ was obtained in the spin disturbance calculation and is given by Eq. (5.322). Therefore, we have

$$
\delta\langle \hat{\tau}^{m}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle = -U_{\ell_1} \sum_{n\sigma} \epsilon_{mn\sigma} \langle m^{n}_{\ell_1}\rangle \delta\langle \hat{S}^{\sigma}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle \ . \tag{5.363}
$$

The components can be written as

$$
\begin{aligned}
\delta\langle \hat{\tau}^{x}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle &= -U_{\ell_1} \left\{ \langle m^{y}_{\ell_1}\rangle \delta\langle \hat{S}^{z}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle - \langle m^{z}_{\ell_1}\rangle \delta\langle \hat{S}^{y}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle \right\} \\
\delta\langle \hat{\tau}^{y}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle &= -U_{\ell_1} \left\{ \langle m^{z}_{\ell_1}\rangle \delta\langle \hat{S}^{x}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle - \langle m^{x}_{\ell_1}\rangle \delta\langle \hat{S}^{z}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle \right\} \ . \\
\delta\langle \hat{\tau}^{z}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle &= -U_{\ell_1} \left\{ \langle m^{x}_{\ell_1}\rangle \delta\langle \hat{S}^{y}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle - \langle m^{y}_{\ell_1}\rangle \delta\langle \hat{S}^{x}_{\ell_1}(\mathbf{q}_\parallel,t)\rangle \right\}
\end{aligned}
\tag{5.364}
$$

5.4.13 External torques

The torque due to the external static field can be calculated using the operator of Eq. (5.20):

$$\delta\langle\hat{\tau}^m_{\ell_1}(\mathbf{q}_\parallel,t)\rangle = \frac{eE_0}{\hbar\omega}\left|\mathcal{T}^m_{\ell_1}(\omega)\right|\sin[\omega t-\phi^m_T(\omega)]\delta_{\mathbf{q}_\parallel 0}\ , \tag{5.365}$$

where

$$\mathcal{T}^m_{\ell_1}(\omega) = \sum_{\substack{\mathbf{k}_\parallel\\ n\sigma\sigma'}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\epsilon_{mn\sigma}\hbar\omega_n\chi^{\sigma\sigma'\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\ . \tag{5.366}$$

Once again, this can be related to the spin disturbance identifying

$$\delta\langle\hat{\tau}^m_{\ell_1}(\mathbf{q}_\parallel,t)\rangle = \frac{eE_0}{\hbar\omega}\sum_{n\sigma}\epsilon_{mn\sigma}\hbar\omega_n\left|\mathcal{T}^\sigma_{\ell_1}(\omega)\right|\sin[\omega t-\phi^m_T(\omega)]\delta_{\mathbf{q}_\parallel 0}\ , \tag{5.367}$$

where

$$\mathcal{T}^\sigma_{\ell_1}(\omega) = \sum_{\substack{\mathbf{k}_\parallel\\ \sigma'}}\sum_{\substack{\ell\ell'\\ \mu\gamma\xi}}\chi^{\sigma\sigma'\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E = \mathcal{D}^\sigma_{\ell_1}(\omega)\ . \tag{5.368}$$

$\mathcal{D}^\sigma_{\ell_1}(\omega)$ was obtained in the spin disturbance calculation and is given by Eq. (5.322). Therefore, we have

$$\begin{aligned}\delta\langle\hat{\tau}^m_{\ell_1}(\mathbf{q}_\parallel,t)\rangle &= \sum_{n\sigma}\epsilon_{mn\sigma}\hbar\omega_n\delta\langle\hat{S}^\sigma_{\ell_1}(\mathbf{q}_\parallel,t)\rangle\\ &= g_S\mu_B\sum_{n\sigma}\epsilon_{mn\sigma}B'^n\delta\langle\hat{S}^\sigma_{\ell_1}(\mathbf{q}_\parallel,t)\rangle\end{aligned}\ , \tag{5.369}$$

which can be seen as the torque between the external field and the induced magnetization. The components can be written as

$$\begin{aligned}\delta\langle\hat{\tau}^x_{\ell_1}(\mathbf{q}_\parallel,t)\rangle &= \hbar\omega_y\delta\langle\hat{S}^z_{\ell_1}(\mathbf{q}_\parallel,t)\rangle - \hbar\omega_z\delta\langle\hat{S}^y_{\ell_1}(\mathbf{q}_\parallel,t)\rangle\\ \delta\langle\hat{\tau}^y_{\ell_1}(\mathbf{q}_\parallel,t)\rangle &= \hbar\omega_z\delta\langle\hat{S}^x_{\ell_1}(\mathbf{q}_\parallel,t)\rangle - \hbar\omega_x\delta\langle\hat{S}^z_{\ell_1}(\mathbf{q}_\parallel,t)\rangle\\ \delta\langle\hat{\tau}^z_{\ell_1}(\mathbf{q}_\parallel,t)\rangle &= \hbar\omega_x\delta\langle\hat{S}^y_{\ell_1}(\mathbf{q}_\parallel,t)\rangle - \hbar\omega_y\delta\langle\hat{S}^x_{\ell_1}(\mathbf{q}_\parallel,t)\rangle\end{aligned}\ . \tag{5.370}$$

$$t_{ij}^{\mu\nu} = t_{ji}^{\nu\mu}$$

Appendix A

Momentum operator in terms of hopping integrals

To calculate $\mathbf{p}_{k\ell}^{\gamma\xi}(\mathbf{q}_{\parallel})$, consider the commutator

$$\begin{aligned}
\left[\hat{H}_0, e^{i\mathbf{q}\cdot\hat{\mathbf{r}}}\right] &= \left[\frac{\hat{p}^2}{2m} + U(\hat{\mathbf{r}}), e^{i\mathbf{q}\cdot\hat{\mathbf{r}}}\right] \\
&= \left[\frac{\hat{p}^2}{2m}, e^{i\mathbf{q}\cdot\hat{\mathbf{r}}}\right] \\
&= \frac{1}{2m}\sum_{\alpha}\left[\hat{p}^{\alpha}\hat{p}^{\alpha}, e^{i\mathbf{q}\cdot\hat{\mathbf{r}}}\right] \\
&= \frac{1}{2m}\sum_{\alpha}\left\{\hat{p}^{\alpha}\left[\hat{p}^{\alpha}, e^{i\mathbf{q}\cdot\hat{\mathbf{r}}}\right] + \left[\hat{p}^{\alpha}, e^{i\mathbf{q}\cdot\hat{\mathbf{r}}}\right]\hat{p}^{\alpha}\right\} \\
&= \frac{1}{2m}\sum_{\alpha}\left\{\hat{p}^{\alpha}\frac{\hbar}{i}iq^{\alpha}e^{i\mathbf{q}\cdot\hat{\mathbf{r}}} + \frac{\hbar}{i}iq^{\alpha}e^{i\mathbf{q}\cdot\hat{\mathbf{r}}}\hat{p}^{\alpha}\right\} \\
&= \frac{\hbar}{2m}\sum_{\alpha}\left\{\hat{p}^{\alpha}q^{\alpha}e^{i\mathbf{q}\cdot\hat{\mathbf{r}}} + e^{i\mathbf{q}\cdot\hat{\mathbf{r}}}q^{\alpha}\hat{p}^{\alpha}\right\} \\
&= \frac{\hbar}{2m}\left\{\hat{\mathbf{p}}\cdot\mathbf{q}e^{i\mathbf{q}\cdot\hat{\mathbf{r}}} + e^{i\mathbf{q}\cdot\hat{\mathbf{r}}}\hat{\mathbf{p}}\cdot\mathbf{q}\right\} \ .
\end{aligned} \tag{A.1}$$

where we have used $[\hat{p}^{\alpha}, F(r^{\alpha})] = \frac{\hbar}{i}\frac{\partial F(r^{\alpha})}{\partial r^{\alpha}}$. If we take $\mathbf{q} = q^{\alpha}\hat{\mathbf{e}}^{\alpha}$, where $\alpha = x, y$ or z, we obtain

$$\lim_{q^{\alpha}\to 0}\frac{1}{q^{\alpha}}\left[\hat{H}_0, e^{iq^{\alpha}\hat{r}^{\alpha}}\right] = \frac{\hbar}{m}\hat{p}^{\alpha} \ . \tag{A.2}$$

Within tight-binding, we have

$$\hat{H}_0 = \sum_{\ell\ell'}\sum_{\mu\nu}\sum_{\mathbf{k}_{\parallel}}\sum_{\sigma} t_{\ell\ell'}^{\mu\nu}(\mathbf{k}_{\parallel})c_{\ell\mu\sigma}^{\dagger}(\mathbf{k}_{\parallel})c_{\ell'\nu\sigma}(\mathbf{k}_{\parallel}) \ . \tag{A.3}$$

In second quantization,

$$e^{i\mathbf{q}_\parallel\cdot\hat{\mathbf{r}}_\parallel} = \sum_{\ell\ell'}\sum_{\mu\nu}\sum_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\sum_{\sigma\sigma'}\langle\ell\mu\mathbf{k}_\parallel\sigma|e^{i\mathbf{q}_\parallel\cdot\hat{\mathbf{r}}_\parallel}|\ell'\nu\mathbf{k}'_\parallel\sigma'\rangle c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel)c_{\ell'\nu\sigma'}(\mathbf{k}'_\parallel)\ . \tag{A.4}$$

But

$$\begin{aligned}\langle\ell\mu\mathbf{k}_\parallel\sigma|e^{i\mathbf{q}_\parallel\cdot\hat{\mathbf{r}}_\parallel}|\ell'\nu\mathbf{k}'_\parallel\sigma'\rangle =&\langle\ell\mu\mathbf{k}_\parallel\sigma|\ell\nu\mathbf{k}'_\parallel+\mathbf{q}_\parallel\sigma'\rangle\\ =&\delta_{\ell\ell'}\delta_{\mu\nu}\delta_{\mathbf{k}_\parallel\mathbf{k}'_\parallel+\mathbf{q}_\parallel}\delta_{\sigma\sigma'}\ .\end{aligned} \tag{A.5}$$

So

$$e^{i\mathbf{q}_\parallel\cdot\hat{\mathbf{r}}_\parallel} = \sum_{\ell\mu\mathbf{k}_\parallel\sigma} c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)c_{\ell\mu\sigma}(\mathbf{k}_\parallel)\ . \tag{A.6}$$

Now we need to calculate the commutator

$$\left[\hat{H}_0, e^{i\mathbf{q}_\parallel\cdot\hat{\mathbf{r}}_\parallel}\right] = \sum_{\ell\ell'}\sum_{\mu\nu}\sum_{\mathbf{k}_\parallel}\sum_{\sigma} t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel)\sum_{m\mu'\mathbf{k}'_\parallel\sigma'}\left[c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel)c_{\ell'\nu\sigma}(\mathbf{k}_\parallel), c^\dagger_{m\mu'\sigma'}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel)c_{m\mu'\sigma'}(\mathbf{k}'_\parallel)\right]\ . \tag{A.7}$$

Since

$$\begin{aligned}\left[c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel)c_{\ell'\nu\sigma}(\mathbf{k}_\parallel), c^\dagger_{m\mu'\sigma'}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel)c_{m\mu'\sigma'}(\mathbf{k}'_\parallel)\right] =&c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel)c_{m\mu'\sigma'}(\mathbf{k}'_\parallel)\delta_{\ell'm}\delta_{\nu\mu'}\delta_{\sigma\sigma'}\delta_{\mathbf{k}_\parallel\mathbf{k}'_\parallel+\mathbf{q}_\parallel}\\ &- c^\dagger_{m\mu'\sigma'}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel)c_{\ell'\nu\sigma}(\mathbf{k}_\parallel)\delta_{\ell m}\delta_{\mu\mu'}\delta_{\sigma\sigma'}\delta_{\mathbf{k}_\parallel\mathbf{k}'_\parallel}\ ,\end{aligned} \tag{A.8}$$

we obtain

$$\left[\hat{H}_0, e^{i\mathbf{q}_\parallel\cdot\hat{\mathbf{r}}_\parallel}\right] = \sum_{\ell\ell'}\sum_{\mu\nu}\sum_{\mathbf{k}'_\parallel}\sum_{\sigma}\left\{t^{\mu\nu}_{\ell\ell'}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel) - t^{\mu\nu}_{\ell\ell'}(\mathbf{k}'_\parallel)\right\}c^\dagger_{\ell\mu\sigma}(\mathbf{k}'_\parallel+\mathbf{q}_\parallel)c_{\ell'\nu\sigma}(\mathbf{k}'_\parallel)\ . \tag{A.9}$$

Finally,

$$\begin{aligned}\frac{\hbar}{m}\hat{p}^\alpha =& \lim_{q^\alpha\to 0}\frac{1}{q^\alpha}\left[\hat{H}_0, e^{iq^\alpha\hat{r}^\alpha}\right]\\ =& \lim_{q^\alpha\to 0}\sum_{\substack{\ell\ell'\\ \mu\nu}}\sum_{\mathbf{k}_\parallel\sigma}\frac{t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel+q^\alpha\hat{\mathbf{e}}^\alpha) - t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel)}{q^\alpha}c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel+q^\alpha\hat{\mathbf{e}}^\alpha)c_{\ell'\nu\sigma}(\mathbf{k}_\parallel)\\ =&\sum_{\substack{\ell\ell'\\ \mu\nu}}\sum_{\mathbf{k}_\parallel\sigma}\frac{\partial t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel)}{\partial k^\alpha}c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel)c_{\ell'\nu\sigma}(\mathbf{k}_\parallel)\ ,\end{aligned} \tag{A.10}$$

or

$$\frac{\hbar}{m}\hat{p}^\alpha = \sum_{\substack{\ell\ell'\\ \mu\nu}}\sum_{\mathbf{k}_\parallel\sigma}\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{e}}^\alpha c^\dagger_{\ell\mu\sigma}(\mathbf{k}_\parallel)c_{\ell'\nu\sigma}(\mathbf{k}_\parallel)\ . \tag{A.11}$$

Projecting on the basis:

$$\frac{\hbar}{m}p^{\alpha\,\gamma\xi}_{k\ell}(\mathbf{q}_\parallel) = \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{k\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{e}}^{\alpha}\Big|_{\mathbf{k}_\parallel=\mathbf{q}_\parallel} \quad , \tag{A.12}$$

or

$$\frac{\hbar}{m}\mathbf{p}^{\gamma\xi}_{k\ell}(\mathbf{q}_\parallel) = \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{k\ell}(\mathbf{k}_\parallel)\Big|_{\mathbf{k}_\parallel=\mathbf{q}_\parallel} \quad . \tag{A.13}$$

The derivative of the hopping can be calculated using Eq. (5.80), as

$$\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\mu\nu}_{ij}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{e}}^{\alpha} = \sum_{\mathbf{R}_\parallel} i(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)\cdot\hat{\mathbf{e}}^{\alpha} e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)} t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel) \tag{A.14}$$

$$\frac{\hbar}{m}\mathbf{p}(\mathbf{k}_\parallel) = \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t(\mathbf{k}_\parallel) \ . \tag{A.15}$$

$$\begin{aligned}
\frac{1}{N}\sum_{\mathbf{k}_\parallel}\frac{p^2(\mathbf{k}_\parallel)}{2m} =& \frac{1}{N}\sum_{\mathbf{k}_\parallel}\frac{m}{2\hbar^2}\left\{\sum_{\mathbf{R}'_\parallel} i(\mathbf{R}'_\parallel)e^{i\mathbf{k}_\parallel\cdot\mathbf{R}'_\parallel}\underline{t}(\mathbf{R}'_\parallel)\right\}\left\{\sum_{\mathbf{R}_\parallel} i(-\mathbf{R}_\parallel)e^{-i\mathbf{k}_\parallel\cdot\mathbf{R}_\parallel}\underline{t}(-\mathbf{R}_\parallel)\right\}\\
=& -\frac{1}{N}\sum_{\mathbf{k}_\parallel}\frac{m}{2\hbar^2}\sum_{\mathbf{R}_\parallel,\mathbf{R}'_\parallel} e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)}\left\{(\mathbf{R}'_\parallel)\underline{t}(\mathbf{R}'_\parallel)\right\}\left\{(-\mathbf{R}_\parallel)\underline{t}(-\mathbf{R}_\parallel)\right\}\\
=& -\frac{m}{2\hbar^2}\sum_{\mathbf{R}_\parallel}\left\{(\mathbf{R}_\parallel)\underline{t}(\mathbf{R}_\parallel)\right\}\left\{(-\mathbf{R}_\parallel)\underline{t}^{T}(\mathbf{R}_\parallel)\right\}\\
=& \frac{m}{2\hbar^2}\sum_{\mathbf{R}_\parallel}\left\{\mathbf{R}^2_\parallel\underline{t}(\mathbf{R}_\parallel)\underline{t}^{T}(\mathbf{R}_\parallel)\right\} \ .
\end{aligned} \tag{A.16}$$

Appendix B

SHE for small frequencies

The results we have obtained for the spin Hall effect involves a $\frac{1}{\omega}$ pre-factor. When $\omega = 0$ we should obtain the static SHE, which is the one usually measured in experiments. It is then convenient to expand the results for $\omega \to 0$. For example, the spin disturbance obtained in Eq. (5.318) is given by

$$\delta\langle \hat{S}^{m}_{\ell_1}(t)\rangle = \frac{ie}{m\omega}\sum_{\substack{\mathbf{k}_\parallel \\ \sigma}}\sum_{\substack{\ell\ell' \\ \mu\gamma\xi}}\sum_{\alpha=1}^{3}\frac{\partial t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)}{\partial k_\alpha}E^{\alpha}_0 e^{-i\omega t}\chi^{m\sigma}_{\ell_1\ell_1\ell\ell'}{}^{\mu\mu\gamma\xi}(\mathbf{k}_\parallel,\omega) \ . \tag{B.1}$$

The most general way we can write the susceptibilities is

$$\begin{aligned}\chi^{mnrs}_{(0)ijkl}{}^{\mu\nu\gamma\xi}(\omega) = \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\Big(&G^{nr}_{jk}{}^{\nu\gamma}(\omega'+\omega)\left\{G^{sm}_{li}{}^{\xi\mu}(\omega') - \left[G^{ms}_{il}{}^{\mu\xi}(\omega')\right]^*\right\} \\ &+ \left[G^{ms}_{il}{}^{\mu\xi}(\omega'-\omega)\right]^*\left\{G^{nr}_{jk}{}^{\nu\gamma}(\omega') - \left[G^{rn}_{kj}{}^{\gamma\nu}(\omega')\right]^*\right\}\Big) \ ,\end{aligned} \tag{B.2}$$

where the spin indices m, n are related to the first spin index from the susceptibilities we have written before, and r, s are related to the second spin index. For $\omega \to 0$, we have

$$G^{nr}_{jk}{}^{\nu\gamma}(\omega'+\omega) \simeq G^{nr}_{jk}{}^{\nu\gamma}(\omega') + \omega\frac{dG^{nr}_{jk}{}^{\nu\gamma}(\omega')}{d\omega'} \ . \tag{B.3}$$

So

$$\begin{aligned}\chi^{mnrs}_{(0)ijkl}{}^{\mu\nu\gamma\xi}(\omega) = \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\Bigg(&\left[G^{nr}_{jk}{}^{\nu\gamma}(\omega') + \omega\frac{dG^{nr}_{jk}{}^{\nu\gamma}(\omega')}{d\omega'}\right]\left\{G^{sm}_{li}{}^{\xi\mu}(\omega') - \left[G^{ms}_{il}{}^{\mu\xi}(\omega')\right]^*\right\} \\ &+ \left[G^{ms}_{il}{}^{\mu\xi}(\omega') - \omega\frac{dG^{ms}_{il}{}^{\mu\xi}(\omega')}{d\omega'}\right]^*\left\{G^{nr}_{jk}{}^{\nu\gamma}(\omega') - \left[G^{rn}_{kj}{}^{\gamma\nu}(\omega')\right]^*\right\}\Bigg) \ ,\end{aligned} \tag{B.4}$$

The zeroth order terms in ω are

$$
\begin{aligned}
\chi_{(0)ijkl}^{\;\;mnrs\;\;\mu\nu\gamma\xi}(\omega) &= \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\left(G_{jk}^{nr\;\nu\gamma}(\omega')\left\{G_{li}^{sm\;\xi\mu}(\omega') - \left[G_{il}^{ms\;\mu\xi}(\omega')\right]^*\right\}\right. \\
&\qquad\left. + \left[G_{il}^{ms\;\mu\xi}(\omega')\right]^*\left\{G_{jk}^{nr\;\nu\gamma}(\omega') - \left[G_{kj}^{rn\;\gamma\nu}(\omega')\right]^*\right\}\right) \\
&= \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\left(G_{jk}^{nr\;\nu\gamma}(\omega')G_{li}^{sm\;\xi\mu}(\omega') - \left[G_{il}^{ms\;\mu\xi}(\omega')\right]^*\left[G_{kj}^{rn\;\gamma\nu}(\omega')\right]^*\right) \\
&= \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\left\{G_{jk}^{nr\;\nu\gamma}(\omega')G_{li}^{sm\;\xi\mu}(\omega') - G_{li}^{-sm\;\xi\mu}(\omega')G_{jk}^{-nr\;\nu\gamma}(\omega')\right\}\;,
\end{aligned}
\tag{B.5}
$$

The relation between HF and RPA susceptibilities (for the one we need) is

$$
\chi_{iikl}^{\;mnrs\;\mu\mu\gamma\xi}(\omega) = \chi_{(0)iikl}^{\;mnrs\;\mu\mu\gamma\xi}(\omega) + \chi_{(0)iijj}^{\;mnpq\;\mu\mu\alpha\beta}(\omega)U_j^{pqvw\;\alpha\beta\lambda\rho}\chi_{jjkl}^{\;vwrs\;\lambda\rho\gamma\xi}(\omega)\;, \tag{B.6}
$$

Using Lowde-Windsor, we substitute Eq. (3.183). Thus

$$
\chi_{iikl}^{\;mnrs\;\mu\mu\gamma\xi}(\omega) = \chi_{(0)iikl}^{\;mnrs\;\mu\mu\gamma\xi}(\omega) + \chi_{(0)iijj}^{\;mnpq\;\mu\mu\alpha\beta}(\omega)U_j^{pqvw}\chi_{jjkl}^{\;vwrs\;\beta\alpha\gamma\xi}(\omega)\;, \tag{B.7}
$$

Appendix C

Hartree-Fock Susceptibility Matrix

We need to calculate the susceptibility matrix within Hartree-Fock approximation, given by Eq. (3.179). We can write the generalized susceptibility in time domain as

$$\chi_{(0)ijk\ell}^{\alpha\beta\;\mu\nu\gamma\xi}(t) = -\frac{i}{\hbar}\Theta(t)\langle[S_{ij}^{\alpha\,\mu\nu}(t), S_{k\ell}^{\beta\,\gamma\xi}(0)]\rangle \ , \tag{C.1}$$

The commutator is given by

$$[S_{ij}^{\alpha\,\mu\nu}(t), S_{k\ell}^{\beta\,\gamma\xi}(0)] = \frac{1}{4}\left\{c_{i\mu s_1}^{\dagger}(t)\sigma_{s_1s_2}^{\alpha}c_{j\nu s_2}(t)c_{k\gamma s_3}^{\dagger}\sigma_{s_3s_4}^{\beta}c_{\ell\xi s_4} - c_{k\gamma s_3}^{\dagger}\sigma_{s_3s_4}^{\beta}c_{\ell\xi s_4}c_{i\mu s_1}^{\dagger}(t)\sigma_{s_1s_2}^{\alpha}c_{j\nu s_2}(t)\right\} \ . \tag{C.2}$$

Using Wick's theorem, we can substitute the product of 4 operators by

$$c_1^{\dagger}(t)c_2(t)c_3^{\dagger}c_4 \rightarrow \langle c_1^{\dagger}(t)c_2(t)\rangle c_3^{\dagger}c_4 + \langle c_1^{\dagger}(t)c_4\rangle c_2(t)c_3^{\dagger} + \langle c_3^{\dagger}c_4\rangle c_1^{\dagger}(t)c_2(t) + \langle c_2(t)c_3^{\dagger}\rangle c_1^{\dagger}(t)c_4 \ . \tag{C.3}$$

The second term is

$$c_3^{\dagger}c_4c_1^{\dagger}(t)c_2(t) \rightarrow \langle c_3^{\dagger}c_4\rangle c_1^{\dagger}(t)c_2(t) + \langle c_3^{\dagger}c_2(t)\rangle c_4c_1^{\dagger}(t) + \langle c_1^{\dagger}(t)c_2(t)\rangle c_3^{\dagger}c_4 + \langle c_4c_1^{\dagger}(t)\rangle c_3^{\dagger}c_2(t) \ . \tag{C.4}$$

The expectation value of the first and third terms cancel and we the expectation value of the commutator give

$$\begin{aligned}\langle[S_{ij}^{\alpha\,\mu\nu}(t), S_{k\ell}^{\beta\,\gamma\xi}(0)]\rangle = \frac{1}{4}\Big\{&\langle c_{i\mu s_1}^{\dagger}(t)c_{\ell\xi s_4}\rangle\sigma_{s_1s_2}^{\alpha}\langle c_{j\nu s_2}(t)c_{k\gamma s_3}^{\dagger}\rangle\sigma_{s_3s_4}^{\beta} \\ &-\langle c_{k\gamma s_3}^{\dagger}c_{j\nu s_2}(t)\rangle\sigma_{s_3s_4}^{\beta}\langle c_{\ell\xi s_4}c_{i\mu s_1}^{\dagger}(t)\rangle\sigma_{s_1s_2}^{\alpha}\Big\} \ .\end{aligned} \tag{C.5}$$

Adding and subtracting the term $\langle c^{\dagger}_{i\mu s_1}(t)c_{\ell\xi s_4}\rangle\sigma^{\alpha}_{s_1 s_2}\langle c^{\dagger}_{k\gamma s_3}c_{j\nu s_2}(t)\rangle\sigma^{\beta}_{s_3 s_4}$, we have

$$
\begin{aligned}
4\langle[S_{ij}^{\alpha\,\mu\nu}(t),S_{k\ell}^{\beta\,\gamma\xi}(0)]\rangle =&\langle c^{\dagger}_{i\mu s_1}(t)c_{\ell\xi s_4}\rangle\sigma^{\alpha}_{s_1 s_2}\langle c_{j\nu s_2}(t)c^{\dagger}_{k\gamma s_3}\rangle\sigma^{\beta}_{s_3 s_4}\\
&+\langle c^{\dagger}_{i\mu s_1}(t)c_{\ell\xi s_4}\rangle\sigma^{\alpha}_{s_1 s_2}\langle c^{\dagger}_{k\gamma s_3}c_{j\nu s_2}(t)\rangle\sigma^{\beta}_{s_3 s_4}\\
&-\langle c^{\dagger}_{i\mu s_1}(t)c_{\ell\xi s_4}\rangle\sigma^{\alpha}_{s_1 s_2}\langle c^{\dagger}_{k\gamma s_3}c_{j\nu s_2}(t)\rangle\sigma^{\beta}_{s_3 s_4}\\
&-\langle c^{\dagger}_{k\gamma s_3}c_{j\nu s_2}(t)\rangle\sigma^{\beta}_{s_3 s_4}\langle c_{\ell\xi s_4}c^{\dagger}_{i\mu s_1}(t)\rangle\sigma^{\alpha}_{s_1 s_2}\\
=&\langle c^{\dagger}_{i\mu s_1}(t)c_{\ell\xi s_4}\rangle\sigma^{\alpha}_{s_1 s_2}\langle\{c_{j\nu s_2}(t),c^{\dagger}_{k\gamma s_3}\}\rangle\sigma^{\beta}_{s_3 s_4}\\
&-\langle c^{\dagger}_{k\gamma s_3}c_{j\nu s_2}(t)\rangle\sigma^{\beta}_{s_3 s_4}\langle\{c^{\dagger}_{i\mu s_1}(t),c_{\ell\xi s_4}\}\rangle\sigma^{\alpha}_{s_1 s_2}\ .
\end{aligned}
\tag{C.6}
$$

Identifying the monoeletronic retarded Green function

$$
G_{jk}^{s_2 s_3\,\nu\gamma}(t)=-\frac{i}{\hbar}\Theta(t)\langle\{c_{j\nu s_2}(t),c^{\dagger}_{k\gamma s_3}\}\rangle \tag{C.7}
$$

and it's conjugate

$$
\begin{aligned}
[G_{i\ell}^{s_1 s_4\,\mu\xi}(t)]^{*}=&\frac{i}{\hbar}\Theta(t)\langle\{c^{\dagger}_{i\mu s_1}(t),c_{\ell\xi s_4}\}\rangle\\
=&G_{\ell i}^{-s_4 s_1\,\xi\mu}(-t)\ ,
\end{aligned}
\tag{C.8}
$$

where G^{-} denotes the advanced Green function, we can write

$$
\chi_{(0)\,ijk\ell}^{\alpha\beta\,\mu\nu\gamma\xi}(t)=\frac{1}{4}\left\{\langle c^{\dagger}_{i\mu s_1}(t)c_{\ell\xi s_4}\rangle\sigma^{\alpha}_{s_1 s_2}G_{jk}^{s_2 s_3\,\nu\gamma}(t)\sigma^{\beta}_{s_3 s_4}+\langle c^{\dagger}_{k\gamma s_3}c_{j\nu s_2}(t)\rangle\sigma^{\beta}_{s_3 s_4}G_{\ell i}^{-s_4 s_1\,\xi\mu}(-t)\sigma^{\alpha}_{s_1 s_2}\right\}\ . \tag{C.9}
$$

We can use Eq. (1.54) to obtain

$$
\langle\hat{c}^{\dagger}_{j\nu\sigma'}\hat{c}_{i\mu\sigma}(t)\rangle=-\frac{\hbar}{\pi}\int d\omega e^{-i\omega t}f(\omega)\Im G_{ij}^{\sigma\sigma'\,\mu\nu}(\omega)\ . \tag{C.10}
$$

and

$$
\langle\hat{c}^{\dagger}_{j\nu\sigma'}(t)\hat{c}_{i\mu\sigma}\rangle=-\frac{\hbar}{\pi}\int d\omega e^{i\omega t}f(\omega)\Im G_{ij}^{\sigma\sigma'\,\mu\nu}(\omega)\ . \tag{C.11}
$$

where

$$
\Im G_{ij}^{\sigma\sigma'\,\mu\nu}(\omega)=\frac{1}{2i}\left[G_{ij}^{\sigma\sigma'\,\mu\nu}(\omega+i\eta)-G_{ij}^{\sigma\sigma'\,\mu\nu}(\omega-i\eta)\right]\ . \tag{C.12}
$$

Substituting back in the susceptibility and reorganising the terms,

$$
\begin{aligned}
\chi_{(0)\,ijk\ell}^{\alpha\beta\,\mu\nu\gamma\xi}(t)=-\frac{\hbar}{4\pi}\int d\omega' f(\omega')\Big\{&e^{i\omega' t}\sigma^{\alpha}_{s_1 s_2}G_{jk}^{s_2 s_3\,\nu\gamma}(t)\sigma^{\beta}_{s_3 s_4}\Im G_{\ell i}^{s_4 s_1\,\xi\mu}(\omega')\\
&+e^{-i\omega' t}\sigma^{\alpha}_{s_1 s_2}\Im G_{jk}^{s_2 s_3\,\nu\gamma}(\omega')\sigma^{\beta}_{s_3 s_4}G_{\ell i}^{-s_4 s_1\,\xi\mu}(-t)\Big\}\ ,
\end{aligned}
\tag{C.13}
$$

The Fourier transform (given by Eq. (5.113)) of the susceptibility is,

$$
\begin{aligned}
\chi^{\alpha\beta}_{(0)ijk\ell}{}^{\mu\nu\gamma\xi}(\omega) &= \int dt e^{i\omega t}\chi^{\alpha\beta}_{(0)ijk\ell}{}^{\mu\nu\gamma\xi}(t)\\
&= -\frac{\hbar}{4\pi}\int d\omega' f(\omega')\left\{\sigma^{\alpha}_{s_1s_2}\int dt e^{i(\omega+\omega')t}G^{s_2s_3}_{jk}{}^{\nu\gamma}(t)\sigma^{\beta}_{s_3s_4}\Im G^{s_4s_1}_{\ell i}{}^{\xi\mu}(\omega')\right.\\
&\qquad\left.+\sigma^{\alpha}_{s_1s_2}\Im G^{s_2s_3}_{jk}{}^{\nu\gamma}(\omega')\sigma^{\beta}_{s_3s_4}\int dt e^{i(\omega-\omega')t}G^{-s_4s_1}_{\ell i}{}^{\xi\mu}(-t)\right\}\\
&= -\frac{\hbar}{4\pi}\int d\omega' f(\omega')\left\{\sigma^{\alpha}_{s_1s_2}G^{s_2s_3}_{jk}{}^{\nu\gamma}(\omega'+\omega+i\eta)\sigma^{\beta}_{s_3s_4}\Im G^{s_4s_1}_{\ell i}{}^{\xi\mu}(\omega')\right.\\
&\qquad\left.+\sigma^{\alpha}_{s_1s_2}\Im G^{s_2s_3}_{jk}{}^{\nu\gamma}(\omega')\sigma^{\beta}_{s_3s_4}G^{s_4s_1}_{\ell i}{}^{\xi\mu}(\omega'-\omega-i\eta)\right\} \ .
\end{aligned}
\tag{C.14}
$$

Finally, we can write this using 2×2 matrices in the spin space as

$$
\begin{aligned}
\chi^{\alpha\beta}_{(0)ijk\ell}{}^{\mu\nu\gamma\xi}(\omega) &= -\frac{\hbar}{4\pi}\int d\omega' f(\omega')\mathrm{Tr}\left\{\sigma^{\alpha}G^{\nu\gamma}_{jk}(\omega'+\omega+i\eta)\sigma^{\beta}\Im G^{\xi\mu}_{\ell i}(\omega')+\sigma^{\alpha}\Im G^{\nu\gamma}_{jk}(\omega')\sigma^{\beta}G^{\xi\mu}_{\ell i}(\omega'-\omega-i\eta)\right\}\\
&= -\frac{\hbar}{8i\pi}\int d\omega' f(\omega')\mathrm{Tr}\left\{\sigma^{\alpha}G^{\nu\gamma}_{jk}(\omega'+\omega+i\eta)\sigma^{\beta}\left[G^{\xi\mu}_{\ell i}(\omega'+i\eta)-G^{\xi\mu}_{\ell i}(\omega'-i\eta)\right]\right.\\
&\qquad\left.+\sigma^{\alpha}\left[G^{\nu\gamma}_{jk}(\omega'+i\eta)-G^{\nu\gamma}_{jk}(\omega'-i\eta)\right]\sigma^{\beta}G^{\xi\mu}_{\ell i}(\omega'-\omega-i\eta)\right\}\\
&= -\frac{\hbar}{8i\pi}\int d\omega' f(\omega')\mathrm{Tr}\left\{\sigma^{\alpha}G^{\nu\gamma}_{jk}(\omega'+\omega+i\eta)\sigma^{\beta}G^{\xi\mu}_{\ell i}(\omega'+i\eta)-\sigma^{\alpha}G^{\nu\gamma}_{jk}(\omega'-i\eta)\sigma^{\beta}G^{\xi\mu}_{\ell i}(\omega'-\omega-i\eta\right.\\
&\qquad -\sigma^{\alpha}G^{\nu\gamma}_{jk}(\omega'+\omega+i\eta)\sigma^{\beta}G^{\xi\mu}_{\ell i}(\omega'-i\eta)+\sigma^{\alpha}G^{\nu\gamma}_{jk}(\omega'+i\eta)\sigma^{\beta}G^{\xi\mu}_{\ell i}(\omega'-\omega-i\eta
\end{aligned}
\tag{C.15}
$$

In the $\{+,\uparrow,\downarrow,-\}$ basis, we can also write in terms of four spin indices as

$$
\begin{aligned}
\chi^{mnrs}_{(0)iijj}{}^{\mu\nu\gamma\xi}(\omega) = \frac{i\hbar}{2\pi}\int d\omega' f(\omega')&\left(G^{nr}_{ij}{}^{\nu\gamma}(\omega'+\omega)\left\{G^{sm}_{ji}{}^{\xi\mu}(\omega')-\left[G^{ms}_{ij}{}^{\mu\xi}(\omega')\right]^*\right\}\right.\\
&\left.+\left[G^{ms}_{ij}{}^{\mu\xi}(\omega'-\omega)\right]^*\left\{G^{nr}_{ij}{}^{\nu\gamma}(\omega')-\left[G^{rn}_{ji}{}^{\gamma\nu}(\omega')\right]^*\right\}\right) \ ,
\end{aligned}
\tag{C.16}
$$

since, in our convention, the Pauli matrices for these components have a factor 2 on each of the elements. This is the same result as the one obtained in Eq. (3.89). It can be transformed to the $\{0,x,y,z\}$ basis using the results on Appendix S.

So, the elements of the matrix are given by:

$$
\begin{aligned}
\chi_{(0)ij}^{++\mu\nu\gamma\xi}(\omega) = \chi_{(0)ij}^{\uparrow\downarrow\uparrow\downarrow\mu\nu\gamma\xi}(\omega) = \frac{i\hbar}{2\pi}\int d\omega' f(\omega') \Big(& G_{ij}^{\downarrow\uparrow\nu\gamma}(\omega'+\omega)\left\{G_{ji}^{\downarrow\uparrow\xi\mu}(\omega') - \left[G_{ij}^{\uparrow\downarrow\mu\xi}(\omega')\right]^*\right\} \\
& + \left[G_{ij}^{\uparrow\downarrow\mu\xi}(\omega'-\omega)\right]^*\left\{G_{ij}^{\downarrow\uparrow\nu\gamma}(\omega') - \left[G_{ji}^{\uparrow\downarrow\gamma\nu}(\omega')\right]^*\right\}\Big) \\
= \frac{i\hbar}{2\pi}\int d\omega' f(\omega') & \left\{G_{ji}^{\downarrow\uparrow\xi\mu}(\omega')G_{ij}^{\downarrow\uparrow\nu\gamma}(\omega'+\omega) - \left[G_{ji}^{\uparrow\downarrow\gamma\nu}(\omega')G_{ij}^{\uparrow\downarrow\mu\xi}(\omega'-\omega)\right]^*\right\} \\
- \frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega' & \left[G_{ij}^{\uparrow\downarrow\mu\xi}(\omega')\right]^* G_{ij}^{\downarrow\uparrow\nu\gamma}(\omega'+\omega)\ ,
\end{aligned}
\tag{C.17}
$$

In multilayers,

$$
\begin{aligned}
\chi_{(0)\ell\ell'}^{\uparrow\downarrow\uparrow\downarrow\mu\nu\gamma\xi}(\mathbf{q}_\parallel,\omega) = & \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel} G_{\ell'\ell}^{\downarrow\uparrow\xi\mu}(\mathbf{k}_\parallel,\omega')G_{\ell\ell'}^{\downarrow\uparrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega) \\
& - \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell'\ell}^{\uparrow\downarrow\gamma\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega')G_{\ell\ell'}^{\uparrow\downarrow\mu\xi}(\mathbf{k}_\parallel,\omega'-\omega)\right]^* \\
& - \frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega'\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell\ell'}^{\uparrow\downarrow\mu\xi}(\mathbf{k}_\parallel,\omega')\right]^* G_{\ell\ell'}^{\downarrow\uparrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)\ ,
\end{aligned}
\tag{C.18}
$$

$$
\begin{aligned}
\chi_{(0)\ell\ell'}^{\uparrow\downarrow\uparrow\uparrow\mu\nu\gamma\xi}(\mathbf{q}_\parallel,\omega) = & \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel} G_{\ell'\ell}^{\uparrow\uparrow\xi\mu}(\mathbf{k}_\parallel,\omega')G_{\ell\ell'}^{\downarrow\uparrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega) \\
& - \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell'\ell}^{\uparrow\downarrow\gamma\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega')G_{\ell\ell'}^{\uparrow\uparrow\mu\xi}(\mathbf{k}_\parallel,\omega'-\omega)\right]^* \\
& - \frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega'\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell\ell'}^{\uparrow\uparrow\mu\xi}(\mathbf{k}_\parallel,\omega')\right]^* G_{\ell\ell'}^{\downarrow\uparrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)\ ,
\end{aligned}
\tag{C.19}
$$

$$
\begin{aligned}
\chi_{(0)\ell\ell'}^{\uparrow\downarrow\downarrow\downarrow\mu\nu\gamma\xi}(\mathbf{q}_\parallel,\omega) = & \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel} G_{\ell'\ell}^{\downarrow\uparrow\xi\mu}(\mathbf{k}_\parallel,\omega')G_{\ell\ell'}^{\downarrow\downarrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega) \\
& - \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell'\ell}^{\downarrow\downarrow\gamma\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega')G_{\ell\ell'}^{\uparrow\downarrow\mu\xi}(\mathbf{k}_\parallel,\omega'-\omega)\right]^* \\
& - \frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega'\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell\ell'}^{\uparrow\downarrow\mu\xi}(\mathbf{k}_\parallel,\omega')\right]^* G_{\ell\ell'}^{\downarrow\downarrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)\ ,
\end{aligned}
\tag{C.20}
$$

$$
\begin{aligned}
\chi_{(0)\ell\ell'}^{\uparrow\downarrow\downarrow\uparrow\mu\nu\gamma\xi}(\mathbf{q}_\parallel,\omega) =& \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel} G_{\ell'\ell}^{\uparrow\uparrow\xi\mu}(\mathbf{k}_\parallel,\omega')G_{\ell\ell'}^{\downarrow\downarrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega) \\
&-\frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell'\ell}^{\downarrow\downarrow\gamma\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega')G_{\ell\ell'}^{\uparrow\uparrow\mu\xi}(\mathbf{k}_\parallel,\omega'-\omega)\right]^* \\
&-\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega'\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell\ell'}^{\uparrow\uparrow\mu\xi}(\mathbf{k}_\parallel,\omega')\right]^* G_{\ell\ell'}^{\downarrow\downarrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)\ ,
\end{aligned}
\tag{C.21}
$$

The elements of the second line are

$$
\begin{aligned}
\chi_{(0)\ell\ell'}^{\uparrow\uparrow\uparrow\downarrow\mu\nu\gamma\xi}(\mathbf{q}_\parallel,\omega) =& \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel} G_{\ell'\ell}^{\downarrow\uparrow\xi\mu}(\mathbf{k}_\parallel,\omega')G_{\ell\ell'}^{\uparrow\uparrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega) \\
&-\frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell'\ell}^{\uparrow\uparrow\gamma\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega')G_{\ell\ell'}^{\uparrow\downarrow\mu\xi}(\mathbf{k}_\parallel,\omega'-\omega)\right]^* \\
&-\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega'\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell\ell'}^{\uparrow\downarrow\mu\xi}(\mathbf{k}_\parallel,\omega')\right]^* G_{\ell\ell'}^{\uparrow\uparrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)\ ,
\end{aligned}
\tag{C.22}
$$

$$
\begin{aligned}
\chi_{(0)\ell\ell'}^{\uparrow\uparrow\uparrow\uparrow\mu\nu\gamma\xi}(\mathbf{q}_\parallel,\omega) =& \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel} G_{\ell'\ell}^{\uparrow\uparrow\xi\mu}(\mathbf{k}_\parallel,\omega')G_{\ell\ell'}^{\uparrow\uparrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega) \\
&-\frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell'\ell}^{\uparrow\uparrow\gamma\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega')G_{\ell\ell'}^{\uparrow\uparrow\mu\xi}(\mathbf{k}_\parallel,\omega'-\omega)\right]^* \\
&-\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega'\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell\ell'}^{\uparrow\uparrow\mu\xi}(\mathbf{k}_\parallel,\omega')\right]^* G_{\ell\ell'}^{\uparrow\uparrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)\ ,
\end{aligned}
\tag{C.23}
$$

$$
\begin{aligned}
\chi_{(0)\ell\ell'}^{\uparrow\uparrow\downarrow\downarrow\mu\nu\gamma\xi}(\mathbf{q}_\parallel,\omega) =& \frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel} G_{\ell'\ell}^{\downarrow\uparrow\xi\mu}(\mathbf{k}_\parallel,\omega')G_{\ell\ell'}^{\uparrow\downarrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega) \\
&-\frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell'\ell}^{\downarrow\uparrow\gamma\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega')G_{\ell\ell'}^{\uparrow\downarrow\mu\xi}(\mathbf{k}_\parallel,\omega'-\omega)\right]^* \\
&-\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega'\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell\ell'}^{\uparrow\downarrow\mu\xi}(\mathbf{k}_\parallel,\omega')\right]^* G_{\ell\ell'}^{\uparrow\downarrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)\ ,
\end{aligned}
\tag{C.24}
$$

$$
\begin{aligned}
\chi_{(0)\ell\ell'}^{\uparrow\uparrow\downarrow\uparrow\mu\nu\gamma\xi}(\mathbf{q}_\parallel,\omega) =&\frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel} G_{\ell'\ell}^{\uparrow\uparrow\xi\mu}(\mathbf{k}_\parallel,\omega')G_{\ell\ell'}^{\uparrow\downarrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)\\
&-\frac{i\hbar}{2\pi}\int d\omega' f(\omega')\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell'\ell}^{\downarrow\uparrow\gamma\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega')G_{\ell\ell'}^{\uparrow\uparrow\mu\xi}(\mathbf{k}_\parallel,\omega'-\omega)\right]^*\\
&-\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega'\frac{1}{N}\sum_{\mathbf{k}_\parallel}\left[G_{\ell\ell'}^{\uparrow\uparrow\mu\xi}(\mathbf{k}_\parallel,\omega')\right]^* G_{\ell\ell'}^{\uparrow\downarrow\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)\ ,
\end{aligned}
\tag{C.25}
$$

In general, we can write

$$
\begin{aligned}
\chi_{(0)ijk\ell}^{mnrs\,\mu\nu\gamma\xi}(\omega) =& \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F} d\omega'\left\{G_{\ell i}^{sm\,\xi\mu}(\omega')G_{jk}^{nr\,\nu\gamma}(\omega'+\omega)-\left[G_{kj}^{rn\,\gamma\nu}(\omega')G_{i\ell}^{ms\,\mu\xi}(\omega'-\omega)\right]^*\right\}\\
&-\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega'\left[G_{i\ell}^{ms\,\mu\xi}(\omega')\right]^* G_{jk}^{nr\,\nu\gamma}(\omega'+\omega)\ .
\end{aligned}
\tag{C.26}
$$

C.1 Complex plane integral

Extending to the complex plane, we can write $\omega' \to z = \omega' \pm iy$ (the signal is chosen in accordance to the analyticity of the Green functions on the upper or lower half plane). The first integral can be done in an imaginary axis for $z = \omega_F \pm iy \to dz = \pm i dy$. Therefore,

$$
\begin{aligned}
\chi_{(0)ijk\ell}^{mnrs\,\mu\nu\gamma\xi}(\omega) =& \frac{\hbar}{2\pi}\int_{\eta}^{\infty} dy\left\{G_{\ell i}^{sm\,\xi\mu}(\omega_F+iy)G_{jk}^{nr\,\nu\gamma}(\omega_F+\omega+iy)+\left[G_{kj}^{rn\,\gamma\nu}(\omega_F+iy)G_{i\ell}^{ms\,\mu\xi}(\omega_F-\omega+iy)\right]^*\right\}\\
&-\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega'\left[G_{i\ell}^{ms\,\mu\xi}(\omega'+i\eta)\right]^* G_{jk}^{nr\,\nu\gamma}(\omega'+\omega+i\eta)\ .
\end{aligned}
\tag{C.27}
$$

where the change of sign comes from the fact that $I_r + I_i + I_{R\to\infty} = 0 \to I_r = -I_i$.

If we change variables $\omega'' = \omega' - \omega$ on the second term of the first integral,

$$\begin{aligned}
\chi_{(0)ijk\ell}^{mnrs\;\mu\nu\gamma\xi}(\omega) = \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F} d\omega' {G_{\ell i}^{sm}}^{\xi\mu}(\omega'){G_{jk}^{nr}}^{\nu\gamma}(\omega'+\omega) \\
-\frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F} d\omega' \left[{G_{kj}^{rn}}^{\gamma\nu}(\omega'){G_{i\ell}^{ms}}^{\mu\xi}(\omega'-\omega)\right]^* \\
-\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega' \left[{G_{i\ell}^{ms}}^{\mu\xi}(\omega')\right]^* {G_{jk}^{nr}}^{\nu\gamma}(\omega'+\omega) \\
= \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F} d\omega' {G_{\ell i}^{sm}}^{\xi\mu}(\omega'){G_{jk}^{nr}}^{\nu\gamma}(\omega'+\omega) \\
-\frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F-\omega} d\omega' \left[{G_{kj}^{rn}}^{\gamma\nu}(\omega'+\omega){G_{i\ell}^{ms}}^{\mu\xi}(\omega')\right]^* \\
-\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega' \left[{G_{i\ell}^{ms}}^{\mu\xi}(\omega')\right]^* {G_{jk}^{nr}}^{\nu\gamma}(\omega'+\omega)\ .
\end{aligned} \tag{C.28}$$

Rewriting the second term so the integral is extended to ω_F, we have

$$\begin{aligned}
\chi_{(0)ijk\ell}^{mnrs\;\mu\nu\gamma\xi}(\omega) = \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F} d\omega' {G_{\ell i}^{sm}}^{\xi\mu}(\omega'){G_{jk}^{nr}}^{\nu\gamma}(\omega'+\omega) \\
-\frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F-\omega} d\omega' \left[{G_{kj}^{rn}}^{\gamma\nu}(\omega'+\omega){G_{i\ell}^{ms}}^{\mu\xi}(\omega')\right]^* \\
-\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega' \left[{G_{kj}^{rn}}^{\gamma\nu}(\omega'+\omega){G_{i\ell}^{ms}}^{\mu\xi}(\omega')\right]^* \\
+\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega' \left[{G_{kj}^{rn}}^{\gamma\nu}(\omega'+\omega){G_{i\ell}^{ms}}^{\mu\xi}(\omega')\right]^* \\
-\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega' \left[{G_{i\ell}^{ms}}^{\mu\xi}(\omega')\right]^* {G_{jk}^{nr}}^{\nu\gamma}(\omega'+\omega) \\
= \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F} d\omega' \left\{{G_{\ell i}^{sm}}^{\xi\mu}(\omega'){G_{jk}^{nr}}^{\nu\gamma}(\omega'+\omega) - \left[{G_{kj}^{rn}}^{\gamma\nu}(\omega'+\omega){G_{i\ell}^{ms}}^{\mu\xi}(\omega')\right]^*\right\} \\
-\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega' \left[{G_{i\ell}^{ms}}^{\mu\xi}(\omega')\right]^* \left\{{G_{jk}^{nr}}^{\nu\gamma}(\omega'+\omega) - \left[{G_{kj}^{rn}}^{\gamma\nu}(\omega'+\omega)\right]^*\right\}\ .
\end{aligned} \tag{C.29}$$

In multilayers,

$$\begin{aligned}
\chi_{(0)ijk\ell}^{mnrs\;\mu\nu\gamma\xi}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega) = \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F} d\omega' \left\{{G_{\ell i}^{sm}}^{\xi\mu}(\mathbf{k}_\parallel,\omega'){G_{jk}^{nr}}^{\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega) - \left[{G_{kj}^{rn}}^{\gamma\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)G_{i}^{n}\right.\right. \\
-\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega' \left[{G_{i\ell}^{ms}}^{\mu\xi}(\mathbf{k}_\parallel,\omega')\right]^* \left\{{G_{jk}^{nr}}^{\nu\gamma}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega) - \left[{G_{kj}^{rn}}^{\gamma\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega\right.\right.
\end{aligned} \tag{C.30}$$

Finally, extending the first integral to the complex plane,

$$\begin{aligned}\chi^{mnrs\,\mu\nu\gamma\xi}_{(0)ijk\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega) = {} & \frac{\hbar}{2\pi}\int_\eta^\infty dy G^{sm\,\xi\mu}_{\ell i}(\mathbf{k}_\parallel,\omega_F+iy)G^{nr\,\nu\gamma}_{jk}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega_F+\omega+iy)\\ & +\frac{\hbar}{2\pi}\int_\eta^\infty dy\left[G^{rn\,\gamma\nu}_{kj}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega_F+\omega+iy)G^{ms\,\mu\xi}_{i\ell}(\mathbf{k}_\parallel,\omega_F+iy)\right]^*\\ & -\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega'\left[G^{ms\,\mu\xi}_{i\ell}(\mathbf{k}_\parallel,\omega')\right]^*\left\{G^{nr\,\nu\gamma}_{jk}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)-\left[G^{rn\,\gamma\nu}_{kj}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)\right]\right.\end{aligned} \tag{C.31}$$

$$\begin{aligned}= {} & \frac{\hbar}{2\pi}\int_\eta^\infty dy G^{sm\,\xi\mu}_{\ell i}(\mathbf{k}_\parallel,\omega_F+iy)G^{nr\,\nu\gamma}_{jk}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega_F+\omega+iy)\\ & +\frac{\hbar}{2\pi}\int_\eta^\infty dy\left[G^{rn\,\gamma\nu}_{kj}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega_F+\omega+iy)G^{ms\,\mu\xi}_{i\ell}(\mathbf{k}_\parallel,\omega_F+iy)\right]^*\\ & -\frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega'\left[G^{ms\,\mu\xi}_{i\ell}(\mathbf{k}_\parallel,\omega')\right]^*\left\{G^{nr\,\nu\gamma}_{jk}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)-\left[G^{rn\,\gamma\nu}_{kj}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega'+\omega)\right]^*\right\}\ .\end{aligned} \tag{C.32}$$

Note that the first two terms satisfy the following relation

$$\begin{aligned}\left[\chi^{mnrs\,\mu\nu\gamma\xi}_{(1+2)ijk\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega)\right]^* = {} & \frac{\hbar}{2\pi}\int_\eta^\infty dy\left[G^{sm\,\xi\mu}_{\ell i}(\mathbf{k}_\parallel,\omega_F+iy)G^{nr\,\nu\gamma}_{jk}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega_F+\omega+iy)\right]^*\\ & +\frac{\hbar}{2\pi}\int_\eta^\infty dy G^{rn\,\gamma\nu}_{kj}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega_F+\omega+iy)G^{ms\,\mu\xi}_{i\ell}(\mathbf{k}_\parallel,\omega_F+iy)\\ = {} & \frac{\hbar}{2\pi}\int_\eta^\infty dy G^{ms\,\mu\xi}_{i\ell}(\mathbf{k}_\parallel,\omega_F+iy)G^{rn\,\gamma\nu}_{kj}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega_F+\omega+iy) \qquad ,\\ & +\frac{\hbar}{2\pi}\int_\eta^\infty dy\left[G^{nr\,\nu\gamma}_{jk}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega_F+\omega+iy)G^{sm\,\xi\mu}_{\ell i}(\mathbf{k}_\parallel,\omega_F+iy)\right]^*\\ = {} & \chi^{srnm\,\xi\gamma\nu\mu}_{(1+2)\ell kji}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega)\end{aligned} \tag{C.33}$$

such that

$$\begin{aligned}\sum_{\mu\nu}\left[\chi^{+-\,\mu\nu}_{(1+2)ii}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega)\right]^* & = \sum_{\mu\nu}\left[\chi^{\uparrow\downarrow\downarrow\uparrow\,\mu\mu\nu\nu}_{(1+2)iiii}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega)\right]^*\\ & = \sum_{\mu\nu}\chi^{\uparrow\downarrow\downarrow\uparrow\,\nu\nu\mu\mu}_{(1+2)iiii}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega)\ .\\ & = \sum_{\mu\nu}\chi^{+-\,\mu\nu}_{(1+2)ii}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel,\omega)\end{aligned} \tag{C.34}$$

Finally, the integral from η to ∞ can be calculated numerically by changing the variables to $y = \frac{\eta+x}{1-x}$, such that

$$
\begin{aligned}
x =& \frac{y-\eta}{y+1} \quad \text{and} \\
dy =& \frac{dx}{1-x} + \frac{\eta + x}{(1-x)^2} dx \ , \\
=& \frac{1+\eta}{(1-x)^2} dx
\end{aligned}
\tag{C.35}
$$

with the limits of integration changed to

$$
\begin{aligned}
y = \eta \quad &\Rightarrow \quad x = 0 \\
y \to \infty \quad &\Rightarrow \quad x \to 1
\end{aligned} \ .
\tag{C.36}
$$

Another possibility is to remove the $\omega_F - \omega$ to ω_F from the first term of Eq. (C.28), so we end up with

$$
\begin{aligned}
\chi_{(0)ijk\ell}^{mnrs\;\mu\nu\gamma\xi}(\omega) =& \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F-\omega} d\omega' G_{\ell i}^{sm\;\xi\mu}(\omega') G_{jk}^{nr\;\nu\gamma}(\omega'+\omega) \\
&+ \frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega' G_{\ell i}^{sm\;\xi\mu}(\omega') G_{jk}^{nr\;\nu\gamma}(\omega'+\omega) \\
&- \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F-\omega} d\omega' \left[G_{kj}^{rn\;\gamma\nu}(\omega'+\omega) G_{i\ell}^{ms\;\mu\xi}(\omega') \right]^* \\
&- \frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega' \left[G_{i\ell}^{ms\;\mu\xi}(\omega') \right]^* G_{jk}^{nr\;\nu\gamma}(\omega'+\omega) \\
=& \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F-\omega} d\omega' \left\{ G_{\ell i}^{sm\;\xi\mu}(\omega') G_{jk}^{nr\;\nu\gamma}(\omega'+\omega) - \left[G_{kj}^{rn\;\gamma\nu}(\omega'+\omega) G_{i\ell}^{ms\;\mu\xi}(\omega') \right]^* \right\} \\
&+ \frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega' \left\{ G_{\ell i}^{sm\;\xi\mu}(\omega') - \left[G_{i\ell}^{ms\;\mu\xi}(\omega') \right]^* \right\} G_{jk}^{nr\;\nu\gamma}(\omega'+\omega) \ .
\end{aligned}
\tag{C.37}
$$

C.2 Excluding η from the transformation

Depending if the broadening is added to the argument of the GF or the susceptibility, different imaginary parts have to be added on the Green functions. To take this into account, we first do the transformation $y' = y - \eta \Rightarrow dy' = dy$ and the integral from $y = \eta$ to $y = \infty$ becomes an integral from $y' = 0$ to $y' = \infty$:

$$
\begin{aligned}
\chi_{(0)ijk\ell}^{mnrs\;\mu\nu\gamma\xi}(\mathbf{k}_\parallel, \mathbf{k}_\parallel + \mathbf{q}_\parallel, \omega) =& \frac{\hbar}{2\pi}\int_0^\infty dy' G_{\ell i}^{sm\;\xi\mu}(\mathbf{k}_\parallel, \omega_F + i(y'+\eta')) G_{jk}^{nr\;\nu\gamma}(\mathbf{k}_\parallel + \mathbf{q}_\parallel, \omega_F + \omega + i(y'+\eta)) \\
&+ \frac{\hbar}{2\pi}\int_0^\infty dy' \left[G_{kj}^{rn\;\gamma\nu}(\mathbf{k}_\parallel + \mathbf{q}_\parallel, \omega_F + \omega + i(y'+\eta)) G_{i\ell}^{ms\;\mu\xi}(\mathbf{k}_\parallel, \omega_F + i(y'+\eta')) \right]^* \\
&- \frac{i\hbar}{2\pi}\int_{\omega_F-\omega}^{\omega_F} d\omega' \left[G_{i\ell}^{ms\;\mu\xi}(\mathbf{k}_\parallel, \omega' + i\eta') \right]^* \left\{ G_{jk}^{nr\;\nu\gamma}(\mathbf{k}_\parallel + \mathbf{q}_\parallel, \omega' + \omega + i\eta) - \left[G_{kj}^{rn\;\gamma\nu}(\mathbf{k}_\parallel + \right.\right.
\end{aligned}
\tag{C.38}
$$

To broaden the energy levels, $\eta' = \eta$, while to move away from the real axis in ω, $\eta' \to 0^+$.

We then use $y' = \frac{x}{1-x}$, such that

$$\begin{aligned} x =& \frac{y'}{y'+1} \quad \text{and} \\ dy' =& \frac{dx}{1-x} + \frac{x}{(1-x)^2}dx \; . \\ =& \frac{1}{(1-x)^2}dx \end{aligned} \tag{C.39}$$

On the limits of the integral from 0 to ∞,

$$\begin{aligned} y' = 0 \quad &\Rightarrow \quad x = 0 \\ y \to \infty \quad &\Rightarrow \quad x \to 1 \end{aligned} \; . \tag{C.40}$$

C.3 Low frequency limit

For $\omega = 0$, we can write Eq. C.15 as

$$\begin{aligned} \chi^{\alpha\beta\,\mu\nu\gamma\xi}_{(0)ijk\ell}(0) =& -\frac{\hbar}{4\pi}\int d\omega' f(\omega')\mathrm{Tr}\left\{\sigma^\alpha G^{\nu\gamma}_{jk}(\omega'+i\eta)\sigma^\beta \Im G^{\xi\mu}_{\ell i}(\omega') + \sigma^\alpha \Im G^{\nu\gamma}_{jk}(\omega')\sigma^\beta G^{\xi\mu}_{\ell i}(\omega'-i\eta)\right\} \\ =& -\frac{\hbar}{8i\pi}\int d\omega' f(\omega')\mathrm{Tr}\left\{\sigma^\alpha G^{\nu\gamma}_{jk}(\omega'+i\eta)\sigma^\beta \left[G^{\xi\mu}_{\ell i}(\omega'+i\eta) - G^{\xi\mu}_{\ell i}(\omega'-i\eta)\right]\right. \\ & \left. +\sigma^\alpha \left[G^{\nu\gamma}_{jk}(\omega'+i\eta) - G^{\nu\gamma}_{jk}(\omega'-i\eta)\right]\sigma^\beta G^{\xi\mu}_{\ell i}(\omega'-i\eta)\right\} \\ =& -\frac{\hbar}{8i\pi}\int d\omega' f(\omega')\mathrm{Tr}\left\{\sigma^\alpha G^{\nu\gamma}_{jk}(\omega'+i\eta)\sigma^\beta G^{\xi\mu}_{\ell i}(\omega'+i\eta) - \sigma^\alpha G^{\nu\gamma}_{jk}(\omega'-i\eta)\sigma^\beta G^{\xi\mu}_{\ell i}(\omega'-i\eta)\right\} \; . \end{aligned} \tag{C.41}$$

$$\chi^{\alpha\beta\,\mu\nu\gamma\xi}_{(0)ijk\ell}(0) = -\frac{\hbar}{8i\pi}\int d\omega' f(\omega')\mathrm{Tr}\left\{\sigma^\alpha G^{\nu\gamma}_{jk}(\omega'+i\eta)\sigma^\beta G^{\xi\mu}_{\ell i}(\omega'+i\eta) - \sigma^\alpha G^{\nu\gamma}_{jk}(\omega'-i\eta)\sigma^\beta G^{\xi\mu}_{\ell i}(\omega'-i\eta)\right\} \; . \tag{C.42}$$

Using Eq. (C.12), we can write

$$\chi^{\alpha\beta\,\mu\nu\gamma\xi}_{(0)ijk\ell}(0) = -\frac{\hbar}{4\pi}\Im\int d\omega' f(\omega')\mathrm{Tr}\left\{\sigma^\alpha G^{\nu\gamma}_{jk}(\omega')\sigma^\beta G^{\xi\mu}_{\ell i}(\omega')\right\} \; . \tag{C.43}$$

C.4 Intra- and interband contributions

We can also calculate the susceptibilities using the eigenstates of the Hamiltonian. With that intent, we calculate the total transverse magnetic susceptibility as

$$\chi^{+-} = \sum_{\substack{ij\\ \mu\nu}} \chi^{\mu\nu}_{ij} \tag{C.44}$$

Appendix D

Angular momentum $\mathbf{L}$ and $\mathbf{L}\cdot\mathbf{S}$ matrices

To obtain the angular momentum matrices, we need the cubic harmonics given in term of the harmonics spherics by

- s orbital

$$s = X_{00} = \frac{1}{\sqrt{4\pi}} = Y_0^0 \ , \tag{D.1}$$

- p orbitals

$$\begin{aligned} p_x =& N_1^{\mathrm{C}} \frac{x}{r} = \frac{1}{\sqrt{2}} \left(Y_1^{-1} - Y_1^1 \right) \\ p_y =& N_1^{\mathrm{C}} \frac{y}{r} = i \frac{1}{\sqrt{2}} \left(Y_1^{-1} + Y_1^1 \right) \ , \\ p_z =& N_1^{\mathrm{C}} \frac{z}{r} = Y_1^0 \end{aligned} \tag{D.2}$$

- d orbitals

$$\begin{aligned} d_{xy} =& N_2^{\mathrm{C}} \frac{xy}{r^2} = \frac{i}{\sqrt{2}} \left(Y_2^{-2} - Y_2^2 \right) \\ d_{yz} =& N_2^{\mathrm{C}} \frac{yz}{r^2} = \frac{i}{\sqrt{2}} \left(Y_2^{-1} + Y_2^1 \right) \\ d_{zx} =& N_2^{\mathrm{C}} \frac{xz}{r^2} = \frac{1}{\sqrt{2}} \left(Y_2^{-1} - Y_2^1 \right) \qquad , \\ d_{x^2-y^2} =& N_2^{\mathrm{C}} \frac{x^2-y^2}{2r^2} = \frac{1}{\sqrt{2}} \left(Y_2^{-2} + Y_2^2 \right) \\ d_{3z^2-r^2} =& N_2^{\mathrm{C}} \frac{3z^2-r^2}{2r^2\sqrt{3}} = Y_2^0 \end{aligned} \tag{D.3}$$

where $N_1^{\mathrm{C}} = \left(\frac{3}{4\pi}\right)^{1/2}$ and $N_2^{\mathrm{C}} = \left(\frac{15}{4\pi}\right)^{1/2}$.

To calculate the matrix $L^z_{\mu\nu}$, we use the fact that

$$\hat{L}^z|\ell m\rangle = \hbar m|\ell m\rangle \ , \tag{D.4}$$

where $\langle\theta,\phi|\ell,m\rangle = Y_\ell^m(\theta,\phi)$. This way, we can write (in units of $\hbar$)

$$\begin{aligned}
\hat{L}^z s =& 0\\
\hat{L}^z p_x =& -\frac{1}{\sqrt{2}}\left(Y_1^1 + Y_1^{-1}\right) = ip_y\\
\hat{L}^z p_y =& -i\frac{1}{\sqrt{2}}\left(Y_1^1 - Y_1^{-1}\right) = -ip_x\\
\hat{L}^z p_z =& 0\\
\hat{L}^z d_{xy} =& -2i\frac{1}{\sqrt{2}}\left(Y_2^2 + Y_2^{-2}\right) = -2id_{x^2-y^2}\\
\hat{L}^z d_{yz} =& \frac{i}{\sqrt{2}}\left(Y_2^1 - Y_2^{-1}\right) = -id_{zx}\\
\hat{L}^z d_{zx} =& -\frac{1}{\sqrt{2}}\left(Y_2^1 + Y_2^{-1}\right) = id_{yz}\\
\hat{L}^z d_{x^2-y^2} =& 2\frac{1}{\sqrt{2}}\left(Y_2^2 - Y_2^{-2}\right) = 2id_{xy}\\
\hat{L}^z d_{3z^2-r^2} =& 0
\end{aligned} \ . \tag{D.5}$$

From this results, it is easy to obtain the matrix elements $\langle\mu|L^z|\nu\rangle$ to write

$$L^z = \hbar\begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & -i & 0 & 0 & 0 & 0 & 0 & 0\\
0 & i & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 2i & 0\\
0 & 0 & 0 & 0 & 0 & 0 & i & 0 & 0\\
0 & 0 & 0 & 0 & 0 & -i & 0 & 0 & 0\\
0 & 0 & 0 & 0 & -2i & 0 & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix} \ . \tag{D.6}$$

The transverse components can be obtained from the ladder operators $\hat{L}^+$ and $\hat{L}^-$. Since $\langle\mu|L^-|\nu\rangle = [\langle\nu|L^+|\mu\rangle]^*$, we can obtain the latest in matrix form in terms of the former.

To obtain the matrix elements of L^+, we use the relation

$$\hat{L}^+|\ell,m\rangle = \hbar\sqrt{\ell(\ell+1) - m(m+1)}|\ell,m+1\rangle \tag{D.7}$$

to obtain (in units of $\hbar$)

$$\begin{aligned}
\hat{L}^{+}s =&0\\
\hat{L}^{+}p_x =&-\sqrt{2}\frac{1}{\sqrt{2}}\left(-Y_1^0\right)=p_z\\
\hat{L}^{+}p_y =&\sqrt{2}i\frac{1}{\sqrt{2}}\left(Y_1^0\right)=ip_z\\
\hat{L}^{+}p_z =&\sqrt{2}Y_1^1=-p_x-ip_y\\
\hat{L}^{+}d_{xy} =&\sqrt{4}\frac{i}{\sqrt{2}}\left(Y_2^{-1}\right)=d_{yz}+id_{zx}\\
\hat{L}^{+}d_{yz} =&\frac{i}{\sqrt{2}}\left(\sqrt{4}Y_2^2+\sqrt{6}Y_2^0\right)=-d_{xy}+id_{x^2-y^2}+\sqrt{3}id_{3z^2-r^2}\\
\hat{L}^{+}d_{zx} =&-\frac{1}{\sqrt{2}}\left(\sqrt{4}Y_2^2-\sqrt{6}Y_2^{-1}\right)=-id_{xy}-d_{x^2-y^2}+\sqrt{3}d_{3z^2-r^2}\\
\hat{L}^{+}d_{x^2-y^2} =&\sqrt{4}\frac{1}{\sqrt{2}}\left(Y_2^{-1}\right)=-id_{yz}+d_{zx}\\
\hat{L}^{+}d_{3z^2-r^2} =&\sqrt{6}Y_2^1=-\sqrt{3}id_{yz}-\sqrt{3}d_{zx}
\end{aligned}, \tag{D.8}$$

Thus, in matrix form,

$$L^{+}=\hbar\begin{pmatrix}
0&0&0&0&0&0&0&0&0\\
0&0&0&-1&0&0&0&0&0\\
0&0&0&-i&0&0&0&0&0\\
0&1&i&0&0&0&0&0&0\\
0&0&0&0&0&-1&-i&0&0\\
0&0&0&0&1&0&0&-i&-\sqrt{3}i\\
0&0&0&0&i&0&0&1&-\sqrt{3}\\
0&0&0&0&0&i&-1&0&0\\
0&0&0&0&0&\sqrt{3}i&\sqrt{3}&0&0
\end{pmatrix} \tag{D.9}$$

and we can easily obtain

$$L^{-}=\hbar\begin{pmatrix}
0&0&0&0&0&0&0&0&0\\
0&0&0&1&0&0&0&0&0\\
0&0&0&-i&0&0&0&0&0\\
0&-1&i&0&0&0&0&0&0\\
0&0&0&0&0&1&-i&0&0\\
0&0&0&0&-1&0&0&-i&-\sqrt{3}i\\
0&0&0&0&i&0&0&-1&\sqrt{3}\\
0&0&0&0&0&i&1&0&0\\
0&0&0&0&0&\sqrt{3}i&-\sqrt{3}&0&0
\end{pmatrix}. \tag{D.10}$$

The x and y components can also be calculated. Therefore

$$L^x = \frac{1}{2}\left(L^+ + L^-\right) = \hbar \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -i & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & i & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -i & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & -i & -\sqrt{3}i \\ 0 & 0 & 0 & 0 & i & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & \sqrt{3}i & 0 & 0 & 0 \end{pmatrix} , \tag{D.11}$$

$$L^y = \frac{1}{2i}\left(L^+ - L^-\right) = \hbar \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & i & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -i & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -i & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & -i & \sqrt{3}i \\ 0 & 0 & 0 & 0 & 0 & 0 & i & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -\sqrt{3}i & 0 & 0 \end{pmatrix} \tag{D.12}$$

Since the orbital angular momentum is defined in terms of the cubic axis $\{x, y, z\}$ and the spin has its own set of axis $\{x', y', z'\}$, we need to rotate one of them to obtain the product $\lambda\mathbf{L}\cdot\mathbf{S}$. We include the $\hbar$ of both angular momenta in the λ coefficient, so it has the units of energy. If we call the rotation angles around z and y axis as ϕ and θ, respectively, we have

$$\mathbf{S} = R_z(\phi)R_y(\theta)\mathbf{S}' , \tag{D.13}$$

where $\mathbf{S}'$ is the spin operator in $\{x', y', z'\}$ and the rotation matrices around y and z axis are given by

$$R_y(\theta) = \begin{pmatrix} \cos(\theta) & 0 & \sin(\theta) \\ 0 & 1 & 0 \\ -\sin(\theta) & 0 & \cos(\theta) \end{pmatrix} , \text{ and} \tag{D.14}$$

$$R_z(\phi) = \begin{pmatrix} \cos(\phi) & -\sin(\phi) & 0 \\ \sin(\phi) & \cos(\phi) & 0 \\ 0 & 0 & 1 \end{pmatrix} . \tag{D.15}$$

We can also rotate the orbital angular momentum vector as

$$\mathbf{L}' = R_y(-\theta)R_z(-\phi)\mathbf{L} . \tag{D.16}$$

The product $R_z(\phi)R_y(\theta)$ is

$$\begin{aligned}
R_z(\phi)R_y(\theta) &= \begin{pmatrix} \cos(\phi) & -\sin(\phi) & 0 \\ \sin(\phi) & \cos(\phi) & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} \cos(\theta) & 0 & \sin(\theta) \\ 0 & 1 & 0 \\ -\sin(\theta) & 0 & \cos(\theta) \end{pmatrix} \\
&= \begin{pmatrix} \cos(\theta)\cos(\phi) & -\sin(\phi) & \sin(\theta)\cos(\phi) \\ \cos(\theta)\sin(\phi) & \cos(\phi) & \sin(\theta)\sin(\phi) \\ -\sin(\theta) & 0 & \cos(\theta) \end{pmatrix} ,
\end{aligned} \tag{D.17}$$

while

$$\begin{aligned}
R_y(-\theta)R_z(-\phi) =& [R_z(\phi)R_y(\theta)]^T \\
=& \begin{pmatrix} \cos(\theta) & 0 & -\sin(\theta) \\ 0 & 1 & 0 \\ \sin(\theta) & 0 & \cos(\theta) \end{pmatrix} \begin{pmatrix} \cos(\phi) & \sin(\phi) & 0 \\ -\sin(\phi) & \cos(\phi) & 0 \\ 0 & 0 & 1 \end{pmatrix} \\
=& \begin{pmatrix} \cos(\theta)\cos(\phi) & \cos(\theta)\sin(\phi) & -\sin(\theta) \\ -\sin(\phi) & \cos(\phi) & 0 \\ \sin(\theta)\cos(\phi) & \sin(\theta)\sin(\phi) & \cos(\theta) \end{pmatrix} .
\end{aligned} \tag{D.18}$$

The spin vector written in the cubic coordinates is

$$\begin{aligned}
\mathbf{S} &= \begin{pmatrix} \cos(\theta)\cos(\phi) & -\sin(\phi) & \sin(\theta)\cos(\phi) \\ \cos(\theta)\sin(\phi) & \cos(\phi) & \sin(\theta)\sin(\phi) \\ -\sin(\theta) & 0 & \cos(\theta) \end{pmatrix} \begin{pmatrix} S'^x \\ S'^y \\ S'^z \end{pmatrix} \\
&= \begin{pmatrix} S'^x\cos(\theta)\cos(\phi) - S'^y\sin(\phi) + S'^z\sin(\theta)\cos(\phi) \\ S'^x\cos(\theta)\sin(\phi) + S'^y\cos(\phi) + S'^z\sin(\theta)\sin(\phi) \\ -S'^x\sin(\theta) + S'^z\cos(\theta) \end{pmatrix} .
\end{aligned} \tag{D.19}$$

In terms of S'^+, S'^- and S'^z, this vector becomes

$$\begin{aligned}
\mathbf{S} &= \begin{pmatrix} \frac{1}{2}(S'^+ + S'^-)\cos(\theta)\cos(\phi) + \frac{i}{2}(S'^+ - S'^-)\sin(\phi) + S'^z\sin(\theta)\cos(\phi) \\ \frac{1}{2}(S'^+ + S'^-)\cos(\theta)\sin(\phi) - \frac{i}{2}(S'^+ - S'^-)\cos(\phi) + S'^z\sin(\theta)\sin(\phi) \\ -\frac{1}{2}(S'^+ + S'^-)\sin(\theta) + S'^z\cos(\theta) \end{pmatrix} \\
&= \begin{pmatrix} \frac{1}{2}S'^+[\cos(\theta)\cos(\phi) + i\sin(\phi)] + \frac{1}{2}S'^-[\cos(\theta)\cos(\phi) - i\sin(\phi)] + S'^z\sin(\theta)\cos(\phi) \\ \frac{1}{2}S'^+[\cos(\theta)\sin(\phi) - i\cos(\phi)] + \frac{1}{2}S'^-[\cos(\theta)\sin(\phi) + i\cos(\phi)] + S'^z\sin(\theta)\sin(\phi) \\ -\frac{1}{2}S'^+\sin(\theta) - \frac{1}{2}S'^-\sin(\theta) + S'^z\cos(\theta) \end{pmatrix} .
\end{aligned} \tag{D.20}$$

The orbital angular momentum vector in the spin system of coordinates is

$$\begin{aligned}
\mathbf{L}' &= \begin{pmatrix} \cos(\theta)\cos(\phi) & \cos(\theta)\sin(\phi) & -\sin(\theta) \\ -\sin(\phi) & \cos(\phi) & 0 \\ \sin(\theta)\cos(\phi) & \sin(\theta)\sin(\phi) & \cos(\theta) \end{pmatrix} \begin{pmatrix} L^x \\ L^y \\ L^z \end{pmatrix} \\
&= \begin{pmatrix} L^x\cos(\theta)\cos(\phi) + L^y\cos(\theta)\sin(\phi) - L^z\sin(\theta) \\ -L^x\sin(\phi) + L^y\cos(\phi) \\ L^x\sin(\theta)\cos(\phi) + L^y\sin(\theta)\sin(\phi) + L^z\cos(\theta) \end{pmatrix} .
\end{aligned} \tag{D.21}$$

The product $\mathbf{L}\cdot\mathbf{S}$ may be written as

$$\begin{aligned}
\mathbf{L}\cdot\mathbf{S}=\mathbf{L}'\cdot\mathbf{S}' =& L^x S^x + L^y S^y + L^z S^z\\
=& \frac{1}{2}L^- S^+ + \frac{1}{2}L^+ S^- + L^z S^z\\
=& \left\{L^x\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right]+L^y\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right]-L^z\sin(\theta)\right\}\frac{1}{2}S'^+\\
&+\left\{L^x\left[\cos(\theta)\cos(\phi)-i\sin(\phi)\right]+L^y\left[\cos(\theta)\sin(\phi)+i\cos(\phi)\right]-L^z\sin(\theta)\right\}\frac{1}{2}S'^-\\
&+\left\{L^x\sin(\theta)\cos(\phi)+L^y\sin(\theta)\sin(\phi)+L^z\cos(\theta)\right\}S'^z\ .
\end{aligned} \tag{D.22}$$

In second quantization, we need the matrix elements

$$\begin{aligned}
&\left\langle m\alpha s\left|\mathbf{L}_m\cdot\mathbf{S}_m\right| m\beta s'\right\rangle\\
&=\frac{1}{2}L'^-_{\alpha\beta}S^+_{ss'}+\frac{1}{2}L'^+_{\alpha\beta}S^-_{ss'}+L'^z_{\alpha\beta}S^z_{ss'}\\
&=\frac{1}{2}\left\{L^x_{\alpha\beta}\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right]+L^y_{\alpha\beta}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right]-L^z_{\alpha\beta}\sin(\theta)\right\}S'^+_{ss'}\\
&+\frac{1}{2}\left\{L^x_{\alpha\beta}\left[\cos(\theta)\cos(\phi)-i\sin(\phi)\right]+L^y_{\alpha\beta}\left[\cos(\theta)\sin(\phi)+i\cos(\phi)\right]-L^z_{\alpha\beta}\sin(\theta)\right\}S'^-_{ss'}\\
&+\left\{L^x_{\alpha\beta}\sin(\theta)\cos(\phi)+L^y_{\alpha\beta}\sin(\theta)\sin(\phi)+L^z_{\alpha\beta}\cos(\theta)\right\}S'^z_{ss'}\ .
\end{aligned} \tag{D.23}$$

In the hamiltonian

$$\begin{aligned}
&\hat{H}_{\mathrm{so}}=\sum_{\substack{m\alpha\beta\\ ss'}}\lambda_m\left\langle m\alpha s\left|\mathbf{L}'_m\cdot\mathbf{S}'_m\right| m\beta s'\right\rangle c^\dagger_{m\alpha s}c_{m\beta s'}\\
&=\sum_{m\alpha\beta}\frac{\lambda_m}{2}\left\{\hat{L}'^-_{\alpha\beta}c^\dagger_{m\alpha\uparrow}c_{m\beta\downarrow}+\hat{L}'^+_{\alpha\beta}c^\dagger_{m\alpha\downarrow}c_{m\beta\uparrow}+\hat{L}'^z_{\alpha\beta}\left(c^\dagger_{m\alpha\uparrow}c_{m\beta\uparrow}-c^\dagger_{m\alpha\downarrow}c_{m\beta\downarrow}\right)\right\}\ .
\end{aligned} \tag{D.24}$$

The first term on the right hand side of the equation above can be written in matrix form using the matrices L^x, L^y and L^z in the combination of the first term of Eq. (D.23). Therefore

$$
\begin{aligned}
\frac{1}{2}\hat{L}'^{-}_{\alpha\beta} =& \frac{1}{2}\begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & -i\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & i\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & -i\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & -i\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & -\sqrt{3}i\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] \\
0 & 0 & 0 & 0 & i\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & i\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & \sqrt{3}i\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0 & 0
\end{pmatrix} \\
&+\frac{1}{2}\begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & i\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -i\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & i\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -i\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & -i\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & \sqrt{3}i\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] \\
0 & 0 & 0 & 0 & 0 & 0 & i\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & -\sqrt{3}i\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0
\end{pmatrix} \\
&+\frac{1}{2}\begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & i\sin(\theta) & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -i\sin(\theta) & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & -2i\sin(\theta) & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & -i\sin(\theta) & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & i\sin(\theta) & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 2i\sin(\theta) & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix} . \\
=& \begin{array}{c}
 \\ s \\ p_x \\ p_y \\ p_z \\ d_{xy} \\ d_{yz} \\ d_{zx} \\ d_{x^2-y^2} \\ d_{3z^2-r^2}
\end{array}
\begin{array}{c}
\begin{array}{c|ccc|ccccc}
s & p_x & p_y & p_z & d_{xy} & d_{yz} & d_{zx} & d_{x^2-y^2} & d_{3z^2-r^2}
\end{array} \\
\left(\begin{array}{c|ccc|ccccc}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\hline
0 & 0 & \frac{1}{2}i\sin(\theta) & \frac{1}{2}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{1}{2}i\sin(\theta) & 0 & \frac{1}{2}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{1}{2}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & -\frac{1}{2}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & 0 & 0 & 0 & 0 & 0 & 0 \\
\hline
0 & 0 & 0 & 0 & 0 & \frac{1}{2}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & \frac{1}{2}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & -i\sin(\theta) & 0 \\
0 & 0 & 0 & 0 & -\frac{1}{2}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & 0 & -\frac{1}{2}i\sin(\theta) & \frac{1}{2}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & \frac{\sqrt{3}}{2}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] \\
0 & 0 & 0 & 0 & -\frac{1}{2}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & \frac{1}{2}i\sin(\theta) & 0 & -\frac{1}{2}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & \frac{\sqrt{3}}{2}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] \\
0 & 0 & 0 & 0 & i\sin(\theta) & -\frac{1}{2}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & \frac{1}{2}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & -\frac{\sqrt{3}}{2}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & -\frac{\sqrt{3}}{2}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & 0 & 0
\end{array}\right)
\end{array}
\end{aligned}
\tag{D.25}
$$

The second term is just the transpose conjugate of this matrix. The third term can also be obtained following the same steps, and we find

$$
\begin{aligned}
\frac{1}{2}\hat{L}'^{z}_{\alpha\beta} = & \frac{1}{2}\begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & -i\sin(\theta)\cos(\phi) & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & i\sin(\theta)\cos(\phi) & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & -i\sin(\theta)\cos(\phi) & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & -i\sin(\theta)\cos(\phi) & -\sqrt{3}i\sin(\theta)\cos(\phi) \\
0 & 0 & 0 & 0 & i\sin(\theta)\cos(\phi) & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & i\sin(\theta)\cos(\phi) & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & \sqrt{3}i\sin(\theta)\cos(\phi) & 0 & 0 & 0
\end{pmatrix} \\
& + \frac{1}{2}\begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & i\sin(\theta)\sin(\phi) & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & -i\sin(\theta)\sin(\phi) & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & i\sin(\theta)\sin(\phi) & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -i\sin(\theta)\sin(\phi) & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & -i\sin(\theta)\sin(\phi) & \sqrt{3}i\sin(\theta)\sin(\phi) \\
0 & 0 & 0 & 0 & 0 & 0 & i\sin(\theta)\sin(\phi) & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & -\sqrt{3}i\sin(\theta)\sin(\phi) & 0 & 0
\end{pmatrix} \\
& + \frac{1}{2}\begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & -i\cos(\theta) & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & i\cos(\theta) & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 2i\cos(\theta) & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & i\cos(\theta) & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & -i\cos(\theta) & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -2i\cos(\theta) & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{pmatrix} \\
= & \begin{array}{c} \\ s \\ p_x \\ p_y \\ p_z \\ d_{xy} \\ d_{yz} \\ d_{zx} \\ d_{x^2-y^2} \\ d_{3z^2-r^2} \end{array}
\begin{array}{c}
\begin{array}{c|ccc|ccccc} s & p_x & p_y & p_z & d_{xy} & d_{yz} & d_{zx} & d_{x^2-y^2} & d_{3z^2-r^2} \end{array} \\
\left(\begin{array}{c|ccc|ccccc}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
\hline
0 & 0 & -\frac{1}{2}i\cos(\theta) & \frac{1}{2}i\sin(\theta)\sin(\phi) & 0 & 0 & 0 & 0 & 0 \\
0 & \frac{1}{2}i\cos(\theta) & 0 & -\frac{1}{2}i\sin(\theta)\cos(\phi) & 0 & 0 & 0 & 0 & 0 \\
0 & -\frac{1}{2}i\sin(\theta)\sin(\phi) & \frac{1}{2}i\sin(\theta)\cos(\phi) & 0 & 0 & 0 & 0 & 0 & 0 \\
\hline
0 & 0 & 0 & 0 & 0 & \frac{1}{2}i\sin(\theta)\sin(\phi) & -\frac{1}{2}i\sin(\theta)\cos(\phi) & i\cos(\theta) & 0 \\
0 & 0 & 0 & 0 & -\frac{1}{2}i\sin(\theta)\sin(\phi) & 0 & \frac{1}{2}i\cos(\theta) & -\frac{1}{2}i\sin(\theta)\cos(\phi) & -\frac{\sqrt{3}}{2}i\sin(\theta)\cos(\phi) \\
0 & 0 & 0 & 0 & \frac{1}{2}i\sin(\theta)\cos(\phi) & -\frac{1}{2}i\cos(\theta) & 0 & -\frac{1}{2}i\sin(\theta)\sin(\phi) & \frac{\sqrt{3}}{2}i\sin(\theta)\sin(\phi) \\
0 & 0 & 0 & 0 & -i\cos(\theta) & \frac{1}{2}i\sin(\theta)\cos(\phi) & \frac{1}{2}i\sin(\theta)\sin(\phi) & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & \frac{\sqrt{3}}{2}i\sin(\theta)\cos(\phi) & -\frac{\sqrt{3}}{2}i\sin(\theta)\sin(\phi) & 0 & 0
\end{array}\right)
\end{array}.
\end{aligned}
\tag{D.26}
$$

The components S^+, S^- and S^z written in terms of S'^+, S'^- and S'^z are

$$
\begin{aligned}
S^+ =& S^x + iS^y \\
=& S'^x \cos(\theta)e^{i\phi} + iS'^y e^{i\phi} + S'^z \sin(\theta)e^{i\phi} \\
=& \frac{1}{2}\left(S'^+ + S^-\right)\cos(\theta)e^{i\phi} + \frac{1}{2}\left(S'^+ - S^-\right)e^{i\phi} + S'^z \sin(\theta)e^{i\phi} \\
=& \frac{1}{2}S'^+\left[\cos(\theta)+1\right]e^{i\phi} + \frac{1}{2}S^-\left[\cos(\theta)-1\right]e^{i\phi} + S'^z \sin(\theta)e^{i\phi} \\
S^- =& \left(S^+\right)^\dagger \\
=& \frac{1}{2}S'^-\left[\cos(\theta)+1\right]e^{-i\phi} + \frac{1}{2}S^+\left[\cos(\theta)-1\right]e^{-i\phi} + S'^z \sin(\theta)e^{-i\phi} \\
S^z =& -\frac{1}{2}\left(S'^+ + S'^-\right)\sin(\theta) + S'^z\cos(\theta)\ .
\end{aligned}
\tag{D.27}
$$

The product $\mathbf{L}\cdot\mathbf{S}$ may be written as

$$
\begin{aligned}
\mathbf{L}\cdot\mathbf{S} =& L^x S^x + L^y S^y + L^z S^z \\
=& \frac{1}{2}\left(L^+S^- + L^-S^+ + 2L^zS^z\right)\ .
\end{aligned}
\tag{D.28}
$$

Thus, substituting the rotated spins,

$$
\begin{aligned}
\mathbf{L}\cdot\mathbf{S} =& \frac{1}{2}\left(L^+S^- + L^-S^+ + 2L^zS^z\right) \\
=& \frac{1}{2}\left\{L^+\left[\frac{1}{2}S'^-\left[\cos(\theta)+1\right]e^{-i\phi} + \frac{1}{2}S^+\left[\cos(\theta)-1\right]e^{-i\phi} + S'^z\sin(\theta)e^{-i\phi}\right]\right. \\
&+ L^-\left[\frac{1}{2}S'^+\left[\cos(\theta)+1\right]e^{i\phi} + \frac{1}{2}S^-\left[\cos(\theta)-1\right]e^{i\phi} + S'^z\sin(\theta)e^{i\phi}\right] \\
&\left.+2L^z\left[-\frac{1}{2}\left(S'^+ + S'^-\right)\sin(\theta) + S'^z\cos(\theta)\right]\right\}\ .
\end{aligned}
\tag{D.29}
$$

Putting together the terms for each spin operator

$$
\begin{aligned}
\mathbf{L}\cdot\mathbf{S} =& S'^+ \\
=& \frac{1}{2}\left\{L^+\left[\frac{1}{2}S'^-\left[\cos(\theta)+1\right]e^{-i\phi} + \frac{1}{2}S^+\left[\cos(\theta)-1\right]e^{-i\phi} + S'^z\sin(\theta)e^{-i\phi}\right]\right. \\
&+ L^-\left[\frac{1}{2}S'^+\left[\cos(\theta)+1\right]e^{i\phi} + \frac{1}{2}S^-\left[\cos(\theta)-1\right]e^{i\phi} + S'^z\sin(\theta)e^{i\phi}\right] \\
&\left.+2L^z\left[-\frac{1}{2}\left(S'^+ + S'^-\right)\sin(\theta) + S'^z\cos(\theta)\right]\right\}\ .
\end{aligned}
\tag{D.30}
$$

$$
L^{+} = \begin{array}{cc}
 & \begin{array}{ccccccccc} s & p_x & p_y & p_z & d_{xy} & d_{yz} & d_{zx} & d_{x^2-y^2} & d_{3z^2-r^2} \end{array} \\
\begin{array}{c} s \\ p_x \\ p_y \\ p_z \\ d_{xy} \\ d_{yz} \\ d_{zx} \\ d_{x^2-y^2} \\ d_{3z^2-r^2} \end{array} &
\begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & -i & 0 & 0 & 0 & 0 & 0 \\
0 & -1 & i & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & -1 & -i & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 & -i & -\sqrt{3}i \\
0 & 0 & 0 & 0 & i & 0 & 0 & 1 & -\sqrt{3} \\
0 & 0 & 0 & 0 & 0 & i & -1 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & \sqrt{3}i & \sqrt{3} & 0 & 0
\end{pmatrix}
\end{array} \tag{D.31}
$$

$$
L^{+} = \begin{array}{cc}
 & \begin{array}{c|ccc|ccccc} s & p_x & p_y & p_z & d_{xy} & d_{yz} & d_{zx} & d_{x^2-y^2} & d_{3z^2-r^2} \end{array} \\
\begin{array}{c} s \\ p_x \\ p_y \\ p_z \\ d_{xy} \\ d_{yz} \\ d_{zx} \\ d_{x^2-y^2} \\ d_{3z^2-r^2} \end{array} &
\left(\begin{array}{c|ccc|ccccc}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ \hline
0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & -i & 0 & 0 & 0 & 0 & 0 \\
0 & -1 & i & 0 & 0 & 0 & 0 & 0 & 0 \\ \hline
0 & 0 & 0 & 0 & 0 & -1 & -i & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 & -i & -\sqrt{3}i \\
0 & 0 & 0 & 0 & i & 0 & 0 & 1 & -\sqrt{3} \\
0 & 0 & 0 & 0 & 0 & i & -1 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & \sqrt{3}i & \sqrt{3} & 0 & 0
\end{array}\right)
\end{array} \tag{D.32}
$$

D.1 f orbitals

TO DO: CORRECT THE ORDER OF THE OPERATORS, AND ALSO THE NORMALIZATION FACTORS FROM WIKIPEDIA'S REAL HARMONIC SPHERICS

The cubic harmonics given in term of the harmonics spherics for the $f-$orbitals are given by

$$\begin{aligned}
f_{z^3} =& N_3^{\mathrm{C}} \frac{z(2z^2-3x^2-3y^2)}{2r^3\sqrt{15}} = Y_3^0 \\
f_{xz^2} =& N_3^{\mathrm{C}} \frac{x(4z^2-x^2-y^2)}{2r^3\sqrt{5}} = \frac{1}{\sqrt{2}}\left(Y_3^1 - Y_3^{-1}\right) \\
f_{yz^2} =& N_3^{\mathrm{C}} \frac{y(4z^2-x^2-y^2)}{r^3\sqrt{5}} = \frac{1}{i\sqrt{2}}\left(Y_3^1 + Y_3^{-1}\right) \\
f_{xyz} =& N_3^{\mathrm{C}} \frac{xyz}{r^3} = \frac{1}{i\sqrt{2}}\left(Y_3^2 - Y_3^{-2}\right) \\
f_{z(x^2-y^2)} =& N_3^{\mathrm{C}} \frac{z\left(x^2-y^2\right)}{2r^3} = \frac{1}{\sqrt{2}}\left(Y_3^2 + Y_3^{-2}\right) \\
f_{x(x^2-3y^2)} =& N_3^{\mathrm{C}} \frac{x\left(x^2-3y^2\right)}{2r^3\sqrt{3}} = \frac{1}{\sqrt{2}}\left(Y_3^3 - Y_3^{-3}\right) \\
f_{y(3x^2-y^2)} =& N_3^{\mathrm{C}} \frac{y\left(3x^2-y^2\right)}{2r^3\sqrt{3}} = \frac{1}{i\sqrt{2}}\left(Y_3^3 + Y_3^{-3}\right)
\end{aligned} \quad , \tag{D.33}$$

where $N_3^{\mathrm{C}} = \left(\frac{105}{4\pi}\right)^{1/2}$.

Calculating $\hat{L}^z|\ell m\rangle = m|\ell m\rangle$ for these orbitals, we have

$$\begin{aligned}
\hat{L}^z f_{z^3} =& 0 \\
\hat{L}^z f_{xz^2} =& \frac{1}{\sqrt{2}}\left(Y_3^1 + Y_3^{-1}\right) = i f_{yz^2} \\
\hat{L}^z f_{yz^2} =& \frac{1}{i\sqrt{2}}\left(Y_3^1 - Y_3^{-1}\right) = -i f_{xz^2} \\
\hat{L}^z f_{xyz} =& \frac{2}{i\sqrt{2}}\left(Y_3^2 + Y_3^{-2}\right) = -2i f_{z(x^2-y^2)} \\
\hat{L}^z f_{z(x^2-y^2)} =& \frac{2}{\sqrt{2}}\left(Y_3^2 - Y_3^{-2}\right) = 2i f_{xyz} \\
\hat{L}^z f_{x(x^2-3y^2)} =& \frac{3}{\sqrt{2}}\left(Y_3^3 + Y_3^{-3}\right) = 3i f_{y(3x^2-y^2)} \\
\hat{L}^z f_{y(3x^2-y^2)} =& \frac{3}{i\sqrt{2}}\left(Y_3^3 - Y_3^{-3}\right) = -3i f_{x(x^2-3y^2)}
\end{aligned} \quad . \tag{D.34}$$

The f-orbital block from L^z matrix is

$$L^z = \begin{array}{c} \\ f_{z^3} \\ f_{xz^2} \\ f_{yz^2} \\ f_{xyz} \\ f_{z(x^2-y^2)} \\ f_{x(x^2-3y^2)} \\ f_{y(3x^2-y^2)} \end{array} \begin{array}{c} \begin{array}{ccccccc} f_{z^3} & f_{xz^2} & f_{yz^2} & f_{xyz} & f_{z(x^2-y^2)} & f_{x(x^2-3y^2)} & f_{y(3x^2-y^2)} \end{array} \\ \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -i & 0 & 0 & 0 & 0 \\ 0 & i & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 2i & 0 & 0 \\ 0 & 0 & 0 & -2i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -3i \\ 0 & 0 & 0 & 0 & 0 & 3i & 0 \end{pmatrix} \end{array} \tag{D.35}$$

To obtain the L^+ matrix, we use the relation $\hat{L}^+|\ell, m\rangle = \sqrt{\ell(\ell+1) - m(m+1)}|\ell, m+1\rangle$ to calculate

$$\begin{aligned}
\hat{L}^+ f_{z^3} &= \sqrt{12} Y_3^1 = \sqrt{6} f_{xz^2} + i\sqrt{6} f_{yz^2} \\
\hat{L}^+ f_{xz^2} &= \sqrt{5} Y_3^2 - \sqrt{6} Y_3^0 = -\sqrt{6} f_{z^3} + i\sqrt{\frac{5}{2}} f_{xyz} + \sqrt{\frac{5}{2}} f_{z(x^2-y^2)} \\
\hat{L}^+ f_{yz^2} &= -i\left(\sqrt{5} Y_3^2 + \sqrt{6} Y_3^0\right) = -i\sqrt{6} f_{z^3} + \sqrt{\frac{5}{2}} f_{xyz} - i\sqrt{\frac{5}{2}} f_{z(x^2-y^2)} \\
\hat{L}^+ f_{xyz} &= -i\left(\sqrt{3} Y_3^3 - \sqrt{5} Y_3^{-1}\right) = -i\sqrt{\frac{5}{2}} f_{xz^2} - \sqrt{\frac{5}{2}} f_{yz^2} - i\sqrt{\frac{3}{2}} f_{x(x^2-3y^2)} + \sqrt{\frac{3}{2}} f_{y(3x^2-y^2)} \ . \\
\hat{L}^+ f_{z(x^2-y^2)} &= \sqrt{3} Y_3^3 + \sqrt{5} Y_3^{-1} = -\sqrt{\frac{5}{2}} f_{xz^2} + i\sqrt{\frac{5}{2}} f_{yz^2} + \sqrt{\frac{3}{2}} f_{x(x^2-3y^2)} + i\sqrt{\frac{3}{2}} f_{y(3x^2-y^2)} \\
\hat{L}^+ f_{x(x^2-3y^2)} &= -\sqrt{3} Y_3^{-2} = i\sqrt{\frac{3}{2}} f_{xyz} - \sqrt{\frac{3}{2}} f_{z(x^2-y^2)} \\
\hat{L}^+ f_{y(3x^2-y^2)} &= -i\sqrt{3} Y_3^{-2} = -\sqrt{\frac{3}{2}} f_{xyz} - i\sqrt{\frac{3}{2}} f_{z(x^2-y^2)}
\end{aligned} \tag{D.36}$$

So, we have

$$L^+ = \begin{array}{c} \\ f_{z^3} \\ f_{xz^2} \\ f_{yz^2} \\ f_{xyz} \\ f_{z(x^2-y^2)} \\ f_{x(x^2-3y^2)} \\ f_{y(3x^2-y^2)} \end{array} \begin{array}{c} \begin{array}{ccccccc} f_{z^3} & f_{xz^2} & f_{yz^2} & f_{xyz} & f_{z(x^2-y^2)} & f_{x(x^2-3y^2)} & f_{y(3x^2-y^2)} \end{array} \\ \begin{pmatrix} 0 & -\sqrt{6} & -i\sqrt{6} & 0 & 0 & 0 & 0 \\ \sqrt{6} & 0 & 0 & -i\sqrt{\frac{5}{2}} & -\sqrt{\frac{5}{2}} & 0 & 0 \\ i\sqrt{6} & 0 & 0 & -\sqrt{\frac{5}{2}} & i\sqrt{\frac{5}{2}} & 0 & 0 \\ 0 & i\sqrt{\frac{5}{2}} & \sqrt{\frac{5}{2}} & 0 & 0 & i\sqrt{\frac{3}{2}} & -\sqrt{\frac{3}{2}} \\ 0 & \sqrt{\frac{5}{2}} & -i\sqrt{\frac{5}{2}} & 0 & 0 & -\sqrt{\frac{3}{2}} & -i\sqrt{\frac{3}{2}} \\ 0 & 0 & 0 & -i\sqrt{\frac{3}{2}} & \sqrt{\frac{3}{2}} & 0 & 0 \\ 0 & 0 & 0 & \sqrt{\frac{3}{2}} & i\sqrt{\frac{3}{2}} & 0 & 0 \end{pmatrix} \end{array} \tag{D.37}$$

and $L^- = [L^+]^\dagger$, i.e.,

$$L^- = \begin{array}{c} \\ f_{z^3} \\ f_{xz^2} \\ f_{yz^2} \\ f_{xyz} \\ f_{z(x^2-y^2)} \\ f_{x(x^2-3y^2)} \\ f_{y(3x^2-y^2)} \end{array} \begin{array}{c} \begin{array}{ccccccc} f_{z^3} & f_{xz^2} & f_{yz^2} & f_{xyz} & f_{z(x^2-y^2)} & f_{x(x^2-3y^2)} & f_{y(3x^2-y^2)} \end{array} \\ \begin{pmatrix} 0 & \sqrt{6} & -i\sqrt{6} & 0 & 0 & 0 & 0 \\ -\sqrt{6} & 0 & 0 & -i\sqrt{\frac{5}{2}} & \sqrt{\frac{5}{2}} & 0 & 0 \\ i\sqrt{6} & 0 & 0 & \sqrt{\frac{5}{2}} & i\sqrt{\frac{5}{2}} & 0 & 0 \\ 0 & i\sqrt{\frac{5}{2}} & -\sqrt{\frac{5}{2}} & 0 & 0 & i\sqrt{\frac{3}{2}} & \sqrt{\frac{3}{2}} \\ 0 & -\sqrt{\frac{5}{2}} & -i\sqrt{\frac{5}{2}} & 0 & 0 & \sqrt{\frac{3}{2}} & -i\sqrt{\frac{3}{2}} \\ 0 & 0 & 0 & -i\sqrt{\frac{3}{2}} & -\sqrt{\frac{3}{2}} & 0 & 0 \\ 0 & 0 & 0 & -\sqrt{\frac{3}{2}} & i\sqrt{\frac{3}{2}} & 0 & 0 \end{pmatrix} \end{array} \tag{D.38}$$

The matrices of the $x-$ and $y-$components of the angular momentum are given by

$$\begin{aligned} L^x =& \frac{1}{2}\left(L^+ + L^-\right) \\ =& \begin{array}{c} \\ f_{z^3} \\ f_{xz^2} \\ f_{yz^2} \\ f_{xyz} \\ f_{z(x^2-y^2)} \\ f_{x(x^2-3y^2)} \\ f_{y(3x^2-y^2)} \end{array} \begin{array}{c} \begin{array}{ccccccc} f_{z^3} & f_{xz^2} & f_{yz^2} & f_{xyz} & f_{z(x^2-y^2)} & f_{x(x^2-3y^2)} & f_{y(3x^2-y^2)} \end{array} \\ \begin{pmatrix} 0 & 0 & -i\sqrt{6} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -i\sqrt{\frac{5}{2}} & 0 & 0 & 0 \\ i\sqrt{6} & 0 & 0 & 0 & i\sqrt{\frac{5}{2}} & 0 & 0 \\ 0 & i\sqrt{\frac{5}{2}} & 0 & 0 & 0 & i\sqrt{\frac{3}{2}} & 0 \\ 0 & 0 & -i\sqrt{\frac{5}{2}} & 0 & 0 & 0 & -i\sqrt{\frac{3}{2}} \\ 0 & 0 & 0 & -i\sqrt{\frac{3}{2}} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & i\sqrt{\frac{3}{2}} & 0 & 0 \end{pmatrix} \end{array} \end{aligned} \tag{D.39}$$

and

$$L^y = \frac{1}{2i}\left(L^+ - L^-\right)$$

$$= \begin{array}{c} \\ f_{z^3} \\ f_{xz^2} \\ f_{yz^2} \\ f_{xyz} \\ f_{z(x^2-y^2)} \\ f_{x(x^2-3y^2)} \\ f_{y(3x^2-y^2)} \end{array} \begin{array}{c} \begin{array}{ccccccc} f_{z^3} & f_{xz^2} & f_{yz^2} & f_{xyz} & f_{z(x^2-y^2)} & f_{x(x^2-3y^2)} & f_{y(3x^2-y^2)} \end{array} \\ \begin{pmatrix} 0 & i\sqrt{6} & 0 & 0 & 0 & 0 & 0 \\ -i\sqrt{6} & 0 & 0 & 0 & i\sqrt{\frac{5}{2}} & 0 & 0 \\ 0 & 0 & 0 & i\sqrt{\frac{5}{2}} & 0 & 0 & 0 \\ 0 & 0 & -i\sqrt{\frac{5}{2}} & 0 & 0 & 0 & i\sqrt{\frac{3}{2}} \\ 0 & -i\sqrt{\frac{5}{2}} & 0 & 0 & 0 & i\sqrt{\frac{3}{2}} & 0 \\ 0 & 0 & 0 & 0 & -i\sqrt{\frac{3}{2}} & 0 & 0 \\ 0 & 0 & 0 & -i\sqrt{\frac{3}{2}} & 0 & 0 & 0 \end{pmatrix} \end{array} \tag{D.40}$$

So, the hamiltonian is composed by the three terms showed in Eq. (D.23). To write them, we need $\frac{1}{2}L'^{-}_{\alpha\beta}$, which is given by

$$
\begin{aligned}
\frac{1}{2}\hat{L}'^{-}_{\alpha\beta} =& \frac{1}{2}L^x\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right]+\frac{1}{2}L^y\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right]-\frac{1}{2}L^z\sin(\theta)\\
=&\frac{1}{2}\begin{pmatrix}
0 & 0 & -i\sqrt{6}\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0 & 0 & 0\\
0 & 0 & 0 & -i\sqrt{\frac{5}{2}}\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0 & 0\\
i\sqrt{6}\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0 & 0 & i\sqrt{\frac{5}{2}}\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0\\
0 & i\sqrt{\frac{5}{2}}\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0 & 0 & i\sqrt{\frac{3}{2}}\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0\\
0 & 0 & -i\sqrt{\frac{5}{2}}\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0 & 0 & -i\sqrt{\frac{3}{2}}\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right]\\
0 & 0 & 0 & -i\sqrt{\frac{3}{2}}\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0 & 0\\
0 & 0 & 0 & 0 & i\sqrt{\frac{3}{2}}\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right] & 0 & 0
\end{pmatrix}\\
&+\frac{1}{2}\begin{pmatrix}
0 & i\sqrt{6}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0 & 0 & 0\\
-i\sqrt{6}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0 & i\sqrt{\frac{5}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0\\
0 & 0 & 0 & i\sqrt{\frac{5}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0\\
0 & 0 & -i\sqrt{\frac{5}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0 & i\sqrt{\frac{3}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right]\\
0 & -i\sqrt{\frac{5}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0 & i\sqrt{\frac{3}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0\\
0 & 0 & 0 & 0 & -i\sqrt{\frac{3}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0\\
0 & 0 & 0 & -i\sqrt{\frac{3}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0
\end{pmatrix}\\
&+\frac{1}{2}\begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & i\sin(\theta) & 0 & 0 & 0 & 0\\
0 & -i\sin(\theta) & 0 & 0 & 0 & 0 & 0\\
0 & 0 & 0 & 0 & -2i\sin(\theta) & 0 & 0\\
0 & 0 & 0 & 2i\sin(\theta) & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 0 & 0 & 3i\sin(\theta)\\
0 & 0 & 0 & 0 & 0 & -3i\sin(\theta) & 0
\end{pmatrix}\\
=&\frac{1}{2}\begin{pmatrix}
0 & \sqrt{6}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & \sqrt{6}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & 0 & 0 & 0 & 0\\
-\sqrt{6}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & 0 & i\sin(\theta) & \sqrt{\frac{5}{2}}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & \sqrt{\frac{5}{2}}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & 0 & 0\\
-\sqrt{6}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & -i\sin(\theta) & 0 & \sqrt{\frac{5}{2}}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & -\sqrt{\frac{5}{2}}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & 0 & 0\\
0 & -\sqrt{\frac{5}{2}}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & -\sqrt{\frac{5}{2}}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & 0 & -2i\sin(\theta) & -\sqrt{\frac{3}{2}}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & \sqrt{\frac{3}{2}}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right]\\
0 & -\sqrt{\frac{5}{2}}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & \sqrt{\frac{5}{2}}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & 2i\sin(\theta) & 0 & \sqrt{\frac{3}{2}}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & \sqrt{\frac{3}{2}}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right]\\
0 & 0 & 0 & \sqrt{\frac{3}{2}}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & -\sqrt{\frac{3}{2}}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & 0 & 3i\sin(\theta)\\
0 & 0 & 0 & -\sqrt{\frac{3}{2}}\left[\cos(\phi)+i\cos(\theta)\sin(\phi)\right] & -\sqrt{\frac{3}{2}}\left[\sin(\phi)-i\cos(\theta)\cos(\phi)\right] & -3i\sin(\theta) & 0
\end{pmatrix}
\end{aligned}
\tag{D.41}
$$

The second term is related to $\frac{1}{2}L'^{+}_{\alpha\beta}$, that can be obtained by the transpose conjugate of the term above. Finally, $L'^{z}_{\alpha\beta}$ is given by (the $\frac{1}{2}$ factor comes from S^z)

$$
\begin{aligned}
\frac{1}{2}\hat{L}'^{z}_{\alpha\beta} =& \frac{1}{2}L^{x}_{\alpha\beta}\sin(\theta)\cos(\phi) + \frac{1}{2}L^{y}_{\alpha\beta}\sin(\theta)\sin(\phi) + \frac{1}{2}L^{z}_{\alpha\beta}\cos(\theta) \\
=& \frac{1}{2}\begin{pmatrix}
0 & 0 & -i\sqrt{6}\sin(\theta)\cos(\phi) & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & -i\sqrt{\frac{5}{2}}\sin(\theta)\cos(\phi) & 0 & 0 & 0 \\
i\sqrt{6}\sin(\theta)\cos(\phi) & 0 & 0 & 0 & i\sqrt{\frac{5}{2}}\sin(\theta)\cos(\phi) & 0 & 0 \\
0 & i\sqrt{\frac{5}{2}}\sin(\theta)\cos(\phi) & 0 & 0 & 0 & i\sqrt{\frac{3}{2}}\sin(\theta)\cos(\phi) & 0 \\
0 & 0 & -i\sqrt{\frac{5}{2}}\sin(\theta)\cos(\phi) & 0 & 0 & 0 & -i\sqrt{\frac{3}{2}}\sin(\theta)\cos(\phi) \\
0 & 0 & 0 & -i\sqrt{\frac{3}{2}}\sin(\theta)\cos(\phi) & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & i\sqrt{\frac{3}{2}}\sin(\theta)\cos(\phi) & 0 & 0
\end{pmatrix} \\
&+ \frac{1}{2}\begin{pmatrix}
0 & i\sqrt{6}\sin(\theta)\sin(\phi) & 0 & 0 & 0 & 0 & 0 \\
-i\sqrt{6}\sin(\theta)\sin(\phi) & 0 & 0 & 0 & i\sqrt{\frac{5}{2}}\sin(\theta)\sin(\phi) & 0 & 0 \\
0 & 0 & 0 & i\sqrt{\frac{5}{2}}\sin(\theta)\sin(\phi) & 0 & 0 & 0 \\
0 & 0 & -i\sqrt{\frac{5}{2}}\sin(\theta)\sin(\phi) & 0 & 0 & 0 & i\sqrt{\frac{3}{2}}\sin(\theta)\sin(\phi) \\
0 & -i\sqrt{\frac{5}{2}}\sin(\theta)\sin(\phi) & 0 & 0 & 0 & i\sqrt{\frac{3}{2}}\sin(\theta)\sin(\phi) & 0 \\
0 & 0 & 0 & 0 & -i\sqrt{\frac{3}{2}}\sin(\theta)\sin(\phi) & 0 & 0 \\
0 & 0 & 0 & -i\sqrt{\frac{3}{2}}\sin(\theta)\sin(\phi) & 0 & 0 & 0
\end{pmatrix} \\
&+ \frac{1}{2}\begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & -i\cos(\theta) & 0 & 0 & 0 & 0 \\
0 & i\cos(\theta) & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 2i\cos(\theta) & 0 & 0 \\
0 & 0 & 0 & -2i\cos(\theta) & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & -3i\cos(\theta) \\
0 & 0 & 0 & 0 & 0 & 3i\cos(\theta) & 0
\end{pmatrix} \\
=& \frac{1}{2}\begin{pmatrix}
0 & i\sqrt{6}\sin(\theta)\sin(\phi) & -i\sqrt{6}\sin(\theta)\cos(\phi) & 0 & 0 & 0 & 0 \\
-i\sqrt{6}\sin(\theta)\sin(\phi) & 0 & -i\cos(\theta) & -i\sqrt{\frac{5}{2}}\sin(\theta)\cos(\phi) & i\sqrt{\frac{5}{2}}\sin(\theta)\sin(\phi) & 0 & 0 \\
i\sqrt{6}\sin(\theta)\cos(\phi) & i\cos(\theta) & 0 & i\sqrt{\frac{5}{2}}\sin(\theta)\sin(\phi) & i\sqrt{\frac{5}{2}}\sin(\theta)\cos(\phi) & 0 & 0 \\
0 & i\sqrt{\frac{5}{2}}\sin(\theta)\cos(\phi) & -i\sqrt{\frac{5}{2}}\sin(\theta)\sin(\phi) & 0 & 2i\cos(\theta) & i\sqrt{\frac{3}{2}}\sin(\theta)\cos(\phi) & i\sqrt{\frac{3}{2}}\sin(\theta)\sin(\phi) \\
0 & -i\sqrt{\frac{5}{2}}\sin(\theta)\sin(\phi) & -i\sqrt{\frac{5}{2}}\sin(\theta)\cos(\phi) & -2i\cos(\theta) & 0 & i\sqrt{\frac{3}{2}}\sin(\theta)\sin(\phi) & -i\sqrt{\frac{3}{2}}\sin(\theta)\cos(\phi) \\
0 & 0 & 0 & -i\sqrt{\frac{3}{2}}\sin(\theta)\cos(\phi) & -i\sqrt{\frac{3}{2}}\sin(\theta)\sin(\phi) & 0 & -3i\cos(\theta) \\
0 & 0 & 0 & -i\sqrt{\frac{3}{2}}\sin(\theta)\sin(\phi) & i\sqrt{\frac{3}{2}}\sin(\theta)\cos(\phi) & 3i\cos(\theta) & 0
\end{pmatrix}
\end{aligned}
\tag{D.42}
$$

$$
\frac{1}{2}L^{y}\left[\cos(\theta)\cos(\phi)+i\sin(\phi)\right]=\frac{1}{2}\begin{pmatrix}
0 & i\sqrt{6}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0 & 0 & 0\\
-i\sqrt{6}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0 & i\sqrt{\frac{5}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0\\
0 & 0 & 0 & i\sqrt{\frac{5}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0\\
0 & 0 & -i\sqrt{\frac{5}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0 & i\sqrt{\frac{3}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right]\\
0 & -i\sqrt{\frac{5}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0 & i\sqrt{\frac{3}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0\\
0 & 0 & 0 & 0 & -i\sqrt{\frac{3}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0\\
0 & 0 & 0 & -i\sqrt{\frac{3}{2}}\left[\cos(\theta)\sin(\phi)-i\cos(\phi)\right] & 0 & 0 & 0
\end{pmatrix}
\tag{D.43}
$$

$$
-\frac{1}{2}L^{z}\sin(\theta)=\frac{1}{2}\begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & i\sin(\theta) & 0 & 0 & 0 & 0\\
0 & -i\sin(\theta) & 0 & 0 & 0 & 0 & 0\\
0 & 0 & 0 & 0 & -2i\sin(\theta) & 0 & 0\\
0 & 0 & 0 & 2i\sin(\theta) & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 0 & 0 & 3i\sin(\theta)\\
0 & 0 & 0 & 0 & 0 & -3i\sin(\theta) & 0
\end{pmatrix}
\tag{D.44}
$$

The full matrix L^z can be written as

$$
L^{z}=\begin{array}{cc}
 & \begin{array}{c|ccc|ccccc|ccccccc}
s & p_x & p_y & p_z & d_{xy} & d_{yz} & d_{zx} & d_{x^2-y^2} & d_{3z^2-r^2} & f_{z^3} & f_{xz^2} & f_{yz^2} & f_{xyz} & f_{z(x^2-y^2)} & f_{x(x^2-3y^2)} & f_{y(3x^2-y^2)}
\end{array}\\
\begin{array}{c}
s\\ p_x\\ p_y\\ p_z\\ d_{xy}\\ d_{yz}\\ d_{zx}\\ d_{x^2-y^2}\\ d_{3z^2-r^2}\\ f_{z^3}\\ f_{xz^2}\\ f_{yz^2}\\ f_{xyz}\\ f_{z(x^2-y^2)}\\ f_{x(x^2-3y^2)}\\ f_{y(3x^2-y^2)}
\end{array} &
\left(\begin{array}{c|ccc|ccccc|ccccccc}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
\hline
0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & 0 & -i & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & -1 & i & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
\hline
0 & 0 & 0 & 0 & 0 & -1 & -i & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 1 & 0 & 0 & -i & -\sqrt{3}i & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & 0 & 0 & i & 0 & 0 & 1 & -\sqrt{3} & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 0 & i & -1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 0 & \sqrt{3}i & \sqrt{3} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
\hline
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -i & 0 & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & i & 0 & 0 & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 2i & 0 & 0\\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -2i & 0 & 0 & 0\\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -3i\\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 3i & 0
\end{array}\right)
\end{array}
\tag{D.45}
$$

Appendix E

Drafts

E.1 Electric Field Perturbation

To describe the Spin Hall Effect, we are going to consider an applied electric field, which couples to the charge density as

$$\hat{H}_E(t) = \int d\mathbf{r}\rho(\mathbf{r},t)\phi(\mathbf{r},t) \ , \tag{E.1}$$

where $\phi(\mathbf{r},t)$ is the scalar potential and is related to the applied electric field as $\mathbf{E}(\mathbf{r},t) = -\nabla\phi(\mathbf{r},t)$. For a collection of particles ℓ, the classical density is given by

$$\rho(\mathbf{r},t) = \sum_{\ell} e_\ell \delta(\mathbf{r} - \mathbf{r}_\ell(t)) \ . \tag{E.2}$$

Let's consider an oscillatory electric field, uniform in space, applied in the $\hat{x}$ direction. It can be written as

$$\mathbf{E}(t) = E\cos(\omega t)\hat{x} \ . \tag{E.3}$$

The scalar potential that gives this electric field may be written as

$$\phi(\mathbf{r},t) = -E\cos(\omega t)x \ . \tag{E.4}$$

So, the hamiltonian is given by

$$H_E(t) = -E\cos(\omega t)\sum_{\ell} e_\ell x_\ell(t) \ , \tag{E.5}$$

where C is a constant, and x_ℓ is the x-component of the position of particle ℓ. With this interaction the current density is

$$
\begin{aligned}
\langle \hat{J}^m_{ij} \rangle (t) =& -\frac{i}{\hbar} \int dt' \Theta(t-t') \left\langle \left[\hat{J}^m_{ij}(t), \hat{H}_E(t') \right] \right\rangle \\
=& \frac{i}{\hbar} \int dt' \Theta(t-t') \left\langle \left[\hat{J}^m_{ij}(t), \sum_\ell e_\ell x_\ell(t') \right] \right\rangle E \cos(\omega t') \\
=& \frac{i}{\hbar} \int dt' \Theta(t-t') Tr \left\{ \rho \left[\hat{J}^m_{ij}(t), \sum_\ell e_\ell x_\ell(t') \right] \right\} E \cos(\omega t') \\
=& -\frac{i}{\hbar} \int dt' \Theta(t-t') Tr \left\{ \left[\rho, \sum_\ell e_\ell x_\ell(t') \right] \hat{J}^m_{ij}(t) \right\} E \cos(\omega t') \ .
\end{aligned} \tag{E.6}
$$

since $Tr\{A[B,C]\} = Tr\{ABC - ACB\} = Tr\{CAB - ACB\} = -Tr\{[A,C]B\}$.

Now we need Kubo's identity

$$
[\rho, \hat{A}(t)] = i\hbar\rho \int_0^\beta d\lambda \dot{\hat{A}}(t - i\lambda\hbar) \ , \tag{E.7}
$$

where $\hat{A}(t)$ is an operator in Heisenberg's picture, i.e.,

$$
\hat{A}(t) = e^{\frac{i\hat{H}t}{\hbar}} \hat{A}(0) e^{-\frac{i\hat{H}t}{\hbar}} \ , \tag{E.8}
$$

and ρ is the density operator

$$
\rho = \frac{e^{-\beta\hat{H}}}{Tr\left\{e^{-\beta\hat{H}}\right\}} \ . \tag{E.9}
$$

This result can be obtained as follows

$$
\begin{aligned}
i\hbar\rho \int_0^\beta d\lambda \dot{\hat{A}}(t - i\lambda\hbar) =& \rho \int_0^\beta d\lambda \left[\hat{A}(t - i\lambda\hbar), \hat{H} \right] \\
=& \rho \int_0^\beta d\lambda \left[e^{\frac{i\hat{H}(-i\lambda\hbar)}{\hbar}} \hat{A}(t) e^{-\frac{i\hat{H}(-i\lambda\hbar)}{\hbar}}, \hat{H} \right] \\
=& \rho \int_0^\beta d\lambda \left[e^{\lambda\hat{H}} \hat{A}(t) e^{-\lambda\hat{H}}, \hat{H} \right] \\
=& \rho \int_0^\beta d\lambda \left\{ e^{\lambda\hat{H}} \hat{A}(t) e^{-\lambda\hat{H}} \hat{H} - \hat{H} e^{\lambda\hat{H}} \hat{A}(t) e^{-\lambda\hat{H}} \right\} \\
=& -\rho \int_0^\beta d\lambda \frac{d}{d\lambda} \left\{ e^{\lambda\hat{H}} \hat{A}(t) e^{-\lambda\hat{H}} \right\} \\
=& -\rho e^{\beta\hat{H}} \hat{A}(t) e^{-\beta\hat{H}} + \rho\hat{A}(t) \\
=& -\hat{A}(t)\rho + \rho\hat{A}(t) \\
=& [\rho, \hat{A}(t)] \ .
\end{aligned} \tag{E.10}
$$

Substituting this result in Eq. (E.6), we get

$$\begin{aligned}\langle \hat{J}_{ij}^m \rangle(t) &= \int dt' \Theta(t-t') \int_0^\beta d\lambda Tr \left\{ \rho \sum_\ell e_\ell \dot{x}_\ell (t' - i\lambda\hbar) \hat{J}_{ij}^m(t) \right\} E \cos(\omega t') \\ &= \int dt' \Theta(t-t') \int_0^\beta d\lambda Tr \left\{ \rho J^{\mathrm{C}} (t' - i\lambda\hbar) \hat{J}_{ij}^m(t) \right\} E \cos(\omega t') \\ &= \int dt' \Theta(t-t') \int_0^\beta d\lambda \left\langle J^{\mathrm{C}} (t' - i\lambda\hbar) \hat{J}_{ij}^m(t) \right\rangle E \cos(\omega t') \ ,\end{aligned} \tag{E.11}$$

where $J^{\mathrm{C}}(t) = \sum_\ell e_\ell \dot{x}_\ell(t)$.

E.1.1 László calculations

Following László Szunyogh calculations, we have:

$$\hat{H}'(t) = \int d\mathbf{r} \hat{\rho}(\mathbf{r}, t) \phi(\mathbf{r}, t), \tag{E.12}$$

where $-\nabla\phi(\mathbf{r}, t) = \mathbf{E}(\mathbf{r}, t)$ and $\hat{\rho}(\mathbf{r}, t)$ is the charge density operator.

Kubo's formula, that we usually write as

$$\delta\langle A \rangle(t) = -\frac{i}{\hbar} \int dt' \Theta(t-t') \left\langle \left[A(t), \hat{H}'(t') \right] \right\rangle \ , \tag{E.13}$$

can also be written as

$$\delta\langle A \rangle(t) = -\frac{i}{\hbar} \int dt' \Theta(t-t') \mathrm{Tr} \left\{ \left[\hat{H}'(t'), \rho_0 \right] A(t) \right\} \ , \tag{E.14}$$

where ρ_0 is the unperturbed density operator.

Using Eq. (E.7), this equation becomes

$$\begin{aligned}\delta\langle A \rangle(t) &= - \int dt' \Theta(t-t') \int_0^\beta d\lambda \mathrm{Tr} \left\{ \rho_0 \dot{\hat{H}}'(t' - i\lambda\hbar) A(t) \right\} \\ - &= \int dt' \Theta(t-t') \int_0^\beta d\lambda \mathrm{Tr} \left\{ \rho_0 \dot{\hat{H}}' A(t - t' + i\lambda\hbar) \right\} \ ,\end{aligned} \tag{E.15}$$

For the same electric field considered before

$$\mathbf{E}(t) = E\cos(\omega t)\hat{x} \ , \tag{E.16}$$

the potential vector can be chosen as

$$\mathbf{A}(t) = -\frac{E}{\omega}\sin(\omega t)\hat{x} \ . \tag{E.17}$$

So, the perturbation hamiltonian given by Eq. (5.210) can be written as

$$\begin{aligned}\hat{H}_A(t) &= -\frac{E}{\omega}\sin(\omega t)\sum_{\ell} e_\ell \dot{x}_\ell \\ &= -\frac{E}{\omega}\sin(\omega t)J^{\mathrm{C}} \ ,\end{aligned} \tag{E.18}$$

E.1.2 Electric Field in tight-binding

If we neglect variations of the electric field on an intra-atomic scale (which is a good approximation, since the electric field is already relatively small), the perturbation can be added in the diagonal elements of the tight-binding hamiltonian as

$$\begin{aligned}\hat{H}_E(t) =& - e\sum_{\substack{ij \\ \sigma}} \phi(\mathbf{R}_i, t)\delta_{ij}c^\dagger_{i\sigma}c_{j\sigma} \\ =& - eE\cos(\omega t)\sum_{i\sigma} x_i c^\dagger_{i\sigma}c_{i\sigma} \ ,\end{aligned} \tag{E.19}$$

Thus, the current density in linear response is given by

$$\begin{aligned}\langle \hat{J}^m_{ij}\rangle(t) =& -\frac{i}{\hbar}\int dt'\Theta(t-t')\left\langle\left[\hat{J}^m_{ij}(t), \hat{H}_E(t')\right]\right\rangle \\ =&\frac{i}{\hbar}\int dt'\Theta(t-t')\sum_{\ell\sigma} e x_\ell \left\langle\left[\hat{J}^m_{ij}(t), c^\dagger_{\ell\sigma}(t')c_{\ell\sigma}(t')\right]\right\rangle E\cos(\omega t') \\ =&\frac{i}{\hbar}\int dt'\Theta(t-t')\sum_{\ell\sigma} e x_\ell \left\langle\left[i\left\{t_{ij}S^m_{ij}(t) - t_{ji}S^m_{ji}(t)\right\}, c^\dagger_{\ell\sigma}(t')c_{\ell\sigma}(t')\right]\right\rangle E\cos(\omega t') \\ =& - i\sum_{\ell\sigma} e x_\ell \int dt' \left\{t_{ij}\chi^{m\sigma}_{ij\ell\ell}(t-t') - t_{ji}\chi^{m\sigma}_{ji\ell\ell}(t-t')\right\} E\cos(\omega t') \ .\end{aligned} \tag{E.20}$$

$$\rho(\mathbf{r}) = e\sum_{\sigma}\psi^{\dagger}_{\sigma}(\mathbf{r})\psi_{\sigma}(\mathbf{r}) \tag{E.21}$$

What's the relation between $\rho(\mathbf{r})$ and ρ_i?

$$\begin{aligned}\psi^{\dagger}_{\sigma}(\mathbf{r}) &= \sum_i \phi^*_i(\mathbf{r})c_{i\sigma} \ , \\ \psi_{\sigma}(\mathbf{r}) &= \sum_j \phi_j(\mathbf{r})c_{j\sigma}\end{aligned} \tag{E.22}$$

$$\begin{aligned}\rho(\mathbf{r}) =& e\sum_{\substack{ij\\ \sigma}} \phi^*_i(\mathbf{r})\phi_j(\mathbf{r})c_{i\sigma}c_{j\sigma} \\ \int \rho(\mathbf{r})d\mathbf{r} =& e\sum_{\substack{ij\\ \sigma}} \int \phi^*_i(\mathbf{r})\phi_j(\mathbf{r})d\mathbf{r}c_{i\sigma}c_{j\sigma} \\ =& e\sum_{\substack{ij\\ \sigma}} \delta_{ij}c_{i\sigma}c_{j\sigma} \\ =& e\sum_{\substack{i\\ \sigma}} c_{i\sigma}c_{i\sigma} \\ =& \sum_i \rho_i\end{aligned} \tag{E.23}$$

$$\begin{aligned}\int_{V_m} \rho(\mathbf{r})d\mathbf{r} =& e\sum_{\substack{ij\\ \sigma}} \int_{V_m} \phi^*_i(\mathbf{r})\phi_j(\mathbf{r})d\mathbf{r}c_{i\sigma}c_{j\sigma} \\ =& e\sum_{\substack{ij\\ \sigma}} \delta_{ij}\delta_{im}c_{i\sigma}c_{j\sigma} \\ =& e\sum_{\sigma} c_{m\sigma}c_{m\sigma} \\ =& \rho_m\end{aligned} \tag{E.24}$$

Continuity equations:

$$\frac{d\rho(\mathbf{r})}{dt} + \nabla\cdot\mathbf{J}^{\mathrm{C}}(\mathbf{r}) = 0 \tag{E.25}$$

$$\frac{d\rho_i}{dt} + \sum_m J^{\mathrm{C}}_{im} = 0 \tag{E.26}$$

$$\begin{aligned}\hat{H}_0 =& \sum_{ij} t_{ij} c_i^\dagger c_j \\ =& \frac{\hat{\mathbf{p}}^2}{2m} + U(\hat{\mathbf{r}})\end{aligned} \tag{E.27}$$

$$\begin{aligned}[\hat{\mathbf{r}}, \hat{H}_0] = \hat{\mathbf{r}}\hat{H}_0 - \hat{H}_0\hat{\mathbf{r}} =& \frac{[\hat{\mathbf{r}}, \hat{\mathbf{p}}^2]}{2m} \\ =& 2\hat{\mathbf{p}}\frac{[\hat{\mathbf{r}}, \hat{\mathbf{p}}]}{2m} \\ =& i\hbar\frac{\hat{\mathbf{p}}}{m}\end{aligned} \tag{E.28}$$

$$\begin{aligned}\langle m|\hat{\mathbf{r}}\hat{H}_0|n\rangle - \langle m|\hat{H}_0\hat{\mathbf{r}}|n\rangle =& i\hbar\frac{\langle m|\hat{\mathbf{p}}|n\rangle}{m} \\ \sum_{ij} t_{ij}\delta_{jn}\langle m|\hat{\mathbf{r}}|i\rangle - \sum_{ij} t_{ij}\delta_{mi}\langle j|\hat{\mathbf{r}}|n\rangle =& i\hbar\frac{\langle m|\hat{\mathbf{p}}|n\rangle}{m} \\ \sum_i \{t_{in}\langle m|\hat{\mathbf{r}}|i\rangle - t_{mi}\langle i|\hat{\mathbf{r}}|n\rangle\} =& i\hbar\frac{\langle m|\hat{\mathbf{p}}|n\rangle}{m}\end{aligned} \tag{E.29}$$

$$
\begin{aligned}
|\mu,\mathbf{k}\rangle &= \sum_{\ell} e^{i\mathbf{k}\cdot\mathbf{R}_\ell}|\mu,\mathbf{R}_\ell\rangle \\
|\mu,\mathbf{k}+\mathbf{q}\rangle &= \sum_{\ell} e^{i(\mathbf{k}+\mathbf{q})\cdot\mathbf{R}_\ell}|\mu,\mathbf{R}_\ell\rangle
\end{aligned}
\tag{E.30}
$$

$$
\begin{aligned}
\langle\mathbf{r}|\mu,\mathbf{k}\rangle &= \phi^{\mu}_{\mathbf{k}}(\mathbf{r}) = \sum_{\ell} e^{i\mathbf{k}\cdot\mathbf{R}_\ell}\phi^{\mu}(\mathbf{r}-\mathbf{R}_\ell) \\
\langle\mathbf{r}|\mu,\mathbf{k}+\mathbf{q}\rangle &= \phi^{\mu}_{\mathbf{k}+\mathbf{q}}(\mathbf{r}) = \sum_{\ell} e^{i(\mathbf{k}+\mathbf{q})\cdot\mathbf{R}_\ell}\phi^{\mu}(\mathbf{r}-\mathbf{R}_\ell)
\end{aligned}
\tag{E.31}
$$

$$
\begin{aligned}
\phi^{\mu}_{\mathbf{k}}(\mathbf{r}+\mathbf{R}) &= \sum_{\ell} e^{i\mathbf{k}\cdot\mathbf{R}_\ell}\phi^{\mu}(\mathbf{r}-(\mathbf{R}_\ell-\mathbf{R})) \\
&= \sum_{\ell'} e^{i\mathbf{k}\cdot(\mathbf{R}_{\ell'}+\mathbf{R})}\phi^{\mu}(\mathbf{r}-\mathbf{R}_{\ell'}) \\
&= e^{i\mathbf{k}\cdot\mathbf{R}}\sum_{\ell'} e^{i\mathbf{k}\cdot\mathbf{R}_{\ell'}}\phi^{\mu}(\mathbf{r}-\mathbf{R}_{\ell'}) \\
&= e^{i\mathbf{k}\cdot\mathbf{R}}\phi^{\mu}_{\mathbf{k}}(\mathbf{r})
\end{aligned}
\tag{E.32}
$$

$$
\begin{aligned}
\phi^{\mu}_{\mathbf{k}+\mathbf{q}}(\mathbf{r}+\mathbf{R}) &= \sum_{\ell} e^{i(\mathbf{k}+\mathbf{q})\cdot\mathbf{R}_\ell}\phi^{\mu}(\mathbf{r}-(\mathbf{R}_\ell-\mathbf{R})) \\
&= \sum_{\ell'} e^{i(\mathbf{k}+\mathbf{q})\cdot(\mathbf{R}_{\ell'}+\mathbf{R})}\phi^{\mu}(\mathbf{r}-\mathbf{R}_{\ell'}) \\
&= e^{i(\mathbf{k}+\mathbf{q})\cdot\mathbf{R}}\sum_{\ell'} e^{i(\mathbf{k}+\mathbf{q})\cdot\mathbf{R}_{\ell'}}\phi^{\mu}(\mathbf{r}-\mathbf{R}_{\ell'}) \\
&= e^{i(\mathbf{k}+\mathbf{q})\cdot\mathbf{R}}\phi^{\mu}_{\mathbf{k}+\mathbf{q}}(\mathbf{r})
\end{aligned}
\tag{E.33}
$$

$$
\begin{aligned}
\psi^{\mu}_{?}(\mathbf{r}) &= \langle\mathbf{r}|e^{i\mathbf{q}\cdot\hat{\mathbf{r}}}|\mu,\mathbf{k}\rangle \\
&= e^{i\mathbf{q}\cdot\mathbf{r}}\phi^{\mu}_{\mathbf{k}}(\mathbf{r}) \\
&= e^{i\mathbf{q}\cdot\mathbf{r}}\sum_{\ell} e^{i\mathbf{k}\cdot\mathbf{R}_\ell}\phi^{\mu}(\mathbf{r}-\mathbf{R}_\ell) \\
&= \sum_{\ell} e^{i\mathbf{q}\cdot(\mathbf{r}-\mathbf{R}_\ell)}e^{i(\mathbf{k}+\mathbf{q})\cdot\mathbf{R}_\ell}\phi^{\mu}(\mathbf{r}-\mathbf{R}_\ell)
\end{aligned}
\tag{E.34}
$$

$$
\begin{aligned}
\psi^{\mu}_{?}(\mathbf{r}+\mathbf{R}) &= e^{i\mathbf{q}\cdot(\mathbf{r}+\mathbf{R})}\phi^{\mu}_{\mathbf{k}}(\mathbf{r}+\mathbf{R}) \\
&= e^{i\mathbf{q}\cdot\mathbf{R}}e^{i\mathbf{q}\cdot\mathbf{r}}e^{i\mathbf{k}\cdot\mathbf{R}}\phi^{\mu}_{\mathbf{k}}(\mathbf{r}) \\
&= e^{i(\mathbf{k}+\mathbf{q})\cdot\mathbf{R}}e^{i\mathbf{q}\cdot\mathbf{r}}\phi^{\mu}_{\mathbf{k}}(\mathbf{r}) \\
&= e^{i(\mathbf{k}+\mathbf{q})\cdot\mathbf{R}}\psi^{\mu}_{?}(\mathbf{r})
\end{aligned}
\tag{E.35}
$$

$$
\Rightarrow e^{i\mathbf{q}\cdot\hat{\mathbf{r}}}|\mu,\mathbf{k}\rangle = |\mu,\mathbf{k}+\mathbf{q}\rangle \tag{E.36}
$$

$$
\begin{aligned}
\chi^{+-\mu\nu\gamma\xi}_{(0)ijkl}(\mathbf{k},\mathbf{k}+\mathbf{Q};\omega) = \frac{i\hbar}{2\pi}\int d\omega' f(\omega') \Big(& G^{\downarrow\downarrow\nu\gamma}_{jk}(\mathbf{k}+\mathbf{Q},\omega'+\omega)\left\{G^{\uparrow\uparrow\xi\mu}_{li}(\mathbf{k},\omega') - \left[G^{\uparrow\uparrow\mu\xi}_{il}(\mathbf{k},\omega')\right]^*\right\} \\
& + \left[G^{\uparrow\uparrow\mu\xi}_{il}(\mathbf{k},\omega'-\omega)\right]^*\left\{G^{\downarrow\downarrow\nu\gamma}_{jk}(\mathbf{k}+\mathbf{Q},\omega') - \left[G^{\downarrow\downarrow\gamma\nu}_{kj}(\mathbf{k}+\mathbf{Q},\omega')\right]^*\right\}\Big) \ .
\end{aligned}
\tag{E.37}
$$

$$
\begin{aligned}
\left[\chi^{+-\mu\nu\gamma\xi}_{(0)ijkl}(\mathbf{k},\mathbf{k}+\mathbf{Q};\omega)\right]^* &= \frac{i\hbar}{2\pi}\int d\omega' f(\omega') \Big(G^{\uparrow\uparrow\mu\xi}_{il}(\mathbf{k},\omega'-\omega)\left\{G^{\downarrow\downarrow\gamma\nu}_{kj}(\mathbf{k}+\mathbf{Q},\omega') - \left[G^{\downarrow\downarrow\nu\gamma}_{jk}(\mathbf{k}+\mathbf{Q},\omega')\right]^*\right\} \\
&\qquad + \left[G^{\downarrow\downarrow\nu\gamma}_{jk}(\mathbf{k}+\mathbf{Q},\omega'+\omega)\right]^*\left\{G^{\uparrow\uparrow\mu\xi}_{il}(\mathbf{k},\omega') - \left[G^{\uparrow\uparrow\xi\mu}_{li}(\mathbf{k},\omega')\right]^*\right\}\Big) \\
&= \chi^{-+\nu\mu\xi\gamma}_{(0)jilk}(\mathbf{k}+\mathbf{Q},\mathbf{k};-\omega) \ .
\end{aligned}
\tag{E.38}
$$

$$
\begin{aligned}
\chi^{\sigma\sigma' ss'\mu\nu\gamma\xi}_{(0)ijkl}(\mathbf{k},\mathbf{k}+\mathbf{Q};0) = \frac{i\hbar}{2\pi}\int d\omega' f(\omega') \Big\{ & G^{\sigma' s\nu\gamma}_{jk}(\mathbf{k}+\mathbf{Q},\omega') G^{s'\sigma\xi\mu}_{li}(\mathbf{k},\omega') \\
& - \left[G^{\sigma s'\mu\xi}_{il}(\mathbf{k},\omega') G^{s\sigma'\gamma\nu}_{kj}(\mathbf{k}+\mathbf{Q},\omega')\right]^*\Big\} \ .
\end{aligned}
\tag{E.39}
$$

The local susceptibility for $\mathbf{Q}=0$ is

$$
\chi^{\sigma\sigma' ss'\mu\nu\gamma\xi}_{(0)ijji}(\mathbf{k},\mathbf{k};0) = \frac{i\hbar}{2\pi}\int d\omega' f(\omega') \left\{ G^{\sigma' s\nu\gamma}_{jj}(\mathbf{k},\omega') G^{s'\sigma\xi\mu}_{ii}(\mathbf{k},\omega') - \left[G^{\sigma s'\mu\xi}_{ii}(\mathbf{k},\omega') G^{s\sigma'\gamma\nu}_{jj}(\mathbf{k},\omega')\right]^*\right\} \ .
\tag{E.40}
$$

Appendix F

Equation of motion

TO DO: I NEED TO CHECK THIS CALCULATIONS - HOW TO WRITE THE EQUATION OF MOTION IF I USE THE TIME-DEPENCENCY ON THE SECOND OPERATOR

Let's write the generalized transverse susceptibility as

$$\chi_{ijk\ell}^{+-\mu\nu\gamma\xi}(t) = \langle\langle S_{ij}^{+\mu\nu}(0); S_{k\ell}^{-\gamma\xi}(-t)\rangle\rangle = -\frac{i}{\hbar}\Theta(t)\langle[S_{ij}^{+\mu\nu}(0), S_{k\ell}^{-\gamma\xi}(-t)]\rangle \ . \tag{F.1}$$

Using Heisenberg equation of motion,

$$\begin{aligned} i\hbar\frac{d}{dt}S_{k\ell}^{-\gamma\xi}(-t) =& - i\hbar\frac{d}{d(-t)}S_{k\ell}^{-\gamma\xi}(-t) \\ =& - [S_{k\ell}^{-\gamma\xi}(-t), \hat{H}] \end{aligned} \tag{F.2}$$

we can obtain

$$\begin{aligned} i\hbar\frac{d}{dt}\chi_{ijk\ell}^{\mu\nu\gamma\xi}(t) =&\delta(t)\langle[S_{ij}^{+\mu\nu}(t), S_{k\ell}^{-\gamma\xi}(0)]\rangle + \frac{i}{\hbar}\Theta(t)\langle[S_{ij}^{+\mu\nu}(0), [S_{k\ell}^{-\gamma\xi}, \hat{H}](-t)]\rangle \\ =&\delta(t)\langle[S_{ij}^{+\mu\nu}(0), S_{k\ell}^{-\gamma\xi}(0)]\rangle + \frac{i}{\hbar}\Theta(t)\langle[S_{ij}^{+\mu\nu}(t), [S_{k\ell}^{-\gamma\xi}, \hat{H}](0)]\rangle \\ =&\delta(t)\langle[S_{ij}^{+\mu\nu}(0), S_{k\ell}^{-\gamma\xi}(0)]\rangle - \langle\langle S_{ij}^{+\mu\nu}; [S_{k\ell}^{-\gamma\xi}, \hat{H}]\rangle\rangle \ . \end{aligned} \tag{F.3}$$

The first term on the right hand side is the same as before, and we write it as

$$[S_{ij}^{+\mu\nu}, S_{k\ell}^{-\gamma\xi}] = D_{ijk\ell}^{\mu\nu\gamma\xi} \ . \tag{F.4}$$

The commutator with the hamiltonian is

$$[S_{k\ell}^{-\gamma\xi}, \hat{H}] = [S_{k\ell}^{-\gamma\xi}, \hat{H}_0] + [S_{k\ell}^{-\gamma\xi}, \hat{H}_{\text{int}}] + [S_{k\ell}^{-\gamma\xi}, \hat{H}_{\text{Z}}] \ . \tag{F.5}$$

The first one, involving $\hat{H}_0$, is

$$[S_{k\ell}^{-\gamma\xi}, \hat{H}_0] = \sum_{\substack{mn\\ \sigma}}\sum_{\alpha\beta} t_{mn}^{\alpha\beta}[c_{k\gamma\downarrow}^{\dagger}c_{\ell\xi\uparrow}, c_{m\alpha\sigma}^{\dagger}c_{n\beta\sigma}] \ , \tag{F.6}$$

where the commutator is

$$[c^\dagger_{k\gamma\downarrow}c_{\ell\xi\uparrow}, c^\dagger_{m\alpha\sigma}c_{n\beta\sigma}] = c^\dagger_{k\gamma\downarrow}c_{n\beta\sigma}\delta_{\ell m}\delta_{\xi\alpha}\delta_{\sigma\uparrow} - c^\dagger_{m\alpha\sigma}c_{\ell\xi\uparrow}\delta_{kn}\delta_{\gamma\beta}\delta_{\sigma\downarrow}\ . \tag{F.7}$$

So, we have

$$\begin{aligned}[][S^{-\gamma\xi}_{k\ell}, \hat{H}_0] &= \sum_{\substack{mn\\ \sigma}}\sum_{\alpha\beta} t^{\alpha\beta}_{mn}\left(c^\dagger_{k\gamma\downarrow}c_{n\beta\sigma}\delta_{\ell m}\delta_{\xi\alpha}\delta_{\sigma\uparrow} - c^\dagger_{m\alpha\sigma}c_{\ell\xi\uparrow}\delta_{kn}\delta_{\gamma\beta}\delta_{\sigma\downarrow}\right)\\ &= \sum_{n\beta} t^{\xi\beta}_{\ell n}c^\dagger_{k\gamma\downarrow}c_{n\beta\uparrow} - \sum_{m\alpha} t^{\alpha\gamma}_{mk}c^\dagger_{m\alpha\downarrow}c_{\ell\xi\uparrow}\ .\end{aligned} \tag{F.8}$$

This term can be written in a more convenient way as

$$[S^{-\gamma\xi}_{k\ell}, \hat{H}_0] = \sum_{m\alpha}\sum_{n\beta}\left(t^{\xi\beta}_{kn}\delta_{mk}\delta_{\gamma\alpha} - t^{\alpha\gamma}_{mk}\delta_{\ell n}\delta_{\xi\beta}\right)S^{-\alpha\beta}_{mn}\ . \tag{F.9}$$

The term involving $\hat{H}_{\text{int}}$ is

$$[S^{-\gamma\xi}_{k\ell}, \hat{H}_{\text{int}}] = \frac{1}{2}\sum_{\substack{m\\ \sigma,\sigma'}}\sum_{\alpha\beta\lambda\eta} U^m_{\alpha\beta\lambda\eta}\left[c^\dagger_{k\gamma\downarrow}c_{\ell\xi\uparrow}, c^\dagger_{m\alpha\sigma}c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}c_{m\lambda\sigma}\right]\ , \tag{F.10}$$

where

$$\begin{aligned}\left[c^\dagger_{m\gamma\downarrow}c_{\ell\xi\uparrow}, c^\dagger_{m\alpha\sigma}c^\dagger_{m\beta\sigma'}c_{m\xi\sigma'}c_{m\gamma\sigma}\right] = {} & c^\dagger_{k\gamma\downarrow}c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}c_{m\lambda\sigma}\delta_{m\ell}\delta_{\alpha\xi}\delta_{\sigma\uparrow}\\ & + c^\dagger_{m\alpha\sigma}c^\dagger_{k\gamma\downarrow}c_{m\eta\sigma'}c_{m\lambda\sigma}\delta_{m\ell}\delta_{\beta\xi}\delta_{\sigma'\uparrow}\\ & - c^\dagger_{m\alpha\sigma}c^\dagger_{m\beta\sigma'}c_{\ell\xi\uparrow}c_{m\lambda\sigma}\delta_{km}\delta_{\gamma\eta}\delta_{\sigma'\downarrow}\\ & - c^\dagger_{m\alpha\sigma}c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}c_{\ell\xi\uparrow}\delta_{km}\delta_{\gamma\lambda}\delta_{\sigma\downarrow}.\end{aligned} \tag{F.11}$$

Therefore

$$\begin{aligned}[][S^{-\gamma\xi}_{k\ell}, \hat{H}_{\text{int}}] = {} & \frac{1}{2}\sum_{\substack{\beta\lambda\eta\\ \sigma'}} U^\ell_{\xi\beta\lambda\eta}c^\dagger_{k\gamma\downarrow}c^\dagger_{\ell\beta\sigma'}c_{\ell\eta\sigma'}c_{\ell\lambda\uparrow} + \frac{1}{2}\sum_{\substack{\alpha\lambda\eta\\ \sigma}} U^\ell_{\alpha\xi\lambda\eta}c^\dagger_{k\gamma\downarrow}c^\dagger_{\ell\alpha\sigma}c_{\ell\lambda\sigma}c_{\ell\eta\uparrow}\\ & - \frac{1}{2}\sum_{\substack{\alpha\beta\lambda\\ \sigma}} U^k_{\alpha\beta\lambda\gamma}c^\dagger_{k\beta\downarrow}c^\dagger_{k\alpha\sigma}c_{k\lambda\sigma}c_{\ell\xi\uparrow} - \frac{1}{2}\sum_{\substack{\alpha\beta\eta\\ \sigma'}} U^k_{\alpha\beta\gamma\eta}c^\dagger_{k\alpha\downarrow}c^\dagger_{k\beta\sigma'}c_{k\eta\sigma'}c_{\ell\xi\uparrow}\ .\end{aligned} \tag{F.12}$$

Changing variables names, we can write

$$\begin{aligned}[][S^{-\gamma\xi}_{k\ell}, \hat{H}_{\text{int}}] = \frac{1}{2}\sum_{\substack{\alpha\beta\lambda\\ \sigma}} \Big(& U^\ell_{\xi\beta\lambda\alpha}c^\dagger_{k\gamma\downarrow}c^\dagger_{\ell\beta\sigma}c_{\ell\alpha\sigma}c_{\ell\lambda\uparrow} + U^\ell_{\alpha\xi\lambda\beta}c^\dagger_{k\gamma\downarrow}c^\dagger_{\ell\alpha\sigma}c_{\ell\lambda\sigma}c_{\ell\beta\uparrow}\\ & -U^k_{\alpha\beta\lambda\gamma}c^\dagger_{k\beta\downarrow}c^\dagger_{k\alpha\sigma}c_{k\lambda\sigma}c_{\ell\xi\uparrow} - U^k_{\alpha\beta\gamma\lambda}c^\dagger_{k\alpha\downarrow}c^\dagger_{k\beta\sigma}c_{k\lambda\sigma}c_{\ell\xi\uparrow}\Big)\ ,\end{aligned} \tag{F.13}$$

or

$$[S^{-\gamma\xi}_{k\ell}, \hat{H}_{\rm int}] = \frac{1}{2}\sum_{\substack{\alpha\beta\lambda\\ \sigma}} \left[\left(U^{\ell}_{\xi\alpha\lambda\beta} + U^{\ell}_{\alpha\xi\beta\lambda}\right) c^\dagger_{k\gamma\downarrow} c^\dagger_{\ell\alpha\sigma} c_{\ell\beta\sigma} c_{\ell\lambda\uparrow} - \left(U^{k}_{\beta\alpha\lambda\gamma} + U^{k}_{\alpha\beta\gamma\lambda}\right) c^\dagger_{k\alpha\downarrow} c^\dagger_{k\beta\sigma} c_{k\lambda\sigma} c_{\ell\xi\uparrow}\right] . \tag{F.14}$$

Using the symmetry of U, i.e.,

$$U_{1234} = U_{2143} \ , \tag{F.15}$$

we can write

$$[S^{-\gamma\xi}_{k\ell}, \hat{H}_{\rm int}] = \sum_{\substack{\alpha\beta\lambda\\ \sigma}} \left(U^{\ell}_{\xi\alpha\lambda\beta} c^\dagger_{k\gamma\downarrow} c^\dagger_{\ell\alpha\sigma} c_{\ell\beta\sigma} c_{\ell\lambda\uparrow} - U^{k}_{\beta\alpha\lambda\gamma} c^\dagger_{k\alpha\downarrow} c^\dagger_{k\beta\sigma} c_{k\lambda\sigma} c_{\ell\xi\uparrow}\right) \ . \tag{F.16}$$

The Zeeman contribution is

$$\begin{aligned}
[S^{-\gamma\xi}_{k\ell}, \hat{H}_{\rm Z}] =& \frac{g\mu_B B}{2} \sum_m \sum_\lambda \left[c^\dagger_{k\gamma\downarrow} c_{\ell\xi\uparrow}, c^\dagger_{m\lambda\uparrow} c_{m\lambda\uparrow} - c^\dagger_{m\lambda\downarrow} c_{m\lambda\downarrow}\right] \\
=& \frac{g\mu_B B}{2} \sum_m \sum_\lambda \left[c^\dagger_{k\gamma\downarrow} c_{m\lambda\uparrow} \delta_{\ell m}\delta_{\xi\lambda} + c^\dagger_{m\lambda\downarrow} c_{\ell\xi\uparrow} \delta_{km}\delta_{\gamma\lambda}\right] \\
=& \hbar\omega_0 S^{-\gamma\xi}_{k\ell} \ ,
\end{aligned} \tag{F.17}$$

where $\hbar\omega_0 = g\mu_B B$.

Gathering all these results in the equation of motion, we obtain

$$\begin{aligned}
i\hbar\frac{d}{dt}\chi^{\mu\nu\gamma\xi}_{ijkl}(t) =& \delta(t)\langle c^\dagger_{i\mu\uparrow} c_{l\xi\uparrow}\delta_{jk}\delta_{\nu\gamma} - c^\dagger_{k\gamma\downarrow} c_{j\nu\downarrow}\delta_{il}\delta_{\mu\xi}\rangle \\
&- \sum_{m\alpha}\sum_{n\beta} \chi^{\mu\nu\alpha\beta}_{ijmn} \left(t^{\xi\beta}_{kn}\delta_{mk}\delta_{\gamma\alpha} - t^{\alpha\gamma}_{mk}\delta_{\ell n}\delta_{\xi\beta}\right) \\
&- \sum_{\substack{\alpha\beta\lambda\\ \sigma}} \langle\langle S^{+\mu\nu}_{ij}; U^{\ell}_{\xi\alpha\lambda\beta} c^\dagger_{k\gamma\downarrow} c^\dagger_{\ell\alpha\sigma} c_{\ell\beta\sigma} c_{\ell\lambda\uparrow} - U^{k}_{\beta\alpha\lambda\gamma} c^\dagger_{k\alpha\downarrow} c^\dagger_{k\beta\sigma} c_{k\lambda\sigma} c_{\ell\xi\uparrow}\rangle\rangle \\
&- \hbar\omega_0 \chi^{\mu\nu\gamma\xi}_{ijkl}(t) \ .
\end{aligned} \tag{F.18}$$

F.1 Random Phase Approximation (RPA)

To obtain the equation of motion in this approximation, we write the interaction term as

$$\begin{aligned}
&\sum_{\substack{\alpha\beta\lambda\\ \sigma}} \left(U^{\ell}_{\xi\alpha\lambda\beta} c^\dagger_{k\gamma\downarrow} c^\dagger_{\ell\alpha\sigma} c_{\ell\beta\sigma} c_{\ell\lambda\uparrow} - U^{k}_{\beta\alpha\lambda\gamma} c^\dagger_{k\alpha\downarrow} c^\dagger_{k\beta\sigma} c_{k\lambda\sigma} c_{\ell\xi\uparrow}\right) \\
=& \sum_{\substack{\alpha\beta\lambda\\ \sigma}} \left(U^{k}_{\beta\alpha\lambda\gamma} c^\dagger_{k\beta\sigma} c_{\ell\xi\uparrow} c^\dagger_{k\alpha\downarrow} c_{k\lambda\sigma} - U^{\ell}_{\xi\alpha\lambda\beta} c^\dagger_{\ell\alpha\sigma} c_{\ell\lambda\uparrow} c^\dagger_{k\gamma\downarrow} c_{\ell\beta\sigma}\right) \ .
\end{aligned} \tag{F.19}$$

Now we linearize this term using

$$c_1^\dagger c_2 c_3^\dagger c_4 \to \langle c_1^\dagger c_2\rangle c_3^\dagger c_4 - \langle c_1^\dagger c_4\rangle c_3^\dagger c_2 + \langle c_3^\dagger c_4\rangle c_1^\dagger c_2 - \langle c_3^\dagger c_2\rangle c_1^\dagger c_4 \ . \tag{F.20}$$

So,

$$\begin{aligned}
&\sum_{\substack{\alpha\beta\lambda\\ \sigma}} U^k_{\beta\alpha\lambda\gamma} c^\dagger_{k\beta\sigma} c_{\ell\xi\uparrow} c^\dagger_{k\alpha\downarrow} c_{k\lambda\sigma} - U^\ell_{\xi\alpha\lambda\beta} c^\dagger_{\ell\alpha\sigma} c_{\ell\lambda\uparrow} c^\dagger_{k\gamma\downarrow} c_{\ell\beta\sigma} \\
&\to \sum_{\alpha\beta\lambda} U^k_{\beta\alpha\lambda\gamma} \left\{ \langle c^\dagger_{k\beta\uparrow} c_{\ell\xi\uparrow}\rangle c^\dagger_{k\alpha\downarrow} c_{k\lambda\uparrow} - \langle c^\dagger_{k\beta\downarrow} c_{k\lambda\downarrow}\rangle c^\dagger_{k\alpha\downarrow} c_{\ell\xi\uparrow} - \langle c^\dagger_{k\beta\uparrow} c_{k\lambda\uparrow}\rangle c^\dagger_{k\alpha\downarrow} c_{\ell\xi\uparrow} + \langle c^\dagger_{k\alpha\downarrow} c_{k\lambda\downarrow}\rangle c^\dagger_{k\beta\downarrow} c_{\ell\xi\uparrow} \right\} \\
&- U^\ell_{\xi\alpha\lambda\beta} \left\{ \langle c^\dagger_{\ell\alpha\uparrow} c_{\ell\lambda\uparrow}\rangle c^\dagger_{k\gamma\downarrow} c_{\ell\beta\uparrow} - \langle c^\dagger_{\ell\alpha\downarrow} c_{\ell\beta\downarrow}\rangle c^\dagger_{k\gamma\downarrow} c_{\ell\lambda\uparrow} - \langle c^\dagger_{\ell\alpha\uparrow} c_{\ell\beta\uparrow}\rangle c^\dagger_{k\gamma\downarrow} c_{\ell\lambda\uparrow} + \langle c^\dagger_{k\gamma\downarrow} c_{\ell\beta\downarrow}\rangle c^\dagger_{\ell\alpha\downarrow} c_{\ell\lambda\uparrow} \right\} \ .
\end{aligned} \tag{F.21}$$

Note that the expectation values of opposite spins operators vanish. To explicit the susceptibility $\chi^{\mu\nu\alpha\beta}_{ijmn}$, we write

$$\begin{aligned}
\sum_{\eta\rho\lambda}\sum_{mn\alpha\beta} U^k_{\rho\eta\lambda\gamma} \Big\{ &\langle c^\dagger_{k\rho\uparrow} c_{\ell\xi\uparrow}\rangle \delta_{mk}\delta_{nk}\delta_{\alpha\eta}\delta_{\beta\lambda} - \langle c^\dagger_{k\rho\downarrow} c_{k\lambda\downarrow}\rangle \delta_{mk}\delta_{n\ell}\delta_{\alpha\eta}\delta_{\beta\xi} \\
&-\langle c^\dagger_{k\rho\uparrow} c_{k\lambda\uparrow}\rangle \delta_{mk}\delta_{n\ell}\delta_{\alpha\eta}\delta_{\beta\xi} + \langle c^\dagger_{k\eta\downarrow} c_{k\lambda\downarrow}\rangle \delta_{mk}\delta_{n\ell}\delta_{\alpha\rho}\delta_{\beta\xi} \Big\} S^{-\alpha\beta}_{mn} \\
-U^\ell_{\xi\eta\lambda\rho} \Big\{ &\langle c^\dagger_{\ell\eta\uparrow} c_{\ell\lambda\uparrow}\rangle \delta_{mk}\delta_{n\ell}\delta_{\alpha\gamma}\delta_{\beta\rho} - \langle c^\dagger_{\ell\eta\downarrow} c_{\ell\rho\downarrow}\rangle \delta_{mk}\delta_{n\ell}\delta_{\alpha\gamma}\delta_{\beta\lambda} \\
&-\langle c^\dagger_{\ell\eta\uparrow} c_{\ell\rho\uparrow}\rangle \delta_{mk}\delta_{n\ell}\delta_{\alpha\gamma}\delta_{\beta\lambda} + \langle c^\dagger_{k\gamma\downarrow} c_{\ell\rho\downarrow}\rangle \delta_{m\ell}\delta_{n\ell}\delta_{\alpha\eta}\delta_{\beta\lambda} \Big\} S^{-\alpha\beta}_{mn} \ .
\end{aligned} \tag{F.22}$$

Reorganizing the terms,

$$\begin{aligned}
&\sum_{\eta\rho\lambda}\sum_{mn\alpha\beta} S^{-\alpha\beta}_{mn} \Big\{ U^k_{\rho\eta\lambda\gamma} \langle c^\dagger_{k\rho\uparrow} c_{\ell\xi\uparrow}\rangle \delta_{mk}\delta_{nk}\delta_{\alpha\eta}\delta_{\beta\lambda} - U^\ell_{\xi\eta\lambda\rho} \langle c^\dagger_{k\gamma\downarrow} c_{\ell\rho\downarrow}\rangle \delta_{m\ell}\delta_{n\ell}\delta_{\alpha\eta}\delta_{\beta\lambda} \\
&- U^k_{\rho\eta\lambda\gamma} \left[\langle c^\dagger_{k\rho\downarrow} c_{k\lambda\downarrow}\rangle \delta_{mk}\delta_{n\ell}\delta_{\alpha\eta}\delta_{\beta\xi} + \langle c^\dagger_{k\rho\uparrow} c_{k\lambda\uparrow}\rangle \delta_{mk}\delta_{n\ell}\delta_{\alpha\eta}\delta_{\beta\xi} - \langle c^\dagger_{k\eta\downarrow} c_{k\lambda\downarrow}\rangle \delta_{mk}\delta_{n\ell}\delta_{\alpha\rho}\delta_{\beta\xi} \right] \\
&-U^\ell_{\xi\eta\lambda\rho} \left[\langle c^\dagger_{\ell\eta\uparrow} c_{\ell\lambda\uparrow}\rangle \delta_{mk}\delta_{n\ell}\delta_{\alpha\gamma}\delta_{\beta\rho} - \langle c^\dagger_{\ell\eta\downarrow} c_{\ell\rho\downarrow}\rangle \delta_{mk}\delta_{n\ell}\delta_{\alpha\gamma}\delta_{\beta\lambda} - \langle c^\dagger_{\ell\eta\uparrow} c_{\ell\rho\uparrow}\rangle \delta_{mk}\delta_{n\ell}\delta_{\alpha\gamma}\delta_{\beta\lambda} \right] \Big\} \ .
\end{aligned} \tag{F.23}$$

The first two terms can be written as

$$\sum_{\rho}\sum_{mn\alpha\beta} S^{-\alpha\beta}_{mn} \left\{ U^k_{\rho\alpha\beta\gamma} \langle c^\dagger_{k\rho\uparrow} c_{\ell\xi\uparrow}\rangle \delta_{mk}\delta_{nk} - U^\ell_{\xi\alpha\beta\rho} \langle c^\dagger_{k\gamma\downarrow} c_{\ell\rho\downarrow}\rangle \delta_{m\ell}\delta_{n\ell} \right\} \ . \tag{F.24}$$

Rewriting the remaining terms,

$$\sum_{\eta\rho\lambda}\sum_{mn\alpha\beta} S^{-\alpha\beta}_{mn}\Big\{U^n_{\xi\eta\lambda\rho}\delta_{mk}\delta_{n\ell}\delta_{\alpha\gamma}\Big[\langle c^\dagger_{n\eta\uparrow}c_{n\rho\uparrow}\rangle\delta_{\beta\lambda}+\langle c^\dagger_{n\eta\downarrow}c_{n\rho\downarrow}\rangle\delta_{\beta\lambda}-\langle c^\dagger_{n\eta\downarrow}c_{n\lambda\downarrow}\rangle\delta_{\beta\rho}\Big]$$
$$-\,U^m_{\rho\eta\lambda\gamma}\delta_{mk}\delta_{n\ell}\delta_{\beta\xi}\Big[\langle c^\dagger_{m\rho\downarrow}c_{m\lambda\downarrow}\rangle\delta_{\alpha\eta}+\langle c^\dagger_{m\rho\uparrow}c_{m\lambda\uparrow}\rangle\delta_{\alpha\eta}-\langle c^\dagger_{m\eta\uparrow}c_{m\lambda\uparrow}\rangle\delta_{\alpha\rho}\Big]\Big\}$$
$$=\sum_{mn}\sum_{\alpha\beta\rho\lambda} S^{-\alpha\beta}_{mn}\Big\{\delta_{mk}\delta_{n\ell}\delta_{\alpha\gamma}\Big[U^n_{\xi\rho\beta\lambda}\langle c^\dagger_{n\rho\uparrow}c_{n\lambda\uparrow}\rangle+U^n_{\xi\rho\beta\lambda}\langle c^\dagger_{n\rho\downarrow}c_{n\lambda\downarrow}\rangle-U^n_{\xi\rho\lambda\beta}\langle c^\dagger_{n\rho\downarrow}c_{n\lambda\downarrow}\rangle\Big]$$
$$-\,\delta_{mk}\delta_{n\ell}\delta_{\beta\xi}\Big[U^m_{\rho\alpha\lambda\gamma}\langle c^\dagger_{m\rho\downarrow}c_{m\lambda\downarrow}\rangle+U^m_{\rho\alpha\lambda\gamma}\langle c^\dagger_{m\rho\uparrow}c_{m\lambda\uparrow}\rangle-U^m_{\alpha\rho\lambda\gamma}\langle c^\dagger_{m\rho\uparrow}c_{m\lambda\uparrow}\rangle\Big]\Big\}\ . \tag{F.25}$$

Using the symmetry of U,

$$\sum_{mn}\sum_{\alpha\beta\rho\lambda} S^{-\alpha\beta}_{mn}\Big\{\delta_{mk}\delta_{n\ell}\delta_{\alpha\gamma}\Big[U^n_{\xi\rho\beta\lambda}\langle c^\dagger_{n\rho\uparrow}c_{n\lambda\uparrow}\rangle+\left(U^n_{\xi\rho\beta\lambda}-U^n_{\rho\xi\beta\lambda}\right)\langle c^\dagger_{n\rho\downarrow}c_{n\lambda\downarrow}\rangle\Big]$$
$$-\delta_{mk}\delta_{n\ell}\delta_{\beta\xi}\Big[U^m_{\alpha\rho\gamma\lambda}\langle c^\dagger_{m\rho\downarrow}c_{m\lambda\downarrow}\rangle+\left(U^m_{\alpha\rho\gamma\lambda}-U^m_{\rho\alpha\gamma\lambda}\right)\langle c^\dagger_{m\rho\uparrow}c_{m\lambda\uparrow}\rangle\Big]\Big\}\ . \tag{F.26}$$

The equation of motion within RPA becomes

$$\left(i\hbar\frac{d}{dt}+\hbar\omega_0\right)\chi^{\mu\nu\gamma\xi}_{ijk\ell}(t)=\delta(t)D^{\mu\nu\gamma\xi}_{ijk\ell}-\sum_{\substack{mn\\ \alpha\beta}}\chi^{\mu\nu\alpha\beta}_{ijmn}\left(K^{\alpha\beta\gamma\xi}_{mnk\ell}+J^{\alpha\beta\gamma\xi}_{mnk\ell}+J'^{\alpha\beta\gamma\xi}_{mnk\ell}\right)\ , \tag{F.27}$$

where

$$D^{\mu\nu\gamma\xi}_{ijk\ell}=\langle c^\dagger_{i\mu\uparrow}c_{\ell\xi\uparrow}\delta_{jk}\delta_{\nu\gamma}-c^\dagger_{k\gamma\downarrow}c_{j\nu\downarrow}\delta_{i\ell}\delta_{\mu\xi}\rangle\ , \tag{F.28a}$$

$$K^{\alpha\beta\gamma\xi}_{mnk\ell}=t^{\xi\beta}_{kn}\delta_{mk}\delta_{\gamma\alpha}-t^{\alpha\gamma}_{mk}\delta_{\ell n}\delta_{\xi\beta}\ , \tag{F.28b}$$

$$J^{\alpha\beta\gamma\xi}_{mnk\ell}=\sum_\rho\Big\{U^k_{\rho\alpha\beta\gamma}\langle c^\dagger_{k\rho\uparrow}c_{\ell\xi\uparrow}\rangle\delta_{mk}\delta_{nk}-U^\ell_{\xi\alpha\beta\rho}\langle c^\dagger_{k\gamma\downarrow}c_{\ell\rho\downarrow}\rangle\delta_{m\ell}\delta_{n\ell}\Big\}\ . \tag{F.28c}$$

$$J'^{\alpha\beta\gamma\xi}_{mnk\ell}=\sum_{\rho\lambda}\delta_{mk}\delta_{n\ell}\delta_{\alpha\gamma}\Big[U^n_{\xi\rho\beta\lambda}\langle c^\dagger_{n\rho\uparrow}c_{n\lambda\uparrow}\rangle+\left(U^n_{\xi\rho\beta\lambda}-U^n_{\rho\xi\beta\lambda}\right)\langle c^\dagger_{n\rho\downarrow}c_{n\lambda\downarrow}\rangle\Big]$$
$$-\,\delta_{mk}\delta_{n\ell}\delta_{\beta\xi}\Big[U^m_{\alpha\rho\gamma\lambda}\langle c^\dagger_{m\rho\downarrow}c_{m\lambda\downarrow}\rangle+\left(U^m_{\alpha\rho\gamma\lambda}-U^m_{\rho\alpha\gamma\lambda}\right)\langle c^\dagger_{m\rho\uparrow}c_{m\lambda\uparrow}\rangle\Big]\ . \tag{F.28d}$$

F.2 Hartree-Fock Approximation

Using the linearization in the hamiltonian

$$\hat H_{\rm int}=\frac{1}{2}\sum_{\substack{m\\ \sigma,\sigma'}}\sum_{\alpha\beta\lambda\eta}U^m_{\alpha\beta\lambda\eta}c^\dagger_{m\alpha\sigma}c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}c_{m\lambda\sigma}\ , \tag{F.29}$$

given by

$$c^\dagger_{m\alpha\sigma}c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}c_{m\lambda\sigma} \rightarrow \langle c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}\rangle c^\dagger_{m\alpha\sigma}c_{m\lambda\sigma} + \langle c^\dagger_{m\alpha\sigma}c_{m\lambda\sigma}\rangle c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'} - \langle c^\dagger_{m\alpha\sigma}c_{m\eta\sigma'}\rangle \delta_{\sigma\sigma'} c^\dagger_{m\beta\sigma'}c_{m\lambda\sigma} - \langle c^\dagger_{m\beta\sigma'}c_{m\lambda\sigma}\rangle \delta_{\sigma\sigma'} c^\dagger_{m\alpha\sigma}c_{m\eta\sigma'} \ , \tag{F.30}$$

we obtain

$$\hat{H}^{\mathrm{HF}}_{\mathrm{int}} = \frac{1}{2}\sum_{\substack{m\\ \sigma,\sigma'}}\sum_{\alpha\beta\lambda\eta} U^m_{\alpha\beta\lambda\eta}\Big\{\langle c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}\rangle c^\dagger_{m\alpha\sigma}c_{m\lambda\sigma} + \langle c^\dagger_{m\alpha\sigma}c_{m\lambda\sigma}\rangle c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'} - \langle c^\dagger_{m\alpha\sigma}c_{m\eta\sigma'}\rangle \delta_{\sigma\sigma'} c^\dagger_{m\beta\sigma'}c_{m\lambda\sigma} - \langle c^\dagger_{m\beta\sigma'}c_{m\lambda\sigma}\rangle \delta_{\sigma\sigma'} c^\dagger_{m\alpha\sigma}c_{m\eta\sigma'}\Big\} \ . \tag{F.31}$$

Changing indices, we can write

$$\hat{H}^{\mathrm{HF}}_{\mathrm{int}} = \frac{1}{2}\sum_{\substack{m\\ \sigma,\sigma'}}\sum_{\alpha\beta\lambda\eta} \Big\{U^m_{\alpha\beta\lambda\eta}\langle c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}\rangle c^\dagger_{m\alpha\sigma}c_{m\lambda\sigma} + U^m_{\beta\alpha\eta\lambda}\langle c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}\rangle c^\dagger_{m\alpha\sigma}c_{m\lambda\sigma} - U^m_{\alpha\beta\lambda\eta}\langle c^\dagger_{m\alpha\sigma'}c_{m\eta\sigma'}\rangle \delta_{\sigma\sigma'} c^\dagger_{m\beta\sigma}c_{m\lambda\sigma} - U^m_{\beta\alpha\eta\lambda}\langle c^\dagger_{m\alpha\sigma'}c_{m\eta\sigma'}\rangle \delta_{\sigma\sigma'} c^\dagger_{m\beta\sigma}c_{m\lambda\sigma}\Big\} \ . \tag{F.32}$$

Using the symmetry of U and changing more indices,

$$\hat{H}^{\mathrm{HF}}_{\mathrm{int}} = \frac{1}{2}\sum_{\substack{m\\ \sigma,\sigma'}}\sum_{\alpha\beta\lambda\eta} \Big\{U^m_{\alpha\beta\lambda\eta}\langle c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}\rangle c^\dagger_{m\alpha\sigma}c_{m\lambda\sigma} + U^m_{\alpha\beta\lambda\eta}\langle c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}\rangle c^\dagger_{m\alpha\sigma}c_{m\lambda\sigma} - U^m_{\beta\alpha\lambda\eta}\langle c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}\rangle \delta_{\sigma\sigma'} c^\dagger_{m\alpha\sigma}c_{m\lambda\sigma} - U^m_{\beta\alpha\lambda\eta}\langle c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}\rangle \delta_{\sigma\sigma'} c^\dagger_{m\alpha\sigma}c_{m\lambda\sigma}\Big\} \ . \tag{F.33}$$

Finally, we end up with

$$\hat{H}^{\mathrm{HF}}_{\mathrm{int}} = \sum_{\substack{m\\ \sigma,\sigma'}}\sum_{\alpha\beta\lambda\eta} \left(U^m_{\alpha\beta\lambda\eta} - U^m_{\beta\alpha\lambda\eta}\delta_{\sigma\sigma'}\right)\langle c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}\rangle c^\dagger_{m\alpha\sigma}c_{m\lambda\sigma} \ . \tag{F.34}$$

We need to calculate

$$[S^{-\gamma\xi}_{k\ell}, \hat{H}^{\mathrm{HF}}_{\mathrm{int}}] = \sum_{\substack{m\\ \sigma,\sigma'}}\sum_{\alpha\beta\lambda\eta} \left(U^m_{\alpha\beta\lambda\eta} - U^m_{\beta\alpha\lambda\eta}\delta_{\sigma\sigma'}\right)\langle c^\dagger_{m\beta\sigma'}c_{m\eta\sigma'}\rangle [c^\dagger_{k\gamma\downarrow}c_{\ell\xi\uparrow}, c^\dagger_{m\alpha\sigma}c_{m\lambda\sigma}] \ . \tag{F.35}$$

The commutator is

$$[c^\dagger_{k\gamma\downarrow}c_{\ell\xi\uparrow}, c^\dagger_{m\alpha\sigma}c_{m\lambda\sigma}] = c^\dagger_{k\gamma\downarrow}c_{m\lambda\sigma}\delta_{m\ell}\delta_{\xi\alpha}\delta_{\sigma\uparrow} - c^\dagger_{m\alpha\sigma}c_{\ell\xi\uparrow}\delta_{mk}\delta_{\gamma\lambda}\delta_{\sigma\downarrow} \ , \tag{F.36}$$

which gives us

$$
\begin{aligned}
[S^{-\gamma\xi}_{k\ell}, \hat{H}^{\rm HF}_{\rm int}] = \sum_{\substack{m\\ \sigma'}} \sum_{\alpha\beta\lambda\eta} \Big\{ & \left(U^m_{\alpha\beta\lambda\eta} - U^m_{\beta\alpha\lambda\eta}\delta_{\uparrow\sigma'}\right) \langle c^\dagger_{m\beta\sigma'} c_{m\eta\sigma'}\rangle c^\dagger_{k\gamma\downarrow} c_{m\lambda\uparrow}\delta_{m\ell}\delta_{\xi\alpha} \\
& - \left(U^m_{\alpha\beta\lambda\eta} - U^m_{\beta\alpha\lambda\eta}\delta_{\downarrow\sigma'}\right) \langle c^\dagger_{m\beta\sigma'} c_{m\eta\sigma'}\rangle c^\dagger_{m\alpha\downarrow} c_{\ell\xi\uparrow}\delta_{mk}\delta_{\gamma\lambda} \Big\} \ .
\end{aligned}
\tag{F.37}
$$

Summing in orbitals

$$
\begin{aligned}
[S^{-\gamma\xi}_{k\ell}, \hat{H}^{\rm HF}_{\rm int}] = \sum_{\substack{m\\ \sigma'}} \sum_{\alpha\beta\eta} \Big\{ & \left(U^m_{\xi\beta\alpha\eta} - U^m_{\beta\xi\alpha\eta}\delta_{\uparrow\sigma'}\right) \langle c^\dagger_{m\beta\sigma'} c_{m\eta\sigma'}\rangle c^\dagger_{k\gamma\downarrow} c_{m\alpha\uparrow}\delta_{m\ell} \\
& - \left(U^m_{\alpha\beta\gamma\eta} - U^m_{\beta\alpha\gamma\eta}\delta_{\downarrow\sigma'}\right) \langle c^\dagger_{m\beta\sigma'} c_{m\eta\sigma'}\rangle c^\dagger_{m\alpha\downarrow} c_{\ell\xi\uparrow}\delta_{mk} \Big\} \ .
\end{aligned}
\tag{F.38}
$$

Summing in spins and rewriting

$$
\begin{aligned}
[S^{-\gamma\xi}_{k\ell}, \hat{H}^{\rm HF}_{\rm int}] &= \sum_{\substack{m\\ \alpha\beta\eta}} \Big\{ U^m_{\xi\beta\alpha\eta}\langle c^\dagger_{m\beta\downarrow} c_{m\eta\downarrow}\rangle + \left(U^m_{\xi\beta\alpha\eta} - U^m_{\beta\xi\alpha\eta}\right) \langle c^\dagger_{m\beta\uparrow} c_{m\eta\uparrow}\rangle \Big\}\, c^\dagger_{k\gamma\downarrow} c_{m\alpha\uparrow}\delta_{m\ell} \\
&\quad - \Big\{ U^m_{\alpha\beta\gamma\eta}\langle c^\dagger_{m\beta\uparrow} c_{m\eta\uparrow}\rangle + \left(U^m_{\alpha\beta\gamma\eta} - U^m_{\beta\alpha\gamma\eta}\right) \langle c^\dagger_{m\beta\downarrow} c_{m\eta\downarrow}\rangle \Big\}\, c^\dagger_{m\alpha\downarrow} c_{\ell\xi\uparrow}\delta_{mk} \\
&= \sum_{\substack{mn\\ \alpha\beta\lambda\eta}} S^{-\alpha\beta}_{mn} \Big\{ \Big[U^n_{\xi\lambda\beta\eta}\langle c^\dagger_{n\lambda\downarrow} c_{n\eta\downarrow}\rangle + \left(U^n_{\xi\lambda\beta\eta} - U^n_{\lambda\xi\beta\eta}\right) \langle c^\dagger_{n\lambda\uparrow} c_{n\eta\uparrow}\rangle \Big] \delta_{n\ell}\delta_{km}\delta_{\gamma\alpha} \\
&\quad - \Big[U^m_{\alpha\lambda\gamma\eta}\langle c^\dagger_{m\lambda\uparrow} c_{m\eta\uparrow}\rangle + \left(U^m_{\alpha\lambda\gamma\eta} - U^m_{\lambda\alpha\gamma\eta}\right) \langle c^\dagger_{m\lambda\downarrow} c_{m\eta\downarrow}\rangle \Big] \delta_{mk}\delta_{n\ell}\delta_{\xi\beta} \Big\} \\
&= \sum_{\substack{mn\\ \alpha\beta}} S^{-\alpha\beta}_{mn} J'^{\alpha\beta\gamma\xi}_{mnk\ell} \ .
\end{aligned}
\tag{F.39}
$$

where $J'^{\alpha\beta\gamma\xi}_{mnk\ell}$ is given by Eq. (F.28d). So, the equation of motion within Hartree-Fock approximation is given by

$$
\left(i\hbar\frac{d}{dt} + \hbar\omega_0\right) {}^{0}\chi^{\mu\nu\gamma\xi}_{ijkl}(t) = \delta(t) D^{\mu\nu\gamma\xi}_{ijkl} - \sum_{\substack{mn\\ \alpha\beta}} \left(K^{\mu\nu\alpha\beta}_{ijmn} + J'^{\mu\nu\alpha\beta}_{ijmn}\right) {}^{0}\chi^{\alpha\beta\gamma\xi}_{mnkl}, \tag{F.40}
$$

where χ^0 represents the susceptibility calculated in this approximation, and the elements of D, K and J' are given by Eqs. F.28a, F.28b and F.28d, respectively.

F.3 Relation between RPA and HF susceptibilities

So, the equations of motion for both approximations can be written as

$$
\left(i\hbar\frac{d}{dt} - \hbar\omega_0\right)\chi(t) = \delta(t)D - \chi(t)\left(K + J + J'\right) \text{ , and} \tag{F.41a}
$$

$$
\left(i\hbar\frac{d}{dt} - \hbar\omega_0\right)\chi^{0}(t) = \delta(t)D - \chi^0(t)\left(K + J'\right) \ . \tag{F.41b}
$$

Fourier transforming these equations

$$\hbar\,(\omega-\omega_0)\,\chi(\omega) = D - \chi(\omega)\left(K+J+J'\right)\ , \tag{F.42a}$$
$$\hbar\,(\omega-\omega_0)\,\chi^0(\omega) = D - \chi^0(\omega)\left(K+J'\right)\ . \tag{F.42b}$$

From them, we can obtain

$$\begin{aligned}\chi(\omega) &= D\left[\hbar\,(\omega-\omega_0)+K+J'\right]^{-1} - \chi(\omega)J\left[\hbar\,(\omega-\omega_0)+K+J'\right]^{-1}\ ,\\ \overset{0}{\chi}(\omega) &= D\left[\hbar\,(\omega+\omega_0)+K+J'\right]^{-1}\ .\end{aligned} \tag{F.43}$$

Substituting the second on the first,

$$\chi(\omega) = \overset{0}{\chi}(\omega) - \chi(\omega)P\overset{0}{\chi}(\omega)\ , \tag{F.44}$$

where $P = JD^{-1} \Rightarrow PD = J$ or

$$\sum_{\substack{ij\\ \mu\nu}} P^{\alpha\beta\mu\nu}_{mnij}\,D^{\mu\nu\gamma\xi}_{ijkl} = J^{\alpha\beta\gamma\xi}_{mnkl}\ . \tag{F.45}$$

Using D and J obtained in Eqs. F.28a and F.28c, we have

$$\begin{aligned}&\sum_{\substack{ij\\ \mu\nu}} P^{\alpha\beta\mu\nu}_{mnij}\,\langle c^\dagger_{i\mu\uparrow}c_{\ell\xi\uparrow}\delta_{jk}\delta_{\nu\gamma} - c^\dagger_{k\gamma\downarrow}c_{j\nu\downarrow}\delta_{i\ell}\delta_{\mu\xi}\rangle\\ &=\sum_{\rho}\left\{U^k_{\rho\alpha\beta\gamma}\langle c^\dagger_{k\rho\uparrow}c_{\ell\xi\uparrow}\rangle\delta_{mk}\delta_{nk} - U^\ell_{\xi\alpha\beta\rho}\langle c^\dagger_{k\gamma\downarrow}c_{\ell\rho\downarrow}\rangle\delta_{m\ell}\delta_{n\ell}\right\}\ .\end{aligned} \tag{F.46}$$

To obtain P, it is easier to rewrite both sides as

$$\begin{aligned}&\sum_{i\mu} P^{\alpha\beta\mu\gamma}_{mnik}\langle c^\dagger_{i\mu\uparrow}c_{\ell\xi\uparrow}\rangle - \sum_{j\nu} P^{\alpha\beta\xi\nu}_{mn\ell j}\langle c^\dagger_{k\gamma\downarrow}c_{j\nu\downarrow}\rangle\\ &=\sum_{i\rho} P^{\alpha\beta\rho\gamma}_{mnik}\langle c^\dagger_{i\rho\uparrow}c_{\ell\xi\uparrow}\rangle - \sum_{j\rho} P^{\alpha\beta\xi\rho}_{mn\ell j}\langle c^\dagger_{k\gamma\downarrow}c_{j\rho\downarrow}\rangle\\ &=\sum_{\rho}\left\{U^k_{\rho\alpha\beta\gamma}\langle c^\dagger_{k\rho\uparrow}c_{\ell\xi\uparrow}\rangle\delta_{mk}\delta_{nk} - U^\ell_{\xi\alpha\beta\rho}\langle c^\dagger_{k\gamma\downarrow}c_{\ell\rho\downarrow}\rangle\delta_{m\ell}\delta_{n\ell}\right\}\\ &=\sum_{i\rho} U^k_{\rho\alpha\beta\gamma}\langle c^\dagger_{i\rho\uparrow}c_{\ell\xi\uparrow}\rangle\delta_{mk}\delta_{nk}\delta_{ik} - \sum_{j\rho} U^\ell_{\xi\alpha\beta\rho}\langle c^\dagger_{k\gamma\downarrow}c_{j\rho\downarrow}\rangle\delta_{m\ell}\delta_{n\ell}\delta_{j\ell}\ .\end{aligned} \tag{F.47}$$

Now we can see that

$$P^{\alpha\beta\mu\nu}_{mnij} = U^j_{\mu\alpha\beta\nu}\delta_{mj}\delta_{nj}\delta_{ij} \tag{F.48}$$

This results in

$$\begin{aligned}\chi^{\mu\nu\gamma\xi}_{ijkl}(\omega) &= \overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{ijkl}(\omega) - \sum_{\substack{mnrs\\ \alpha\beta\eta\rho}} \chi^{\mu\nu\alpha\beta}_{ijmn}(\omega)P^{\alpha\beta\eta\rho}_{mnrs}\overset{0}{\chi}{}^{\eta\rho\gamma\xi}_{rskl}(\omega)\\ &= \overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{ijkl}(\omega) - \sum_{\substack{m\\ \alpha\beta\eta\rho}} \chi^{\mu\nu\alpha\beta}_{ijmm}(\omega)U^m_{\eta\alpha\beta\rho}\overset{0}{\chi}{}^{\eta\rho\gamma\xi}_{mmkl}(\omega)\ .\end{aligned} \tag{F.49}$$

Appendix G

Sum rules

In general, the hamiltonian can be written as:

$$\hat{H} = \begin{pmatrix} H^{\uparrow\uparrow} & H^{\uparrow\downarrow} \\ H^{\downarrow\uparrow} & H^{\downarrow\downarrow} \end{pmatrix} \;, \tag{G.1}$$

or

$$\begin{aligned} \hat{H} =& \hat{H}^0 + \vec{\sigma} \cdot \hat{\vec{H}} \\ =& \sigma^0 \hat{H}^0 + \sigma^x \hat{\mathcal{X}} + \sigma^y \hat{\mathcal{Y}} + \sigma^z \hat{\mathcal{Z}} \;, \end{aligned} \tag{G.2}$$

where σ^0 is the 2×2 identity matrix and $\hat{H}_0$ is the spin-independent. Each component of the hamiltonian can be written as

$$\hat{H}^0 = \frac{\hat{H}^{\uparrow\uparrow} + \hat{H}^{\downarrow\downarrow}}{2} \,,\, \hat{\mathcal{X}} = \frac{\hat{H}^{\uparrow\downarrow} + \hat{H}^{\downarrow\uparrow}}{2} \,,\, \hat{\mathcal{Y}} = i\frac{\hat{H}^{\uparrow\downarrow} - \hat{H}^{\downarrow\uparrow}}{2} \,,\, \hat{\mathcal{Z}} = \frac{\hat{H}^{\uparrow\uparrow} - \hat{H}^{\downarrow\downarrow}}{2} \;. \tag{G.3}$$

We can also write the hamiltonian as

$$\begin{aligned} \hat{H} =& \frac{1}{2}\left(\sigma^{\uparrow} + \sigma^{\downarrow}\right) \hat{H}^0 + \frac{1}{2}\left(\sigma^+ + \sigma^-\right) \hat{\mathcal{X}} + \frac{1}{2i}\left(\sigma^+ - \sigma^-\right) \hat{\mathcal{Y}} + \frac{1}{2}\left(\sigma^{\uparrow} - \sigma^{\downarrow}\right) \hat{\mathcal{Z}} \\ =& \sigma^{\uparrow}\frac{1}{2}\left(\hat{H}^0 + \hat{\mathcal{Z}}\right) + \sigma^{+}\frac{1}{2}\left(\hat{\mathcal{X}} - i\hat{\mathcal{Y}}\right) + \sigma^{-}\frac{1}{2}\left(\hat{\mathcal{X}} + i\hat{\mathcal{Y}}\right) + \sigma^{\downarrow}\frac{1}{2}\left(\hat{H}^0 - \hat{\mathcal{Z}}\right) \\ =& \sigma^+ \hat{\mathcal{P}} + \sigma^{\uparrow} \hat{\mathcal{U}} + \sigma^{\downarrow} \hat{\mathcal{D}} + \sigma^- \hat{\mathcal{M}} \;, \end{aligned} \tag{G.4}$$

where

$$\sigma^{\uparrow} = \begin{pmatrix} 2 & 0 \\ 0 & 0 \end{pmatrix} ,\, \sigma^{+} = \begin{pmatrix} 0 & 2 \\ 0 & 0 \end{pmatrix} ,\, \sigma^{-} = \begin{pmatrix} 0 & 0 \\ 2 & 0 \end{pmatrix} ,\, \sigma^{\downarrow} = \begin{pmatrix} 0 & 0 \\ 0 & 2 \end{pmatrix} . \tag{G.5}$$

$$\hat{\mathcal{P}} = \frac{\hat{H}^{\uparrow\downarrow}}{2} \,,\, \hat{\mathcal{U}} = \frac{\hat{H}^{\uparrow\uparrow}}{2} \,,\, \hat{\mathcal{D}} = \frac{\hat{H}^{\downarrow\downarrow}}{2} \,,\, \hat{\mathcal{M}} = \frac{\hat{H}^{\downarrow\uparrow}}{2} \;. \tag{G.6}$$

We can also write the hamiltonian as

$$\begin{aligned}\hat{H}_{ij}^{\mu\nu} =& H_{ij}^{0\,\mu\nu}\sigma^0 + \boldsymbol{\sigma}\cdot\mathbf{B}_{\mathrm{xc},i}^{\mu\nu}\delta_{ij} + \boldsymbol{\sigma}\cdot\mathbf{B}_{\mathrm{soc},i}^{\mu\nu}\delta_{ij} + \boldsymbol{\sigma}\cdot\mathbf{B}_{\mathrm{ext},i}^{\mu\nu}\delta_{ij}\\ =& H_{ij}^{0\,\mu\nu}\sigma^0 + \boldsymbol{\sigma}\cdot\mathbf{B}_{\mathrm{eff},i}^{\mu\nu}\delta_{ij}\\ =& H_{ij}^{0\,\mu\nu}\sigma^0 + \left[\sigma^x B_{\mathrm{eff},i}^{x\,\mu\nu} + \sigma^y B_{\mathrm{eff},i}^{y\,\mu\nu} + \sigma^z B_{\mathrm{eff},i}^{z\,\mu\nu}\right]\delta_{ij}\ .\ .\end{aligned} \tag{G.7}$$

where $\mathbf{B}_{\mathrm{xc},i}^{\mu\nu} = -\delta_{\mu\nu}\frac{U^i}{2}\langle\mathbf{m}_i\rangle$, with μ and ν restricted to the d orbitals (where $U^i \neq 0$); $\mathbf{B}_{\mathrm{soc},i}^{\mu\nu} = \frac{\lambda_i}{2}\mathbf{L}'_{\mu\nu}$ and $\mathbf{B}_{\mathrm{ext},i}^{\mu\nu} = \frac{g_S\mu_B}{2}\delta_{\mu\nu}\mathbf{B}'$. Comparing with Eq. (G.2), we note that $\hat{\mathcal{X}}_{ij}^{\mu\nu} = B_{\mathrm{eff},i}^{x\,\mu\nu}\delta_{ij}$; $\hat{\mathcal{Y}}_{ij}^{\mu\nu} = B_{\mathrm{eff},i}^{y\,\mu\nu}\delta_{ij}$ and $\hat{\mathcal{Z}}_{ij}^{\mu\nu} = B_{\mathrm{eff},i}^{z\,\mu\nu}\delta_{ij}$.

If we apply a unitary transformation given by the Pauli matrix σ^x (remember that $[\sigma^\mu]^\dagger = \sigma^\mu$), we have

$$\begin{aligned}\overline{\hat{H}}^x =& \sigma^x\hat{H}\sigma^x\\ =& \begin{pmatrix} 0 & 1\\ 1 & 0\end{pmatrix}\begin{pmatrix} H^{\uparrow\uparrow} & H^{\uparrow\downarrow}\\ H^{\downarrow\uparrow} & H^{\downarrow\downarrow}\end{pmatrix}\begin{pmatrix} 0 & 1\\ 1 & 0\end{pmatrix}\\ =& \begin{pmatrix} H^{\downarrow\downarrow} & H^{\downarrow\uparrow}\\ H^{\uparrow\downarrow} & H^{\uparrow\uparrow}\end{pmatrix}\ .\end{aligned} \tag{G.8}$$

If the transformation is given by the $y-$component of the Pauli matrix,

$$\begin{aligned}\overline{\hat{H}}^y =& \sigma^y\hat{H}\sigma^y\\ =& \begin{pmatrix} 0 & -i\\ i & 0\end{pmatrix}\begin{pmatrix} H^{\uparrow\uparrow} & H^{\uparrow\downarrow}\\ H^{\downarrow\uparrow} & H^{\downarrow\downarrow}\end{pmatrix}\begin{pmatrix} 0 & -i\\ i & 0\end{pmatrix}\\ =& \begin{pmatrix} H^{\downarrow\downarrow} & -H^{\downarrow\uparrow}\\ -H^{\uparrow\downarrow} & H^{\uparrow\uparrow}\end{pmatrix}\ .\end{aligned} \tag{G.9}$$

And for σ^z

$$\begin{aligned}\overline{\hat{H}}^z =& \sigma^z\hat{H}\sigma^z\\ =& \begin{pmatrix} 1 & 0\\ 0 & -1\end{pmatrix}\begin{pmatrix} H^{\uparrow\uparrow} & H^{\uparrow\downarrow}\\ H^{\downarrow\uparrow} & H^{\downarrow\downarrow}\end{pmatrix}\begin{pmatrix} 1 & 0\\ 0 & -1\end{pmatrix}\\ =& \begin{pmatrix} H^{\uparrow\uparrow} & -H^{\uparrow\downarrow}\\ -H^{\downarrow\uparrow} & H^{\downarrow\downarrow}\end{pmatrix}\ .\end{aligned} \tag{G.10}$$

The Green functions are given by

$$G = \left(E - \hat{H}\right)^{-1} = \begin{pmatrix} E - H^{\uparrow\uparrow} & -H^{\uparrow\downarrow}\\ -H^{\downarrow\uparrow} & E - H^{\downarrow\downarrow}\end{pmatrix}^{-1}\ . \tag{G.11}$$

If we transform it using σ^x, we obtain

$$\begin{aligned}
\overline{G}^x =&\sigma^x G \sigma^x = \left(E - \hat{\overline{H}}^x\right)^{-1} \\
=&\begin{pmatrix} E - H^{\downarrow\downarrow} & -H^{\downarrow\uparrow} \\ -H^{\uparrow\downarrow} & E - H^{\uparrow\uparrow} \end{pmatrix}^{-1} \\
=&\begin{pmatrix} G^{\downarrow\downarrow} & G^{\downarrow\uparrow} \\ G^{\uparrow\downarrow} & G^{\uparrow\uparrow} \end{pmatrix} .
\end{aligned} \tag{G.12}$$

For σ^y

$$\begin{aligned}
\overline{G}^y =&\sigma^y G \sigma^y = \left(E - \hat{\overline{H}}^y\right)^{-1} \\
=&\begin{pmatrix} E - H^{\downarrow\downarrow} & H^{\downarrow\uparrow} \\ H^{\uparrow\downarrow} & E - H^{\uparrow\uparrow} \end{pmatrix}^{-1} \\
=&\begin{pmatrix} G^{\downarrow\downarrow} & -G^{\downarrow\uparrow} \\ -G^{\uparrow\downarrow} & G^{\uparrow\uparrow} \end{pmatrix} .
\end{aligned} \tag{G.13}$$

and σ^z

$$\begin{aligned}
\overline{G}^z =&\sigma^z G \sigma^z = \left(E - \hat{\overline{H}}^z\right)^{-1} \\
=&\begin{pmatrix} E - H^{\uparrow\uparrow} & H^{\uparrow\downarrow} \\ H^{\downarrow\uparrow} & E - H^{\downarrow\downarrow} \end{pmatrix}^{-1} \\
=&\begin{pmatrix} G^{\uparrow\uparrow} & -G^{\uparrow\downarrow} \\ -G^{\downarrow\uparrow} & G^{\downarrow\downarrow} \end{pmatrix} .
\end{aligned} \tag{G.14}$$

The difference between each transformed hamiltonian and the original one are

$$\begin{aligned}
\Delta H^x =&\hat{\overline{H}}^x - \hat{H} \\
=&\sigma^x \hat{H} \sigma^x - \hat{H} \\
=&\sigma^x \left[\hat{H}, \sigma^x\right] \\
=&-2i\sigma^x \left(\sigma^z \hat{\mathcal{Y}} - \sigma^y \hat{\mathcal{Z}}\right) \\
=&-2\left(\sigma^y \hat{\mathcal{Y}} + \sigma^z \hat{\mathcal{Z}}\right) ,
\end{aligned} \tag{G.15}$$

or, alternatively,

$$\begin{aligned}
\Delta H^x =&\hat{\overline{H}}^x - \hat{H} \\
=&\begin{pmatrix} H^{\downarrow\downarrow} & H^{\downarrow\uparrow} \\ H^{\uparrow\downarrow} & H^{\uparrow\uparrow} \end{pmatrix} - \begin{pmatrix} H^{\uparrow\uparrow} & H^{\uparrow\downarrow} \\ H^{\downarrow\uparrow} & H^{\downarrow\downarrow} \end{pmatrix} \\
=&\begin{pmatrix} H^{\downarrow\downarrow} - H^{\uparrow\uparrow} & H^{\downarrow\uparrow} - H^{\uparrow\downarrow} \\ H^{\uparrow\downarrow} - H^{\downarrow\uparrow} & H^{\uparrow\uparrow} - H^{\downarrow\downarrow} \end{pmatrix} \\
=&\begin{pmatrix} -\hat{\mathcal{Z}} & i\hat{\mathcal{Y}} \\ -i\hat{\mathcal{Y}} & \hat{\mathcal{Z}} \end{pmatrix} \\
=&-2\hat{\mathcal{Y}}\sigma^y - 2\hat{\mathcal{Z}}\sigma^z ,
\end{aligned} \tag{G.16}$$

$$
\begin{aligned}
\Delta H^y =&\hat{\overline{H}}^y - \hat{H} \\
=&\begin{pmatrix} H^{\downarrow\downarrow} & -H^{\downarrow\uparrow} \\ -H^{\uparrow\downarrow} & H^{\uparrow\uparrow} \end{pmatrix} - \begin{pmatrix} H^{\uparrow\uparrow} & H^{\uparrow\downarrow} \\ H^{\downarrow\uparrow} & H^{\downarrow\downarrow} \end{pmatrix} \\
=&\begin{pmatrix} H^{\downarrow\downarrow} - H^{\uparrow\uparrow} & -H^{\downarrow\uparrow} - H^{\uparrow\downarrow} \\ -H^{\uparrow\downarrow} - H^{\downarrow\uparrow} & H^{\uparrow\uparrow} - H^{\downarrow\downarrow} \end{pmatrix} \\
=&\begin{pmatrix} -\hat{\mathcal{Z}} & -\hat{\mathcal{X}} \\ -\hat{\mathcal{X}} & \hat{\mathcal{Z}} \end{pmatrix} \\
=&-2\hat{\mathcal{X}}\sigma^x - 2\hat{\mathcal{Z}}\sigma^z \ ,
\end{aligned}
\tag{G.17}
$$

$$
\begin{aligned}
\Delta H^z =&\hat{\overline{H}}^z - \hat{H} \\
=&\begin{pmatrix} H^{\uparrow\uparrow} & -H^{\uparrow\downarrow} \\ -H^{\downarrow\uparrow} & H^{\downarrow\downarrow} \end{pmatrix} - \begin{pmatrix} H^{\uparrow\uparrow} & H^{\uparrow\downarrow} \\ H^{\downarrow\uparrow} & H^{\downarrow\downarrow} \end{pmatrix} \\
=&\begin{pmatrix} 0 & -2H^{\uparrow\downarrow} \\ -2H^{\downarrow\uparrow} & 0 \end{pmatrix} \\
=&\begin{pmatrix} 0 & -\hat{\mathcal{X}} + i\hat{\mathcal{Y}} \\ -\hat{\mathcal{X}} - i\hat{\mathcal{Y}} & 0 \end{pmatrix} \\
=&-2\hat{\mathcal{X}}\sigma^x - 2\hat{\mathcal{Y}}\sigma^y \ .
\end{aligned}
\tag{G.18}
$$

In our model, the matrices above are given by

$$
\begin{aligned}
\hat{\mathcal{Z}}_m^{\alpha\beta} &= \frac{1}{2}\left(H^{\uparrow\uparrow} - H^{\downarrow\downarrow}\right)_m^{\alpha\beta} \\
&= \frac{\lambda_m}{2} L'^z_{\alpha\beta} - \frac{U^m}{2}\delta_{\alpha\beta}\langle m_m^z\rangle + \delta_{\alpha\beta}\frac{\hbar\omega_z}{2} \\
&= \left(B^{z\,\mathrm{soc}} + B^{z\,\mathrm{xc}} + B^{z\,\mathrm{ext}}\right)_m^{\alpha\beta} \ ,
\end{aligned}
\tag{G.19}
$$

$$
\begin{aligned}
\hat{\mathcal{Y}}_m^{\alpha\beta} =&\frac{i}{2}\left(H^{\uparrow\downarrow} - H^{\downarrow\uparrow}\right)_m^{\alpha\beta} \\
=&i\frac{\lambda_m}{4} L'^-_{\alpha\beta} - \frac{i}{2}U^m\delta_{\alpha\beta}\langle S_m^-\rangle - i\frac{\lambda_m}{4} L'^+_{\alpha\beta} + \frac{i}{2}U^m\delta_{\alpha\beta}\langle S_m^+\rangle \\
=&\frac{\lambda_m}{2} L'^y_{\alpha\beta} - \frac{U^m}{2}\delta_{\alpha\beta}\langle m_m^y\rangle + \delta_{\alpha\beta}\frac{\hbar\omega_y}{2} \\
=&\left(B^{y\,\mathrm{soc}} + B^{y\,\mathrm{xc}} + B^{y\,\mathrm{ext}}\right)_m^{\alpha\beta} \ ,
\end{aligned}
\tag{G.20}
$$

and

$$\begin{aligned}\hat{\mathcal{X}}_m^{\alpha\beta} =&\frac{1}{2}\left(H^{\uparrow\downarrow}+H^{\downarrow\uparrow}\right)_m^{\alpha\beta}\\ =&\frac{\lambda_m}{4}L'^{+}_{\alpha\beta}-\frac{1}{2}U^m\delta_{\alpha\beta}\langle S_m^+\rangle+\frac{\lambda_m}{4}L'^{-}_{\alpha\beta}-\frac{1}{2}U^m\delta_{\alpha\beta}\langle S_m^-\rangle\\ =&\frac{\lambda_m}{2}L'^{x}_{\alpha\beta}-\frac{U^m}{2}\delta_{\alpha\beta}\langle m_m^x\rangle+\delta_{\alpha\beta}\frac{\hbar\omega_x}{2}\\ =&\left(B^{x\text{soc}}+B^{x\text{xc}}+B^{x\text{ext}}\right)_m^{\alpha\beta}\ .\end{aligned} \tag{G.21}$$

It's easy to see that $\left(\hat{\mathcal{Z}}_m^{\beta\alpha}\right)^*=\hat{\mathcal{Z}}_m^{\alpha\beta}$, $\left(\hat{\mathcal{Y}}_m^{\beta\alpha}\right)^*=\hat{\mathcal{Y}}_m^{\alpha\beta}$ and $\left(\hat{\mathcal{X}}_m^{\beta\alpha}\right)^*=\hat{\mathcal{X}}_m^{\alpha\beta}$.

Using Dyson's equation:

$$\begin{aligned}G =&\overline{G}^{\mu}-\overline{G}^{\mu}\Delta H^{\mu}G\\ G-\overline{G}^{\mu} =&-\overline{G}^{\mu}(\overline{H}^{\mu}-H^{\mu})G\\ G-\sigma^{\mu}G\sigma^{\mu} =&\sigma^{\mu}G\sigma^{\mu}\left(H-\sigma^{\mu}H\sigma^{\mu}\right)G\end{aligned}\ . \tag{G.22}$$

Multiplying by σ^μ from the left side (and taking into account that $(\sigma^\mu)^2=1$),

$$\sigma^{\mu}G-G\sigma^{\mu}=G\left(\sigma^{\mu}H-H\sigma^{\mu}\right)G\ . \tag{G.23}$$

Which can be written as

$$[\sigma^{\mu},G]=G\left[\sigma^{\mu},H\right]G\ . \tag{G.24}$$

If instead we use Dyson's equation written as:

$$G=\overline{G}^{\mu}-G\Delta H^{\mu}\overline{G}^{\mu}\ , \tag{G.25}$$

we have

$$\begin{aligned}G-\sigma^{\mu}G\sigma^{\mu} =&G\left(H-\sigma^{\mu}H\sigma^{\mu}\right)\sigma^{\mu}G\sigma^{\mu}\\ G-\sigma^{\mu}G\sigma^{\mu} =&G\left(H\sigma^{\mu}-\sigma^{\mu}H\right)G\sigma^{\mu}\end{aligned}\ . \tag{G.26}$$

Multiplying by σ^μ from the right side this time,

$$G\sigma^{\mu}-\sigma^{\mu}G=G\left(H\sigma^{\mu}-\sigma^{\mu}H\right)G\ , \tag{G.27}$$

or

$$[G,\sigma^{\mu}]=G\left[H,\sigma^{\mu}\right]G\ . \tag{G.28}$$

This is the same result as Eq. (G.24).

The Green function can be decomposed in terms of the Pauli matrices and the identity σ^0 as

$$\begin{aligned}G =&\begin{pmatrix}G^{\uparrow\uparrow} & G^{\uparrow\downarrow}\\ G^{\downarrow\uparrow} & G^{\downarrow\downarrow}\end{pmatrix}\\ =&\sigma^0G^0+\sigma^xG^x+\sigma^yG^y+\sigma^zG^z\end{aligned}\ , \tag{G.29}$$

where

$$\begin{aligned} G^0 &= \frac{G^{\uparrow\uparrow} + G^{\downarrow\downarrow}}{2} \\ G^x &= \frac{G^{\uparrow\downarrow} + G^{\downarrow\uparrow}}{2} \\ G^y &= i\frac{G^{\uparrow\downarrow} - G^{\downarrow\uparrow}}{2} \\ G^z &= \frac{G^{\uparrow\uparrow} - G^{\downarrow\downarrow}}{2} \end{aligned} . \tag{G.30}$$

Substituting in Eq. (G.24),

$$\begin{aligned} &\left[\sigma^x, \sigma^0 G^0 + \sigma^x G^x + \sigma^y G^y + \sigma^z G^z\right] = \\ &\qquad 2i\left(\sigma^z G^y - \sigma^y G^z\right) = G\left[\sigma^x, H\right] G \\ &\left[\sigma^y, \sigma^0 G^0 + \sigma^x G^x + \sigma^y G^y + \sigma^z G^z\right] = \\ &\qquad 2i\left(-\sigma^z G^x + \sigma^x G^z\right) = G\left[\sigma^y, H\right] G \\ &\left[\sigma^z, \sigma^0 G^0 + \sigma^x G^x + \sigma^y G^y + \sigma^z G^z\right] = \\ &\qquad 2i\left(\sigma^y G^x - \sigma^x G^y\right) = G\left[\sigma^z, H\right] G \end{aligned} . \tag{G.31}$$

Expanding the commutators on the right side, we can obtain

$$\begin{aligned} \left(\sigma^z G^y - \sigma^y G^z\right) &= G\left(\sigma^z \hat{\mathcal{Y}} - \sigma^y \hat{\mathcal{Z}}\right) G \\ \left(\sigma^z G^x - \sigma^x G^z\right) &= G\left(\sigma^z \hat{\mathcal{X}} - \sigma^x \hat{\mathcal{Z}}\right) G \\ \left(\sigma^y G^x - \sigma^x G^y\right) &= G\left(\sigma^y \hat{\mathcal{X}} - \sigma^x \hat{\mathcal{Y}}\right) G \end{aligned} . \tag{G.32}$$

These sum rules can be used to obtain relations for the magnetization components with static limit of the susceptibilities. To obtain each component of the magnetization, we can multiply by the Pauli matrix that multiplies the Green function of the desired magnetization component and use the identity

$$\sigma^a \sigma^b = i \sum_c \varepsilon_{abc}\, \sigma^c + \delta_{ab} \sigma^0 \ , \tag{G.33}$$

taking the trace of the resulting equation. For example, to obtain the $z-$component of the magnetization, we can either multiply the first equation by σ^y to get

$$\left(i\sigma^x G^y - \sigma^0 G^z\right) = \sigma^y G\left(\sigma^z \hat{\mathcal{Y}} - \sigma^y \hat{\mathcal{Z}}\right) G \ , \tag{G.34}$$

or we can multiply the second equation by σ^x, obtaining

$$\left(-i\sigma^y G^x - \sigma^0 G^z\right) = \sigma^x G\left(\sigma^z \hat{\mathcal{X}} - \sigma^x \hat{\mathcal{Z}}\right) G \ . \tag{G.35}$$

Taking the trace of the equations above, we have

$$\begin{aligned}-2G^z &= \mathrm{Tr}\left\{\sigma^y G\left(\sigma^z\hat{\mathcal{Y}} - \sigma^y\hat{\mathcal{Z}}\right)G\right\}\\ -2G^z &= \mathrm{Tr}\left\{\sigma^x G\left(\sigma^z\hat{\mathcal{X}} - \sigma^x\hat{\mathcal{Z}}\right)G\right\}\end{aligned}. \tag{G.36}$$

For the $y-$component of the magnetization, the equations that we have are

$$\begin{aligned}\left(\sigma^0 G^y + i\sigma^x G^z\right) =&\sigma^z G\left(\sigma^z\hat{\mathcal{Y}} - \sigma^y\hat{\mathcal{Z}}\right)G\\ \left(i\sigma^z G^x - \sigma^0 G^y\right) =&\sigma^x G\left(\sigma^y\hat{\mathcal{X}} - \sigma^x\hat{\mathcal{Y}}\right)G\end{aligned}. \tag{G.37}$$

Taking the trace

$$\begin{aligned}2G^y &= \mathrm{Tr}\left\{\sigma^z G\left(\sigma^z\hat{\mathcal{Y}} - \sigma^y\hat{\mathcal{Z}}\right)G\right\}\\ -2G^y &= \mathrm{Tr}\left\{\sigma^x G\left(\sigma^y\hat{\mathcal{X}} - \sigma^x\hat{\mathcal{Y}}\right)G\right\}\end{aligned}. \tag{G.38}$$

Finally, to obtain the $x-$component of the magnetization, we can either multiply the third equation by σ^y or the second equation by σ^z to get

$$\begin{aligned}\left(\sigma^0 G^x + i\sigma^z G^y\right) =&\sigma^y G\left(\sigma^y\hat{\mathcal{X}} - \sigma^x\hat{\mathcal{Y}}\right)G\\ \left(\sigma^0 G^x - i\sigma^y G^z\right) =&\sigma^z G\left(\sigma^z\hat{\mathcal{X}} - \sigma^x\hat{\mathcal{Z}}\right)G\end{aligned}. \tag{G.39}$$

Tracing over spins,

$$\begin{aligned}2G^x =&\mathrm{Tr}\left\{\sigma^y G\sigma^y\hat{\mathcal{X}}G\right\} - \mathrm{Tr}\left\{\sigma^y G\sigma^x\hat{\mathcal{Y}}G\right\}\\ 2G^x =&\mathrm{Tr}\left\{\sigma^z G\sigma^z\hat{\mathcal{X}}G\right\} - \mathrm{Tr}\left\{\sigma^z G\sigma^x\hat{\mathcal{Z}}G\right\}\end{aligned}. \tag{G.40}$$

In components, the most general form of the above equations becomes

$$\begin{aligned}2G^{x\,\nu\mu}_{ji} =&\sum_{\substack{k\\ \gamma\xi}}\mathrm{Tr}\left\{\sigma^y G^{\nu\gamma}_{jk}\sigma^y\hat{\mathcal{X}}^{\gamma\xi}_k G^{\xi\mu}_{ki}\right\} - \mathrm{Tr}\left\{\sigma^y G^{\nu\gamma}_{jk}\sigma^x\hat{\mathcal{Y}}^{\gamma\xi}_k G^{\xi\mu}_{ki}\right\}\\ 2G^{x\,\nu\mu}_{ji} =&\sum_{\substack{k\\ \gamma\xi}}\mathrm{Tr}\left\{\sigma^z G^{\nu\gamma}_{jk}\sigma^z\hat{\mathcal{X}}^{\gamma\xi}_k G^{\xi\mu}_{ki}\right\} - \mathrm{Tr}\left\{\sigma^z G^{\nu\gamma}_{jk}\sigma^x\hat{\mathcal{Z}}^{\gamma\xi}_k G^{\xi\mu}_{ki}\right\}\end{aligned} \tag{G.41}$$

where the trace is taken on spin space. This expression is valid for both retarded and advanced Green functions. To relate with the correlation function obtained in Eq. (1.58), we can write the integral of the imaginary part (defined as in Eq. (C.12)) of left-hand side as

$$\begin{aligned}-\frac{\hbar}{2i\pi}\int d\omega' f(\omega')2\left\{G^{x\,\nu\mu}_{ji}(\omega'+i0^+) - G^{x\,\nu\mu}_{ji}(\omega'-i0^+)\right\} =& -\frac{\hbar}{\pi}\Im\int d\omega' f(\omega')2G^{x\,\nu\mu}_{ji}(\omega')\\ =& -\frac{\hbar}{\pi}\Im\int d\omega' f(\omega')\mathrm{Tr}\left[\boldsymbol{\sigma}^x\mathbf{G}^{\nu\mu}_{ji}(\omega')\right]\\ =&2\langle S^{x\,\mu\nu}_{ij}\rangle\end{aligned}. \tag{G.42}$$

On the right side, the first term becomes

$$-\frac{\hbar}{2i\pi}\int d\omega' f(\omega')\sum_{\substack{k\\ \gamma\xi}}\mathrm{Tr}\left\{\sigma^y G^{\nu\gamma}_{jk}(\omega'+i0^+)\sigma^y\hat{\mathcal{X}}^{\gamma\xi}_k G^{\xi\mu}_{ki}(\omega'+i0^+)-\sigma^y G^{\nu\gamma}_{jk}(\omega'-i0^+)\sigma^y\hat{\mathcal{X}}^{\gamma\xi}_k G^{\xi\mu}_{ki}(\omega'-i0^+)\right\}$$

$$\sum_{\substack{k\\ \gamma\xi}}-\frac{\hbar}{\pi}\Im\int d\omega' f(\omega')\mathrm{Tr}\left\{\sigma^y G^{\nu\gamma}_{jk}(\omega')\sigma^y\hat{\mathcal{X}}^{\gamma\xi}_k G^{\xi\mu}_{ki}(\omega')\right\}=4\sum_{\substack{k\\ \gamma\xi}}\chi^{yy\,\mu\nu\gamma\xi}_{ijkk}(0)\hat{\mathcal{X}}^{\gamma\xi}_k \tag{G.43}$$

where we have identified the susceptibility given by Eq. (C.43). Finally, the sum rules for the $x-$component of the spin density can be written as

$$\begin{aligned}\langle S^{x\,\mu\nu}_{ij}\rangle =&2\sum_{\substack{k\\ \gamma\xi}}\left\{\chi^{yy\,\mu\nu\gamma\xi}_{ijkk}(0)\hat{\mathcal{X}}^{\gamma\xi}_k-\chi^{yx\,\mu\nu\gamma\xi}_{ijkk}(0)\hat{\mathcal{Y}}^{\gamma\xi}_k\right\}\\ \langle S^{x\,\mu\nu}_{ij}\rangle =&2\sum_{\substack{k\\ \gamma\xi}}\left\{\chi^{zz\,\mu\nu\gamma\xi}_{ijkk}(0)\hat{\mathcal{X}}^{\gamma\xi}_k-\chi^{zx\,\mu\nu\gamma\xi}_{ijkk}(0)\hat{\mathcal{Z}}^{\gamma\xi}_k\right\}\end{aligned}\ , \tag{G.44}$$

while for $y-$components, they are

$$\begin{aligned}\langle S^{y\,\mu\nu}_{ij}\rangle =&2\sum_{\substack{k\\ \gamma\xi}}\left\{\chi^{zz\,\mu\nu\gamma\xi}_{ijkk}(0)\hat{\mathcal{Y}}^{\gamma\xi}_k-\chi^{zy\,\mu\nu\gamma\xi}_{ijkk}(0)\hat{\mathcal{Z}}^{\gamma\xi}_k\right\}\\ -\langle S^{y\,\mu\nu}_{ij}\rangle =&2\sum_{\substack{k\\ \gamma\xi}}\left\{\chi^{xy\,\mu\nu\gamma\xi}_{ijkk}(0)\hat{\mathcal{X}}^{\gamma\xi}_k-\chi^{xx\,\mu\nu\gamma\xi}_{ijkk}(0)\hat{\mathcal{Y}}^{\gamma\xi}_k\right\}\end{aligned}\ , \tag{G.45}$$

and the $z-$components

$$\begin{aligned}-\langle S^{z\,\mu\nu}_{ij}\rangle =&2\sum_{\substack{k\\ \gamma\xi}}\left\{\chi^{yz\,\mu\nu\gamma\xi}_{ijkk}(0)\hat{\mathcal{Y}}^{\gamma\xi}_k-\chi^{yy\,\mu\nu\gamma\xi}_{ijkk}(0)\hat{\mathcal{Z}}^{\gamma\xi}_k\right\}\\ -\langle S^{z\,\mu\nu}_{ij}\rangle =&2\sum_{\substack{k\\ \gamma\xi}}\left\{\chi^{xz\,\mu\nu\gamma\xi}_{ijkk}(0)\hat{\mathcal{X}}^{\gamma\xi}_k-\chi^{xx\,\mu\nu\gamma\xi}_{ijkk}(0)\hat{\mathcal{Z}}^{\gamma\xi}_k\right\}\end{aligned}\ . \tag{G.46}$$

The spin current operator is given by Eq. (5.6). So, the ground state spin current with $x-$polarization can be written as

$$\begin{aligned}\langle\hat{I}^x_{ij}\rangle =&i\sum_{\mu\nu}\left\{t^{\mu\nu}_{ij}\langle S^{x\,\mu\nu}_{ij}\rangle-t^{\nu\mu}_{ji}\langle S^{x\,\nu\mu}_{ji}\rangle\right\}\\ =&i\sum_{\mu\nu}\left\{t^{\mu\nu}_{ij}\langle S^{m\,\mu\nu}_{ij}\rangle-t^{\nu\mu}_{ji}\langle S^{m\,\nu\mu}_{ji}\rangle\right\}\ ,\end{aligned} \tag{G.47}$$

Using the continuity equation in equilibrium, given by Eq. (5.7),

$$\sum_j \mathbf{I}_{ij} =\boldsymbol{\tau}_i\ , \tag{G.48}$$

where the torque is given by Eq. (5.17). The components of the torque are given by

$$\begin{aligned}
\tau_i^x &= \left(\hat{S}_i^y B_{\text{eff},i}^z - \hat{S}_i^z B_{\text{eff},i}^y\right) \\
\tau_i^y &= \left(\hat{S}_i^z B_{\text{eff},i}^x - \hat{S}_i^x B_{\text{eff},i}^z\right) \ , \\
\tau_i^z &= \left(\hat{S}_i^x B_{\text{eff},i}^y - \hat{S}_i^y B_{\text{eff},i}^x\right)
\end{aligned} \tag{G.49}$$

Therefore, using the results above, we can write

$$\begin{aligned}
\sum_j \langle I_{ij}^x \rangle =& \langle \tau_i^x \rangle \\
=& \langle \hat{S}_i^y \rangle B_{\text{eff},i}^z - \langle \hat{S}_i^z \rangle B_{\text{eff},i}^y \ ,
\end{aligned} \tag{G.50}$$

$$\begin{aligned}
\langle \hat{S}_i^y \rangle = \sum_\mu \langle S_{ii}^{y\,\mu\mu} \rangle =& 2\sum_{j,\mu\nu} \left\{ \chi_{ij}^{zz\,\mu\nu}(0) B_{\text{eff},j}^{y\,\nu\nu} - \chi_{ij}^{zy\,\mu\nu}(0) B_{\text{eff},j}^{z\,\nu\nu} \right\} \\
\langle \hat{S}_i^y \rangle = \sum_\mu \langle S_{ii}^{y\,\mu\mu} \rangle =& 2\sum_{j,\mu\nu} \left\{ \chi_{ij}^{xx\,\mu\nu}(0) B_{\text{eff},j}^{y\,\nu\nu} - \chi_{ij}^{xy\,\mu\nu}(0) B_{\text{eff},j}^{x\,\nu\nu} \right\} \ ,
\end{aligned} \tag{G.51}$$

$$\begin{aligned}
\langle \hat{S}_i^z \rangle = \sum_\mu \langle S_{ii}^{z\,\mu\mu} \rangle =& 2\sum_{j,\mu\nu} \left\{ \chi_{ij}^{yy\,\mu\nu}(0) B_{\text{eff},j}^{z\,\nu\nu} - \chi_{ij}^{yz\,\mu\nu}(0) B_{\text{eff},j}^{y\,\nu\nu} \right\} \\
\langle \hat{S}_i^z \rangle = \sum_\mu \langle S_{ii}^{z\,\mu\mu} \rangle =& 2\sum_{j,\mu\nu} \left\{ \chi_{ij}^{xx\,\mu\nu}(0) B_{\text{eff},j}^{z\,\nu\nu} - \chi_{ij}^{xz\,\mu\nu}(0) B_{\text{eff},j}^{x\,\nu\nu} \right\} \ .
\end{aligned} \tag{G.52}$$

G.1 Local sum rules

To relate the susceptibility calculated at $\omega = 0$ to the spin densities, we use the results above to obtain

$$\begin{aligned}
\langle S_i^x \rangle = \sum_\mu \langle S_{ii}^{x\,\mu\mu} \rangle =& 2\sum_{\substack{k\\ \mu\gamma\xi}} \left\{ \chi_{iikk}^{yy\,\mu\mu\gamma\xi}(0) B_{\text{eff},k}^{x\,\gamma\xi} - \chi_{iikk}^{yx\,\mu\mu\gamma\xi}(0) B_{\text{eff},k}^{y\,\gamma\xi} \right\} \\
\langle S_i^x \rangle = \sum_\mu \langle S_{ii}^{x\,\mu\mu} \rangle =& 2\sum_{\substack{k\\ \mu\gamma\xi}} \left\{ \chi_{iikk}^{zz\,\mu\mu\gamma\xi}(0) B_{\text{eff},k}^{x\,\gamma\xi} - \chi_{iikk}^{zx\,\mu\mu\gamma\xi}(0) B_{\text{eff},k}^{z\,\gamma\xi} \right\} \ ,
\end{aligned} \tag{G.53}$$

$$\begin{aligned}
\langle S_i^y \rangle = \sum_\mu \langle S_{ii}^{y\,\mu\mu} \rangle =& 2\sum_{\substack{k\\ \mu\gamma\xi}} \left\{ \chi_{iikk}^{zz\,\mu\mu\gamma\xi}(0) B_{\text{eff},k}^{y\,\gamma\xi} - \chi_{iikk}^{zy\,\mu\mu\gamma\xi}(0) B_{\text{eff},k}^{z\,\gamma\xi} \right\} \\
\langle S_i^y \rangle = \sum_\mu \langle S_{ii}^{y\,\mu\mu} \rangle =& 2\sum_{\substack{k\\ \mu\gamma\xi}} \left\{ \chi_{iikk}^{xx\,\mu\mu\gamma\xi}(0) B_{\text{eff},k}^{y\,\gamma\xi} - \chi_{iikk}^{xy\,\mu\mu\gamma\xi}(0) B_{\text{eff},k}^{x\,\gamma\xi} \right\} \ ,
\end{aligned} \tag{G.54}$$

$$
\begin{aligned}
\langle S_i^z \rangle = \sum_{\mu} \langle S_{ii}^{z\,\mu\mu} \rangle =& 2 \sum_{\substack{k \\ \mu\gamma\xi}} \left\{ \chi_{iikk}^{yy\,\mu\mu\gamma\xi}(0) B_{\text{eff},k}^{z\,\gamma\xi} - \chi_{iikk}^{yz\,\mu\mu\gamma\xi}(0) B_{\text{eff},k}^{y\,\gamma\xi} \right\} \\
\langle S_i^z \rangle = \sum_{\mu} \langle S_{ii}^{z\,\mu\mu} \rangle =& 2 \sum_{\substack{k \\ \mu\gamma\xi}} \left\{ \chi_{iikk}^{xx\,\mu\mu\gamma\xi}(0) B_{\text{eff},k}^{z\,\gamma\xi} - \chi_{iikk}^{xz\,\mu\mu\gamma\xi}(0) B_{\text{eff},k}^{x\,\gamma\xi} \right\}
\end{aligned} . \tag{G.55}
$$

Using the components of the total field on the hamiltonian,

$$
\begin{aligned}
\mathbf{B}_{\text{eff},i}^{\mu\nu} =& \mathbf{B}_{\text{xc},i}^{\mu\nu} + \mathbf{B}_{\text{ext},i}^{\mu\nu} + \mathbf{B}_{\text{soc},i}^{\mu\nu} \\
=& - \delta_{\mu\nu} \frac{U^i}{2} \mathbf{m}_i^{(d)} + \frac{g_S \mu_B}{2} \delta_{\mu\nu} \mathbf{B}' + \frac{\lambda_i}{2} \mathbf{L}'_{\mu\nu}
\end{aligned} . \tag{G.56}
$$

G.1.1 No spin-orbit coupling

If no spin-orbit coupling is present, $\mathbf{B}_{\text{soc},i}^{\mu\nu} = 0$ and $\mathbf{B}_{\text{xc},i}^{\mu\nu} + \mathbf{B}_{\text{ext},i}^{\mu\nu} \propto \delta_{\mu\nu}$. We can then choose the direction of the magnetization as the z-axis such that $B_{\text{xc},i}^{x\,\mu\nu} = B_{\text{xc},i}^{y\,\mu\nu} = B_{\text{ext},i}^{x\,\mu\nu} = B_{\text{ext},i}^{y\,\mu\nu} = 0$. Thus,

$$
\begin{aligned}
0 =& 2 \sum_{\substack{k \\ \mu\nu}} \chi_{iikk}^{zx\,\mu\mu\nu\nu}(0) \hat{\mathcal{Z}}_k^{\nu\nu} \\
0 =& 2 \sum_{\substack{k \\ \mu\nu}} \chi_{iikk}^{zy\,\mu\mu\nu\nu}(0) \hat{\mathcal{Z}}_k^{\nu\nu} \\
\langle S_i^z \rangle = \sum_{\mu} \langle S_{ii}^{z\,\mu\mu} \rangle =& 2 \sum_{\substack{k \\ \mu\nu}} \chi_{iikk}^{yy\,\mu\mu\nu\nu}(0) \hat{\mathcal{Z}}_k^{\nu\nu} \\
\langle S_i^z \rangle = \sum_{\mu} \langle S_{ii}^{z\,\mu\mu} \rangle =& 2 \sum_{\substack{k \\ \mu\nu}} \chi_{iikk}^{xx\,\mu\mu\nu\nu}(0) \hat{\mathcal{Z}}_k^{\nu\nu}
\end{aligned} . \tag{G.57}
$$

Substituting the exchange correlation and external fields, we obtain that

$$
m_i^z = 2 \langle S_i^z \rangle = 4 \sum_{\substack{k \\ \mu\nu}} \chi_{iikk}^{xx\,\mu\mu\nu\nu}(0) \frac{\left(-U^k m_k^z + \hbar\omega_z \right)}{2} . \tag{G.58}
$$

where the sum over μ and ν are restricted to the d orbitals (i.e., m_i^z is the magnetic moment of these orbitals only, and the external magnetic field should be applied only on the d orbitals). This results in

$$
\frac{1}{2\left(-U^i + \frac{\hbar\omega_z}{m_i^z} \right)} = \sum_{\substack{k \\ \mu\nu}} \chi_{iikk}^{xx\,\mu\mu\nu\nu}(0) . \tag{G.59}
$$

In the $+,-$ basis,

$$\frac{1}{-U^i + \frac{\hbar\omega_z}{m_i^z}} = \frac{1}{2}\sum_{\substack{k\\ \mu\nu}} \left[\chi_{ik}^{+-\,\mu\nu}(0) + \chi_{ik}^{-+\,\mu\nu}(0) \right] . \tag{G.60}$$

If there is only one site in the unit cell,

$$\frac{1}{-U + \frac{\hbar\omega_z}{m^z}} = \mathrm{Re}\left[\sum_{\mu\nu} \chi^{+-\,\mu\nu}(0) \right] . \tag{G.61}$$

G.2 Longitudinal sum rules

Another set of sum rules can be obtained by just using the relation

$$GG = -\frac{dG}{dE} . \tag{G.62}$$

This can be used for a particular combination of the Green functions product on the susceptibilities. Using the general form of the HF susceptibility obtained in Eq. (C.43) applied to the general case of multilayers, it is given by

$$\chi^{\alpha\sigma\;\mu\nu\gamma\gamma}_{(0)\,ij\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel 0) = -\frac{\hbar}{4\pi}\sum_{\sigma} \mathrm{Im}\int d\omega' f(\omega') \mathrm{Tr}\left\{ \sigma^\alpha G^{\nu\gamma}_{j\ell}(\mathbf{k}_\parallel, \omega') G^{\gamma\mu}_{\ell i}(\mathbf{k}_\parallel, \omega') \right\} . \tag{G.63}$$

Integrating by parts, the energy derivative goes to the Fermi function. For $T = 0K$, we obtain a “sum rule” that involve just Green functions at E_{F}.

We can also write Eqs. G.32 in terms of the components $+, -, \uparrow, \downarrow$ as

$$\begin{aligned}\frac{1}{2}\left(\sigma^{\uparrow}-\sigma^{\downarrow}\right) i\frac{G^{\uparrow\downarrow}-G^{\downarrow\uparrow}}{2}-\frac{1}{2}\left(\sigma^{+}-\sigma^{-}\right)\frac{G^{\uparrow\uparrow}-G^{\downarrow\downarrow}}{2} =&G\left(\sigma^{z}\hat{\mathcal{Y}}-\sigma^{y}\hat{\mathcal{Z}}\right)G\\ \left(\sigma^{z}G^{x}-\sigma^{x}G^{z}\right) =&G\left(\sigma^{z}\hat{\mathcal{X}}-\sigma^{x}\hat{\mathcal{Z}}\right)G\,\cdot\\ \left(\sigma^{y}G^{x}-\sigma^{x}G^{y}\right) =&G\left(\sigma^{y}\hat{\mathcal{X}}-\sigma^{x}\hat{\mathcal{Y}}\right)G\end{aligned} \tag{G.64}$$

$$\begin{aligned}\left(\sigma^{z}G^{\downarrow\uparrow}-\sigma^{+}G^{z}\right) =&G\left(\sigma^{z}\hat{H}^{\downarrow\uparrow}-\sigma^{+}\hat{\mathcal{Z}}\right)G\\ \left(\sigma^{z}G^{\uparrow\downarrow}-\sigma^{-}G^{z}\right) =&G\left(\sigma^{z}\hat{H}^{\uparrow\downarrow}-\sigma^{-}\hat{\mathcal{Z}}\right)G\\ \left(\sigma^{+}G^{\uparrow\downarrow}-\sigma^{-}G^{\downarrow\uparrow}\right) =&G\left(\sigma^{+}\hat{H}^{\uparrow\downarrow}-\sigma^{-}\hat{H}^{\downarrow\uparrow}\right)G\\ \left(\sigma^{z}G^{y}-\sigma^{y}G^{z}\right) =&G\left(\sigma^{z}\hat{\mathcal{Y}}-\sigma^{y}\hat{\mathcal{Z}}\right)G\\ \left(\sigma^{z}G^{x}-\sigma^{x}G^{z}\right) =&G\left(\sigma^{z}\hat{\mathcal{X}}-\sigma^{x}\hat{\mathcal{Z}}\right)G\\ \left(\sigma^{y}G^{x}-\sigma^{x}G^{y}\right) =&G\left(\sigma^{y}\hat{\mathcal{X}}-\sigma^{x}\hat{\mathcal{Y}}\right)G\end{aligned}\;. \tag{G.65}$$

or

$$
\begin{aligned}
G =& \sigma^x G \sigma^x + \sigma^x G \sigma^x \left(\hat{\mathcal{Y}}\sigma^y + \hat{\mathcal{Z}}\sigma^z\right) G \\
G =& \sigma^y G \sigma^y + \sigma^y G \sigma^y \left(\hat{\mathcal{Z}}\sigma^z + \hat{\mathcal{X}}\sigma^x\right) G \ . \\
G =& \sigma^z G \sigma^z + \sigma^z G \sigma^z \left(\hat{\mathcal{X}}\sigma^x + \hat{\mathcal{Y}}\sigma^y\right) G
\end{aligned}
\tag{G.66}
$$

$$
\begin{aligned}
\sigma^x G =& G \sigma^x + G \sigma^x \left(\hat{\mathcal{Y}}\sigma^y + \hat{\mathcal{Z}}\sigma^z\right) G \\
\sigma^y G =& G \sigma^y + G \sigma^y \left(\hat{\mathcal{Z}}\sigma^z + \hat{\mathcal{X}}\sigma^x\right) G \ . \\
\sigma^z G =& G \sigma^z + G \sigma^z \left(\hat{\mathcal{X}}\sigma^x + \hat{\mathcal{Y}}\sigma^y\right) G
\end{aligned}
\tag{G.67}
$$

$\hat{\mathcal{X}}$, $\hat{\mathcal{Y}}$ and $\hat{\mathcal{Z}}$ are not matrices in spin space, so

$$
\begin{aligned}
\sigma^x G =& G \sigma^x + G \left(\hat{\mathcal{Y}}\sigma^x\sigma^y + \hat{\mathcal{Z}}\sigma^x\sigma^z\right) G \\
\sigma^y G =& G \sigma^y + G \left(\hat{\mathcal{Z}}\sigma^y\sigma^z + \hat{\mathcal{X}}\sigma^y\sigma^x\right) G \ . \\
\sigma^z G =& G \sigma^z + G \left(\hat{\mathcal{X}}\sigma^z\sigma^x + \hat{\mathcal{Y}}\sigma^z\sigma^y\right) G
\end{aligned}
\tag{G.68}
$$

$$
\begin{aligned}
\sigma^x G =& G \sigma^x + iG \left(\hat{\mathcal{Y}}\sigma^z - \hat{\mathcal{Z}}\sigma^y\right) G \\
\sigma^y G =& G \sigma^y + iG \left(\hat{\mathcal{Z}}\sigma^x - \hat{\mathcal{X}}\sigma^z\right) G \ . \\
\sigma^z G =& G \sigma^z + iG \left(\hat{\mathcal{X}}\sigma^y - \hat{\mathcal{Y}}\sigma^x\right) G
\end{aligned}
\tag{G.69}
$$

which leads us to the following equations, respectively:

$$
\begin{aligned}
\begin{pmatrix} G^{\uparrow\uparrow} & G^{\uparrow\downarrow} \\ G^{\downarrow\uparrow} & G^{\downarrow\downarrow} \end{pmatrix} &= \begin{pmatrix} G^{\downarrow\downarrow} & G^{\downarrow\uparrow} \\ G^{\uparrow\downarrow} & G^{\uparrow\uparrow} \end{pmatrix} - \begin{pmatrix} G^{\downarrow\downarrow} & G^{\downarrow\uparrow} \\ G^{\uparrow\downarrow} & G^{\uparrow\uparrow} \end{pmatrix} \begin{pmatrix} -\hat{\mathcal{Z}} & i\hat{\mathcal{Y}} \\ -i\hat{\mathcal{Y}} & \hat{\mathcal{Z}} \end{pmatrix} \begin{pmatrix} G^{\uparrow\uparrow} & G^{\uparrow\downarrow} \\ G^{\downarrow\uparrow} & G^{\downarrow\downarrow} \end{pmatrix} \\
&= \begin{pmatrix} G^{\downarrow\downarrow} & G^{\downarrow\uparrow} \\ G^{\uparrow\downarrow} & G^{\uparrow\uparrow} \end{pmatrix} - \begin{pmatrix} G^{\downarrow\downarrow} & G^{\downarrow\uparrow} \\ G^{\uparrow\downarrow} & G^{\uparrow\uparrow} \end{pmatrix} \begin{pmatrix} -\hat{\mathcal{Z}}G^{\uparrow\uparrow} + i\hat{\mathcal{Y}}G^{\downarrow\uparrow} & -\hat{\mathcal{Z}}G^{\uparrow\downarrow} + i\hat{\mathcal{Y}}G^{\downarrow\downarrow} \\ -i\hat{\mathcal{Y}}G^{\uparrow\uparrow} + \hat{\mathcal{Z}}G^{\downarrow\uparrow} & -i\hat{\mathcal{Y}}G^{\uparrow\downarrow} + \hat{\mathcal{Z}}G^{\downarrow\downarrow} \end{pmatrix} \\
&= \begin{pmatrix} G^{\downarrow\downarrow} & G^{\downarrow\uparrow} \\ G^{\uparrow\downarrow} & G^{\uparrow\uparrow} \end{pmatrix} - \left(\begin{matrix} -G^{\downarrow\downarrow}\hat{\mathcal{Z}}G^{\uparrow\uparrow} + G^{\downarrow\downarrow}i\hat{\mathcal{Y}}G^{\downarrow\uparrow} - G^{\downarrow\uparrow}i\hat{\mathcal{Y}}G^{\uparrow\uparrow} + G^{\downarrow\uparrow}\hat{\mathcal{Z}}G^{\downarrow\uparrow} \\ -G^{\uparrow\downarrow}\hat{\mathcal{Z}}G^{\uparrow\uparrow} + G^{\uparrow\downarrow}i\hat{\mathcal{Y}}G^{\downarrow\uparrow} - G^{\uparrow\uparrow}i\hat{\mathcal{Y}}G^{\uparrow\uparrow} + G^{\uparrow\uparrow}\hat{\mathcal{Z}}G^{\downarrow\uparrow} \end{matrix} \right. \\
&\qquad\qquad \left. \begin{matrix} -G^{\downarrow\downarrow}\hat{\mathcal{Z}}G^{\uparrow\downarrow} + G^{\downarrow\downarrow}i\hat{\mathcal{Y}}G^{\downarrow\downarrow} - G^{\downarrow\uparrow}i\hat{\mathcal{Y}}G^{\uparrow\downarrow} + G^{\downarrow\uparrow}\hat{\mathcal{Z}}G^{\downarrow\downarrow} \\ -G^{\uparrow\downarrow}\hat{\mathcal{Z}}G^{\uparrow\downarrow} + G^{\uparrow\downarrow}i\hat{\mathcal{Y}}G^{\downarrow\downarrow} - G^{\uparrow\uparrow}i\hat{\mathcal{Y}}G^{\uparrow\downarrow} + G^{\uparrow\uparrow}\hat{\mathcal{Z}}G^{\downarrow\downarrow} \end{matrix} \right) \ .
\end{aligned}
\tag{G.70}
$$

$$
\begin{pmatrix} G^{\uparrow\uparrow} & G^{\uparrow\downarrow} \\ G^{\downarrow\uparrow} & G^{\downarrow\downarrow} \end{pmatrix} = \begin{pmatrix} G^{\downarrow\downarrow} & G^{\downarrow\uparrow} \\ G^{\uparrow\downarrow} & G^{\uparrow\uparrow} \end{pmatrix} - \begin{pmatrix} G^{\downarrow\downarrow} & G^{\downarrow\uparrow} \\ G^{\uparrow\downarrow} & G^{\uparrow\uparrow} \end{pmatrix} \begin{pmatrix} -\hat{\mathcal{Z}} & i\hat{\mathcal{Y}} \\ -i\hat{\mathcal{Y}} & \hat{\mathcal{Z}} \end{pmatrix} \begin{pmatrix} G^{\uparrow\uparrow} & G^{\uparrow\downarrow} \\ G^{\downarrow\uparrow} & G^{\downarrow\downarrow} \end{pmatrix}
$$

$$
= \begin{pmatrix} G^{\downarrow\downarrow} & G^{\downarrow\uparrow} \\ G^{\uparrow\downarrow} & G^{\uparrow\uparrow} \end{pmatrix} - \begin{pmatrix} G^{\downarrow\downarrow} & G^{\downarrow\uparrow} \\ G^{\uparrow\downarrow} & G^{\uparrow\uparrow} \end{pmatrix} \begin{pmatrix} -\hat{\mathcal{Z}}G^{\uparrow\uparrow} + i\hat{\mathcal{Y}}G^{\downarrow\uparrow} & -\hat{\mathcal{Z}}G^{\uparrow\downarrow} + i\hat{\mathcal{Y}}G^{\downarrow\downarrow} \\ -i\hat{\mathcal{Y}}G^{\uparrow\uparrow} + \hat{\mathcal{Z}}G^{\downarrow\uparrow} & -i\hat{\mathcal{Y}}G^{\uparrow\downarrow} + \hat{\mathcal{Z}}G^{\downarrow\downarrow} \end{pmatrix}
$$

$$
= \begin{pmatrix} G^{\downarrow\downarrow} & G^{\downarrow\uparrow} \\ G^{\uparrow\downarrow} & G^{\uparrow\uparrow} \end{pmatrix} - \begin{pmatrix} -G^{\downarrow\downarrow}\hat{\mathcal{Z}}G^{\uparrow\uparrow} + G^{\downarrow\downarrow}i\hat{\mathcal{Y}}G^{\downarrow\uparrow} - G^{\downarrow\uparrow}i\hat{\mathcal{Y}}G^{\uparrow\uparrow} + G^{\downarrow\uparrow}\hat{\mathcal{Z}}G^{\downarrow\uparrow} & -G^{\downarrow\downarrow}\hat{\mathcal{Z}}G^{\uparrow\downarrow} + G^{\downarrow\downarrow}i\hat{\mathcal{Y}}G^{\downarrow\downarrow} - G^{\downarrow\uparrow}i\hat{\mathcal{Y}}G^{\uparrow\downarrow} + G^{\downarrow\uparrow}\hat{\mathcal{Z}}G^{\downarrow\downarrow} \\ -G^{\uparrow\downarrow}\hat{\mathcal{Z}}G^{\uparrow\uparrow} + G^{\uparrow\downarrow}i\hat{\mathcal{Y}}G^{\downarrow\uparrow} - G^{\uparrow\uparrow}i\hat{\mathcal{Y}}G^{\uparrow\uparrow} + G^{\uparrow\uparrow}\hat{\mathcal{Z}}G^{\downarrow\uparrow} & -G^{\uparrow\downarrow}\hat{\mathcal{Z}}G^{\uparrow\downarrow} + G^{\uparrow\downarrow}i\hat{\mathcal{Y}}G^{\downarrow\downarrow} - G^{\uparrow\uparrow}i\hat{\mathcal{Y}}G^{\uparrow\downarrow} + G^{\uparrow\uparrow}\hat{\mathcal{Z}}G^{\downarrow\downarrow} \end{pmatrix} . \tag{G.71}
$$

$$
\begin{aligned}
G^{\uparrow\uparrow} &= G^{\downarrow\downarrow} + G^{\downarrow\downarrow}\hat{\mathcal{Z}}G^{\uparrow\uparrow} - iG^{\downarrow\downarrow}\hat{\mathcal{Y}}G^{\downarrow\uparrow} + iG^{\downarrow\uparrow}\hat{\mathcal{Y}}G^{\uparrow\uparrow} - G^{\downarrow\uparrow}\hat{\mathcal{Z}}G^{\downarrow\uparrow} \\
G^{\uparrow\downarrow} &= G^{\downarrow\uparrow} + G^{\downarrow\downarrow}\hat{\mathcal{Z}}G^{\uparrow\downarrow} - iG^{\downarrow\downarrow}\hat{\mathcal{Y}}G^{\downarrow\downarrow} + iG^{\downarrow\uparrow}\hat{\mathcal{Y}}G^{\uparrow\downarrow} - G^{\downarrow\uparrow}\hat{\mathcal{Z}}G^{\downarrow\downarrow} \\
G^{\downarrow\uparrow} &= G^{\uparrow\downarrow} + G^{\uparrow\downarrow}\hat{\mathcal{Z}}G^{\uparrow\uparrow} - iG^{\uparrow\downarrow}\hat{\mathcal{Y}}G^{\downarrow\uparrow} + iG^{\uparrow\uparrow}\hat{\mathcal{Y}}G^{\uparrow\uparrow} - G^{\uparrow\uparrow}\hat{\mathcal{Z}}G^{\downarrow\uparrow} \\
G^{\downarrow\downarrow} &= G^{\uparrow\uparrow} + G^{\uparrow\downarrow}\hat{\mathcal{Z}}G^{\uparrow\downarrow} - iG^{\uparrow\downarrow}\hat{\mathcal{Y}}G^{\downarrow\downarrow} + iG^{\uparrow\uparrow}\hat{\mathcal{Y}}G^{\uparrow\downarrow} - G^{\uparrow\uparrow}\hat{\mathcal{Z}}G^{\downarrow\downarrow} .
\end{aligned} \tag{G.72}
$$

$(4) - (1)$:

$$
\begin{aligned}
2\left[G^{\downarrow\downarrow} - G^{\uparrow\uparrow}\right] =& G^{\downarrow\downarrow}\Delta G^{\uparrow\uparrow} + G^{\uparrow\uparrow}\Delta G^{\downarrow\downarrow} + G^{\downarrow\downarrow}\Lambda G^{\downarrow\uparrow} - G^{\uparrow\downarrow}\Lambda G^{\downarrow\downarrow} \\
&+ G^{\uparrow\uparrow}\Lambda G^{\uparrow\downarrow} - G^{\downarrow\uparrow}\Lambda G^{\uparrow\uparrow} - G^{\downarrow\uparrow}\Delta G^{\downarrow\uparrow} - G^{\uparrow\downarrow}\Delta G^{\uparrow\downarrow} .
\end{aligned} \tag{G.73}
$$

$(4) + (1)$:

$$
\begin{aligned}
&G^{\downarrow\downarrow}\Delta G^{\uparrow\uparrow} - G^{\uparrow\uparrow}\Delta G^{\downarrow\downarrow} + G^{\downarrow\downarrow}\Lambda G^{\downarrow\uparrow} + G^{\uparrow\downarrow}\Lambda G^{\downarrow\downarrow} \\
&- G^{\uparrow\uparrow}\Lambda G^{\uparrow\downarrow} - G^{\downarrow\uparrow}\Lambda G^{\uparrow\uparrow} - G^{\downarrow\uparrow}\Delta G^{\downarrow\uparrow} + G^{\uparrow\downarrow}\Delta G^{\uparrow\downarrow} = 0 .
\end{aligned} \tag{G.74}
$$

$(2) + [(2)]^*$:

$$
\begin{aligned}
G^{\downarrow\uparrow} - G^{\uparrow\downarrow} - \left[G^{\uparrow\downarrow}\right]^* + \left[G^{\downarrow\uparrow}\right]^* =& G^{\downarrow\downarrow}\Delta G^{\uparrow\downarrow} - \left[G^{\downarrow\uparrow}\Delta G^{\downarrow\downarrow}\right]^* + G^{\downarrow\downarrow}\Lambda G^{\downarrow\downarrow} + \left[G^{\downarrow\downarrow}\Lambda G^{\downarrow\downarrow}\right]^* \\
&- G^{\downarrow\uparrow}\Lambda G^{\uparrow\downarrow} - \left[G^{\downarrow\uparrow}\Lambda G^{\uparrow\downarrow}\right]^* - G^{\downarrow\uparrow}\Delta G^{\downarrow\downarrow} + \left[G^{\downarrow\downarrow}\Delta G^{\uparrow\downarrow}\right]^* .
\end{aligned} \tag{G.75}
$$

$(3) + [(3)]^*$:

$$
\begin{aligned}
G^{\downarrow\uparrow} - G^{\uparrow\downarrow} - \left[G^{\uparrow\downarrow}\right]^* + \left[G^{\downarrow\uparrow}\right]^* =& G^{\uparrow\uparrow}\Delta G^{\downarrow\uparrow} - \left[G^{\uparrow\downarrow}\Delta G^{\uparrow\uparrow}\right]^* + G^{\uparrow\uparrow}\Lambda G^{\uparrow\uparrow} + \left[G^{\uparrow\uparrow}\Lambda G^{\uparrow\uparrow}\right]^* \\
&- G^{\uparrow\downarrow}\Lambda G^{\downarrow\uparrow} - \left[G^{\uparrow\downarrow}\Lambda G^{\downarrow\uparrow}\right]^* - G^{\uparrow\downarrow}\Delta G^{\uparrow\uparrow} + \left[G^{\uparrow\uparrow}\Delta G^{\downarrow\uparrow}\right]^* .
\end{aligned} \tag{G.76}
$$

Consider

$$\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\ \gamma\xi}}\left[\chi^{+-\ \mu\nu\gamma\xi}_{(0)\,iimm}(\mathbf{k},\mathbf{k};0)+\chi^{-+\ \mu\nu\gamma\xi}_{(0)\,iimm}(\mathbf{k},\mathbf{k};0)\right]\Delta^{\gamma\xi}_{m}$$

$$=\frac{i\hbar}{2\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\ \gamma\xi}}\int d\omega' f(\omega')\left\{G^{\downarrow\downarrow\nu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\uparrow\uparrow\xi\mu}_{mi}(\mathbf{k},\omega')-\left[G^{\uparrow\uparrow\mu\xi}_{im}(\mathbf{k},\omega')\Delta^{\xi\gamma}_{m}G^{\downarrow\downarrow\gamma\nu}_{mi}(\mathbf{k},\omega')\right]^{*}\right.$$
$$\left.+G^{\uparrow\uparrow\nu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\downarrow\downarrow\xi\mu}_{mi}(\mathbf{k},\omega')-\left[G^{\downarrow\downarrow\mu\xi}_{im}(\mathbf{k},\omega')\Delta^{\xi\gamma}_{m}G^{\uparrow\uparrow\gamma\nu}_{mi}(\mathbf{k},\omega')\right]^{*}\right\}\ . \tag{G.77}$$

Now, if we take $\mu=\nu$, and change indices $\gamma\rightleftharpoons\xi$ on the second and fourth terms,

$$\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\ \gamma\xi}}\left[\chi^{+-\ \mu\mu\gamma\xi}_{(0)\,iimm}(\mathbf{k},\mathbf{k};0)+\chi^{-+\ \mu\mu\gamma\xi}_{(0)\,iimm}(\mathbf{k},\mathbf{k};0)\right]\Delta^{\gamma\xi}_{m}$$

$$=\frac{i\hbar}{2\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\ \gamma\xi}}\int d\omega' f(\omega')\left\{G^{\downarrow\downarrow\mu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\uparrow\uparrow\xi\mu}_{mi}(\mathbf{k},\omega')-\left[G^{\uparrow\uparrow\mu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\downarrow\downarrow\xi\mu}_{mi}(\mathbf{k},\omega')\right]^{*}\right.$$
$$\left.+G^{\uparrow\uparrow\mu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\downarrow\downarrow\xi\mu}_{mi}(\mathbf{k},\omega')-\left[G^{\downarrow\downarrow\mu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\uparrow\uparrow\xi\mu}_{mi}(\mathbf{k},\omega')\right]^{*}\right\}$$
$$=-\frac{\hbar}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\ \gamma\xi}}\int d\omega' f(\omega')\mathrm{Im}\left\{G^{\downarrow\downarrow\mu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\uparrow\uparrow\xi\mu}_{mi}(\mathbf{k},\omega')+G^{\uparrow\uparrow\mu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\downarrow\downarrow\xi\mu}_{mi}(\mathbf{k},\omega')\right\}\ . \tag{G.78}$$

The same steps can be used to show that

$$\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\ \gamma\xi}}\left[-\chi^{++\ \mu\mu\gamma\xi}_{(0)\,iimm}(\mathbf{k},\mathbf{k};0)-\chi^{--\ \mu\mu\gamma\xi}_{(0)\,iimm}(\mathbf{k},\mathbf{k};0)\right]\Delta^{\gamma\xi}_{m}$$
$$=-\frac{\hbar}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\ \gamma\xi}}\int d\omega' f(\omega')\mathrm{Im}\left\{-G^{\downarrow\uparrow\mu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\downarrow\uparrow\xi\mu}_{mi}(\mathbf{k},\omega')-G^{\uparrow\downarrow\mu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\uparrow\downarrow\xi\mu}_{mi}(\mathbf{k},\omega')\right\}\ . \tag{G.79}$$

Now, taking into account that $\Lambda^{\alpha\beta}_{m}=-(\Lambda^{\beta\alpha}_{m})^{*}$,

$$\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\ \gamma\xi}}\left[\chi^{+\downarrow\ \mu\mu\gamma\xi}_{(0)\,iimm}(\mathbf{k},\mathbf{k};0)-\chi^{-\downarrow\ \mu\mu\gamma\xi}_{(0)\,iimm}(\mathbf{k},\mathbf{k};0)\right]\Lambda^{\gamma\xi}_{m}$$

$$=\frac{i\hbar}{2\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\ \gamma\xi}}\int d\omega' f(\omega')\left\{G^{\downarrow\downarrow\mu\gamma}_{im}(\mathbf{k},\omega')\Lambda^{\gamma\xi}_{m}G^{\downarrow\uparrow\xi\mu}_{mi}(\mathbf{k},\omega')+\left[G^{\uparrow\downarrow\mu\gamma}_{im}(\mathbf{k},\omega')\Lambda^{\gamma\xi}_{m}G^{\downarrow\downarrow\xi\mu}_{mi}(\mathbf{k},\omega')\right]^{*}\right.$$
$$\left.-G^{\uparrow\downarrow\mu\gamma}_{im}(\mathbf{k},\omega')\Lambda^{\gamma\xi}_{m}G^{\downarrow\downarrow\xi\mu}_{mi}(\mathbf{k},\omega')-\left[G^{\downarrow\downarrow\mu\gamma}_{im}(\mathbf{k},\omega')\Lambda^{\gamma\xi}_{m}G^{\downarrow\uparrow\xi\mu}_{mi}(\mathbf{k},\omega')\right]^{*}\right\}$$
$$=-\frac{\hbar}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\ \gamma\xi}}\int d\omega' f(\omega')\mathrm{Im}\left\{G^{\downarrow\downarrow\mu\gamma}_{im}(\mathbf{k},\omega')\Lambda^{\gamma\xi}_{m}G^{\downarrow\uparrow\xi\mu}_{mi}(\mathbf{k},\omega')-G^{\uparrow\downarrow\mu\gamma}_{im}(\mathbf{k},\omega')\Lambda^{\gamma\xi}_{m}G^{\downarrow\downarrow\xi\mu}_{mi}(\mathbf{k},\omega')\right\}\ , \tag{G.80}$$

and

$$\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\\gamma\xi}}\left[\chi^{-\uparrow\,\mu\mu\gamma\xi}_{(0)\,iimm}(\mathbf{k},\mathbf{k};0)-\chi^{+\uparrow\,\mu\mu\gamma\xi}_{(0)\,iimm}(\mathbf{k},\mathbf{k};0)\right]\Lambda^{\gamma\xi}_{m}$$
$$=-\frac{\hbar}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\\gamma\xi}}\int d\omega' f(\omega')\mathrm{Im}\left\{G^{\uparrow\uparrow\,\mu\gamma}_{im}(\mathbf{k},\omega')\Lambda^{\gamma\xi}_{m}G^{\uparrow\downarrow\,\xi\mu}_{mi}(\mathbf{k},\omega')-G^{\downarrow\uparrow\,\mu\gamma}_{im}(\mathbf{k},\omega')\Lambda^{\gamma\xi}_{m}G^{\uparrow\uparrow\,\xi\mu}_{mi}(\mathbf{k},\omega')\right\}\ . \tag{G.81}$$

Finally, summing up all these terms, and using Eq. (G.73), we have

$$\begin{aligned}
&\sum_{\substack{m\\\gamma\xi}}\left[\chi^{+-\,\mu\mu\gamma\xi}_{(0)\,iimm}+\chi^{-+\,\mu\mu\gamma\xi}_{(0)\,iimm}-\chi^{++\,\mu\mu\gamma\xi}_{(0)\,iimm}-\chi^{--\,\mu\mu\gamma\xi}_{(0)\,iimm}\right]\Delta^{\gamma\xi}_{m}\\
&+\sum_{\substack{m\\\gamma\xi}}\left[\chi^{+\downarrow\,\mu\mu\gamma\xi}_{(0)\,iimm}-\chi^{-\downarrow\,\mu\mu\gamma\xi}_{(0)\,iimm}+\chi^{-\uparrow\,\mu\mu\gamma\xi}_{(0)\,iimm}-\chi^{+\uparrow\,\mu\mu\gamma\xi}_{(0)\,iimm}\right]\Lambda^{\gamma\xi}_{m}\\
&=-\frac{2\hbar}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\int d\omega' f(\omega')\mathrm{Im}\left\{G^{\downarrow\downarrow\,\mu\mu}_{ii}(\mathbf{k},\omega')-G^{\uparrow\uparrow\,\mu\mu}_{ii}(\mathbf{k},\omega')\right\}\\
&=-2\hbar\int d\omega' f(\omega')\left\{\rho^{\uparrow\,\mu}_{i}(\omega')-\rho^{\downarrow\,\mu}_{i}(\omega')\right\}\\
&=-2\langle m^{\mu}_{i}\rangle\ .
\end{aligned} \tag{G.82}$$

where the susceptibilities on the left hand side of the equation are already summed in $\mathbf{k}$.

Analogously, we can use Eq. (G.74) to show that

$$\begin{aligned}
&\sum_{\substack{m\\\gamma\xi}}\left[\chi^{+-\,\mu\mu\gamma\xi}_{(0)\,iimm}-\chi^{-+\,\mu\mu\gamma\xi}_{(0)\,iimm}-\chi^{++\,\mu\mu\gamma\xi}_{(0)\,iimm}+\chi^{--\,\mu\mu\gamma\xi}_{(0)\,iimm}\right]\Delta^{\gamma\xi}_{m}\\
&+\sum_{\substack{m\\\gamma\xi}}\left[\chi^{+\downarrow\,\mu\mu\gamma\xi}_{(0)\,iimm}+\chi^{-\downarrow\,\mu\mu\gamma\xi}_{(0)\,iimm}+\chi^{-\uparrow\,\mu\mu\gamma\xi}_{(0)\,iimm}+\chi^{+\uparrow\,\mu\mu\gamma\xi}_{(0)\,iimm}\right]\Lambda^{\gamma\xi}_{m}=0\ .
\end{aligned} \tag{G.83}$$

Now, we can write

$$\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\\gamma\xi}}\left[\chi^{\downarrow-\,\mu\mu\gamma\xi}_{(0)\,iimm}(\mathbf{k},\mathbf{k};0)-\chi^{\downarrow+\,\mu\mu\gamma\xi}_{(0)\,iimm}(\mathbf{k},\mathbf{k};0)\right]\Delta^{\gamma\xi}_{m}$$
$$\begin{aligned}
=\frac{i\hbar}{2\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\\gamma\xi}}\int d\omega' f(\omega')\Big\{&G^{\downarrow\downarrow\,\mu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\uparrow\downarrow\,\xi\mu}_{mi}(\mathbf{k},\omega')-\left[G^{\downarrow\uparrow\,\mu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\downarrow\downarrow\,\xi\mu}_{mi}(\mathbf{k},\omega')\right]^{*}\\
&-G^{\downarrow\uparrow\,\mu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\downarrow\downarrow\,\xi\mu}_{mi}(\mathbf{k},\omega')+\left[G^{\downarrow\downarrow\,\mu\gamma}_{im}(\mathbf{k},\omega')\Delta^{\gamma\xi}_{m}G^{\uparrow\downarrow\,\xi\mu}_{mi}(\mathbf{k},\omega')\right]^{*}\Big\}\ ,
\end{aligned} \tag{G.84}$$

and

$$\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\ \gamma\xi}}\left[\chi^{\downarrow\downarrow\,\mu\mu\gamma\xi}_{(0)iimm}(\mathbf{k},\mathbf{k};0)-\chi^{\downarrow\uparrow\,\mu\mu\gamma\xi}_{(0)iimm}(\mathbf{k},\mathbf{k};0)\right]\Lambda^{\gamma\xi}_{m}$$

$$=\frac{i\hbar}{2\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{m\\ \gamma\xi}}\int d\omega' f(\omega')\left\{G^{\downarrow\downarrow\,\mu\gamma}_{im}(\mathbf{k},\omega')\Lambda^{\gamma\xi}_{m}G^{\downarrow\downarrow\,\xi\mu}_{mi}(\mathbf{k},\omega')+\left[G^{\downarrow\downarrow\,\mu\gamma}_{im}(\mathbf{k},\omega')\Lambda^{\gamma\xi}_{m}G^{\downarrow\downarrow\,\xi\mu}_{mi}(\mathbf{k},\omega')\right]^*\right.$$

$$\left.-\,G^{\downarrow\uparrow\,\mu\gamma}_{im}(\mathbf{k},\omega')\Lambda^{\gamma\xi}_{m}G^{\uparrow\downarrow\,\xi\mu}_{mi}(\mathbf{k},\omega')-\left[G^{\downarrow\uparrow\,\mu\gamma}_{im}(\mathbf{k},\omega')\Lambda^{\gamma\xi}_{m}G^{\uparrow\downarrow\,\xi\mu}_{mi}(\mathbf{k},\omega')\right]^*\right\}\ . \tag{G.85}$$

So, together with Eq. (G.75), we obtain

$$\sum_{\substack{m\\ \gamma\xi}}\left\{\left[\chi^{\downarrow-\,\mu\mu\gamma\xi}_{(0)iimm}(0)-\chi^{\downarrow+\,\mu\mu\gamma\xi}_{(0)iimm}(0)\right]\Delta^{\gamma\xi}_{m}+\left[\chi^{\downarrow\downarrow\,\mu\mu\gamma\xi}_{(0)iimm}(0)-\chi^{\downarrow\uparrow\,\mu\mu\gamma\xi}_{(0)iimm}(0)\right]\Lambda^{\gamma\xi}_{m}\right\}$$

$$=\frac{i\hbar}{2\pi}\frac{1}{N}\sum_{\mathbf{k}}\int d\omega' f(\omega')\left\{G^{\downarrow\uparrow\,\mu\mu}_{ii}(\mathbf{k},\omega')-G^{\uparrow\downarrow\,\mu\mu}_{ii}(\mathbf{k},\omega')-\left[G^{\uparrow\downarrow\,\mu\mu}_{ii}(\mathbf{k},\omega')\right]^*+\left[G^{\downarrow\uparrow\,\mu\mu}_{ii}(\mathbf{k},\omega')\right]^*\right\}\ . \tag{G.86}$$

Using Eqs. 1.59 and 1.60 extended to multilayers, we can finally write

$$\sum_{\substack{m\\ \gamma\xi}}\left\{\left[\chi^{\downarrow-\,\mu\mu\gamma\xi}_{(0)iimm}(0)-\chi^{\downarrow+\,\mu\mu\gamma\xi}_{(0)iimm}(0)\right]\Delta^{\gamma\xi}_{m}+\left[\chi^{\downarrow\downarrow\,\mu\mu\gamma\xi}_{(0)iimm}(0)-\chi^{\downarrow\uparrow\,\mu\mu\gamma\xi}_{(0)iimm}(0)\right]\Lambda^{\gamma\xi}_{m}\right\}=\langle S^+_i\rangle-\langle S^-_i\rangle\ . \tag{G.87}$$

Analogously, for Eq. (G.76), we have

$$\sum_{\substack{m\\ \gamma\xi}}\left\{\left[\chi^{\uparrow+\,\mu\mu\gamma\xi}_{(0)iimm}(0)-\chi^{\uparrow-\,\mu\mu\gamma\xi}_{(0)iimm}(0)\right]\Delta^{\gamma\xi}_{m}+\left[\chi^{\uparrow\uparrow\,\mu\mu\gamma\xi}_{(0)iimm}(0)-\chi^{\uparrow\downarrow\,\mu\mu\gamma\xi}_{(0)iimm}(0)\right]\Lambda^{\gamma\xi}_{m}\right\}=\langle S^+_i\rangle-\langle S^-_i\rangle\ . \tag{G.88}$$

From Eq. (G.73)

$$\begin{aligned}2\sum_{\alpha\beta}L^m_{\alpha\beta}\left[G^{\downarrow\downarrow\beta\alpha}_{ii}(\mathbf{k},\omega')-G^{\uparrow\uparrow\beta\alpha}_{ii}(\mathbf{k},\omega')\right]=&2\sum_{\alpha\beta}\left[L^m_{\alpha\beta}G^{\downarrow\downarrow\beta\alpha}_{ii}(\mathbf{k},\omega')+L^m_{\beta\alpha}G^{\uparrow\uparrow\beta\alpha}_{ii}(\mathbf{k},\omega')\right]\\=&G^{\downarrow\downarrow}\Delta G^{\uparrow\uparrow}+G^{\uparrow\uparrow}\Delta G^{\downarrow\downarrow}+G^{\downarrow\downarrow}\Lambda G^{\downarrow\uparrow}-G^{\uparrow\downarrow}\Lambda G^{\downarrow\downarrow}\\&+G^{\uparrow\uparrow}\Lambda G^{\uparrow\downarrow}-G^{\downarrow\uparrow}\Lambda G^{\uparrow\uparrow}-G^{\downarrow\uparrow}\Delta G^{\downarrow\uparrow}-G^{\uparrow\downarrow}\Delta G^{\uparrow\downarrow}\ .\end{aligned}\tag{G.89}$$

$$\begin{aligned}2\sum_{\alpha\beta}L^m_{\alpha\beta}\left[G^{\downarrow\downarrow\alpha\beta}_{ii}(\mathbf{k},\omega')-G^{\uparrow\uparrow\alpha\beta}_{ii}(\mathbf{k},\omega')\right]^*=&2\sum_{\alpha\beta}\left[L^m_{\beta\alpha}G^{\downarrow\downarrow\alpha\beta}_{ii}(\mathbf{k},\omega')-L^m_{\beta\alpha}G^{\uparrow\uparrow\alpha\beta}_{ii}(\mathbf{k},\omega')\right]^*\\=&G^{\downarrow\downarrow}\Delta G^{\uparrow\uparrow}+G^{\uparrow\uparrow}\Delta G^{\downarrow\downarrow}+G^{\downarrow\downarrow}\Lambda G^{\downarrow\uparrow}-G^{\uparrow\downarrow}\Lambda G^{\downarrow\downarrow}\\&+G^{\uparrow\uparrow}\Lambda G^{\uparrow\downarrow}-G^{\downarrow\uparrow}\Lambda G^{\uparrow\uparrow}-G^{\downarrow\uparrow}\Delta G^{\downarrow\uparrow}-G^{\uparrow\downarrow}\Delta G^{\uparrow\downarrow}\ .\end{aligned}\tag{G.90}$$

$$\begin{aligned}-2\sum_{\alpha\beta}\left[L^m_{\beta\alpha}G^{\downarrow\downarrow\beta\alpha}_{ii}(\mathbf{k},\omega')+L^m_{\alpha\beta}G^{\uparrow\uparrow\beta\alpha}_{ii}(\mathbf{k},\omega')\right]=&G^{\downarrow\downarrow}\Delta G^{\uparrow\uparrow}+G^{\uparrow\uparrow}\Delta G^{\downarrow\downarrow}+G^{\downarrow\downarrow}\Lambda G^{\downarrow\uparrow}-G^{\uparrow\downarrow}\Lambda G^{\downarrow\downarrow}\\&+G^{\uparrow\uparrow}\Lambda G^{\uparrow\downarrow}-G^{\downarrow\uparrow}\Lambda G^{\uparrow\uparrow}-G^{\downarrow\uparrow}\Delta G^{\downarrow\uparrow}-G^{\uparrow\downarrow}\Delta G^{\uparrow\downarrow}\ .\end{aligned}\tag{G.91}$$

Appendix H

Ground state currents

H.1 Charge current

The expectation value of the charge current operator between sites i and j (given by Eq. (5.30)) is

$$\langle \hat{I}^{\mathrm{C}}_{ij}(t)\rangle = \frac{e}{\hbar} i \sum_{\sigma}\sum_{\mu\nu}\left\{ t^{\mu\nu}_{ij}\langle c^{\dagger}_{i\mu\sigma}(t) c_{j\nu\sigma}(t)\rangle - t^{\nu\mu}_{ji}\langle c^{\dagger}_{j\nu\sigma}(t) c_{i\mu\sigma}(t)\rangle \right\} \ . \tag{H.1}$$

For $t = 0$, we can use the correlation function obtained in Eq. (1.56) to write

$$\langle \hat{c}^{\dagger}_{i\mu\sigma}\hat{c}_{j\nu\sigma}\rangle = \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F} d\omega \left\{ G^{\sigma\sigma\,\nu\mu}_{ji}(\omega) - \left[G^{\sigma\sigma\,\mu\nu}_{ij}(\omega)\right]^{*}\right\} \ , \tag{H.2}$$

and

$$\langle \hat{c}^{\dagger}_{j\nu\sigma}\hat{c}_{i\mu\sigma}\rangle = \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F} d\omega \left\{ G^{\sigma\sigma\,\mu\nu}_{ij}(\omega) - \left[G^{\sigma\sigma\,\nu\mu}_{ji}(\omega)\right]^{*}\right\} \ . \tag{H.3}$$

So,

$$\langle \hat{I}^{\mathrm{C}}_{ij}\rangle = -\frac{e}{2\pi}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F} d\omega \; t^{\mu\nu}_{ij}\left\{ G^{\sigma\sigma\,\nu\mu}_{ji}(\omega) - \left[G^{\sigma\sigma\,\mu\nu}_{ij}(\omega)\right]^{*} - G^{\sigma\sigma\,\mu\nu}_{ij}(\omega) + \left[G^{\sigma\sigma\,\nu\mu}_{ji}(\omega)\right]^{*}\right\} \ , \tag{H.4}$$

since, in real space, $t^{\mu\nu}_{ij} = t^{\nu\mu}_{ji}$. This may be simplified to

$$\langle \hat{I}^{\mathrm{C}}_{ij}\rangle = -\frac{e}{\pi}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F} d\omega \; t^{\mu\nu}_{ij}\mathrm{Re}\left\{ G^{\sigma\sigma\,\nu\mu}_{ji}(\omega) - G^{\sigma\sigma\,\mu\nu}_{ij}(\omega)\right\} \ . \tag{H.5}$$

This current is zero in the fundamental state. When we apply a transverse static magnetic field, we have

$$\delta\langle \hat{I}^{\mathrm{C}}_{ij}\rangle = -\frac{e}{\pi}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F} d\omega \; t^{\mu\nu}_{ij}\mathrm{Re}\left\{ \Delta G^{\sigma\sigma\,\nu\mu}_{ji}(\omega) - \Delta G^{\sigma\sigma\,\mu\nu}_{ij}(\omega)\right\} \ , \tag{H.6}$$

where ΔG can be calculated using Dyson's equation.

For multilayers, we can rewrite Eq. (H.1) as

$$\langle \hat{I}^{\mathrm{C}}_{\ell\ell'}(\mathbf{R}_i,\mathbf{R}_j)\rangle = \frac{e}{\hbar}i\sum_{\sigma}\sum_{\mu\nu}\left\{t^{\mu\nu}_{\ell\ell'}(\mathbf{R}_j-\mathbf{R}_i)\langle c^{\dagger}_{\ell\mu\sigma}c_{\ell'\nu\sigma}\rangle(\mathbf{R}_j-\mathbf{R}_i) - t^{\nu\mu}_{\ell'\ell}(\mathbf{R}_i-\mathbf{R}_j)\langle c^{\dagger}_{\ell'\nu\sigma}c_{\ell\mu\sigma}\rangle(\mathbf{R}_i-\mathbf{R}_j)\right\} \ . \tag{H.7}$$

Using Eq. (1.69), i.e.,

$$\langle \hat{c}^{\dagger}_{\ell\mu\sigma}\hat{c}_{\ell'\nu\sigma}\rangle(\mathbf{R}_j-\mathbf{R}_i) = \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F}d\omega\frac{1}{N}\sum_{\mathbf{k}_\parallel}e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)}\left\{G^{\sigma\sigma\,\nu\mu}_{\ell'\ell}(\mathbf{k};\omega) - \left[G^{\sigma\sigma\,\mu\nu}_{\ell\ell'}(\mathbf{k};\omega)\right]^*\right\} \ , \tag{H.8}$$

and

$$\langle \hat{c}^{\dagger}_{\ell'\nu\sigma}\hat{c}_{\ell\mu\sigma}\rangle(\mathbf{R}_i-\mathbf{R}_j) = \frac{i\hbar}{2\pi}\int_{-\infty}^{\omega_F}d\omega\frac{1}{N}\sum_{\mathbf{k}_\parallel}e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\left\{G^{\sigma\sigma\,\mu\nu}_{\ell\ell'}(\mathbf{k};\omega) - \left[G^{\sigma\sigma\,\nu\mu}_{\ell'\ell}(\mathbf{k};\omega)\right]^*\right\} \ , \tag{H.9}$$

we end up with

$$\begin{aligned}\langle \hat{I}^{\mathrm{C}}_{\ell\ell'}(\mathbf{R}_i,\mathbf{R}_j)\rangle = -\frac{e}{2\pi}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F}d\omega\frac{1}{N}\sum_{\mathbf{k}_\parallel}\Big\{&t^{\mu\nu}_{\ell\ell'}(\mathbf{R}_j-\mathbf{R}_i)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)}\left\{G^{\sigma\sigma\,\nu\mu}_{\ell'\ell}(\mathbf{k};\omega) - \left[G^{\sigma\sigma\,\mu\nu}_{\ell\ell'}(\mathbf{k};\omega)\right]^*\right\}\\ &- t^{\nu\mu}_{\ell'\ell}(\mathbf{R}_i-\mathbf{R}_j)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\left\{G^{\sigma\sigma\,\mu\nu}_{\ell\ell'}(\mathbf{k};\omega) - \left[G^{\sigma\sigma\,\nu\mu}_{\ell'\ell}(\mathbf{k};\omega)\right]^*\right\}\Big\} \ .\end{aligned} \tag{H.10}$$

For sites in the same plane,

$$\begin{aligned}\langle \hat{I}^{\mathrm{C}}_{\ell\ell}(\mathbf{R}_i,\mathbf{R}_j)\rangle = -\frac{e}{2\pi}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F}d\omega\frac{1}{N}\sum_{\mathbf{k}_\parallel}\Big\{&t^{\mu\nu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)}\left\{G^{\sigma\sigma\,\nu\mu}_{\ell\ell}(\mathbf{k};\omega) - \left[G^{\sigma\sigma\,\mu\nu}_{\ell\ell}(\mathbf{k};\omega)\right]^*\right\}\\ &- t^{\nu\mu}_{\ell\ell}(\mathbf{R}_i-\mathbf{R}_j)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\left\{G^{\sigma\sigma\,\mu\nu}_{\ell\ell}(\mathbf{k};\omega) - \left[G^{\sigma\sigma\,\nu\mu}_{\ell\ell}(\mathbf{k};\omega)\right]^*\right\}\Big\} \ .\end{aligned} \tag{H.11}$$

Changing $\mu \rightleftharpoons \nu$ in the second line above,

$$\begin{aligned}\langle \hat{I}^{\mathrm{C}}_{\ell\ell}(\mathbf{R}_i,\mathbf{R}_j)\rangle = -\frac{e}{2\pi}\sum_{\sigma}\sum_{\mu\nu}\int_{-\infty}^{\omega_F}d\omega\frac{1}{N}\sum_{\mathbf{k}_\parallel}&\left\{G^{\sigma\sigma\,\nu\mu}_{\ell\ell}(\mathbf{k};\omega) - \left[G^{\sigma\sigma\,\mu\nu}_{\ell\ell}(\mathbf{k};\omega)\right]^*\right\}\\ &\times\left\{t^{\mu\nu}_{\ell\ell}(\mathbf{R}_j-\mathbf{R}_i)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)} - t^{\mu\nu}_{\ell\ell}(\mathbf{R}_i-\mathbf{R}_j)e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j-\mathbf{R}_i)}\right\} \ .\end{aligned} \tag{H.12}$$

The interaction with a transverse magnetic field $\mathbf{b}_x = b_0\hat{\mathbf{x}}$ is described by the hamiltonian

$$\begin{aligned}
\hat{H}_x =& g_S\mu_B \sum_{\ell} \mathbf{b}_x \cdot \hat{\mathbf{S}}_\ell \\
=& g_S\mu_B b_0 \sum_{\ell} \hat{S}^x_\ell \\
=& \frac{g_S\mu_B b_0}{2} \frac{1}{N_\parallel} \sum_{\mathbf{k}_\parallel} \sum_{\ell,\gamma} \left\{ \hat{S}^{+\gamma\gamma}_{\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) + \hat{S}^{-\gamma\gamma}_{\ell\ell}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) \right\} \ .
\end{aligned} \tag{H.13}$$

The current generated by a transverse magnetic field $\mathbf{b} = b_0\hat{\mathbf{x}}$ is given by

$$\begin{aligned}
\delta\langle \hat{I}^{\mathrm{C}}_{\ell\ell}(\mathbf{R}_i, \mathbf{R}_j)\rangle = -\frac{e}{2\pi} \sum_\sigma \sum_{\mu\nu} \int_{-\infty}^{\omega_F} d\omega \frac{1}{N} \sum_{\mathbf{k}_\parallel} \left\{ \Delta G^{\sigma\sigma}_{\ell\ell}{}^{\nu\mu}(\mathbf{k};\omega) - \left[\Delta G^{\sigma\sigma}_{\ell\ell}{}^{\mu\nu}(\mathbf{k};\omega)\right]^* \right\} \\
\times \left\{ t^{\mu\nu}_{\ell\ell}(\mathbf{R}_j - \mathbf{R}_i) e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i - \mathbf{R}_j)} - t^{\mu\nu}_{\ell\ell}(\mathbf{R}_i - \mathbf{R}_j) e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_j - \mathbf{R}_i)} \right\} \ ,
\end{aligned} \tag{H.14}$$

where

$$\Delta G^{\sigma\sigma}_{\ell\ell}{}^{\nu\mu}(\mathbf{k};\omega) = \frac{g_S\mu_B b_0}{2} \sum_{\ell'\gamma} \left\{ G^{\sigma\uparrow}_{\ell\ell'}{}^{\nu\gamma}(\mathbf{k};\omega) G^{\downarrow\sigma}_{\ell'\ell}{}^{\gamma\mu}(\mathbf{k};\omega) + G^{\sigma\downarrow}_{\ell\ell'}{}^{\nu\gamma}(\mathbf{k};\omega) G^{\uparrow\sigma}_{\ell'\ell}{}^{\gamma\mu}(\mathbf{k};\omega) \right\} \ , \tag{H.15}$$

and

$$\left[\Delta G^{\sigma\sigma}_{\ell\ell}{}^{\mu\nu}(\mathbf{k};\omega)\right]^* = \frac{g_S\mu_B b_0}{2} \sum_{\ell'\gamma} \left\{ \left[G^{\sigma\uparrow}_{\ell\ell'}{}^{\mu\gamma}(\mathbf{k};\omega) G^{\downarrow\sigma}_{\ell'\ell}{}^{\gamma\nu}(\mathbf{k};\omega) \right]^* + \left[G^{\sigma\downarrow}_{\ell\ell'}{}^{\mu\gamma}(\mathbf{k};\omega) G^{\uparrow\sigma}_{\ell'\ell}{}^{\gamma\nu}(\mathbf{k};\omega) \right]^* \right\} \ . \tag{H.16}$$

The expectation value for the charge current is

$$\langle \hat{I}^{\mathrm{C}}_{ij}(\mathbf{R}_{\parallel}, \mathbf{R}'_{\parallel}) \rangle = \frac{1}{N_{\parallel}} \sum_{\mathbf{q}_{\parallel}} \left\{ \langle \hat{\mathcal{I}}^{\mathrm{C}}_{ij}(\mathbf{R}'_{\parallel} - \mathbf{R}_{\parallel}, \mathbf{q}_{\parallel}) \rangle e^{i\mathbf{q}_{\parallel}\cdot\mathbf{R}'_{\parallel}} - \langle \hat{\mathcal{I}}^{\mathrm{C}}_{ji}(\mathbf{R}_{\parallel} - \mathbf{R}'_{\parallel}, \mathbf{q}_{\parallel}) e^{i\mathbf{q}_{\parallel}\cdot\mathbf{R}_{\parallel}} \rangle \right\} \ , \tag{H.17}$$

where

$$\langle \hat{\mathcal{I}}^{\mathrm{C}}_{ij}(\mathbf{R}'_{\parallel} - \mathbf{R}_{\parallel}, \mathbf{q}_{\parallel}) \rangle = \frac{ie}{\hbar} \frac{1}{N_{\parallel}} \sum_{\mathbf{k}_{\parallel}} \sum_{\sigma} \sum_{\mu\nu} t^{\mu\nu}_{ij}(\mathbf{R}'_{\parallel} - \mathbf{R}_{\parallel}) \langle c^{\dagger}_{i\mu\sigma}(\mathbf{k}_{\parallel}) c_{j\nu\sigma}(\mathbf{k}_{\parallel} + \mathbf{q}_{\parallel}) \rangle e^{i\mathbf{k}_{\parallel}\cdot(\mathbf{R}'_{\parallel} - \mathbf{R}_{\parallel})} \ . \tag{H.18}$$

H.2 Spin current

$$
\begin{aligned}
\langle \hat{I}_{ij}^{m}(t)\rangle &= i\sum_{\sigma}\sum_{\mu\nu}\left\{ t_{ij}^{\mu\nu}\langle {S_{ij}^{m}}^{\mu\nu}(t)\rangle - t_{ji}^{\nu\mu}\langle {S_{ji}^{m}}^{\nu\mu}(t)\rangle \right\} \\
&= i\sum_{\sigma}\sum_{\mu\nu}\sum_{\alpha\beta}\left\{ t_{ij}^{\mu\nu}\langle c_{i\mu\alpha}^{\dagger}(t)\sigma_{\alpha\beta}^{m}c_{j\nu\beta}(t)\rangle - t_{ji}^{\nu\mu}\langle c_{j\nu\alpha}^{\dagger}(t)\sigma_{\alpha\beta}^{m}c_{i\mu\beta}(t)\rangle \right\} \ .
\end{aligned}
\tag{H.19}
$$

Appendix I

Relation between HF and RPA susceptibilities

In a periodic extended system, the monoelectronic propagators used to calculate the susceptibilities within HF only depends on the indices of sites inside the unit cell and on the relative position between unit cells, i.e.,

$$\begin{aligned}
\overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{R}_i,\mathbf{R}_j,\mathbf{R}_k,\mathbf{R}_l,\omega) =& \frac{i\hbar}{2\pi}\int_{-\infty}^{\infty} d\omega' f(\omega')\times \\
&\times\Big[\widetilde{G}^{\xi\mu\uparrow}_{li}(\mathbf{R}_i-\mathbf{R}_l,\omega')G^{\nu\gamma\downarrow}_{jk}(\mathbf{R}_k-\mathbf{R}_j,\omega'+\omega) \\
&-\widetilde{G}^{-\nu\gamma\downarrow}_{jk}(\mathbf{R}_k-\mathbf{R}_j,\omega')G^{-\xi\mu\uparrow}_{li}(\mathbf{R}_i-\mathbf{R}_l,\omega'-\omega) \\
&+\widetilde{G}^{\nu\gamma\downarrow}_{jk}(\mathbf{R}_k-\mathbf{R}_j,\omega')G^{-\xi\mu\uparrow}_{li}(\mathbf{R}_i-\mathbf{R}_l,\omega'-\omega) \\
&-\widetilde{G}^{-\xi\mu\uparrow}_{li}(\mathbf{R}_i-\mathbf{R}_l,\omega')G^{\nu\gamma\downarrow}_{jk}(\mathbf{R}_k-\mathbf{R}_j,\omega'+\omega)\Big] \\
=&\overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{R}_i-\mathbf{R}_l,\mathbf{R}_k-\mathbf{R}_j)\ ,
\end{aligned} \tag{I.1}$$

where $\{i,j,k,l\}$ are sites inside the unit cell. Its Fourier transform is

$$\begin{aligned}
\overset{0}{\widetilde{\chi}}{}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4) =& \sum_{\substack{\mathbf{R}_i\mathbf{R}_j\\ \mathbf{R}_k\mathbf{R}_l}} e^{i(\mathbf{k}_1\cdot\mathbf{R}_i+\mathbf{k}_2\cdot\mathbf{R}_j+\mathbf{k}_3\cdot\mathbf{R}_k+\mathbf{k}_4\cdot\mathbf{R}_l)}\overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{R}_i,\mathbf{R}_j,\mathbf{R}_k,\mathbf{R}_l) \\
=& \sum_{\substack{\mathbf{R}_i\mathbf{R}_j\\ \mathbf{R}_k\mathbf{R}_l}} e^{i\mathbf{k}_1\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{i\mathbf{k}_3\cdot(\mathbf{R}_k-\mathbf{R}_j)}e^{i(\mathbf{k}_3+\mathbf{k}_2)\cdot\mathbf{R}_j}e^{i(\mathbf{k}_1+\mathbf{k}_4)\cdot\mathbf{R}_l}\times \\
&\times\overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{R}_i-\mathbf{R}_l,\mathbf{R}_k-\mathbf{R}_j) \\
=&N^2\overset{0}{\widetilde{\chi}}{}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4)\ .
\end{aligned} \tag{I.2}$$

The usual magnetic susceptibility is given by

$$
\begin{aligned}
\overset{0}{\chi}{}_{ij}^{\mu\nu}(\mathbf{R}_i,\mathbf{R}_j,\omega) =& \frac{i\hbar}{2\pi}\int_{-\infty}^{\infty} d\omega' f(\omega') \times \\
& \times \Big[\widetilde{G}_{ji}^{\nu\mu\uparrow}(\mathbf{R}_i-\mathbf{R}_j,\omega') G_{ij}^{\mu\nu\downarrow}(\mathbf{R}_j-\mathbf{R}_i,\omega'+\omega) \\
& \quad - \widetilde{G}_{ij}^{-\mu\nu\downarrow}(\mathbf{R}_j-\mathbf{R}_i,\omega') G_{ji}^{-\nu\mu\uparrow}(\mathbf{R}_i-\mathbf{R}_j,\omega'-\omega) \\
& \quad + \widetilde{G}_{ij}^{\mu\nu\downarrow}(\mathbf{R}_j-\mathbf{R}_i,\omega') G_{ji}^{-\nu\mu\uparrow}(\mathbf{R}_i-\mathbf{R}_j,\omega'-\omega) \\
& \quad - \widetilde{G}_{ji}^{-\nu\mu\uparrow}(\mathbf{R}_i-\mathbf{R}_j,\omega') G_{ij}^{\mu\nu\downarrow}(\mathbf{R}_j-\mathbf{R}_i,\omega'+\omega) \Big] \\
=& \overset{0}{\chi}{}_{iijj}^{\mu\mu\nu\nu}(\mathbf{R}_i-\mathbf{R}_j,\mathbf{R}_j-\mathbf{R}_i) \\
=& \overset{0}{\chi}{}_{ij}^{\mu\nu}(\mathbf{R}_j-\mathbf{R}_i)\ ,
\end{aligned} \tag{I.3}
$$

So, the Fourier transform for extended systems is

$$
\begin{aligned}
\overset{0}{\widetilde{\chi}}{}_{ij}^{\mu\nu}(\mathbf{k}_1,\mathbf{k}_2) =& \sum_{\mathbf{R}_i\mathbf{R}_j} e^{i(\mathbf{k}_1\cdot\mathbf{R}_i+\mathbf{k}_2\cdot\mathbf{R}_j)} \overset{0}{\chi}{}_{ij}^{\mu\nu}(\mathbf{R}_i,\mathbf{R}_j) \\
=& \sum_{\mathbf{R}_i\mathbf{R}_j} e^{i\mathbf{k}_2\cdot(\mathbf{R}_j-\mathbf{R}_i)} e^{i(\mathbf{k}_1+\mathbf{k_2})\cdot\mathbf{R}_i} \overset{0}{\chi}{}_{ij}^{\mu\nu}(\mathbf{R}_j-\mathbf{R}_i) \\
=& N \overset{0}{\widetilde{\chi}}{}_{ij}^{\mu\nu}(\mathbf{k}_2)\delta(\mathbf{k}_1+\mathbf{k}_2)\ .
\end{aligned} \tag{I.4}
$$

It can be obtained from the general transformation given by Eq. (I.2) as

$$
\begin{aligned}
\overset{0}{\widetilde{\chi}}{}_{ijkl}^{\mu\nu\gamma\xi}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4) =& \sum_{\substack{\mathbf{R}_i\mathbf{R}_j\\ \mathbf{R}_k\mathbf{R}_l}} e^{i(\mathbf{k}_1\cdot\mathbf{R}_i+\mathbf{k}_2\cdot\mathbf{R}_j+\mathbf{k}_3\cdot\mathbf{R}_k+\mathbf{k}_4\cdot\mathbf{R}_l)} \overset{0}{\chi}{}_{ijkl}^{\mu\nu\gamma\xi}(\mathbf{R}_i,\mathbf{R}_j,\mathbf{R}_k,\mathbf{R}_l) \\
=& \sum_{\substack{\mathbf{R}_i\mathbf{R}_j\\ \mathbf{R}_k\mathbf{R}_l}} e^{i(\mathbf{k}_1\cdot\mathbf{R}_i+\mathbf{k}_2\cdot\mathbf{R}_j+\mathbf{k}_3\cdot\mathbf{R}_k+\mathbf{k}_4\cdot\mathbf{R}_l)} \overset{0}{\chi}{}_{iill}^{\mu\nu\gamma\xi}(\mathbf{R}_i,\mathbf{R}_i,\mathbf{R}_l,\mathbf{R}_l) \\
=& N^2 \sum_{\mathbf{R}_i\mathbf{R}_l} e^{i(\mathbf{k}_1\cdot\mathbf{R}_i+\mathbf{k}_4\cdot\mathbf{R}_l)} \overset{0}{\chi}{}_{iill}^{\mu\nu\gamma\xi}(\mathbf{R}_i,\mathbf{R}_i,\mathbf{R}_l,\mathbf{R}_l)\delta(\mathbf{k}_2)\delta(\mathbf{k}_3) \\
=& N^2 \sum_{\mathbf{R}_i\mathbf{R}_l} e^{i\mathbf{k}_4\cdot(\mathbf{R}_l-\mathbf{R}_i)} e^{i(\mathbf{k}_1+\mathbf{k}_4)\cdot\mathbf{R}_i} \overset{0}{\chi}{}_{iill}^{\mu\nu\gamma\xi}(\mathbf{R}_i-\mathbf{R}_l,\mathbf{R}_l-\mathbf{R}_i)\delta(\mathbf{k}_2)\delta(\mathbf{k}_3) \\
=& N^3 \overset{0}{\chi}{}_{il}^{\mu\nu\gamma\xi}(\mathbf{k}_4)\delta(\mathbf{k}_2)\delta(\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4)\ .
\end{aligned} \tag{I.5}
$$

Here, we defined the Fourier transform of the usual susceptibility as

$$
\begin{aligned}
\chi^{0\,\mu\nu}_{ij}(\mathbf{q},\omega) =& \sum_{\mathbf{R}} e^{i\mathbf{q}\cdot\mathbf{R}} \chi^{0\,\mu\nu}_{ij}(\mathbf{R},\omega) \\
=& \frac{i\hbar}{2\pi}\int_{-\infty}^{\infty} d\omega' f(\omega') \sum_{\mathbf{R}} e^{i\mathbf{q}\cdot\mathbf{R}} \times \\
&\times \Big[\widetilde{G}^{\nu\mu\uparrow}_{ji}(-\mathbf{R},\omega')G^{\mu\nu\downarrow}_{ij}(\mathbf{R},\omega'+\omega) - \widetilde{G}^{-\mu\nu\downarrow}_{ij}(\mathbf{R},\omega')G^{-\nu\mu\uparrow}_{ji}(-\mathbf{R},\omega'-\omega) \\
&\quad + \widetilde{G}^{\mu\nu\downarrow}_{ij}(\mathbf{R},\omega')G^{-\nu\mu\uparrow}_{ji}(-\mathbf{R},\omega'-\omega) - \widetilde{G}^{-\nu\mu\uparrow}_{ji}(-\mathbf{R},\omega')G^{\mu\nu\downarrow}_{ij}(\mathbf{R},\omega'+\omega)\Big] \\
=& \frac{i\hbar}{2\pi}\int_{-\infty}^{\infty} d\omega' f(\omega') \sum_{\mathbf{R}} e^{i\mathbf{q}\cdot\mathbf{R}} \frac{1}{N^2}\sum_{\mathbf{k},\mathbf{k}'} e^{-i\mathbf{k}\cdot\mathbf{R}} e^{i\mathbf{k}'\cdot\mathbf{R}} \\
&\times \Big[\widetilde{G}^{\nu\mu\uparrow}_{ji}(\mathbf{k}',\omega')G^{\mu\nu\downarrow}_{ij}(\mathbf{k},\omega'+\omega) - \widetilde{G}^{-\mu\nu\downarrow}_{ij}(\mathbf{k},\omega')G^{-\nu\mu\uparrow}_{ji}(\mathbf{k}',\omega'-\omega) \\
&\quad + \widetilde{G}^{\mu\nu\downarrow}_{ij}(\mathbf{k},\omega')G^{-\nu\mu\uparrow}_{ji}(\mathbf{k}',\omega'-\omega) - \widetilde{G}^{-\nu\mu\uparrow}_{ji}(\mathbf{k}',\omega')G^{\mu\nu\downarrow}_{ij}(\mathbf{k},\omega'+\omega)\Big] \\
=& \frac{i\hbar}{2\pi}\int_{-\infty}^{\infty} d\omega' f(\omega') \frac{1}{N}\sum_{\mathbf{k},\mathbf{k}'} \delta(\mathbf{q}-\mathbf{k}+\mathbf{k}') \\
&\times \Big[\widetilde{G}^{\nu\mu\uparrow}_{ji}(\mathbf{k}',\omega')G^{\mu\nu\downarrow}_{ij}(\mathbf{k},\omega'+\omega) - \widetilde{G}^{-\mu\nu\downarrow}_{ij}(\mathbf{k},\omega')G^{-\nu\mu\uparrow}_{ji}(\mathbf{k}',\omega'-\omega) \\
&\quad + \widetilde{G}^{\mu\nu\downarrow}_{ij}(\mathbf{k},\omega')G^{-\nu\mu\uparrow}_{ji}(\mathbf{k}',\omega'-\omega) - \widetilde{G}^{-\nu\mu\uparrow}_{ji}(\mathbf{k}',\omega')G^{\mu\nu\downarrow}_{ij}(\mathbf{k},\omega'+\omega)\Big] \\
=& \frac{i\hbar}{2\pi}\int_{-\infty}^{\infty} d\omega' f(\omega') \frac{1}{N}\sum_{\mathbf{k}} \\
&\times \Big[\widetilde{G}^{\nu\mu\uparrow}_{ji}(\mathbf{k}-\mathbf{q},\omega')G^{\mu\nu\downarrow}_{ij}(\mathbf{k},\omega'+\omega) - \widetilde{G}^{-\mu\nu\downarrow}_{ij}(\mathbf{k},\omega')G^{-\nu\mu\uparrow}_{ji}(\mathbf{k}-\mathbf{q},\omega'-\omega) \\
&\quad + \widetilde{G}^{\mu\nu\downarrow}_{ij}(\mathbf{k},\omega')G^{-\nu\mu\uparrow}_{ji}(\mathbf{k}-\mathbf{q},\omega'-\omega) - \widetilde{G}^{-\nu\mu\uparrow}_{ji}(\mathbf{k}-\mathbf{q},\omega')G^{\mu\nu\downarrow}_{ij}(\mathbf{k},\omega'+\omega)\Big] \\
=& \frac{1}{N}\sum_{\mathbf{k}} \chi^{0\,\mu\mu\nu\nu}_{iijj}(\mathbf{k}-\mathbf{q},\mathbf{k}) \ ,
\end{aligned}
\tag{I.6}
$$

When the two last indices are the same, we have

$$\begin{aligned}
\widetilde{\chi}^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4) =& \sum_{\substack{\mathbf{R}_i\mathbf{R}_j\\ \mathbf{R}_k\mathbf{R}_l}} e^{i(\mathbf{k}_1\cdot\mathbf{R}_i+\mathbf{k}_2\cdot\mathbf{R}_j+\mathbf{k}_3\cdot\mathbf{R}_k+\mathbf{k}_4\cdot\mathbf{R}_l)}\chi^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{R}_i,\mathbf{R}_j,\mathbf{R}_k,\mathbf{R}_l)\\
=& \sum_{\substack{\mathbf{R}_i\mathbf{R}_j\\ \mathbf{R}_k\mathbf{R}_l}} e^{i(\mathbf{k}_1\cdot\mathbf{R}_i+\mathbf{k}_2\cdot\mathbf{R}_j+\mathbf{k}_3\cdot\mathbf{R}_k+\mathbf{k}_4\cdot\mathbf{R}_l)}\chi^{0\,\mu\nu\gamma\xi}_{ijll}(\mathbf{R}_i,\mathbf{R}_j,\mathbf{R}_l,\mathbf{R}_l)\\
=& N\sum_{\substack{\mathbf{R}_i\mathbf{R}_j\\ \mathbf{R}_l}} e^{i(\mathbf{k}_1\cdot\mathbf{R}_i+\mathbf{k}_2\cdot\mathbf{R}_j+\mathbf{k}_4\cdot\mathbf{R}_l)}\chi^{0\,\mu\nu\gamma\xi}_{ijll}(\mathbf{R}_i,\mathbf{R}_j,\mathbf{R}_l,\mathbf{R}_l)\delta(\mathbf{k}_3)\\
=& N\sum_{\substack{\mathbf{R}_i\mathbf{R}_j\\ \mathbf{R}_l}} e^{i\mathbf{k}_1\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{i\mathbf{k}_2\cdot(\mathbf{R}_j-\mathbf{R}_l)}e^{i(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_4)\cdot\mathbf{R}_l}\chi^{0\,\mu\nu\gamma\xi}_{ijll}(\mathbf{R}_i-\mathbf{R}_l,\mathbf{R}_l-\mathbf{R}_j)\delta(\mathbf{k}_3)\\
=& N^2\chi^{0\,\mu\nu\gamma\xi}_{ijll}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_4)\delta(\mathbf{k}_3)
\end{aligned}\quad , \tag{I.7}$$

while when the first two indices are equal

$$\begin{aligned}
\widetilde{\chi}^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4) =& \sum_{\substack{\mathbf{R}_i\mathbf{R}_j\\ \mathbf{R}_k\mathbf{R}_l}} e^{i(\mathbf{k}_1\cdot\mathbf{R}_i+\mathbf{k}_2\cdot\mathbf{R}_j+\mathbf{k}_3\cdot\mathbf{R}_k+\mathbf{k}_4\cdot\mathbf{R}_l)}\chi^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{R}_i,\mathbf{R}_j,\mathbf{R}_k,\mathbf{R}_l)\\
=& \sum_{\substack{\mathbf{R}_i\mathbf{R}_j\\ \mathbf{R}_k\mathbf{R}_l}} e^{i(\mathbf{k}_1\cdot\mathbf{R}_i+\mathbf{k}_2\cdot\mathbf{R}_j+\mathbf{k}_3\cdot\mathbf{R}_k+\mathbf{k}_4\cdot\mathbf{R}_l)}\chi^{0\,\mu\nu\gamma\xi}_{iikl}(\mathbf{R}_i,\mathbf{R}_i,\mathbf{R}_k,\mathbf{R}_l)\\
=& N\sum_{\substack{\mathbf{R}_i\\ \mathbf{R}_k\mathbf{R}_l}} e^{i(\mathbf{k}_1\cdot\mathbf{R}_i+\mathbf{k}_3\cdot\mathbf{R}_k+\mathbf{k}_4\cdot\mathbf{R}_l)}\chi^{0\,\mu\nu\gamma\xi}_{iikl}(\mathbf{R}_i,\mathbf{R}_i,\mathbf{R}_k,\mathbf{R}_l)\delta(\mathbf{k}_2)\\
=& N\sum_{\substack{\mathbf{R}_i\\ \mathbf{R}_k\mathbf{R}_l}} e^{i(\mathbf{k}_1+\mathbf{k}_3+\mathbf{k}_4)\cdot\mathbf{R}_i}e^{i\mathbf{k}_3\cdot(\mathbf{R}_k-\mathbf{R}_i)}e^{i\mathbf{k}_4\cdot(\mathbf{R}_l-\mathbf{R}_i)}\chi^{0\,\mu\nu\gamma\xi}_{iikl}(\mathbf{R}_i-\mathbf{R}_l,\mathbf{R}_k-\mathbf{R}_i)\delta(\mathbf{k}_2)\\
=& N^2\chi^{0\,\mu\nu\gamma\xi}_{iikl}(-\mathbf{k}_4,\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_3+\mathbf{k}_4)\delta(\mathbf{k}_2)
\end{aligned}\quad . \tag{I.8}$$

Since the RPA susceptibility is written in terms of the Hartree-Fock, it has the same dependence on the positions and, consequently, on the $\mathbf{k}$ indices. The relation between the RPA and HF susceptibilities is

$$\chi^{\mu\nu\gamma\xi}_{ijkl} = \chi^{0\,\mu\nu\gamma\xi}_{ijkl} - \sum_{\substack{m\\ \eta\lambda\alpha\beta}} \chi^{0\,\mu\nu\eta\lambda}_{ijmm}U^{\eta\alpha\beta\lambda}_{m}\chi^{\alpha\beta\gamma\xi}_{mmkl}\ . \tag{I.9}$$

In extended systems, the RPA susceptibility will also depend on the positions the same way showed in Eq. (I.1). Thus, the relation given in Eq. (I.9) can be rewritten as

$$\chi^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{R}_i-\mathbf{R}_l,\mathbf{R}_k-\mathbf{R}_j) = \overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{R}_i-\mathbf{R}_l,\mathbf{R}_k-\mathbf{R}_j) \\ - \sum_{\substack{m\mathbf{R}_m \\ \eta\lambda\alpha\beta}} \overset{0}{\chi}{}^{\mu\nu\eta\lambda}_{ijmm}(\mathbf{R}_i-\mathbf{R}_m,\mathbf{R}_m-\mathbf{R}_j)U^{\eta\alpha\beta\lambda}_m \chi^{\alpha\beta\gamma\xi}_{mmkl}(\mathbf{R}_m-\mathbf{R}_l,\mathbf{R}_k-\mathbf{R}_m)\ . \tag{I.10}$$

We now make use of the Lowde-Windsor parametrization, i.e., $U^{\eta\alpha\beta\lambda}_m = U^{\lambda\alpha}_m\delta_{\eta\lambda}\delta_{\alpha\beta}$, and also assume that $U^{\lambda\alpha}_m = U_m$ for d orbitals and 0 for s e p orbitals. This means that, for a fixed site m, the matrix $\underline{U}_m$ in the orbital space has the form

$$\underline{U}_m = \begin{pmatrix} 0&0&0&0&0&0&0&0&0\\ 0&0&0&0&0&0&0&0&0\\ 0&0&0&0&0&0&0&0&0\\ 0&0&0&0&0&0&0&0&0\\ 0&0&0&0&U_m&U_m&U_m&U_m&U_m\\ 0&0&0&0&U_m&U_m&U_m&U_m&U_m\\ 0&0&0&0&U_m&U_m&U_m&U_m&U_m\\ 0&0&0&0&U_m&U_m&U_m&U_m&U_m\\ 0&0&0&0&U_m&U_m&U_m&U_m&U_m \end{pmatrix} \tag{I.11}$$

Note that this matrix is not diagonal in orbital space, since $U^{\lambda\alpha}_m \neq \delta_{\lambda\alpha}U_m$. So,

$$\chi^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{R}_i-\mathbf{R}_l,\mathbf{R}_k-\mathbf{R}_j) = \overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{R}_i-\mathbf{R}_l,\mathbf{R}_k-\mathbf{R}_j) \\ - \sum_{\substack{m\mathbf{R}_m \\ \lambda\alpha}} \overset{0}{\chi}{}^{\mu\nu\lambda\lambda}_{ijmm}(\mathbf{R}_i-\mathbf{R}_m,\mathbf{R}_m-\mathbf{R}_j)U^{\lambda\alpha}_m \chi^{\alpha\alpha\gamma\xi}_{mmkl}(\mathbf{R}_m-\mathbf{R}_l,\mathbf{R}_k-\mathbf{R}_m)\ . \tag{I.12}$$

Fourier transforming the position variables inside the planes, $\mathbf{R}$, and dropping the orbital indices for simplicity, we obtain

$$\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi^0_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) \\ - \sum_{m\mathbf{R}_m}\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_m)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_m-\mathbf{R}_j)}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U_m \\ \times\frac{1}{N^2}\sum_{\mathbf{q}_\parallel,\mathbf{q}'_\parallel} e^{-i\mathbf{q}_\parallel\cdot(\mathbf{R}_m-\mathbf{R}_l)}e^{-i\mathbf{q}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_m)}\chi_{mmkl}(\mathbf{q}_\parallel,\mathbf{q}'_\parallel)\ . \tag{I.13}$$

This equation can be rewritten as

$$\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi^0_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)$$
$$-\sum_{m\mathbf{R}_m}\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U_m$$
$$\times\frac{1}{N^2}\sum_{\mathbf{q}_\parallel,\mathbf{q}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_m)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_m-\mathbf{R}_k)}e^{-i\mathbf{q}_\parallel\cdot(\mathbf{R}_m-\mathbf{R}_l)}e^{-i\mathbf{q}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_m)}\chi_{mmkl}(\mathbf{q}_\parallel,\mathbf{q}'_\parallel)\ . \tag{I.14}$$

Now, using $\sum_{\mathbf{R}_m} e^{i(\mathbf{k}_\parallel-\mathbf{k}'_\parallel-\mathbf{q}_\parallel+\mathbf{q}'_\parallel)\cdot\mathbf{R}_m} = N\delta(\mathbf{k}_\parallel-\mathbf{k}'_\parallel-\mathbf{q}_\parallel+\mathbf{q}'_\parallel)$ and removing the common sums and prefactors (and putting back the orbital indices)

$$\chi^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \chi^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) - \sum_m \chi^{0\,\mu\nu\lambda\lambda}_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U^{\lambda\alpha}_m\frac{1}{N}\sum_{\mathbf{q}_\parallel} e^{-i(\mathbf{k}_\parallel-\mathbf{q}_\parallel)\cdot(\mathbf{R}_l-\mathbf{R}_k)}\chi^{\alpha\alpha\gamma\xi}_{mmkl}(\mathbf{q}_\parallel,\mathbf{k}'_\parallel+\mathbf{q}_\parallel-\mathbf{k}_\parallel)\ . \tag{I.15}$$

Since we have symmetry in the reciprocal space, we can make the change $\mathbf{q}_\parallel-\mathbf{k}_\parallel = \mathbf{q}'_\parallel \to \mathbf{q}_\parallel$ to obtain

$$\chi^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \chi^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) - \sum_m \chi^{0\,\mu\nu\lambda\lambda}_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U^{\lambda\alpha}_m\frac{1}{N}\sum_{\mathbf{q}_\parallel} e^{i\mathbf{q}_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_k)}\chi^{\alpha\alpha\gamma\xi}_{mmkl}(\mathbf{k}_\parallel+\mathbf{q}_\parallel,\mathbf{k}'_\parallel+\mathbf{q}_\parallel)\ . \tag{I.16}$$

We can also use the relation obtained in Eq. (F.49), where the last term $\chi^0 U\chi$ whould be changed to $\chi U\chi^0$ in the last term.

Another way to obtain the relation between RPA and HF susceptibilities for extended systems is the following. The Fourier transform of the general relation between the two susceptibilities with four indices is

$$\widetilde{\chi}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,\omega) = \widetilde{\chi}^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,\omega)$$
$$- \frac{1}{N^3}\sum_{\substack{m\\ \eta\lambda\alpha\beta}}\sum_{\substack{\mathbf{k}'_1\mathbf{k}'_2\\ \mathbf{k}'_3\mathbf{k}'_4}} \widetilde{\chi}^{0\,\mu\nu\eta\lambda}_{ijmm}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}'_1,\mathbf{k}'_2,\omega)U^m_{\eta\alpha\beta\lambda}\delta(\mathbf{k}'_1+\mathbf{k}'_2+\mathbf{k}'_3+\mathbf{k}'_4)\widetilde{\chi}^{\alpha\beta\gamma\xi}_{mmkl}(\mathbf{k}'_3,\mathbf{k}'_4,\mathbf{k}_3,\mathbf{k}_4,\omega)\ . \tag{I.17}$$

Here, $\{i,j,k,l,m\}$ also represents planes indices. Substituting Eq. (I.2) in Eq. (I.17) for both susceptibilities, we obtain

$$\widetilde{\chi}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4) = \widetilde{\chi}^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4)$$
$$- \frac{1}{N}\sum_{\substack{m\\ \eta\lambda\alpha\beta}}\sum_{\substack{\mathbf{k}'_1\mathbf{k}'_2\\ \mathbf{k}'_3\mathbf{k}'_4}} \widetilde{\chi}^{0\,\mu\nu\eta\lambda}_{ijmm}(\mathbf{k}_1,\mathbf{k}'_1,\omega)\delta(\mathbf{k}_2+\mathbf{k}'_1)\delta(\mathbf{k}_1+\mathbf{k}'_2)U^m_{\eta\alpha\beta\lambda}\widetilde{\chi}^{\alpha\beta\gamma\xi}_{mmkl}(\mathbf{k}'_3,\mathbf{k}_3,\omega)\delta(\mathbf{k}'_4+\mathbf{k}_3)\delta(\mathbf{k}'_3+\mathbf{k}_4)$$
$$\times\,\delta(\mathbf{k}'_1+\mathbf{k}'_2+\mathbf{k}'_3+\mathbf{k}'_4)\ . \tag{I.18}$$

or

$$\begin{aligned}&\widetilde{\chi}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4) = \widetilde{\chi}^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4)\\ &- \frac{1}{N}\sum_{\substack{m\\ \eta\lambda\alpha\beta}} \widetilde{\chi}^{0\,\mu\nu\eta\lambda}_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2,\omega)U^m_{\eta\alpha\beta\lambda}\widetilde{\chi}^{\alpha\beta\gamma\xi}_{mmkl}(-\mathbf{k}_4,\mathbf{k}_3,\omega)\delta(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3+\mathbf{k}_4)\ .\end{aligned} \tag{I.19}$$

Particularizing for $\mathbf{k}_2 = -\mathbf{k}_3$ and $\mathbf{k}_4 = -\mathbf{k}_1$

$$\widetilde{\chi}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega) = \widetilde{\chi}^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega) - \frac{1}{N}\sum_{\substack{m\\ \eta\lambda\alpha\beta}} \widetilde{\chi}^{0\,\mu\nu\eta\lambda}_{ijmm}(\mathbf{k}_1,\mathbf{k}_3,\omega)U^m_{\eta\alpha\beta\lambda}\widetilde{\chi}^{\alpha\beta\gamma\xi}_{mmkl}(\mathbf{k}_1,\mathbf{k}_3,\omega)\ . \tag{I.20}$$

The usual susceptibility relation can be obtained by substituting the result obtained in Eq. (I.5) into Eq. (I.17):

$$
\begin{aligned}
&N^3\chi_{il}(\mathbf{k}_4)\delta(\mathbf{k}_2)\delta(\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4) = N^3\overset{0}{\chi}_{il}(\mathbf{k}_4)\delta(\mathbf{k}_2)\delta(\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4) \\
&-\frac{1}{N^3}\sum_m \sum_{\substack{\mathbf{k}_1'\mathbf{k}_2'\\ \mathbf{k}_3'\mathbf{k}_4'}} N^3\overset{0}{\chi}_{im}(\mathbf{k}_2')\delta(\mathbf{k}_2)\delta(\mathbf{k}_1')\delta(\mathbf{k}_1+\mathbf{k}_2')U^m\delta(\mathbf{k}_1'+\mathbf{k}_2'+\mathbf{k}_3'+\mathbf{k}_4')N^3\chi_{ml}(\mathbf{k}_4)\delta(\mathbf{k}_4')\delta(\mathbf{k}_3)\delta(\mathbf{k}_3'+\mathbf{k}_4) \\
&\chi_{il}(\mathbf{k}_4)\delta(\mathbf{k}_2)\delta(\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4) = \overset{0}{\chi}_{il}(\mathbf{k}_4)\delta(\mathbf{k}_2)\delta(\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4) \\
&-\sum_m \overset{0}{\chi}_{im}(-\mathbf{k}_1)\delta(\mathbf{k}_2)U^m\delta(-\mathbf{k}_1-\mathbf{k}_4)\chi_{ml}(\mathbf{k}_4)\delta(\mathbf{k}_3) \\
&\chi_{il}(\mathbf{k}_4)\delta(\mathbf{k}_2)\delta(\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4) = \overset{0}{\chi}_{il}(\mathbf{k}_4)\delta(\mathbf{k}_2)\delta(\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4) \\
&-\sum_m \overset{0}{\chi}_{im}(\mathbf{k}_4)\delta(\mathbf{k}_2)U^m\chi_{ml}(\mathbf{k}_4)\delta(\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4) \\
&\chi_{il}(\mathbf{k}_4) = \overset{0}{\chi}_{il}(\mathbf{k}_4) - \sum_m \overset{0}{\chi}_{im}(\mathbf{k}_4)U^m\chi_{ml}(\mathbf{k}_4)\ ,
\end{aligned}
\tag{I.21}
$$

where we have omitted the orbital indices for simplicity.

When the first two indices are the same (e.g., calculation of densities due to external electric fields),

$$
\begin{aligned}
&N^2\chi_{iikl}(-\mathbf{k}_4,\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_3+\mathbf{k}_4)\delta(\mathbf{k}_2) = N^2\chi^0_{iikl}(-\mathbf{k}_4,\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_3+\mathbf{k}_4)\delta(\mathbf{k}_2) \\
&-\frac{1}{N^3}\sum_m \sum_{\substack{\mathbf{k}_1'\mathbf{k}_2'\\ \mathbf{k}_3'\mathbf{k}_4'}} N^3\chi_{im}(\mathbf{k}_2')\delta(\mathbf{k}_2)\delta(\mathbf{k}_1')\delta(\mathbf{k}_1+\mathbf{k}_2')U^m\delta(\mathbf{k}_1'+\mathbf{k}_2'+\mathbf{k}_3'+\mathbf{k}_4')N^2\chi^0_{mmkl}(-\mathbf{k}_4,\mathbf{k}_3)\delta(\mathbf{k}_3'+\mathbf{k}_3+\mathbf{k}_4)\delta \\
&\chi_{iikl}(-\mathbf{k}_4,\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_3+\mathbf{k}_4)\delta(\mathbf{k}_2) = \chi^0_{iikl}(-\mathbf{k}_4,\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_3+\mathbf{k}_4)\delta(\mathbf{k}_2) \\
&-\sum_m \chi_{im}(-\mathbf{k}_1)\delta(\mathbf{k}_2)U^m\delta(-\mathbf{k}_1-\mathbf{k}_3-\mathbf{k}_4)\chi^0_{mmkl}(-\mathbf{k}_4,\mathbf{k}_3)
\end{aligned}
\tag{I.22}
$$

Summing over $\mathbf{k}_2$ and $\mathbf{k}_1$,

$$
\begin{aligned}
\chi_{iikl}(-\mathbf{k}_4,\mathbf{k}_3) &= \chi^0_{iikl}(-\mathbf{k}_4,\mathbf{k}_3) - \sum_m \chi_{im}(\mathbf{k}_3+\mathbf{k}_4)U^m\chi^0_{mmkl}(-\mathbf{k}_4,\mathbf{k}_3) \\
\chi_{iikl}(\mathbf{k}_4,\mathbf{k}_3) &= \chi^0_{iikl}(\mathbf{k}_4,\mathbf{k}_3) - \sum_m \chi_{im}(\mathbf{k}_3-\mathbf{k}_4)U^m\chi^0_{mmkl}(\mathbf{k}_4,\mathbf{k}_3)
\end{aligned}\ .
\tag{I.23}
$$

When the last two indices are the same (e.g., currents generated when a magnetic field is applied),

$$
\begin{aligned}
&N^2\chi_{ijll}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_4)\delta(\mathbf{k}_3) = N^2\chi^0_{ijll}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_4)\delta(\mathbf{k}_3)\\
&-\frac{1}{N^3}\sum_m\sum_{\substack{\mathbf{k}'_1\mathbf{k}'_2\\ \mathbf{k}'_3\mathbf{k}'_4}} N^2\chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}'_2)\delta(\mathbf{k}'_1)U^m\delta(\mathbf{k}'_1+\mathbf{k}'_2+\mathbf{k}'_3+\mathbf{k}'_4)N^3\chi_{ml}(\mathbf{k}_4)\delta(\mathbf{k}'_4)\delta(\mathbf{k}_3)\delta(\mathbf{k}'_3+\\
&\chi_{ijll}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_4)\delta(\mathbf{k}_3) = \chi^0_{ijll}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_4)\delta(\mathbf{k}_3)\\
&-\sum_m \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\delta(-\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_4)\chi_{ml}(\mathbf{k}_4)\delta(\mathbf{k}_3)\ .
\end{aligned}
\tag{I.24}
$$

Summing over $\mathbf{k}_3$ and $\mathbf{k}_4$,

$$
\begin{aligned}
\chi_{ijll}(\mathbf{k}_1,-\mathbf{k}_2) &= \chi^0_{ijll}(\mathbf{k}_1,-\mathbf{k}_2) - \sum_m \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\chi_{ml}(-\mathbf{k}_1-\mathbf{k}_2)\\
\chi_{ijll}(\mathbf{k}_1,\mathbf{k}_2) &= \chi^0_{ijll}(\mathbf{k}_1,\mathbf{k}_2) - \sum_m \chi^0_{ijmm}(\mathbf{k}_1,\mathbf{k}_2)U^m\chi_{ml}(\mathbf{k}_2-\mathbf{k}_1)\ .
\end{aligned}
\tag{I.25}
$$

Finally, the most general result is given by

$$
\begin{aligned}
&N^2\widetilde{\chi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4) = N^2\widetilde{\chi}^0_{ijkl}(\mathbf{k}_1,\mathbf{k}_3)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4)\\
&-\frac{1}{N^3}\sum_m\sum_{\substack{\mathbf{k}'_1\mathbf{k}'_2\\ \mathbf{k}'_3\mathbf{k}'_4}} N^2\chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}'_2)\delta(\mathbf{k}'_1)U^m\delta(\mathbf{k}'_1+\mathbf{k}'_2+\mathbf{k}'_3+\mathbf{k}'_4)N^2\chi_{mmkl}(-\mathbf{k}_4,\mathbf{k}_3)\delta(\mathbf{k}'_3+\mathbf{k}\\
&\widetilde{\chi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4) = \widetilde{\chi}^0_{ijkl}(\mathbf{k}_1,\mathbf{k}_3)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4)\\
&-\frac{1}{N}\sum_m \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\delta(-\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4)\chi_{mmkl}(-\mathbf{k}_4,\mathbf{k}_3)
\end{aligned}
\tag{I.26}
$$

which is the same result as Eq. (I.19).

Substituting Eq. (I.23) into Eq. (I.26),

$$
\begin{aligned}
&\widetilde{\chi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4) = \widetilde{\chi}^0_{ijkl}(\mathbf{k}_1,\mathbf{k}_3)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4)\\
&-\frac{1}{N}\sum_m \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\delta(-\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4)\chi_{mmkl}(-\mathbf{k}_4,\mathbf{k}_3)\\
&\widetilde{\chi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4) = \widetilde{\chi}^0_{ijkl}(\mathbf{k}_1,\mathbf{k}_3)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4)\\
&-\frac{1}{N}\sum_m \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\delta(-\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4)\chi^0_{mmkl}(-\mathbf{k}_4,\mathbf{k}_3)\\
&-\frac{1}{N}\sum_{mn} \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\delta(-\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}_3-\mathbf{k}_4)\chi_{mn}(\mathbf{k}_3+\mathbf{k}_4)U^n\chi^0_{nnkl}(-\mathbf{k}_4,\mathbf{k}_3)
\end{aligned}
\tag{I.27}
$$

Summing over $\mathbf{k}_4$,

$$\begin{aligned}&\widetilde{\chi}_{ijkl}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_2+\mathbf{k}_3)=\widetilde{\chi}^0_{ijkl}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_2+\mathbf{k}_3)\\&-\frac{1}{N}\sum_m \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\chi^0_{mmkl}(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3,\mathbf{k}_3)\\&-\frac{1}{N}\sum_{mn}\chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\chi_{mn}(-\mathbf{k}_1-\mathbf{k}_2)U^n\chi^0_{nnkl}(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3,\mathbf{k}_3)\end{aligned}\quad. \tag{I.28}$$

Particularizing for $\widetilde{\chi}_{ijkl}(\mathbf{k},\mathbf{k})$,

$$\begin{aligned}&\widetilde{\chi}_{ijkl}(\mathbf{k},\mathbf{k})\delta(\mathbf{k}_3-\mathbf{k})=\widetilde{\chi}^0_{ijkl}(\mathbf{k},\mathbf{k})\delta(\mathbf{k}_3-\mathbf{k})\\&-\frac{1}{N}\sum_m \chi^0_{ijmm}(\mathbf{k},\mathbf{k})U^m\chi^0_{mmkl}(\mathbf{k}_3,\mathbf{k}_3)\\&-\frac{1}{N}\sum_{mn}\chi^0_{ijmm}(\mathbf{k},\mathbf{k})U^m\chi_{mn}(0)U^n\chi^0_{nnkl}(\mathbf{k}_3,\mathbf{k}_3)\end{aligned}\quad. \tag{I.29}$$

Particularizing for $\widetilde{\chi}_{ijkl}(\mathbf{k}_1,-\mathbf{k}_2)$,

$$\begin{aligned}&\widetilde{\chi}_{ijkl}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4)=\widetilde{\chi}^0_{ijkl}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_2+\mathbf{k}_3)\delta(\mathbf{k}_1+\mathbf{k}_4)\\&-\frac{1}{N}\sum_m \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\delta(-\mathbf{k}_1-\mathbf{k}_4)\chi^0_{mmkl}(-\mathbf{k}_4,-\mathbf{k}_2)\\&-\frac{1}{N}\sum_{mn}\chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\delta(-\mathbf{k}_1-\mathbf{k}_4)\chi_{mn}(\mathbf{k}_4-\mathbf{k}_2)U^n\chi^0_{nnkl}(-\mathbf{k}_4,-\mathbf{k}_2)\end{aligned}\quad. \tag{I.30}$$

Summing over $\mathbf{k}_3$ and then $\mathbf{k}_4$,

$$\begin{aligned}&\widetilde{\chi}_{ijkl}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_1+\mathbf{k}_4)=\widetilde{\chi}^0_{ijkl}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_1+\mathbf{k}_4)\\&-\sum_m \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\delta(-\mathbf{k}_1-\mathbf{k}_4)\chi^0_{mmkl}(-\mathbf{k}_4,-\mathbf{k}_2)\\&-\sum_{mn}\chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\delta(-\mathbf{k}_1-\mathbf{k}_4)\chi_{mn}(\mathbf{k}_4-\mathbf{k}_2)U^n\chi^0_{nnkl}(-\mathbf{k}_4,-\mathbf{k}_2)\\&\widetilde{\chi}_{ijkl}(\mathbf{k}_1,-\mathbf{k}_2)=\widetilde{\chi}^0_{ijkl}(\mathbf{k}_1,-\mathbf{k}_2)\\&-\sum_m \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\chi^0_{mmkl}(\mathbf{k}_1,-\mathbf{k}_2)\\&-\sum_{mn}\chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\chi_{mn}(-\mathbf{k}_1-\mathbf{k}_2)U^n\chi^0_{nnkl}(\mathbf{k}_1,-\mathbf{k}_2)\end{aligned}\quad, \tag{I.31}$$

$$
\begin{aligned}
&\widetilde{\chi}_{ijkl}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_1+\mathbf{k}_4) = \widetilde{\chi}^0_{ijkl}(\mathbf{k}_1,-\mathbf{k}_2)\delta(\mathbf{k}_1+\mathbf{k}_4)\\
&-\frac{1}{N}\sum_m \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\delta(-\mathbf{k}_1-\mathbf{k}_4)\chi^0_{mmkl}(-\mathbf{k}_4,-\mathbf{k}_2)\\
&-\frac{1}{N}\sum_{mn} \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\delta(-\mathbf{k}_1-\mathbf{k}_4)\chi_{mn}(\mathbf{k}_4-\mathbf{k}_2)U^n\chi^0_{nnkl}(-\mathbf{k}_4,-\mathbf{k}_2)\\
&\widetilde{\chi}_{ijkl}(\mathbf{k}_1,-\mathbf{k}_2) = \widetilde{\chi}^0_{ijkl}(\mathbf{k}_1,-\mathbf{k}_2)\\
&-\frac{1}{N}\sum_m \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\chi^0_{mmkl}(\mathbf{k}_1,-\mathbf{k}_2)\\
&-\frac{1}{N}\sum_{mn} \chi^0_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2)U^m\chi_{mn}(-\mathbf{k}_1-\mathbf{k}_2)U^n\chi^0_{nnkl}(\mathbf{k}_1,-\mathbf{k}_2) \qquad , \qquad (\mathrm{I}.32)\\
&\widetilde{\chi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_2) = \widetilde{\chi}^0_{ijkl}(\mathbf{k}_1,\mathbf{k}_2)\\
&-\frac{1}{N}\sum_m \chi^0_{ijmm}(\mathbf{k}_1,\mathbf{k}_2)U^m\chi^0_{mmkl}(\mathbf{k}_1,\mathbf{k}_2)\\
&-\frac{1}{N}\sum_{mn} \chi^0_{ijmm}(\mathbf{k}_1,\mathbf{k}_2)U^m\chi_{mn}(\mathbf{k}_2-\mathbf{k}_1)U^n\chi^0_{nnkl}(\mathbf{k}_1,\mathbf{k}_2)\\
&\widetilde{\chi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_2) = \widetilde{\chi}^0_{ijkl}(\mathbf{k}_1,\mathbf{k}_2)\\
&-\frac{1}{N}\sum_{mn} \chi^0_{ijmm}(\mathbf{k}_1,\mathbf{k}_2)U^m\left[\delta_{mn}+\chi_{mn}(\mathbf{k}_2-\mathbf{k}_1)U^n\right]\chi^0_{nnkl}(\mathbf{k}_1,\mathbf{k}_2)
\end{aligned}
$$

If instead we sum both sides in $\mathbf{k}_2$ and $\mathbf{k}_4$,

$$\begin{aligned}
\widetilde{\chi}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega) &= \widetilde{\chi}^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega) - \frac{1}{N}\sum_{\substack{m\\ \eta\lambda\alpha\beta}}\sum_{\mathbf{k}'_2}\widetilde{\chi}^{0\,\mu\nu\eta\lambda}_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2,\omega)U^m_{\eta\alpha\beta\lambda}\widetilde{\chi}^{\alpha\beta\gamma\xi}_{mmkl}(\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3,\mathbf{k}_3,\omega)\\
&= \widetilde{\chi}^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega) - \frac{1}{N}\sum_{\substack{m\\ \eta\lambda\alpha\beta}}\sum_{\mathbf{k}'_4}\widetilde{\chi}^{0\,\mu\nu\eta\lambda}_{ijmm}(\mathbf{k}_1,\mathbf{k}_1+\mathbf{k}_3+\mathbf{k}_4,\omega)U^m_{\eta\alpha\beta\lambda}\widetilde{\chi}^{\alpha\beta\gamma\xi}_{mmkl}(-\mathbf{k}_4,\mathbf{k}_3,\omega)\ .
\end{aligned} \tag{I.33}$$

Use of the Lowde-Windsor parametrization once again, i.e., $U^{\eta\alpha\beta\lambda}_m = U^{\lambda\alpha}_m\delta_{\eta\lambda}\delta_{\alpha\beta}$ (and assuming $U^{\lambda\alpha}_m = U_m$ for d orbitals and 0 for s e p orbitals),

$$\widetilde{\chi}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega) = \widetilde{\chi}^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega) - \frac{1}{N}\sum_{\substack{m\\ \alpha\beta}}\widetilde{\chi}^{0\,\mu\nu\alpha\alpha}_{ijmm}(\mathbf{k}_1,\mathbf{k}_3,\omega)U^m_{\alpha\beta}\widetilde{\chi}^{\beta\beta\gamma\xi}_{mmkl}(\mathbf{k}_1,\mathbf{k}_3,\omega)\ , \tag{I.34}$$

where we have changed $\alpha\to\beta$ and $\lambda\to\alpha$.

I.1 Magnetic field perturbation: $\chi_{ij\ell\ell}(\mathbf{k}_{\parallel}, \omega)$

To calculate the spin currents, we need susceptibilities of the form $\chi_{ijll}^{\mu\nu\gamma\gamma}$. In this case, the relation above becomes

$$\chi_{ijll}^{\mu\nu\gamma\gamma}(\mathbf{R}_i - \mathbf{R}_l, \mathbf{R}_l - \mathbf{R}_j) = \overset{0}{\chi}{}_{ijll}^{\mu\nu\gamma\gamma}(\mathbf{R}_i - \mathbf{R}_l, \mathbf{R}_l - \mathbf{R}_j) - \sum_{\substack{m\mathbf{R}_m \\ \eta\lambda\alpha\beta}} \overset{0}{\chi}{}_{ijmm}^{\mu\nu\eta\lambda}(\mathbf{R}_i - \mathbf{R}_m, \mathbf{R}_m - \mathbf{R}_j) U_m^{\eta\alpha\beta\lambda} \chi_{mmll}^{\alpha\beta\gamma\gamma}(\mathbf{R}_l - \mathbf{R}_m) \ , \tag{I.35}$$

Using Lowde-Windsor parametrization, i.e., $U_m^{\eta\alpha\beta\lambda} = U_m^{\lambda\alpha}\delta_{\eta\lambda}\delta_{\alpha\beta}$, where $U_m^{\lambda\alpha} = U_m$ for d orbitals and 0 for s e p orbitals. This means that, for a fixed plane m, the matrix $\underline{U}_m$ in the orbital space has the form

$$\underline{U}_m = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & U_m & U_m & U_m & U_m & U_m \\ 0 & 0 & 0 & 0 & U_m & U_m & U_m & U_m & U_m \\ 0 & 0 & 0 & 0 & U_m & U_m & U_m & U_m & U_m \\ 0 & 0 & 0 & 0 & U_m & U_m & U_m & U_m & U_m \\ 0 & 0 & 0 & 0 & U_m & U_m & U_m & U_m & U_m \end{pmatrix} \tag{I.36}$$

So,

$$\chi_{ijll}^{\mu\nu\gamma\gamma}(\mathbf{R}_i - \mathbf{R}_l, \mathbf{R}_l - \mathbf{R}_j) = \overset{0}{\chi}{}_{ijll}^{\mu\nu\gamma\gamma}(\mathbf{R}_i - \mathbf{R}_l, \mathbf{R}_l - \mathbf{R}_j) - \sum_{\substack{m\mathbf{R}_m \\ \lambda\alpha}} \overset{0}{\chi}{}_{ijmm}^{\mu\nu\lambda\lambda}(\mathbf{R}_i - \mathbf{R}_m, \mathbf{R}_m - \mathbf{R}_j) U_m^{\lambda\alpha} \chi_{ml}^{\alpha\gamma}(\mathbf{R}_l - \mathbf{R}_m) \ , \tag{I.37}$$

where $\chi_{ml}^{\alpha\gamma}(\mathbf{R}_l - \mathbf{R}_m)$ is the usual transverse magnetic susceptibility. Fourier transforming the position variables inside the planes, $\mathbf{R}$, and dropping the orbital indices for simplicity, we obtain

$$\begin{aligned} \frac{1}{N^2}\sum_{\mathbf{k}_\parallel, \mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i - \mathbf{R}_l)} e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l - \mathbf{R}_j)} \chi_{ijll}(\mathbf{k}_\parallel, \mathbf{k}'_\parallel) &= \frac{1}{N^2}\sum_{\mathbf{k}_\parallel, \mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i - \mathbf{R}_l)} e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l - \mathbf{R}_j)} \chi^0_{ijll}(\mathbf{k}_\parallel, \mathbf{k}'_\parallel) \\ &- \sum_{m\mathbf{R}_m} \frac{1}{N^2}\sum_{\mathbf{k}_\parallel, \mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i - \mathbf{R}_m)} e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_m - \mathbf{R}_j)} \chi^0_{ijmm}(\mathbf{k}_\parallel, \mathbf{k}'_\parallel) U_m \\ &\times \frac{1}{N}\sum_{\mathbf{q}_\parallel} e^{-i\mathbf{q}_\parallel\cdot(\mathbf{R}_l - \mathbf{R}_m)} \chi_{ml}(\mathbf{q}_\parallel) \ . \end{aligned} \tag{I.38}$$

This equation can be rewritten as

$$\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi^0_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)$$
$$- \sum_{m\mathbf{R}_m}\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U_m$$
$$\times\frac{1}{N}\sum_{\mathbf{q}_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_m)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_m-\mathbf{R}_l)}e^{-i\mathbf{q}_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_m)}\chi_{ml}(\mathbf{q}_\parallel)\ . \tag{I.39}$$

Now, using $\sum_{\mathbf{R}_m} e^{i(\mathbf{k}_\parallel-\mathbf{k}'_\parallel+\mathbf{q}_\parallel)\cdot\mathbf{R}_m} = N\delta(\mathbf{k}_\parallel-\mathbf{k}'_\parallel+\mathbf{q}_\parallel)$,

$$\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi^0_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)$$
$$- \sum_{m}\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U_m\chi_{ml}(\mathbf{k}'_\parallel-\mathbf{k}_\parallel)\ . \tag{I.40}$$

This way, we get

$$\chi_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \chi^0_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) - \sum_m \chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U_m\chi_{ml}(\mathbf{k}'_\parallel-\mathbf{k}_\parallel)\ . \tag{I.41}$$

Particularizing for $\chi_{ijll}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)$, which is the susceptibilities needed for the calculation of the perpendicular spin currents, and bringing back the orbital indices

$$\chi^{\mu\nu\gamma\gamma}_{ijll}(\mathbf{k}_\parallel,\mathbf{k}_\parallel) = \chi^{0\,\mu\nu\gamma\gamma}_{ijll}(\mathbf{k}_\parallel,\mathbf{k}_\parallel) - \sum_{\substack{m\\ \lambda\alpha}} \chi^{0\,\mu\nu\lambda\lambda}_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)U^{\lambda\alpha}_m\chi^{\alpha\gamma}_{ml}(0)\ . \tag{I.42}$$

To calculate the parallel spin currents, we need $\chi^{\mu\nu\gamma\gamma}_{iill}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel)$, which are given by

$$\chi^{\mu\nu\gamma\gamma}_{iill}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel) = \chi^{0\,\mu\nu\gamma\gamma}_{iill}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel) - \sum_{\substack{m\\ \lambda\alpha}} \chi^{0\,\mu\nu\lambda\lambda}_{iimm}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel)U^{\lambda\alpha}_m\chi^{\alpha\gamma}_{ml}(\mathbf{q}_\parallel)\ . \tag{I.43}$$

The index m covers the planes where the effective electronic interaction is present, and l comprises the planes that generates the spin current. Thus, both of them enumerate the magnetic planes.

$$
\begin{aligned}
&\sum_{\nu}\sum_{\mathbf{k}_\parallel}\left\{t_{ii}^{\mu\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)-t_{ii}^{\mu\nu}(\mathbf{k}_\parallel)\right\}\chi_{iill}^{\mu\nu\gamma\gamma}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel) \\
&=\sum_{\nu}\sum_{\mathbf{k}_\parallel}\left\{t_{ii}^{\mu\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)-t_{ii}^{\mu\nu}(\mathbf{k}_\parallel)\right\}\overset{0}{\chi}{}_{iill}^{\mu\nu\gamma\gamma}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel) \\
&-\sum_{\substack{m\\ \lambda\alpha}}\sum_{\nu}\sum_{\mathbf{k}_\parallel}\left\{t_{ii}^{\mu\nu}(\mathbf{k}_\parallel+\mathbf{q}_\parallel)-t_{ii}^{\mu\nu}(\mathbf{k}_\parallel)\right\}\overset{0}{\chi}{}_{iimm}^{\mu\nu\lambda\lambda}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{q}_\parallel)U_m^{\lambda\alpha}\chi_{ml}^{\alpha\gamma}(\mathbf{q}_\parallel)\ .
\end{aligned}
\tag{I.44}
$$

Fourier transforming the position variables inside the planes, $\mathbf{R}$, and dropping the orbital indices for simplicity, we obtain

$$\begin{aligned}
\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) &= \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi^0_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) \\
&- \sum_{m\mathbf{R}_m}\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_m)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_m-\mathbf{R}_j)}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U_m \\
&\times\frac{1}{N^2}\sum_{\mathbf{k}''_\parallel,\mathbf{k}'''_\parallel} e^{-i\mathbf{k}''_\parallel\cdot(\mathbf{R}_m-\mathbf{R}_l)}e^{-i\mathbf{k}'''_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_m)}\chi_{mmll}(\mathbf{k}''_\parallel,\mathbf{k}'''_\parallel)\ .
\end{aligned} \tag{I.45}$$

This equation can be rewritten as

$$\begin{aligned}
\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) &= \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi^0_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) \\
&- \frac{1}{N^4}\sum_{\substack{\mathbf{k}_\parallel,\mathbf{k}'_\parallel\\ \mathbf{k}''_\parallel,\mathbf{k}'''_\parallel}}\sum_{m\mathbf{R}_m} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U_m \\
&\times e^{i(\mathbf{k}_\parallel-\mathbf{k}''_\parallel)\cdot(\mathbf{R}_m-\mathbf{R}_l)}e^{i(\mathbf{k}'_\parallel-\mathbf{k}'''_\parallel)\cdot(\mathbf{R}_l-\mathbf{R}_m)}\chi_{mmll}(\mathbf{k}''_\parallel,\mathbf{k}'''_\parallel)\ .
\end{aligned} \tag{I.46}$$

Now, using $\sum_{\mathbf{R}_m} e^{i(\mathbf{k}_\parallel-\mathbf{k}''_\parallel-\mathbf{k}'_\parallel+\mathbf{k}'''_\parallel)\cdot\mathbf{R}_m} = N\delta(\mathbf{k}_\parallel-\mathbf{k}''_\parallel-\mathbf{k}'_\parallel+\mathbf{k}'''_\parallel)$,

$$\begin{aligned}
\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) &= \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi^0_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) \\
&- \frac{1}{N^3}\sum_{\substack{\mathbf{k}_\parallel,\mathbf{k}'_\parallel\\ \mathbf{k}''_\parallel}}\sum_{m} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_l-\mathbf{R}_j)}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U_m \\
&\times\chi_{mmll}(\mathbf{k}''_\parallel,\mathbf{k}''_\parallel+\mathbf{k}'_\parallel-\mathbf{k}_\parallel)\ .
\end{aligned} \tag{I.47}$$

This way, we get

$$\chi_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \chi^0_{ijll}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) - \frac{1}{N}\sum_{\mathbf{k}''_\parallel}\sum_m \chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U_m\chi_{mmll}(\mathbf{k}''_\parallel,\mathbf{k}''_\parallel+\mathbf{k}'_\parallel-\mathbf{k}_\parallel)\ . \tag{I.48}$$

Particularizing for $\chi_{ijll}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)$, bringing back the orbital indices and summing over the

γ orbitals,

$$\sum_{\gamma} \chi_{ijll}^{\mu\nu\gamma\gamma}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) = \sum_{\gamma} \chi_{ijll}^{0\,\mu\nu\gamma\gamma}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) - \frac{1}{N} \sum_{\mathbf{k}''_\parallel} \sum_{\substack{m \\ \eta\lambda\alpha\beta\gamma}} \chi_{ijmm}^{0\,\mu\nu\eta\lambda}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) U_m^{\eta\alpha\beta\lambda} \chi_{mmll}^{\alpha\beta\gamma\gamma}(\mathbf{k}''_\parallel, \mathbf{k}''_\parallel) \ . \tag{I.49}$$

Using Lowde-Windsor parametrization, i.e., $U_m^{\eta\alpha\beta\lambda} = U_m^{\lambda\alpha}\delta_{\eta\lambda}\delta_{\alpha\beta}$, where $U_m^{\lambda\alpha} = U_m$ for d orbitals and 0 for s e p orbitals. This means that, for a fixed plane m, the matrix $U_m^{\lambda\alpha}$ has the form

$$\begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & U_m & U_m & U_m & U_m & U_m \\ 0 & 0 & 0 & 0 & U_m & U_m & U_m & U_m & U_m \\ 0 & 0 & 0 & 0 & U_m & U_m & U_m & U_m & U_m \\ 0 & 0 & 0 & 0 & U_m & U_m & U_m & U_m & U_m \\ 0 & 0 & 0 & 0 & U_m & U_m & U_m & U_m & U_m \end{pmatrix} \tag{I.50}$$

So,

$$\sum_{\gamma} \chi_{ijll}^{\mu\nu\gamma\gamma}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) = \sum_{\gamma} \chi_{ijll}^{0\,\mu\nu\gamma\gamma}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) - \frac{1}{N} \sum_{\mathbf{k}''_\parallel} \sum_{\substack{m \\ \langle\lambda\rangle}} \chi_{ijmm}^{0\,\mu\nu\lambda\lambda}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) U_m \sum_{\langle\alpha\rangle\gamma} \chi_{mmll}^{\alpha\alpha\gamma\gamma}(\mathbf{k}''_\parallel, \mathbf{k}''_\parallel) \ , \tag{I.51}$$

where $\sum_{\langle\lambda\alpha\rangle}$ represents a sum over the indices λ and α restricted over orbitals d only. The left-hand side of this equation is the susceptibility we need to calculate the spin current of Eq. (**??**).

To obtain the susceptibility of the right-hand side, we particulare Eq. (I.51) for $i = j$ and $\mu = \nu$, and sum it over $\mathbf{k}_\parallel$ and over the d orbitals of the μ index, i.e.,

$$\begin{aligned} \sum_{\langle\mu\rangle\gamma} \frac{1}{N} \sum_{\mathbf{k}_\parallel} \chi_{il}^{\mu\gamma}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) = & \sum_{\langle\mu\rangle\gamma} \frac{1}{N} \sum_{\mathbf{k}_\parallel} \chi_{il}^{0\,\mu\gamma}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) \\ & - \sum_{\substack{m \\ \langle\mu\lambda\rangle}} \frac{1}{N} \sum_{\mathbf{k}_\parallel} \chi_{im}^{0\,\mu\lambda}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) U_m \sum_{\langle\alpha\rangle\gamma} \frac{1}{N} \sum_{\mathbf{k}''_\parallel} \chi_{ml}^{\alpha\gamma}(\mathbf{k}''_\parallel, \mathbf{k}''_\parallel) \ . \end{aligned} \tag{I.52}$$

If we write the susceptibilities as matrices with components given by the plane indices, we have

$$\begin{aligned} \sum_{\langle\mu\rangle\gamma} \frac{1}{N} \sum_{\mathbf{k}_\parallel} \chi^{\mu\gamma}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) = & \sum_{\langle\mu\rangle\gamma} \frac{1}{N} \sum_{\mathbf{k}_\parallel} \chi^{0\,\mu\gamma}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) \\ & - \sum_{\langle\mu\lambda\rangle} \frac{1}{N} \sum_{\mathbf{k}_\parallel} \chi^{0\,\mu\lambda}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) U \sum_{\langle\alpha\rangle\gamma} \frac{1}{N} \sum_{\mathbf{k}''_\parallel} \chi^{\alpha\gamma}(\mathbf{k}''_\parallel, \mathbf{k}''_\parallel) \ . \end{aligned} \tag{I.53}$$

Now we note that $N^{-1}\sum_{\mathbf{k}''_\parallel}\chi^0(\mathbf{k}''_\parallel,\mathbf{k}''_\parallel) = \chi^0(\mathbf{q}_\parallel = 0)$ is the Fourier transform of the usual Hartree-Fock susceptibility calculated at $\mathbf{q}_\parallel = 0$. For notation simplicity, we define $N^{-1}\sum_{\mathbf{k}_\parallel}\chi(\mathbf{k}_\parallel,\mathbf{k}_\parallel) = \chi$, and expliciting the RPA susceptibility in the equation above we end up with

$$\sum_{\langle\mu\rangle\gamma}\chi^{\mu\gamma} = \left[1 - \sum_{\langle\mu\lambda\rangle}{\chi^{0}}^{\mu\lambda}(0)U\right]^{-1}\sum_{\langle\mu\rangle\gamma}{\chi^{0}}^{\mu\gamma}(0)\ . \tag{I.54}$$

This is the susceptibility we need to calculate the right-hand side of Eq. (I.51). It is also the usual RPA susceptibility summed over the d orbitals in the first index and over all orbitals of the second index, calculated at $\mathbf{q} = 0$. We'll call it

$$\chi'_{ml}(0) = \sum_{\langle\mu\rangle\gamma}\chi^{\mu\gamma}_{ml}(0) = \left\{\left[1 - \sum_{\langle\mu\lambda\rangle}{\chi^{0}}^{\mu\lambda}(0)U\right]^{-1}\sum_{\langle\mu\rangle\gamma}{\chi^{0}}^{\mu\gamma}_{ml}(0)\right\}_{ml}\ . \tag{I.55}$$

Thus, Eq. (I.51) becomes

$$\sum_{\gamma}\chi^{\mu\nu\gamma\gamma}_{ijll}(\mathbf{k}_\parallel,\mathbf{k}_\parallel) = \sum_{\gamma}{\chi^{0}}^{\mu\nu\gamma\gamma}_{ijll}(\mathbf{k}_\parallel,\mathbf{k}_\parallel) - \sum_{\substack{m\\ \langle\lambda\rangle}}{\chi^{0}}^{\mu\nu\lambda\lambda}_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)U_m\chi'_{ml}(0)\ . \tag{I.56}$$

The spin current will be given by this term multiplied by $t^{\mu\nu}_{ij}(\mathbf{k}_\parallel)$, and summed over the orbitals and wave vectors,

$$\begin{aligned}\sum_{\mu\nu\gamma}\frac{1}{N}\sum_{\mathbf{k}_\parallel}t^{\mu\nu}_{ij}(\mathbf{k}_\parallel)\chi^{\mu\nu\gamma\gamma}_{ijll}(\mathbf{k}_\parallel,\mathbf{k}_\parallel) =& \sum_{\mu\nu\gamma}\frac{1}{N}\sum_{\mathbf{k}_\parallel}t^{\mu\nu}_{ij}(\mathbf{k}_\parallel){\chi^{0}}^{\mu\nu\gamma\gamma}_{ijll}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)\\ &- \sum_{\substack{m\\ \mu\nu\langle\lambda\rangle}}\frac{1}{N}\sum_{\mathbf{k}_\parallel}t^{\mu\nu}_{ij}(\mathbf{k}_\parallel){\chi^{0}}^{\mu\nu\lambda\lambda}_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)U_m\chi'_{ml}(0)\ .\end{aligned} \tag{I.57}$$

When we consider second-neighbors hoppings, the term we are calculating at the left-hand side of this equation can be written as a $3\times N_m$ matrix, where each line represents a bond between a site inside the volume V and another outside of V, and N_m is the number of magnetic layers.

Finally, for a system with more than one magnetic plane, we have a sum over l, and the expectation density of current is given by

$$\begin{aligned}\langle J_S\rangle(t) = \frac{\langle I_S\rangle(t)}{N} = \frac{ig\mu_B h_0}{2}e^{-i\omega t}\sum_{\substack{i\in V\\ j\notin V}}\sum_{\substack{l\\ \mu\nu\gamma}}\frac{1}{N}\sum_{\mathbf{k}_\parallel}\Big[&t^{\mu\nu}_{ij}(\mathbf{k}_\parallel)\chi^{\mu\nu\gamma\gamma}_{ijll}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)\\ &-t^{\mu\nu}_{ji}(\mathbf{k}_\parallel)\chi^{\mu\nu\gamma\gamma}_{jill}(\mathbf{k}_\parallel,\mathbf{k}_\parallel,\omega)\Big]\,\delta_{\mathbf{q}_\parallel,0}\ ,\end{aligned} \tag{I.58}$$

The total current density is then the sum over all the elements of the $3\times N_m$ matrix of Eq. (I.57) (for each of the two terms inside de square brackets of the equation above) also summed over the orbitals $\{\mu,\nu,\gamma\}$.

Fourier transforming Eq. (I.10) in the position variables inside the planes $\mathbf{R}$, we have

$$\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi^0_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)$$
$$-\sum_{m\mathbf{R}_m}\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_m)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_m-\mathbf{R}_j)}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U^m$$
$$\times\frac{1}{N^2}\sum_{\mathbf{k}''_\parallel,\mathbf{k}'''_\parallel} e^{-i\mathbf{k}''_\parallel\cdot(\mathbf{R}_m-\mathbf{R}_l)}e^{-i\mathbf{k}'''_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_m)}\chi_{mmkl}(\mathbf{k}''_\parallel,\mathbf{k}'''_\parallel)\ . \tag{I.59}$$

This equation can be rewritten as

$$\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi^0_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)$$
$$-\frac{1}{N^4}\sum_{\substack{\mathbf{k}_\parallel,\mathbf{k}'_\parallel\\ \mathbf{k}''_\parallel,\mathbf{k}'''_\parallel}}\sum_{m\mathbf{R}_m} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U^m$$
$$\times e^{i(\mathbf{k}_\parallel-\mathbf{k}''_\parallel)\cdot(\mathbf{R}_m-\mathbf{R}_l)}e^{i(\mathbf{k}'_\parallel-\mathbf{k}'''_\parallel)\cdot(\mathbf{R}_k-\mathbf{R}_m)}\chi_{mmkl}(\mathbf{k}''_\parallel,\mathbf{k}'''_\parallel)\ , \tag{I.60}$$

or, using that $\sum_{\mathbf{R}_m} e^{i(\mathbf{k}_\parallel-\mathbf{k}''_\parallel-\mathbf{k}'_\parallel+\mathbf{k}'''_\parallel)\cdot\mathbf{R}_m} = N\delta(\mathbf{k}_\parallel-\mathbf{k}''_\parallel-\mathbf{k}'_\parallel+\mathbf{k}'''_\parallel)$,

$$\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi^0_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)$$
$$-\frac{1}{N^3}\sum_{\substack{\mathbf{k}_\parallel,\mathbf{k}'_\parallel\\ \mathbf{k}''_\parallel}}\sum_{m} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U^m$$
$$\times e^{i(\mathbf{k}_\parallel-\mathbf{k}''_\parallel)\cdot(\mathbf{R}_k-\mathbf{R}_l)}\chi_{mmkl}(\mathbf{k}''_\parallel,\mathbf{k}''_\parallel+\mathbf{k}'_\parallel-\mathbf{k}_\parallel)\ . \tag{I.61}$$

Now, to eliminate the pre-factor, we do the transformations $\mathbf{R}=\mathbf{R}_i-\mathbf{R}_l$ and $\mathbf{R}'=\mathbf{R}_k-\mathbf{R}_j$

$$\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot\mathbf{R}}e^{-i\mathbf{k}'_\parallel\cdot\mathbf{R}'}\chi_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot\mathbf{R}}e^{-i\mathbf{k}'_\parallel\cdot\mathbf{R}'}\chi^0_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)$$
$$-\frac{1}{N^3}\sum_{\substack{\mathbf{k}_\parallel,\mathbf{k}'_\parallel\\ \mathbf{k}''_\parallel}}\sum_{m} e^{-i\mathbf{k}_\parallel\cdot\mathbf{R}}e^{-i\mathbf{k}'_\parallel\cdot\mathbf{R}'}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U^m \tag{I.62}$$
$$\times e^{i(\mathbf{k}_\parallel-\mathbf{k}''_\parallel)\cdot(\mathbf{R}'+\mathbf{R}_j-\mathbf{R}_i+\mathbf{R})}\chi_{mmkl}(\mathbf{k}''_\parallel,\mathbf{k}''_\parallel+\mathbf{k}'_\parallel-\mathbf{k}_\parallel)\ .$$

Multiplying by $e^{i\mathbf{k}_1\cdot\mathbf{R}}e^{i\mathbf{k}_2'\cdot\mathbf{R}'}$ and summing over $\mathbf{R}$ and $\mathbf{R}'$, the term on the left hand side and the first term of the right hand side will be of the form

$$\begin{aligned}\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}_\parallel'}\sum_{\mathbf{R},\mathbf{R}'}e^{i(\mathbf{k}_1-\mathbf{k}_\parallel)\cdot\mathbf{R}}e^{i(\mathbf{k}_2-\mathbf{k}_\parallel')\cdot\mathbf{R}'}\chi_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}_\parallel') =& \sum_{\mathbf{k}_\parallel,\mathbf{k}_\parallel'}\delta(\mathbf{k}_1-\mathbf{k}_\parallel)\delta(\mathbf{k}_2-\mathbf{k}_\parallel')\chi_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}_\parallel')\\ =&\chi_{ijkl}(\mathbf{k}_1,\mathbf{k}_2')\end{aligned} \tag{I.63}$$

The last term of the right hand side is

$$\begin{aligned}&-\frac{1}{N^3}\sum_{\substack{\mathbf{k}_\parallel,\mathbf{k}_\parallel'\\ \mathbf{k}_\parallel''}}\sum_m\sum_{\mathbf{R},\mathbf{R}'}e^{i(\mathbf{k}_1-\mathbf{k}_\parallel)\cdot\mathbf{R}}e^{i(\mathbf{k}_2-\mathbf{k}_\parallel')\cdot\mathbf{R}'}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}_\parallel')U^m\\ &\times e^{i(\mathbf{k}_\parallel-\mathbf{k}_\parallel'')\cdot(\mathbf{R}'+\mathbf{R}_j-\mathbf{R}_i+\mathbf{R})}\chi_{mmkl}(\mathbf{k}_\parallel'',\mathbf{k}_\parallel''+\mathbf{k}_\parallel'-\mathbf{k}_\parallel)\\ =&-\frac{1}{N}\sum_{\substack{\mathbf{k}_\parallel,\mathbf{k}_\parallel'\\ \mathbf{k}_\parallel''}}\sum_m\delta(\mathbf{k}_1-\mathbf{k}_\parallel'')\delta(\mathbf{k}_2+\mathbf{k}_\parallel-\mathbf{k}_\parallel'-\mathbf{k}_\parallel'')\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}_\parallel')U^m\\ &\times e^{i(\mathbf{k}_\parallel-\mathbf{k}_\parallel'')\cdot(\mathbf{R}_j-\mathbf{R}_i)}\chi_{mmkl}(\mathbf{k}_\parallel'',\mathbf{k}_\parallel''+\mathbf{k}_\parallel'-\mathbf{k}_\parallel)\\ =&-\frac{1}{N}\sum_{\mathbf{k}_\parallel,\mathbf{k}_\parallel'}\sum_m\delta(\mathbf{k}_2+\mathbf{k}_\parallel-\mathbf{k}_\parallel'-\mathbf{k}_1)\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}_\parallel')U^m\\ &\times e^{i(\mathbf{k}_\parallel-\mathbf{k}_1)\cdot(\mathbf{R}_j-\mathbf{R}_i)}\chi_{mmkl}(\mathbf{k}_1,\mathbf{k}_1+\mathbf{k}_\parallel'-\mathbf{k}_\parallel)\\ =&-\frac{1}{N}\sum_{\mathbf{k}_\parallel}\sum_m\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}_2+\mathbf{k}_\parallel-\mathbf{k}_1)U^me^{i(\mathbf{k}_\parallel-\mathbf{k}_1)\cdot(\mathbf{R}_j-\mathbf{R}_i)}\chi_{mmkl}(\mathbf{k}_1,\mathbf{k}_2)\ .\end{aligned} \tag{I.64}$$

Another way of getting a relation between the susceptibilities: from Eq. (I.61), we can assume fixed values of $\mathbf{R}_k$ and $\mathbf{R}_l$, and multiply by $e^{i\mathbf{k}_1\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{i\mathbf{k}_2\cdot(\mathbf{R}_k-\mathbf{R}_j)}$ and sum over $\mathbf{R}_i$ and $\mathbf{R}_j$. The left hand side and the first term on the right hand side will give us

$$\begin{aligned}\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}_\parallel'}\sum_{\mathbf{R}_i,\mathbf{R}_j}e^{i(\mathbf{k}_1-\mathbf{k}_\parallel)\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{i(\mathbf{k}_2-\mathbf{k}_\parallel')\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}_\parallel') =& \sum_{\mathbf{k}_\parallel,\mathbf{k}_\parallel'}\delta(\mathbf{k}_1-\mathbf{k}_\parallel)\delta(\mathbf{k}_2-\mathbf{k}_\parallel')\chi_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}_\parallel')\\ =&\chi_{ijkl}(\mathbf{k}_1,\mathbf{k}_2)\ .\end{aligned} \tag{I.65}$$

The second term on the right hand side will be

$$
\begin{aligned}
&-\frac{1}{N^3}\sum_{\substack{\mathbf{k}_\parallel,\mathbf{k}'_\parallel\\ \mathbf{k}''_\parallel}}\sum_m\sum_{\mathbf{R}_i,\mathbf{R}_j} e^{i(\mathbf{k}_1-\mathbf{k}_\parallel)\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{i(\mathbf{k}_2-\mathbf{k}'_\parallel)\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U^m\\
&\times e^{i(\mathbf{k}_\parallel-\mathbf{k}''_\parallel)\cdot(\mathbf{R}_k-\mathbf{R}_l)}\chi_{mmkl}(\mathbf{k}''_\parallel,\mathbf{k}''_\parallel+\mathbf{k}'_\parallel-\mathbf{k}_\parallel)\\
=&-\frac{1}{N}\sum_{\substack{\mathbf{k}_\parallel,\mathbf{k}'_\parallel\\ \mathbf{k}''_\parallel}}\sum_m \delta(\mathbf{k}_1-\mathbf{k}_\parallel)\delta(\mathbf{k}_2-\mathbf{k}'_\parallel)\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)U^m \qquad \text{(I.66)}\\
&\times e^{i(\mathbf{k}_\parallel-\mathbf{k}''_\parallel)\cdot(\mathbf{R}_k-\mathbf{R}_l)}\chi_{mmkl}(\mathbf{k}''_\parallel,\mathbf{k}''_\parallel+\mathbf{k}'_\parallel-\mathbf{k}_\parallel)\\
=&-\frac{1}{N}\sum_{\mathbf{k}''_\parallel}\sum_m \chi^0_{ijmm}(\mathbf{k}_1,\mathbf{k}_2)U^m e^{i(\mathbf{k}_1-\mathbf{k}''_\parallel)\cdot(\mathbf{R}_k-\mathbf{R}_l)}\chi_{mmkl}(\mathbf{k}''_\parallel,\mathbf{k}''_\parallel+\mathbf{k}_2-\mathbf{k}_1)\ .
\end{aligned}
$$

The relation that we get is

$$
\chi_{ijkl}(\mathbf{k}_1,\mathbf{k}_2) =\chi^0_{ijkl}(\mathbf{k}_1,\mathbf{k}_2) - \frac{1}{N}\sum_{\mathbf{k}''_\parallel}\sum_m \chi^0_{ijmm}(\mathbf{k}_1,\mathbf{k}_2)U^m e^{i(\mathbf{k}_1-\mathbf{k}''_\parallel)\cdot(\mathbf{R}_k-\mathbf{R}_l)}\chi_{mmkl}(\mathbf{k}''_\parallel,\mathbf{k}''_\parallel+\mathbf{k}_2-\mathbf{k}_1)\ . \tag{I.67}
$$

If we call $\mathbf{R} = \mathbf{R}_k - \mathbf{R}_l$ and sum over $\mathbf{R}$ and divide by N, we end up with

$$
\chi_{ijkl}(\mathbf{k}_1,\mathbf{k}_2) =\chi^0_{ijkl}(\mathbf{k}_1,\mathbf{k}_2) - \frac{1}{N}\sum_{\mathbf{k}''_\parallel}\sum_m \chi^0_{ijmm}(\mathbf{k}_1,\mathbf{k}_2)U^m \delta(\mathbf{k}_1-\mathbf{k}''_\parallel)\chi_{mmkl}(\mathbf{k}''_\parallel,\mathbf{k}''_\parallel+\mathbf{k}_2-\mathbf{k}_1)\ , \tag{I.68}
$$

or

$$
\chi_{ijkl}(\mathbf{k}_1,\mathbf{k}_2) =\chi^0_{ijkl}(\mathbf{k}_1,\mathbf{k}_2) - \frac{1}{N}\sum_m \chi^0_{ijmm}(\mathbf{k}_1,\mathbf{k}_2)U^m \chi_{mmkl}(\mathbf{k}_1,\mathbf{k}_2)\ , \tag{I.69}
$$

Changing the variables $\mathbf{q}_\parallel = \mathbf{k}'_\parallel - \mathbf{k}_\parallel + \mathbf{k}''_\parallel$ in the last sum, we obtain

$$\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi^0_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)$$
$$-\frac{1}{N^3}\sum_{\substack{\mathbf{k}_\parallel,\mathbf{q}_\parallel\\ \mathbf{k}''_\parallel}}\sum_m e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_l)}e^{-i\mathbf{q}_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_j)}\chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{q}_\parallel+\mathbf{k}_\parallel-\mathbf{k}''_\parallel)U^m\chi_{mmkl}(\mathbf{k}''_\parallel,\mathbf{q}_\parallel)\ . \tag{I.70}$$

Finally, changing $\mathbf{q}_\parallel \to \mathbf{k}'_\parallel$,

$$\chi_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \chi^0_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) - \frac{1}{N}\sum_{\mathbf{k}''_\parallel}\sum_m \chi^0_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{k}'_\parallel-\mathbf{k}''_\parallel)U^m\chi_{mmkl}(\mathbf{k}''_\parallel,\mathbf{k}'_\parallel)\ . \tag{I.71}$$

Adding back the orbital indices,

$$\chi^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \chi^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) - \frac{1}{N}\sum_{\mathbf{k}''_\parallel}\sum_{\substack{m\\ \eta\lambda\alpha\beta}} \chi^{0\,\mu\nu\eta\lambda}_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{k}'_\parallel-\mathbf{k}''_\parallel)U^m_{\eta\alpha\beta\lambda}\chi^{\alpha\beta\gamma\xi}_{mmkl}(\mathbf{k}''_\parallel,\mathbf{k}'_\parallel)\ . \tag{I.72}$$

Using Lowde and Windsor parametrization, $U^m_{\eta\alpha\beta\lambda} = U^m\delta_{\eta\lambda}\delta_{\alpha\beta}$, we end up with

$$\chi^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \chi^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) - \frac{1}{N}\sum_{\mathbf{k}''_\parallel}\sum_{\substack{m\\ \lambda\alpha}} \chi^{0\,\mu\nu\lambda\lambda}_{ijmm}(\mathbf{k}_\parallel,\mathbf{k}_\parallel+\mathbf{k}'_\parallel-\mathbf{k}''_\parallel)U^m\chi^{\alpha\alpha\gamma\xi}_{mmkl}(\mathbf{k}''_\parallel,\mathbf{k}'_\parallel)\ . \tag{I.73}$$

Another way to obtain the relation showed in Eq. (I.73) is the following. The general relation between these two susceptibilities with four indices is

$$\begin{aligned}&\widetilde{\chi}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,\omega) = \widetilde{\chi}^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4,\omega)\\ &-\frac{1}{N^3}\sum_{\substack{m\\ \eta\lambda\alpha\beta}}\sum_{\substack{\mathbf{k}'_1\mathbf{k}'_2\\ \mathbf{q}_1\mathbf{q}_2}} \widetilde{\chi}^{0\,\mu\nu\eta\lambda}_{ijmm}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}'_1,\mathbf{k}'_2,\omega)U^m_{\eta\alpha\beta\lambda}\delta_{\mathbf{q}_1+\mathbf{k}'_1,-\mathbf{q}_2-\mathbf{k}'_2}\widetilde{\chi}^{\alpha\beta\gamma\xi}_{mmkl}(\mathbf{q}_1,\mathbf{q}_2,\mathbf{k}_3,\mathbf{k}_4,\omega)\ .\end{aligned} \tag{I.74}$$

Here, $\{i,j,k,l,m\}$ also represents planes indices. Substituting Eq. (I.2) in Eq. (I.74) for both susceptibilities, we obtain

$$\begin{aligned}&\widetilde{\chi}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega)\delta_{-\mathbf{k}_3\mathbf{k}_2}\delta_{-\mathbf{k}_1\mathbf{k}_4} = \widetilde{\chi}^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega)\delta_{-\mathbf{k}_3\mathbf{k}_2}\delta_{-\mathbf{k}_1\mathbf{k}_4}\\ &-\frac{1}{N}\sum_{\substack{m\\ \eta\lambda\alpha\beta}}\sum_{\mathbf{q}_1} \widetilde{\chi}^{0\,\mu\nu\eta\lambda}_{ijmm}(\mathbf{k}_1,-\mathbf{k}_2,\omega)U^m_{\eta\alpha\beta\lambda}\widetilde{\chi}^{\alpha\beta\gamma\xi}_{mmkl}(\mathbf{q}_1,\mathbf{k}_3,\omega)\delta_{-\mathbf{k}_3\mathbf{k}_2+\mathbf{k}_1-\mathbf{q}_1}\delta_{-\mathbf{q}_1\mathbf{k}_4}\ .\end{aligned} \tag{I.75}$$

Summing both sides in $\mathbf{k}_2$ and $\mathbf{k}_4$,

$$\begin{aligned}&\widetilde{\chi}^{\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega) = \widetilde{\chi}^{0\,\mu\nu\gamma\xi}_{ijkl}(\mathbf{k}_1,\mathbf{k}_3,\omega)\\ &- \frac{1}{N}\sum_{\substack{m\\ \eta\lambda\alpha\beta}}\sum_{\mathbf{q}_1}\widetilde{\chi}^{0\,\mu\nu\eta\lambda}_{ijmm}(\mathbf{k}_1,\mathbf{k}_1+\mathbf{k}_3-\mathbf{q}_1,\omega)U^m_{\eta\alpha\beta\lambda}\widetilde{\chi}^{\alpha\beta\gamma\xi}_{mmkl}(\mathbf{q}_1,\mathbf{k}_3,\omega)\ .\end{aligned} \tag{I.76}$$

This relation is equivalent to the one obtained in Eq. (I.72).

Particularizing Eq. (**??**) to the susceptibility required for the spin current calculation,

$$\widetilde{\chi}^{\mu\nu\gamma\gamma}_{ijkk}(\mathbf{k},\mathbf{k}) = \widetilde{\chi}^{0\,\mu\nu\gamma\gamma}_{ijkk}(\mathbf{k},\mathbf{k}) - \frac{1}{N}\sum_{\substack{m\\ \eta\alpha}}\sum_{\mathbf{q}}\widetilde{\chi}^{0\,\mu\nu\eta\eta}_{ijmm}(\mathbf{k},2\mathbf{k}-\mathbf{q})U^m\widetilde{\chi}^{\alpha\alpha\gamma\gamma}_{mmkk}(\mathbf{q},\mathbf{k})\ . \tag{I.77}$$

Changing the variables $\mathbf{q}' = \mathbf{k}-\mathbf{q}$, we have

$$\widetilde{\chi}^{\mu\nu\gamma\gamma}_{ijkk}(\mathbf{k},\mathbf{k}) = \widetilde{\chi}^{0\,\mu\nu\gamma\gamma}_{ijkk}(\mathbf{k},\mathbf{k}) - \frac{1}{N}\sum_{\substack{m\\ \eta\alpha}}\sum_{\mathbf{q}}\widetilde{\chi}^{0\,\mu\nu\eta\eta}_{ijmm}(\mathbf{k},\mathbf{k}+\mathbf{q})U^m\widetilde{\chi}^{\alpha\alpha\gamma\gamma}_{mmkk}(\mathbf{k}-\mathbf{q},\mathbf{k})\ . \tag{I.78}$$

I.2 Eletric field perturbation: $\chi_{iik\ell}(\mathbf{k}_\parallel, \omega)$

If we apply an electric field, we need susceptibilities of the kind $\chi^{\mu\nu\gamma\xi}_{iik\ell}$ to calculate in-plane currents and spin disturbances. In this case, Eq. (I.10) becomes

$$\begin{aligned}\chi^{\mu\nu\gamma\xi}_{iik\ell}(\mathbf{R}_i-\mathbf{R}_\ell,\mathbf{R}_k-\mathbf{R}_i) &= \overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{iik\ell}(\mathbf{R}_i-\mathbf{R}_\ell,\mathbf{R}_k-\mathbf{R}_i)\\ &- \sum_{\substack{m\mathbf{R}_m\\ \eta\lambda\alpha\beta}} \overset{0}{\chi}{}^{\mu\nu\eta\lambda}_{iimm}(\mathbf{R}_i-\mathbf{R}_m,\mathbf{R}_m-\mathbf{R}_i)U^{\eta\alpha\beta\lambda}_m\chi^{\alpha\beta\gamma\xi}_{mmk\ell}(\mathbf{R}_m-\mathbf{R}_\ell,\mathbf{R}_k-\mathbf{R}_m)\ ,\end{aligned} \tag{I.79}$$

We can also use the relation obtained in Eq. (F.49),

$$\begin{aligned}\chi^{\mu\nu\gamma\xi}_{iik\ell}(\mathbf{R}_i-\mathbf{R}_\ell,\mathbf{R}_k-\mathbf{R}_i) &= \overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{iik\ell}(\mathbf{R}_i-\mathbf{R}_\ell,\mathbf{R}_k-\mathbf{R}_i)\\ &- \sum_{\substack{m\mathbf{R}_m\\ \eta\lambda\alpha\beta}} \chi^{\mu\nu\eta\lambda}_{iimm}(\mathbf{R}_i-\mathbf{R}_m)U^{\eta\alpha\beta\lambda}_m\overset{0}{\chi}{}^{\alpha\beta\gamma\xi}_{mmk\ell}(\mathbf{R}_m-\mathbf{R}_\ell,\mathbf{R}_k-\mathbf{R}_m)\ .\end{aligned} \tag{I.80}$$

Fourier transforming the in-plane positions, we obtain

$$\begin{aligned}\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_\ell)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_i)}\chi_{iik\ell}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) &= \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_\ell)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_i)}\overset{0}{\chi}_{iik\ell}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)\\ - \sum_{m\mathbf{R}_m}\frac{1}{N}\sum_{\mathbf{q}_\parallel} e^{-i\mathbf{q}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_m)}\chi_{iimm}(\mathbf{q})U_m&\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_m-\mathbf{R}_\ell)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_m)}\overset{0}{\chi}_{mmk\ell}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)\ ,\end{aligned} \tag{I.81}$$

where we have dropped the orbital indices for simplicity. Now we can rewrite the equation above changing only the second term on the RHS,

$$\begin{aligned}\frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_\ell)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_i)}\chi_{iik\ell}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) &= \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_\ell)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_i)}\overset{0}{\chi}_{iik\ell}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)\\ &- \frac{1}{N^2}\sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_\ell)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_k-\mathbf{R}_i)}\\ &\times \sum_{m\mathbf{R}_m}\frac{1}{N}\sum_{\mathbf{q}_\parallel} e^{-i\mathbf{q}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_m)}e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_m-\mathbf{R}_i)}e^{-i\mathbf{k}'_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_m)}\chi_{iimm}(\mathbf{q})U_m\overset{0}{\chi}_{mmk\ell}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)\ .\end{aligned} \tag{I.82}$$

Identifying $\sum_{\mathbf{R}_m} e^{i(\mathbf{k}'_\parallel-\mathbf{k}_\parallel+\mathbf{q}_\parallel)\cdot\mathbf{R}_m} = N\delta(\mathbf{k}'_\parallel-\mathbf{k}_\parallel+\mathbf{q}_\parallel)$, we obtain the $\mathbf{k}_\parallel, \mathbf{k}'_\parallel$ components of the equation as

$$\chi_{iik\ell}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) = \overset{0}{\chi}_{iik\ell}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel) - \sum_m \chi_{iimm}(\mathbf{k}_\parallel-\mathbf{k}'_\parallel)U_m\overset{0}{\chi}_{mmk\ell}(\mathbf{k}_\parallel,\mathbf{k}'_\parallel)\ . \tag{I.83}$$

Returning with orbital indices and particularizing for $\mathbf{k}_\parallel = \mathbf{k}'_\parallel$,

$$\chi^{\mu\nu\gamma\xi}_{iik\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel) = \overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{iik\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel) - \sum_{\substack{m\\ \eta\lambda\alpha\beta}} \chi^{\mu\nu\eta\lambda}_{im}(0) U^{\eta\alpha\beta\lambda}_m \overset{0}{\chi}{}^{\alpha\beta\gamma\xi}_{mmk\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel) \ . \tag{I.84}$$

Here, $\chi^{\mu\nu\eta\lambda}_{im}(\mathbf{q}_\parallel = 0)$ is closely related to the usual susceptibility - the only difference is that we have kept the four orbital indices.

If we had started with Eq. (I.79) instead, it's easy to see that we'd obtain the relation

$$\chi^{\mu\nu\gamma\xi}_{iik\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel) = \overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{iik\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel) - \sum_{\substack{m\\ \eta\lambda\alpha\beta}} \overset{0}{\chi}{}^{\mu\nu\eta\lambda}_{im}(0) U^{\eta\alpha\beta\lambda}_m \chi^{\alpha\beta\gamma\xi}_{mmk\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel) \ . \tag{I.85}$$

In matrix form, this equation becomes

$$\begin{aligned} \chi(\mathbf{k}_\parallel,\mathbf{k}_\parallel) &= \chi^0(\mathbf{k}_\parallel,\mathbf{k}_\parallel) - \chi^0(0)U\chi(\mathbf{k}_\parallel,\mathbf{k}_\parallel) \\ &= \left[1+\chi^0(0)U\right]^{-1}\chi^0(\mathbf{k}_\parallel,\mathbf{k}_\parallel) \ . \end{aligned} \tag{I.86}$$

Writing in terms of components once again,

$$\chi^{\mu\nu\gamma\xi}_{iik\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel) = \sum_{\substack{m\\ \alpha\beta}} \left[1+\chi^0(0)U\right]^{-1\,\mu\nu\alpha\beta}_{iimm} \overset{0}{\chi}{}^{\alpha\beta\gamma\xi}_{mmk\ell}(\mathbf{k}_\parallel,\mathbf{k}_\parallel) \ . \tag{I.87}$$

I.2.1 Disturbances

To calculate the spin/charge disturbances, we need to calculate terms of the form written in Eq. (5.322). We can multiply the Eq. (I.84) by $\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E$ and sum over $\mathbf{k}_\parallel$ to obtain

$$\begin{aligned} \sum_{\mathbf{k}_\parallel} \chi^{\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E =& \sum_{\mathbf{k}_\parallel} \overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E \\ &- \sum_{\substack{m\\ \eta\lambda\alpha\beta}} \chi^{\mu\nu\eta\lambda}_{\ell_1 m}(0) U^{\eta\alpha\beta\lambda}_m \sum_{\mathbf{k}_\parallel} \overset{0}{\chi}{}^{\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E \ . \end{aligned} \tag{I.88}$$

Computationally, we calculate the multi-orbital susceptibilities $\chi^{\mu\nu\eta\lambda}_{\ell_1 m}(0)$ as well as $\sum_{\mathbf{k}_\parallel} \overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E$ - the latter is a $4.N_{pl}.9.9 \times 4.N_{pl}.N_{pl}.9.9$ matrix with indices $\ell_1,\ell,\ell',\mu,\nu,\gamma,\xi$ (and also σ,σ' due to SOC). Calling these matrices χ and χ^0_t, we can write the expression above in matrix form as

$$\begin{aligned} \chi_t =& \chi^0_t - \chi U \chi^0_t \\ =& \left(1-\chi U\right)\chi^0_t \ , \end{aligned} \tag{I.89}$$

where the matrix U is given by Eq. (3.183). Note that χ is the RPA susceptibility, and already involves an inversion of the relation obtained in the last section, i.e.,

$$\begin{aligned}\chi =& \chi^0 - \chi^0 U \chi = \chi^0 - \chi U \chi^0 \\ &= \left(1 + \chi^0 U\right)^{-1} \chi^0 = \chi^0 \left(1 + U\chi^0\right)^{-1} \ .\end{aligned} \tag{I.90}$$

Therefore, substituting back in Eq. (I.89) we have

$$\begin{aligned}\chi_t &= \left[1 - \left(1 + \chi^0 U\right)^{-1} \chi^0 U\right] \chi_t^0 \\ &= \left[\left(1 + \chi^0 U\right)^{-1} \left(1 + \chi^0 U\right) - \left(1 + \chi^0 U\right)^{-1} \chi^0 U\right] \chi_t^0 \\ &= \left(1 + \chi^0 U\right)^{-1} \left[1 + \chi^0 U - \chi^0 U\right] \chi_t^0 \\ &= \left(1 + \chi^0 U\right)^{-1} \chi_t^0 \ .\end{aligned} \tag{I.91}$$

This result can also be obtained following the same steps above, but starting from Eq. (I.85), i.e.,

$$\begin{aligned}\chi_t =& \chi_t^0 - \chi^0 U \chi_t \\ &= \left(1 + \chi^0 U\right)^{-1} \chi_t^0 \ .\end{aligned} \tag{I.92}$$

I.2.2 Currents

Spin and charge currents are obtained using the terms similar to the one written in Eq. (5.283). Besides multiplying from the right by $\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E := B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)$ as before, we need to multiply by $\left[t^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)} - t^{\nu\mu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)}\right] :=$ $A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{k}_\parallel)$ from the left. Therefore, from Eq. (I.84),

$$\begin{aligned}&\sum_{\mathbf{k}_\parallel} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{k}_\parallel) \chi^{\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) = \sum_{\mathbf{k}_\parallel} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{k}_\parallel) \overset{0}{\chi}{}^{\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \\ &- \sum_{\substack{m \\ \eta\lambda\alpha\beta}} \sum_{\mathbf{k}_\parallel} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \mathbf{k}_\parallel) \chi^{\mu\nu\eta\lambda}_{\ell_1 m}(0) U^{\eta\alpha\beta\lambda}_m \overset{0}{\chi}{}^{\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\parallel, \mathbf{k}_\parallel) B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) \ .\end{aligned} \tag{I.93}$$

Putting A and $\chi^0 B$ in evidence on the right hand side,

$$
\begin{aligned}
&\sum_{\mathbf{k}_\parallel} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{k}_\parallel)\chi^{\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) = \sum_{\mathbf{k}_\parallel}\sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{k}_\parallel)\delta_{\alpha\mu}\delta_{\beta\nu}\delta_{m\ell_1}{\chi^0}^{\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}
\\
&-\sum_{\substack{m\\ \eta\lambda\alpha\beta}}\sum_{\mathbf{k}_\parallel} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{k}_\parallel)\chi^{\mu\nu\eta\lambda}_{\ell_1 m}(0)U^{\eta\alpha\beta\lambda}_{m}{\chi^0}^{\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)
\\
&=\sum_{\mathbf{k}_\parallel}\sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{k}_\parallel)\left\{\delta_{\alpha\mu}\delta_{\beta\nu}\delta_{m\ell_1}-\sum_{\eta\lambda}\chi^{\mu\nu\eta\lambda}_{\ell_1 m}(0)U^{\eta\alpha\beta\lambda}_{m}\right\}{\chi^0}^{\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)
\\
&=\sum_{\mathbf{k}_\parallel}\sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{k}_\parallel)\left\{1-\chi(0)U\right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1 mm}{\chi^0}^{\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)
\\
&=\sum_{\mathbf{k}_\parallel}\sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{k}_\parallel)\left\{\left[1+\chi^0(0)U\right]^{-1}\right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1 mm}{\chi^0}^{\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\ .
\end{aligned}
\tag{I.94}
$$

Considering that we only need the distinction in indices $\mathbf{R}'_\parallel-\mathbf{R}_\parallel$, ℓ and σ, we can sum all the other indices to obtain

$$
\begin{aligned}
&\sum_{\substack{\mu\nu\gamma\xi\\ \ell\ell'}}\sum_{\mathbf{k}_\parallel} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{k}_\parallel)\chi^{\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)
\\
&=\sum_{\mu\nu}\sum_{\mathbf{k}_\parallel}\sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{k}_\parallel)\left\{\left[1+\chi^0(0)U\right]^{-1}\right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1 mm}\sum_{\substack{\gamma\xi\\ \ell\ell'}}{\chi^0}^{\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\ .
\end{aligned}
\tag{I.95}
$$

If, instead, we use Eq. (I.85) to relate the HF and RPA susceptibilities, we obtain

$$
\begin{aligned}
&\sum_{\mathbf{k}_\parallel} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{k}_\parallel)\chi^{\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel) = \sum_{\mathbf{k}_\parallel} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{k}_\parallel){\chi^0}^{\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)
\\
&-\sum_{\substack{m\\ \eta\lambda\alpha\beta}}\sum_{\mathbf{k}_\parallel} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel,\mathbf{k}_\parallel){\chi^0}^{\mu\nu\eta\lambda}_{\ell_1 m}(0)U^{\eta\alpha\beta\lambda}_{m}\chi^{\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\parallel,\mathbf{k}_\parallel)B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\ .
\end{aligned}
\tag{I.96}
$$

In this case, it's not as simple (and probably it may give the same result, if possible) as before, since we cannot transform the χ^0 of the first term on the RHS to be equal as the second term on the RHS (because of the k, ℓ indices).

I.3 From the HF hamiltonian

We can write the hamiltonian obtained in Sec. 1.3.1 as

$$\hat{H} = \sum_{\substack{ij\\ \mu\nu}} \sum_{\alpha} c^{\dagger}_{i\mu\alpha} H^{0\,\mu\nu}_{ij} c_{j\nu\alpha} - \sum_{\substack{i\\ \mu\nu=d}} \sum_{\alpha\beta} c^{\dagger}_{i\mu\alpha} \delta_{\mu\nu} \frac{U^i}{2} \boldsymbol{\sigma}_{\alpha\beta} \cdot \mathbf{m}_i c_{i\nu\beta} - \sum_{\substack{i\\ \mu\nu=d}} \sum_{\alpha} c^{\dagger}_{i\mu\alpha} \delta_{\mu\nu} \frac{U^i}{2} n_i c_{i\nu\alpha} \ . \tag{I.97}$$

where $H^{0\,\mu\nu}_{ij}$ includes the tight-binding contribution, external magnetic fields and the spin-orbit interaction. In linear response, we consider that $n_i(t) = n_i^0 + \delta n_i(t)$ and $\mathbf{m}_i(t) = \mathbf{m}_i^0 + \delta\mathbf{m}_i(t)$ such that the first order change in the hamiltonian is given by

$$\delta\hat{H}[n_i, \vec{m}_i] = \hat{H}_{\text{ext}} + \delta n_i(t) \left.\frac{\delta\hat{H}}{\delta n_i(t)}\right|_{n^0,\mathbf{m}^0} + \delta\mathbf{m}_i(t) \cdot \left.\frac{\delta\hat{H}}{\delta\mathbf{m}_i(t)}\right|_{n^0,\mathbf{m}^0} \ . \tag{I.98}$$

The derivatives are given by

$$\begin{aligned} \left.\frac{\delta\hat{H}}{\delta n_i(t)}\right|_{n^0,\mathbf{m}^0} &= -\sum_{\substack{i\\ \mu\nu=d}} \sum_{\alpha\beta} c^{\dagger}_{i\mu\alpha} \delta_{\mu\nu} \frac{U^i}{2} c_{i\nu\beta} \\ \left.\frac{\delta\hat{H}}{\delta\mathbf{m}_i(t)}\right|_{n^0,\mathbf{m}^0} &= -\sum_{\substack{i\\ \mu\nu=d}} \sum_{\alpha\beta} c^{\dagger}_{i\mu\alpha} \delta_{\mu\nu} \frac{U^i}{2} \boldsymbol{\sigma}_{\alpha\beta} c_{i\nu\beta} \ . \end{aligned} \tag{I.99}$$

When an external field is applied to the system, we have

$$\delta\mathbf{m} = \chi \delta\mathbf{B}_{\text{ext}} \tag{I.100}$$

On the other hand, in a single particle point of view, we have

$$\delta\mathbf{m} = \chi_0 \delta\mathbf{B}_{\text{eff}}, \tag{I.101}$$

where $\mathbf{B}_{\text{eff}} = \mathbf{B}_{\text{ext}} - \frac{U^i}{2}\mathbf{m}_i - \frac{U^i}{2} n_i$.

Appendix J

Linear SOC

Using the SOC in linear order, we can write the monoelectronic Green function as

$$G_{\ell\ell'}^{\sigma'\sigma\,\mu\nu}(\mathbf{k},E) \simeq \left[G_{\ell\ell'}^{\sigma'\sigma\,\mu\nu}(\mathbf{k},E)\right]_{\lambda=0} + \lambda\left[\frac{\partial}{\partial\lambda}G_{\ell\ell'}^{\sigma'\sigma\,\mu\nu}(\mathbf{k},E)\right]_{\lambda=0} \tag{J.1}$$

Since $G = (E-H)^{-1} = (E-H_0-H_{\rm so})^{-1} = (E-H_0-\lambda\mathbf{L}\cdot\mathbf{S})^{-1}$, we have (omitting all indices for simplicity)

$$\begin{aligned} G \simeq & G^0 - \lambda\left[(E-H_0-\lambda\mathbf{L}\cdot\mathbf{S})^{-1}(-\mathbf{L}\cdot\mathbf{S})(E-H_0-\lambda\mathbf{L}\cdot\mathbf{S})^{-1}\right]_{\lambda=0} \\ = & G^0 + G^0 H_{\rm so} G^0 \end{aligned} \tag{J.2}$$

where $G^0 = G(\lambda = 0)$. This is just the first term of the Dyson Equation's expansion. We can further develop this equation to obtain

$$\begin{aligned} G \simeq & G^0(1 + H_{\rm so}G^0) \\ = & G^0[1 + H_{\rm so}(E-H_0)^{-1}] \\ = & G^0(E - H_0 + H_{\rm so})(E-H_0)^{-1} \\ = & G^0(E - H_0 + H_{\rm so})G^0 \end{aligned} \tag{J.3}$$

The number of particles or the magnetization can be calculated as

$$\begin{aligned} \rho^\alpha = & -\frac{1}{\pi}\Im\int dE\,\mathrm{Tr}\,\sigma^\alpha G \\ = & -\frac{1}{\pi}\Im\int dE\,\mathrm{Tr}\,\sigma^\alpha\left[G^0 + G^0 H_{\rm so} G^0\right] \\ = & \rho_0^\alpha - \frac{1}{\pi}\Im\int dE\,\mathrm{Tr}\,\sigma^\alpha G^0 H_{\rm so} G^0 \end{aligned} \tag{J.4}$$

The susceptibilities involve a products of Green functions. To calculate it in the linear SOC approximation, we sum Eq. (I.87) over $\mathbf{k}_\parallel$ and expand to first order in the

SOC parameter λ to obtain

$$\begin{aligned}
&\left[\chi^{\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell}(\mathbf{q}_\parallel=0)+\lambda\frac{\partial}{\partial\lambda}\chi^{\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell}(\mathbf{q}_\parallel=0)\right]_{\lambda=0}=\sum_{\substack{m\\ \alpha\beta}}\left\{\left[1+\chi^0(0)U\right]^{-1}\right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1mm}{}^0\chi^{\alpha\beta\gamma\xi}_{mm\ell\ell}(0)\\
&+\sum_{\substack{m\\ \alpha\beta}}\left\{\lambda\frac{\partial}{\partial\lambda}\left[1+\chi^0(0)U\right]^{-1}\right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1mm}{}^0\chi^{\alpha\beta\gamma\xi}_{mm\ell\ell}(0)+\sum_{\substack{m\\ \alpha\beta}}\left\{\left[1+\chi^0(0)U\right]^{-1}\right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1mm}\lambda\frac{\partial}{\partial\lambda}{}^0\chi^{\alpha\beta\gamma\xi}_{mm\ell\ell}(0)\\
&=\sum_{\substack{m\\ \alpha\beta}}\left\{\left[1+\chi^0(0)U\right]^{-1}\right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1mm}{}^0\chi^{\alpha\beta\gamma\xi}_{mm\ell\ell}(0)+\sum_{\substack{m\\ \alpha\beta}}\left\{\left[1+\chi^0(0)U\right]^{-1}\right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1mm}\lambda\frac{\partial}{\partial\lambda}{}^0\chi^{\alpha\beta\gamma\xi}_{mm\ell\ell}(0)\\
&+\sum_{\substack{m\\ \alpha\beta}}\left\{-\left[1+\chi^0(0)U\right]^{-1}\lambda\frac{\partial}{\partial\lambda}\left[\chi^0(0)\right]U\left[1+\chi^0(0)U\right]^{-1}\right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1mm}{}^0\chi^{\alpha\beta\gamma\xi}_{mm\ell\ell}(0)\\
&=\sum_{\substack{m\\ \alpha\beta}}\left\{\left[1+\chi^0(0)U\right]^{-1}\left[1+\left(\chi^0(0)-\lambda\frac{\partial}{\partial\lambda}\left[\chi^0(0)\right]\right)U\right]\left[1+\chi^0(0)U\right]^{-1}\right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1mm}{}^0\chi^{\alpha\beta\gamma\xi}_{mm\ell\ell}(0)\\
&+\sum_{\substack{m\\ \alpha\beta}}\left\{\left[1+\chi^0(0)U\right]^{-1}\right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1mm}\lambda\frac{\partial}{\partial\lambda}{}^0\chi^{\alpha\beta\gamma\xi}_{mm\ell\ell}(0)\ .
\end{aligned}\tag{J.5}$$

In the current calculation, we can start from Eq. (I.95) and do the same expansion in the susceptibility

$$
\begin{aligned}
&\sum_{\substack{\mu\nu\gamma\xi\\ \ell\ell'}} \sum_{\mathbf{k}_\|} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\| - \mathbf{R}_\|, \mathbf{k}_\|) \left[\chi^{\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\|, \mathbf{k}_\|) + \lambda \, \frac{\partial}{\partial\lambda} \chi^{\mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\|, \mathbf{k}_\|) \bigg|_{\lambda=0} \right] B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\|) \\
&= \sum_{\mu\nu} \sum_{\mathbf{k}_\|} \sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\| - \mathbf{R}_\|, \mathbf{k}_\|) \left\{ \left[1 + \chi^0(0)U\right]^{-1} \right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1 mm} \sum_{\substack{\gamma\xi\\ \ell\ell'}} \chi^{0\,\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\|, \mathbf{k}_\|) B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\|) \\
&+ \sum_{\mu\nu} \sum_{\mathbf{k}_\|} \sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\| - \mathbf{R}_\|, \mathbf{k}_\|) \left\{ \lambda \frac{\partial}{\partial\lambda} \left[1 + \chi^0(0)U\right]^{-1} \right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1 mm} \sum_{\substack{\gamma\xi\\ \ell\ell'}} \chi^{0\,\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\|, \mathbf{k}_\|) B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\|) \\
&+ \sum_{\mu\nu} \sum_{\mathbf{k}_\|} \sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\| - \mathbf{R}_\|, \mathbf{k}_\|) \left\{ \left[1 + \chi^0(0)U\right]^{-1} \right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1 mm} \sum_{\substack{\gamma\xi\\ \ell\ell'}} \lambda \frac{\partial}{\partial\lambda} \chi^{0\,\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\|, \mathbf{k}_\|) B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\|) \\
&= \sum_{\mu\nu} \sum_{\mathbf{k}_\|} \sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\| - \mathbf{R}_\|, \mathbf{k}_\|) \left\{ \left[1 + \chi^0(0)U\right]^{-1} \right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1 mm} \sum_{\substack{\gamma\xi\\ \ell\ell'}} \chi^{0\,\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\|, \mathbf{k}_\|) B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\|) \\
&+ \sum_{\mu\nu} \sum_{\mathbf{k}_\|} \sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\| - \mathbf{R}_\|, \mathbf{k}_\|) \left\{ - \left[1 + \chi^0(0)U\right]^{-1} \lambda \frac{\partial}{\partial\lambda} \left[\chi^0(0)\right] U \left[1 + \chi^0(0)U\right]^{-1} \right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1 mm} \sum_{\substack{\gamma\xi\\ \ell\ell'}} \chi^{0\,\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\|, \\
&+ \sum_{\mu\nu} \sum_{\mathbf{k}_\|} \sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\| - \mathbf{R}_\|, \mathbf{k}_\|) \left\{ \left[1 + \chi^0(0)U\right]^{-1} \right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1 mm} \sum_{\substack{\gamma\xi\\ \ell\ell'}} \lambda \frac{\partial}{\partial\lambda} \chi^{0\,\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\|, \mathbf{k}_\|) B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\|) \\
&= \sum_{\mu\nu} \sum_{\mathbf{k}_\|} \sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\| - \mathbf{R}_\|, \mathbf{k}_\|) \left\{ \left[1 + \chi^0(0)U\right]^{-1} \left[1 + \left(\chi^0(0) - \lambda \frac{\partial}{\partial\lambda} \left[\chi^0(0)\right]\right) U\right] \left[1 + \chi^0(0)U\right]^{-1} \right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1 mm} \\
&+ \sum_{\mu\nu} \sum_{\mathbf{k}_\|} \sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\| - \mathbf{R}_\|, \mathbf{k}_\|) \left\{ \left[1 + \chi^0(0)U\right]^{-1} \right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1 mm} \sum_{\substack{\gamma\xi\\ \ell\ell'}} \lambda \frac{\partial}{\partial\lambda} \chi^{0\,\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\|, \mathbf{k}_\|) B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\|) \\
&= \sum_{\mu\nu} \sum_{\mathbf{k}_\|} \sum_{\substack{m\\ \alpha\beta}} A^{\mu\nu}_{\ell_1\ell_1}(\mathbf{R}'_\| - \mathbf{R}_\|, \mathbf{k}_\|) \\
&\times \left\{ \left[1 + \chi^0(0)U\right]^{-1} \left[1 + \left(\chi^0(0) - \lambda \frac{\partial}{\partial\lambda} \left[\chi^0(0)\right]\right) U\right] \left[1 + \chi^0(0)U\right]^{-1} \right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1 mm} \sum_{\substack{\gamma\xi\\ \ell\ell'}} \chi^{0\,\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\|, \mathbf{k}_\|) \\
&+ \left\{ \left[1 + \chi^0(0)U\right]^{-1} \right\}^{\mu\nu\alpha\beta}_{\ell_1\ell_1 mm} \sum_{\substack{\gamma\xi\\ \ell\ell'}} \lambda \frac{\partial}{\partial\lambda} \chi^{0\,\alpha\beta\gamma\xi}_{mm\ell\ell'}(\mathbf{k}_\|, \mathbf{k}_\|) \\
&\times B^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\|) \ .
\end{aligned}
\tag{J.6}
$$

Appendix K

Self-consistency

K.1 Changing center of the bands (fixed magnetization)

Our magnetic self-consistency is done in the following way: we vary the center of the d-orbitals in each layer such that the number of particles per layer from a DFT calculation is obtained. We only change the center of d-orbitals because its density of states is larger, which gives a smaller changes in the center of the bands (compared to s and p orbitals, that have small density of states). This results in less unexpected hybridizations.

Therefore, for a system with N layers, the set of N equations that we need to solve to obtain the number of particles of layer i, n^i, by varying of the center of the bands ϵ_i are given by

$$n^i(\epsilon_1 \cdots \epsilon_N) = n_0^i \Rightarrow n^i(\epsilon_1 \cdots \epsilon_N) - n_0^i = 0 \ , \tag{K.1}$$

for $i = 1 \cdots N$. The number of particles is obtained from the Green functions as

$$\begin{aligned} n^i(\epsilon_1 \cdots \epsilon_N) &= -\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}} \int_{-\infty}^{E_\mathrm{F}} dE \,\mathrm{Im}\,\mathrm{Tr}\, G_{ii}(\mathbf{k}, E + i\eta; \epsilon_1 \cdots \epsilon_N) \\ &= -\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\sigma\mu} \int_{-\infty}^{E_\mathrm{F}} dE \,\mathrm{Im}\, G_{ii}^{\sigma\sigma\,\mu\mu}(\mathbf{k}, E + i\eta; \epsilon_1 \cdots \epsilon_N) \\ &= \frac{1}{N}\sum_{\mathbf{k}}\sum_{\sigma\mu} \left\{ \frac{1}{2} + \frac{1}{\pi}\int_{\eta}^{\infty} dy \,\mathrm{Re}\, G_{ii}^{\sigma\sigma\,\mu\mu}(\mathbf{k}, E_\mathrm{F} + iy; \epsilon_1 \cdots \epsilon_N) \right\} \end{aligned} \tag{K.2}$$

where G_{ii} is a matrix in spin and orbital spaces and the calculation should be done in the limit of $\eta \to 0$. Eq. (K.1) encloses N equations (for the number of particles in each layer) with N unknowns (ϵ_i). The Jacobian is obtained by taking the derivative with respect to ϵ_j, i.e.,

$$\begin{aligned}\frac{\partial n^i(\epsilon_1\cdots\epsilon_N)}{\partial\epsilon_j} &= \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\sigma\mu}\int_\eta^\infty dy\,\mathrm{Re}\left\{\frac{\partial G_{ii}^{\sigma\sigma\,\mu\mu}(\mathbf{k},E_\mathrm{F}+iy;\epsilon_1\cdots\epsilon_N)}{\partial\epsilon_j}\right\}\\ &=\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\sigma\mu}\int_\eta^\infty dy\,\mathrm{Re}\left\{\frac{\partial}{\partial\epsilon_j}\langle i\mu\sigma|\left[E_\mathrm{F}+iy-H(\mathbf{k},\epsilon_1\cdots\epsilon_N)\right]^{-1}|i\mu\sigma\rangle\right\}\\ &=\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\sigma\mu}\int_\eta^\infty dy\,\mathrm{Re}\left\{\left\langle i\mu\sigma\left|\frac{\partial\left[E_\mathrm{F}+iy-H(\mathbf{k},\epsilon_1\cdots\epsilon_N)\right]^{-1}}{\partial\epsilon_j}\right|i\mu\sigma\right\rangle\right\}\ .\end{aligned} \tag{K.3}$$

To obtain the derivative of the inverse of a matrix $\mathbf{U}$, we use the fact that $\mathbf{U}\mathbf{U}^{-1}=\mathbf{1}$ to obtain

$$\frac{\partial\mathbf{U}^{-1}}{\partial x}=-\mathbf{U}^{-1}\frac{\partial\mathbf{U}}{\partial x}\mathbf{U}^{-1}\ , \tag{K.4}$$

or

$$\frac{\partial\mathbf{G}}{\partial x}=\mathbf{G}\frac{\partial\mathbf{H}}{\partial x}\mathbf{G}\ . \tag{K.5}$$

Therefore,

$$\frac{\partial n^i(\epsilon_1\cdots\epsilon_N)}{\partial\epsilon_j}=\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\sigma\mu}\int_\eta^\infty dy\,\mathrm{Re}\left\{\left\langle i\mu\sigma\left|G(\mathbf{k},E_\mathrm{F}+iy)\frac{\partial H(\mathbf{k},\epsilon_1\cdots\epsilon_N)}{\partial\epsilon_j}G(\mathbf{k},E_\mathrm{F}+iy)\right|i\mu\sigma\right\rangle\right\}\ . \tag{K.6}$$

Since we only change the center of the d-orbitals (and this values are in the diagonal of the Hamiltonian), the derivative of the Hamiltonian with respect to ϵ_j is a diagonal matrix with 1 only in the d-orbitals of layer j. So,

$$\frac{\partial n^i(\epsilon_1\cdots\epsilon_N)}{\partial\epsilon_j}=\frac{1}{\pi}\int_\eta^\infty dy\,\mathrm{Re}\left\{\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{\sigma\sigma'\mu\\ \nu=5,9}}G_{ij}^{\sigma\sigma'\,\mu\nu}(\mathbf{k},E_\mathrm{F}+iy)G_{ji}^{\sigma'\sigma\,\nu\mu}(\mathbf{k},E_\mathrm{F}+iy)\right\}\ . \tag{K.7}$$

K.2 Linear SOC

If we use SOC in linear approximation, as in Appendix J, the Green function is given by Eq. (J.2). Therefore, Eq. (K.3) becomes

$$\begin{aligned}\frac{\partial n^i(\epsilon_1\cdots\epsilon_N)}{\partial\epsilon_j}=\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\sigma\mu}\int_\eta^\infty dy\,\mathrm{Re}\Bigg\{\frac{\partial}{\partial\epsilon_j}\Big[&G^0(\mathbf{k},E_\mathrm{F}+iy;\epsilon_1\cdots\epsilon_N)\\ &+G^0(\mathbf{k},E_\mathrm{F}+iy;\epsilon_1\cdots\epsilon_N)\hat{H}_\mathrm{so}G^0(\mathbf{k},E_\mathrm{F}+iy;\epsilon_1\cdots\epsilon_N)\Big]_{ii}^{\sigma\sigma\,\mu\mu}\Bigg\}\ .\end{aligned} \tag{K.8}$$

The first term inside the curly brackets ($\frac{\partial G^0}{\partial \epsilon_j}$) will give rise to a term exactly as K.7 calculated with the Green functions with no SOC ($\lambda = 0$). The second term is given by

$$\frac{\partial}{\partial \epsilon_j}\left[G^0 \hat{H}_{\rm so} G^0\right] = \frac{\partial G^0}{\partial \epsilon_j}\hat{H}_{\rm so} G^0 + G^0 \hat{H}_{\rm so}\frac{\partial G^0}{\partial \epsilon_j} \ . \tag{K.9}$$

Using the results above and bringing the indices back,

$$\sum_{\sigma\mu}\frac{\partial}{\partial \epsilon_j}\left[G\hat{H}_{\rm so}G\right]^{\sigma\sigma\,\mu\mu}_{ii} = \sum_{\substack{\sigma\sigma'\mu \\ \nu=5,9}}\left\{G^{\sigma\sigma'\,\mu\nu}_{ij}\left[G\hat{H}_{\rm so}G\right]^{\sigma'\sigma\,\nu\mu}_{ji} + \left[G\hat{H}_{\rm so}G\right]^{\sigma\sigma'\,\mu\nu}_{ij} G^{\sigma'\sigma\,\nu\mu}_{ji}\right\} \ . \tag{K.10}$$

where now all the Green functions are calculated with no SOC. Putting these results together and comparing with Eq. (K.7), we see that we can also obtain it by substituting the Green functions given by Eq. (J.2) into Eq. (K.7) and dropping the second order term. So,

$$\begin{aligned}\frac{\partial n^i(\epsilon_1\cdots\epsilon_N)}{\partial \epsilon_j} = \frac{1}{\pi}\int_\eta^\infty dy\,{\rm Re}\Bigg\{&\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{\sigma\sigma'\mu \\ \nu=5,9}} G^{\sigma\sigma'\,\mu\nu}_{ij}(\mathbf{k},E_{\rm F}+iy)G^{\sigma'\sigma\,\nu\mu}_{ji}(\mathbf{k},E_{\rm F}+iy) \\ &- \left[G\hat{H}_{\rm so}G\right]^{\sigma\sigma'\,\mu\nu}_{ij}\left[G\hat{H}_{\rm so}G\right]^{\sigma'\sigma\,\nu\mu}_{ji}\Bigg\} \ .\end{aligned} \tag{K.11}$$

K.3 Changing spin density

We can also do the self-consistency on the center of the bands together with the magnetization $\mathbf{m} = m^x\hat{\mathbf{x}} + m^y\hat{\mathbf{y}} + m^z\hat{\mathbf{z}}$. The number of particles is given by Eq. (K.2). Taking into account that the number of particles depends also on the magnetization, we need to complete the system of equations with the calculation of the magnetization components in each layer. Self-consistency is achieved when

$$\left\{\begin{array}{c} n_\ell(\{\epsilon_i\},\{\mathbf{m}_i\}) = n^0_\ell \\ m^{x,{\rm out}}_\ell(\{\epsilon_i\},\{\mathbf{m}_i\}) = m^{x,{\rm in}}_\ell \\ m^{y,{\rm out}}_\ell(\{\epsilon_i\},\{\mathbf{m}_i\}) = m^{y,{\rm in}}_\ell \\ m^{z,{\rm out}}_\ell(\{\epsilon_i\},\{\mathbf{m}_i\}) = m^{z,{\rm in}}_\ell \end{array}\right. \Rightarrow \left\{\begin{array}{c} n^i(\{\epsilon_i\},\{\mathbf{m}_i\}) - n^i_0 = 0 \\ m^{x,{\rm out}}_\ell(\{\epsilon_i\},\{\mathbf{m}_i\}) - m^{x,{\rm in}}_\ell = 0 \\ m^{y,{\rm out}}_\ell(\{\epsilon_i\},\{\mathbf{m}_i\}) - m^{y,{\rm in}}_\ell = 0 \\ m^{z,{\rm out}}_\ell(\{\epsilon_i\},\{\mathbf{m}_i\}) - m^{z,{\rm in}}_\ell = 0 \end{array}\right. . \tag{K.12}$$

Now the system has $4\times N$ equations with $4\times N$ unknowns, where N is the number of planes. The magnetization densities above can be obtained using Eq. (1.72). The transverse components m^x_ℓ and m^y_ℓ are calculated from the real and imaginary part of

m_ℓ^+, i.e.,

$$\begin{aligned}\langle m_\ell^+\rangle = 2\langle S_\ell^+\rangle &= \sum_{\mu=5,9} 2\langle \hat{c}^\dagger_{\ell\mu\uparrow}\hat{c}_{\ell\mu\downarrow}\rangle(\mathbf{R}_\parallel = 0) \\ &= \frac{1}{\pi}\int_\eta^\infty dy \frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\mu=5,9}\left\{\widetilde{G}^{\downarrow\uparrow\mu\mu}_{\ell\ell}(\mathbf{k}_\parallel; E_\mathrm{F}+iy) + \left[\widetilde{G}^{\uparrow\downarrow\mu\mu}_{\ell\ell}(\mathbf{k}_\parallel; E_\mathrm{F}+iy)\right]^*\right\} \ .\end{aligned} \tag{K.13}$$

and the m^z component is given by

$$\begin{aligned}\langle m_\ell^z\rangle =&2\langle S_\ell^z\rangle = \langle n_\ell^\uparrow\rangle - \langle n_\ell^\downarrow\rangle \\ &= \sum_{\mu=5,9}\left\{\langle \hat{c}^\dagger_{\ell\mu\uparrow}\hat{c}_{\ell\mu\uparrow}\rangle(\mathbf{R}_\parallel = 0) - \langle \hat{c}^\dagger_{\ell\mu\downarrow}\hat{c}_{\ell\mu\downarrow}\rangle(\mathbf{R}_\parallel = 0)\right\} \ ,\end{aligned} \tag{K.14}$$

where

$$\langle \hat{c}^\dagger_{\ell\mu\sigma}\hat{c}_{\ell\mu\sigma}\rangle(\mathbf{R}_\parallel = 0) = \frac{1}{2} + \frac{1}{\pi}\,\mathrm{Re}\int_\eta^\infty dy \frac{1}{N_\parallel}\sum_{\mathbf{k}_\parallel}\sum_{\mu=5,9}\widetilde{G}^{\sigma\sigma\mu\mu}_{\ell\ell}(\mathbf{k}_\parallel; E_\mathrm{F}+iy) \ . \tag{K.15}$$

To complete the Jacobian, we first calculate the derivative of the number of particles w.r.t. the magnetization components

$$\frac{\partial n_\ell(\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial m^\alpha_{\ell'}} = \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\sigma\mu}\int_\eta^\infty dy\,\mathrm{Re}\left\{\left\langle \ell\mu\sigma \left| G(\mathbf{k}, E_\mathrm{F}+iy)\frac{\partial H(\mathbf{k},\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial m^\alpha_{\ell'}} G(\mathbf{k}, E_\mathrm{F}+iy)\right| \ell\mu\sigma\right\rangle\right\} \ . \tag{K.16}$$

For this, we need the following derivatives:

$$\begin{aligned}\frac{\partial H(\mathbf{k},\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial m^x_{\ell'}} &= -\frac{U^{\ell'}}{2}\sum_{\mu=5,9}\left\{c^\dagger_{\ell'\mu\uparrow}c_{\ell'\mu\downarrow} + c^\dagger_{\ell'\mu\downarrow}c_{\ell'\mu\uparrow}\right\} \\ \frac{\partial H(\mathbf{k},\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial m^y_{\ell'}} &= -\frac{U^{\ell'}}{2}\sum_{\mu=5,9}\left\{-ic^\dagger_{\ell'\mu\uparrow}c_{\ell'\mu\downarrow} + ic^\dagger_{\ell'\mu\downarrow}c_{\ell'\mu\uparrow}\right\} \\ \frac{\partial H(\mathbf{k},\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial m^z_{\ell'}} &= -\frac{U^{\ell'}}{2}\sum_{\mu=5,9}\left\{c^\dagger_{\ell'\mu\uparrow}c_{\ell'\mu\uparrow} - c^\dagger_{\ell'\mu\downarrow}c_{\ell'\mu\downarrow}\right\} \ .\end{aligned} \tag{K.17}$$

where we take into account only d orbitals (i.e., all these terms vanish for s and p orbitals). These equations can be synthesized as

$$\frac{\partial H(\mathbf{k},\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial m^m_{\ell'}} = -\frac{U^{\ell'}}{2}\sum_{\substack{\mu=5,9\\ \alpha\beta}} c^\dagger_{\ell'\mu\alpha}\sigma^m_{\alpha\beta}c_{\ell'\mu\beta} \ . \tag{K.18}$$

Together with $\frac{\partial H(\mathbf{k},\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial \epsilon_{\ell'}} = \sum_{\mu=5,9}\sum_{\alpha\beta} c^\dagger_{\ell'\mu\alpha}\sigma^0_{\alpha\beta}c_{\ell'\mu\beta}$, where σ^0 is the identity matrix, these terms give all the components we need to obtain the jacobian. All components involve different elements of the product

$$\frac{\partial G(\mathbf{k}, E_\mathrm{F}+iy)}{\partial x_{\ell'}} = G(\mathbf{k}, E_\mathrm{F}+iy)\frac{\partial H(\mathbf{k},\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial x_{\ell'}}G(\mathbf{k}, E_\mathrm{F}+iy) \tag{K.19}$$

where $x_{\ell'} = \{\epsilon_{\ell'}, m^x_{\ell'}, m^y_{\ell'}, m^z_{\ell'}\}$.

The derivatives for each component are

$$\frac{\partial n_\ell(\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial m^\alpha_{\ell'}} = \frac{1}{\pi}\int_\eta^\infty dy\,\mathrm{Re}\left\{\frac{1}{N}\sum_{\mathbf{k}}\sum_{\substack{\sigma\mu\\ \nu=5,9}} G^{\sigma\uparrow\mu\nu}_{\ell\ell'}(\mathbf{k},E_\mathrm{F}+iy)G^{\downarrow\sigma\nu\mu}_{\ell'\ell}(\mathbf{k},E_\mathrm{F}+iy)\right.$$
$$\left. + G^{\sigma\downarrow\mu\nu}_{\ell\ell'}(\mathbf{k},E_\mathrm{F}+iy)G^{\uparrow\sigma\nu\mu}_{ji}(\mathbf{k},E_\mathrm{F}+iy)\right\}\ . \tag{K.20}$$

$$\frac{\partial n_\ell(\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial m^\alpha_{\ell'}} = \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\sigma\mu}\int_\eta^\infty dy\,\mathrm{Re}\left\{\left\langle \ell\mu\sigma\left| G(\mathbf{k},E_\mathrm{F}+iy)\frac{\partial H(\mathbf{k},\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial m^\alpha_{\ell'}}G(\mathbf{k},E_\mathrm{F}+iy)\right|\ell\mu\sigma\right\rangle\right\}\ . \tag{K.21}$$

Another way to obtain the densities (charge and magnetic) is using the equation

$$\begin{aligned}\rho^\alpha_\ell(\{\epsilon_i\},\{\mathbf{m}_i\}) &= -\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\int_{-\infty}^{E_\mathrm{F}} dE\,\mathrm{Im}\,\mathrm{Tr}\,\sigma^\alpha G_{\ell\ell}(\mathbf{k},E+i\eta;\{\epsilon_i\},\{\mathbf{m}_i\})\\
&= \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\mathrm{Im}\,\mathrm{Tr}\left\{i\frac{\pi}{2}\,\sigma^\alpha + \int_\eta^\infty idy\,\sigma^\alpha G_{\ell\ell}(\mathbf{k},E_\mathrm{F}+iy;\{\epsilon_i\},\{\mathbf{m}_i\})\right\}\\
&= \frac{1}{N}\sum_{\mathbf{k}}\mathrm{Re}\,\mathrm{Tr}\left\{\frac{1}{2}\,\sigma^\alpha + \frac{1}{\pi}\int_\eta^\infty dy\,\sigma^\alpha G_{\ell\ell}(\mathbf{k},E_\mathrm{F}+iy;\{\epsilon_i\},\{\mathbf{m}_i\})\right\}\end{aligned} \tag{K.22}$$

The trace is taken only over d orbitals for the magnetic densities ($\alpha = x,y,z$). The system of equations that we have to solve can be written as

$$\rho^\alpha_\ell(\{\epsilon_i\},\{\mathbf{m}_i\}) - \rho_\ell^{\mathrm{in}\,\alpha} = 0\ , \tag{K.23}$$

where $\alpha = 0,x,y,z$. Taking the derivative with respect to $\rho^\beta_{\ell'} = \epsilon_{\ell'}, m^x_{\ell'}, m^y_{\ell'}, m^z_{\ell'}$, we have

$$\frac{\partial\rho^\alpha_\ell(\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial\rho^\beta_{\ell'}} = \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\mathrm{Re}\,\mathrm{Tr}\int_\eta^\infty dy\,\sigma^\alpha\frac{\partial G_{\ell\ell}(\mathbf{k},E_\mathrm{F}+iy;\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial\rho^\beta_{\ell'}} \tag{K.24}$$

Using Eq. (K.19), we obtain

$$\frac{\partial \rho^{\alpha}_{\ell}(\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial \rho^{\beta}_{\ell'}} = \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}} \mathrm{Re}\,\mathrm{Tr}\int_{\eta}^{\infty} dy\, \langle \ell|\sigma^{\alpha} G(\mathbf{k},E_{\mathrm{F}}+iy;\{\epsilon_i\},\{\mathbf{m}_i\}) \frac{\partial H(\mathbf{k},\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial \rho^{\beta}_{\ell'}} G(\mathbf{k},E_{\mathrm{F}}+iy;\{\epsilon_i \tag{K.25}$$

Defining

$$A^{\alpha\beta}_{\ell\ell'} = \langle \ell|\sigma^{\alpha} G(\mathbf{k},E_{\mathrm{F}}+iy;\{\epsilon_i\},\{\mathbf{m}_i\}) \frac{\partial H(\mathbf{k},\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial \rho^{\beta}_{\ell'}} G(\mathbf{k},E_{\mathrm{F}}+iy;\{\epsilon_i\},\{\mathbf{m}_i\})|\ell\rangle \ , \tag{K.26}$$

we end up with

$$\begin{aligned} \frac{\partial \rho^{\alpha}_{\ell}(\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial \rho^{\beta}_{\ell'}} &= \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}} \mathrm{Re}\,\mathrm{Tr}\int_{\eta}^{\infty} dy\, A^{\alpha\beta}_{\ell\ell'} \\ &= \frac{1}{2\pi}\frac{1}{N}\sum_{\mathbf{k}} \mathrm{Tr}\int_{\eta}^{\infty} dy\, \left\{ A^{\alpha\beta}_{\ell\ell'} + \left[A^{\beta\alpha}_{\ell'\ell}\right]^{\dagger} \right\} \end{aligned} \tag{K.27}$$

where

$$\frac{\partial H(\mathbf{k},\{\epsilon_i\},\{\mathbf{m}_i\})}{\partial \rho^{m}_{\ell'}} = -\frac{U^{\ell'}}{2} \sum_{\substack{\mu=5,9\\ \alpha\beta}} c^{\dagger}_{\ell'\mu\alpha}\sigma^{\beta}_{\alpha\beta}c_{\ell'\mu\beta} \ . \tag{K.28}$$

K.4 Including charge self-consistency

Let us now write a general self-consistency including the charge and magnetic parts. The center of the bands are no longer varying. The number of particles is given by

$$\begin{aligned} n_i &= -\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_{\sigma\mu}\int_{-\infty}^{E_{\mathrm{F}}} dE\, \mathrm{Im}\, G^{\sigma\sigma\,\mu\mu}_{ii}(\mathbf{k},E+i\eta) \\ &= \frac{1}{N}\sum_{\mathbf{k}}\sum_{\sigma\mu}\left\{\frac{1}{2}+\frac{1}{\pi}\int_{\eta}^{\infty} dy\, \mathrm{Re}\, G^{\sigma\sigma\,\mu\mu}_{ii}(\mathbf{k},E_{\mathrm{F}}+iy)\right\} \end{aligned} \tag{K.29}$$

The hamiltonian depends on the density due to the $Un/2$ term on Eq. (1.33).

The complete set of equations including charge and magnetization components in each site of the unit cell is

$$\left\{ \begin{aligned} n_i^{\mathrm{out}}(\{n_i,\mathbf{m}_i\}) &= n_i^{\mathrm{in}} \\ m_i^{x,\mathrm{out}}(\{n_i,\mathbf{m}_i\}) &= m_i^{x,\mathrm{in}} \\ m_i^{y,\mathrm{out}}(\{n_i,\mathbf{m}_i\}) &= m_i^{y,\mathrm{in}} \\ m_i^{z,\mathrm{out}}(\{n_i,\mathbf{m}_i\}) &= m_i^{z,\mathrm{in}} \end{aligned} \right. \Rightarrow \left\{ \begin{aligned} n_i^{\mathrm{out}}(\{n_i,\mathbf{m}_i\}) - n_i^{\mathrm{in}} &= 0 \\ m_i^{x,\mathrm{out}}(\{n_i,\mathbf{m}_i\}) - m_i^{x,\mathrm{in}} &= 0 \\ m_i^{y,\mathrm{out}}(\{n_i,\mathbf{m}_i\}) - m_i^{y,\mathrm{in}} &= 0 \\ m_i^{z,\mathrm{out}}(\{n_i,\mathbf{m}_i\}) - m_i^{z,\mathrm{in}} &= 0 \end{aligned} \right. . \tag{K.30}$$

Another way to obtain these densities (charge and magnetic) is

$$\begin{aligned}
\rho_i^\alpha(\{n_i,\mathbf{m}_i\}) &= -\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\int_{-\infty}^{E_\mathrm{F}} dE\,\mathrm{Im}\,\mathrm{Tr}\,\sigma^\alpha G_{ii}(\mathbf{k},E+i\eta;\{n_i,\mathbf{m}_i\}) \\
&= \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\mathrm{Im}\,\mathrm{Tr}\left\{i\frac{\pi}{2}\,\sigma^\alpha + \int_\eta^\infty idy\,\sigma^\alpha G_{ii}(\mathbf{k},E_\mathrm{F}+iy;\{n_i,\mathbf{m}_i\})\right\} \\
&= \frac{1}{N}\sum_{\mathbf{k}}\mathrm{Re}\,\mathrm{Tr}\left\{\frac{1}{2}\,\sigma^\alpha + \frac{1}{\pi}\int_\eta^\infty dy\,\sigma^\alpha G_{ii}(\mathbf{k},E_\mathrm{F}+iy;\{n_i,\mathbf{m}_i\})\right\}
\end{aligned} \tag{K.31}$$

where $\alpha = 0,x,y,z$. The trace is taken over s,p and d orbitals. The system of equations that we have to solve can be written as

$$\rho_i^{\alpha,\mathrm{out}}(\{n_i,\mathbf{m}_i\}) - \rho_i^{\alpha,\mathrm{in}} = 0\ . \tag{K.32}$$

Taking the derivative with respect to $\rho_j^{\beta,\mathrm{in}} = n_j^\mathrm{in}, m_j^{x,\mathrm{in}}, m_j^{y,\mathrm{in}}, m_j^{z,\mathrm{in}}$, we have

$$\frac{\partial\rho_i^{\alpha,\mathrm{out}}(\{n^\mathrm{in},\mathbf{m}^\mathrm{in}\})}{\partial\rho_j^{\beta,\mathrm{in}}} = \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\mathrm{Re}\,\mathrm{Tr}\int_\eta^\infty dy\,\sigma^\alpha\frac{\partial G_{ii}(\mathbf{k},E_\mathrm{F}+iy;\{n^\mathrm{in},\mathbf{m}^\mathrm{in}\})}{\partial\rho_j^{\beta,\mathrm{in}}} - \delta_{ij}\delta_{\alpha\beta} \tag{K.33}$$

Using Eq. (K.19), we obtain

$$\begin{aligned}
\frac{\partial\rho_i^{\alpha,\mathrm{out}}(\{n^\mathrm{in},\mathbf{m}^\mathrm{in}\})}{\partial\rho_j^{\beta,\mathrm{in}}} =& \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\mathrm{Re}\,\mathrm{Tr}\int_\eta^\infty dy \\
& \langle i|\sigma^\alpha G(\mathbf{k},E_\mathrm{F}+iy;\{n^\mathrm{in},\mathbf{m}^\mathrm{in}\})\frac{\partial H(\mathbf{k},\{n^\mathrm{in},\mathbf{m}^\mathrm{in}\})}{\partial\rho_j^{\beta,\mathrm{in}}}G(\mathbf{k},E_\mathrm{F}+iy;\{n^\mathrm{in},\mathbf{m}^\mathrm{in}\})|i\rangle - \delta_{ij}\delta_{\alpha\beta}
\end{aligned} \tag{K.34}$$

Defining

$$A_{ij}^{\alpha\beta} = \langle i|\sigma^\alpha G(\mathbf{k},E_\mathrm{F}+iy;\{n^\mathrm{in},\mathbf{m}^\mathrm{in}\})\frac{\partial H(\mathbf{k},\{n^\mathrm{in},\mathbf{m}^\mathrm{in}\})}{\partial\rho_j^{\beta,\mathrm{in}}}G(\mathbf{k},E_\mathrm{F}+iy;\{n^\mathrm{in},\mathbf{m}^\mathrm{in}\})|i\rangle\ , \tag{K.35}$$

where

$$\frac{\partial H(\mathbf{k},\{n^\mathrm{in},\mathbf{m}^\mathrm{in}\})}{\partial\rho_j^{m,\mathrm{in}}} = -\frac{U^j}{2}\sum_{\substack{\mu=5,9\\ \alpha\beta}} c_{j\mu\alpha}^\dagger\sigma_{\alpha\beta}^m c_{j\mu\beta}\ , \tag{K.36}$$

we end up with

$$\begin{aligned}
\frac{\partial\rho_i^{\alpha,\mathrm{out}}(\{n^\mathrm{in},\mathbf{m}^\mathrm{in}\})}{\partial\rho_j^{\beta,\mathrm{in}}} &= \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\mathrm{Re}\,\mathrm{Tr}\int_\eta^\infty dy\,A_{ij}^{\alpha\beta} \\
&= \frac{1}{2\pi}\frac{1}{N}\sum_{\mathbf{k}}\mathrm{Tr}\int_\eta^\infty dy\,\left\{A_{ij}^{\alpha\beta} + \left[A_{ji}^{\beta\alpha}\right]^\dagger\right\}
\end{aligned} \tag{K.37}$$

K.5 Varying the Fermi level

When performing (extra) approximations on the tight-binding parameters obtained for a certain system, the electronic structure will change. These changes may not conserve the total number of charges on the system. For example, if the parameters were obtained taking into account a certain number of nearest neighbors and a different number is used to perform a new calculation; or if the number of k-points is not well converged; or also if some material parameters obtained for a certain geometry (e.g., bulk) is used in a different structure (e.g., slabs). On top of that, by adding the electron-electron interaction (in a Hubbard model, for example) and allowing the initial paramagnetic system to become magnetic, it may also cause changes on the electronic structure.

In this sense, it is important to enforce total charge neutrality to the system, since it can move from site to site but it cannot be created or depleted. This can be done by varying the Fermi level of the system. Adding an extra equation and variable to the self-consistency equations written in Eq. (K.23):

$$\begin{aligned} n_i^{\text{out},d}(\{n^{\text{in},d},\mathbf{m}^{\text{in},d}\},E_\text{F}) - n_i^{\text{in},d} &=0 \\ \mathbf{m}_i^{\text{out},d}(\{n^{\text{in},d},\mathbf{m}^{\text{in},d}\},E_\text{F}) - \mathbf{m}_i^{\text{in},d} &=0 \\ \sum_i n_i^{\text{out}}(\{n^{\text{in}},\mathbf{m}^{\text{in}}\},E_\text{F}) - N_\text{el} &=0 \end{aligned} \,, \tag{K.38}$$

where N_el is the total charge of the system and the Fermi level E_F is a new unknown to be found. Since the Hamiltonian only depends on the d-orbital densities (due to the terms $-\frac{U\mathbf{m}\cdot\boldsymbol{\sigma}}{2}$ and $-\frac{Un}{2}$ contributing only to the d-block), we find them self-consistently. The last equation enforces total charge neutrality on the system. Thus,

$$\begin{aligned} \rho_i^{\text{out},d}(\{n^{\text{in},d},\mathbf{m}^{\text{in},d}\},E_\text{F}) - \rho_i^{\text{in},d} = 0 \\ \sum_i \left[n_i^{\text{out},d}(\{n^{\text{in},sp},\mathbf{m}^{\text{in},d}\},E_\text{F}) + n_i^{\text{out},d}(\{n^{\text{in},d},\mathbf{m}^{\text{in},d}\},E_\text{F})\right] - N_\text{el} = 0 \end{aligned} \,. \tag{K.39}$$

The densities of the system can be calculated for $T = 0K$ as

$$\rho_i^{\alpha,\text{out}}(\{n^{\text{in}},\mathbf{m}^{\text{in}}\},E_\text{F}) = -\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\int_{-\infty}^{E_\text{F}} dE\, \text{Im}\,\text{Tr}\,\sigma^\alpha G_{ii}(\mathbf{k},E+i\eta;\{n^{\text{in}},\mathbf{m}^{\text{in}}\}) \,. \tag{K.40}$$

where $\alpha = 0,x,y,z$. The derivatives with respect to the densities were already calculated in the previous section (assuming that the Fermi level does not depend on $\rho^{\alpha,\text{in}}$ — they are independent variables). We also need the derivative with respect to the Fermi level,

$$\frac{\partial\rho_i^{\alpha,\text{out}}(\{n^{\text{in}},\mathbf{m}^{\text{in}}\},E_\text{F})}{\partial E_\text{F}} = -\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}} \text{Im}\,\text{Tr}\,\sigma^\alpha G_{ii}(\mathbf{k},E_\text{F}+i\eta;\{n^{\text{in}},\mathbf{m}^{\text{in}}\}) \,. \tag{K.41}$$

The derivatives of the last equation with respect to the densities can be written as

$$\sum_i \frac{\partial n_i^{\text{out}}(\{n^{\text{in}},\mathbf{m}^{\text{in}}\},E_\text{F})}{\partial\rho_j^{\beta,\text{in}}} = \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_i \text{Re}\,\text{Tr}\int_\eta^\infty dy\, \frac{\partial G_{ii}(\mathbf{k},E_\text{F}+iy;\{n^{\text{in}},\mathbf{m}^{\text{in}}\})}{\partial\rho_j^{\beta,\text{in}}} \,, \tag{K.42}$$

This expression is similar to the one obtained before, on Eqs. (K.35)-(K.37) with $\alpha = 0$, including the sum over i in the trace and no δs from the input charge (i.e., the total charge is constant).

Finally, the derivative of the last equation with respect to the Fermi level is similar to Eq. (K.41) (with $\alpha = 0$) also including the sum over sites.

$$\sum_i \frac{\partial n_i^{\text{out}}(\{n^{\text{in}}, \mathbf{m}^{\text{in}}\}, E_{\text{F}})}{\partial E_{\text{F}}} = -\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_i \operatorname{Im}\operatorname{Tr} G_{ii}(\mathbf{k}, E_{\text{F}} + i\eta; \{n^{\text{in}}, \mathbf{m}^{\text{in}}\}) \ . \tag{K.43}$$

K.6 Orbital dependent occupations

To obtain the expectation values required in the hamiltonian given in Eq. (1.33), we need the orbital dependent densities. The self-consistency equations are then

$$\begin{aligned} n_{i\mu}^{\text{out}}(\{n^{\text{in},d}, \mathbf{m}^{\text{in},d}\}, E_{\text{F}}) - n_{i\mu}^{\text{in}} =& 0 \\ \mathbf{m}_i^{\text{out},d}(\{n^{\text{in},d}, \mathbf{m}^{\text{in},d}\}, E_{\text{F}}) - \mathbf{m}_i^{\text{in},d} =& 0 \\ \sum_{i\mu} n_{i\mu}^{\text{out}}(\{n^{\text{in}}, \mathbf{m}^{\text{in}}\}, E_{\text{F}}) - N_{\text{el}} =& 0 \end{aligned} \ , \tag{K.44}$$

where μ cover the d orbitals. Therefore, the system now have 5 orbitals +3 directions of the magnetization for each site, and an extra equation for the total charge neutrality varying E_{F}. This way, at the end of the self-consistency, we obtain the unknowns: $\{n_{i\mu}^d, \mathbf{m}_i^d, E_{\text{F}}\}$.

The output charge and magnetic densities are calculated using

$$\rho_{i\mu}^{\alpha,\text{out}} = -\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}} \int_{-\infty}^{E_{\text{F}}} dE \operatorname{Im}\operatorname{tr} \sigma^\alpha G_{ii}^{\mu\mu}(\mathbf{k}, E + i\eta) \ , \tag{K.45}$$

where tr is the trace over spins only.

K.6.1 Jacobian

The Jacobian is obtained by taking the derivatives of the Eq. (K.44) with respect to the unknowns $\{n_{i\mu}^d, \mathbf{m}_i^d, E_{\text{F}}\}$.

First, taking the derivative of the equations with respect to the orbital density,

$$\frac{\partial \rho_{i\mu}^{\alpha,\text{out}}}{\partial \rho_{j\nu}^{\beta,\text{in}}} - \delta_{ij}\delta_{\mu\nu}\delta_{\alpha\beta} = \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}\sigma}\int_\eta^\infty dy \operatorname{Re}\left\langle i\mu\sigma \left| \sigma^\alpha G(\mathbf{k}, E_{\text{F}} + iy) \frac{\partial H(\mathbf{k})}{\partial \rho_{j\nu}^{\beta,\text{in}}} G(\mathbf{k}, E_{\text{F}} + iy) \right| i\mu\sigma \right\rangle \ . \tag{K.46}$$

Here, $\alpha, \beta = 0, x, y, z$, and the magnetic density $\mathbf{m}^d$ can be obtained by summing over the d orbitals.

The following derivatives are needed

$$\begin{aligned}\frac{\partial H(\mathbf{k})}{\partial n_{j\nu}^{\text{in}}} &= -\frac{U^j}{2}\sum_{\mu=5,9}\sum_{s}\left(1-2\delta_{\mu\nu}\right)c_{j\mu s}^{\dagger}c_{j\mu s} \\ \frac{\partial H(\mathbf{k})}{\partial m_j^{\beta,\text{in},d}} &= -\frac{U^j}{2}\sum_{\mu=5,9}\sum_{ss'}c_{j\mu s}^{\dagger}\sigma_{ss'}^{\beta}c_{j\mu s'}\ .\end{aligned} \tag{K.47}$$

The first matrix is given by $-\frac{U^j}{2}$ in the diagonal of the d orbitals, and U^j should be added to the orbital component for which the derivative is being taken. For the magnetization components, the trace over d orbitals should then be taken (whilst for the charge densities, each d component is necessary). The last line of the Jacobian can be obtained by summing the charge components — including also the s and p orbitals.

The derivative of densities with respect to the Fermi level are given by

$$\frac{\partial \rho_{i\mu}^{\alpha,\text{out}}}{\partial E_{\text{F}}} = -\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\text{Im}\,\text{Tr}\,\sigma^{\alpha}G_{ii}^{\mu\mu}(\mathbf{k},E_{\text{F}}+i\eta)\ . \tag{K.48}$$

K.6.2 Susceptibility

We can rewrite Eq. K.49 to make the susceptibility equation showing up explicitly. For this, we have

$$\frac{\partial \rho_{i\mu}^{\alpha,\text{out}}}{\partial \rho_{j\nu}^{\beta,\text{in}}} = \delta_{ij}\delta_{\mu\nu}\delta_{\alpha\beta} + \frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}\sigma}\int_{\eta}^{\infty}dy\,\text{Re}\sum_{k\gamma}\text{tr}\left\{\sigma^{\alpha}G_{ik}^{\mu\gamma}(\mathbf{k},E_{\text{F}}+iy)\left[\frac{\partial H(\mathbf{k})}{\partial \rho_{j\nu}^{\beta,\text{in}}}\right]_{kk}^{\gamma\gamma}G_{ki}^{\gamma\mu}(\mathbf{k},E_{\text{F}}+iy)\right\}\ . \tag{K.49}$$

Since the derivatives are local in site and orbital, as shown in Eq. K.47. σ and G are matrices in spin space, and the trace is taken over this degree of freedom.

where N is the total charge of the system and the Fermi level E_{F} is a new unknown to be found. It also changes when n_i or $\mathbf{m}_i$ is varied, i.e., $E_{\mathrm{F}} = E_{\mathrm{F}}(\{n_i, \mathbf{m}_i\})$.

The densities of the system can be calculated as

$$\rho_i^\alpha(\{n_i, \mathbf{m}_i\}, E_{\mathrm{F}}) = -\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\int_{-\infty}^{\infty} dE f\left(E - E_{\mathrm{F}}(\{n_i, \mathbf{m}_i\})\right) \operatorname{Im}\operatorname{Tr}\sigma^\alpha G_{ii}(\mathbf{k}, E + i\eta; \{n_i, \mathbf{m}_i\}) \ . \tag{K.50}$$

where $\alpha = 0, x, y, z$ and $f(E - E_{\mathrm{F}})$ is the Fermi-Dirac distribution. For $T = 0K$, this equation can be calculated as shown in Eq. K.31.

Taking the derivative with respect to the densities (charge or magnetization)

$$\begin{aligned}\frac{\partial \rho_i^\alpha(\{n_i, \mathbf{m}_i\}, E_{\mathrm{F}})}{\partial \rho_j^\beta} = &-\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\int_{-\infty}^{\infty} dE f\left(E - E_{\mathrm{F}}(\{n_i, \mathbf{m}_i\})\right) \operatorname{Im}\operatorname{Tr}\sigma^\alpha \frac{\partial G_{ii}(\mathbf{k}, E + i\eta; \{n_i, \mathbf{m}_i\})}{\partial \rho_j^\beta} \\ &-\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\int_{-\infty}^{\infty} dE \frac{\partial f\left(E - E_{\mathrm{F}}(\{n_i, \mathbf{m}_i\})\right)}{\partial \rho_j^\beta} \operatorname{Im}\operatorname{Tr}\sigma^\alpha G_{ii}(\mathbf{k}, E + i\eta; \{n_i, \mathbf{m}_i\}) \ .\end{aligned} \tag{K.51}$$

For $T = 0K$, the first term recovers Eq. (K.32). The second term can be written as

$$-\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\int_{-\infty}^{\infty} dE \frac{\partial f\left(E - E_{\mathrm{F}}\right)}{\partial E_{\mathrm{F}}}\frac{\partial E_{\mathrm{F}}(\{n_i, \mathbf{m}_i\})}{\partial \rho_j^\beta} \operatorname{Im}\operatorname{Tr}\sigma^\alpha G_{ii}(\mathbf{k}, E + i\eta; \{n_i, \mathbf{m}_i\}) \ . \tag{K.52}$$

Taking $T = 0K$, $\frac{\partial f(E - E_{\mathrm{F}})}{\partial E_{\mathrm{F}}} = -\frac{\partial f(E - E_{\mathrm{F}})}{\partial \epsilon} = \delta(E - E_{\mathrm{F}})$, and this term becomes

$$-\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\frac{\partial E_{\mathrm{F}}(\{n_i, \mathbf{m}_i\})}{\partial \rho_j^\beta} \operatorname{Im}\operatorname{Tr}\sigma^\alpha G_{ii}(\mathbf{k}, E_{\mathrm{F}} + i\eta; \{n_i, \mathbf{m}_i\}) \ . \tag{K.53}$$

The derivative of the Fermi level can be obtained from the last equation in Eq. (K.38)

$$\sum_i \frac{\partial n_i(\{n_i, \mathbf{m}_i\}, E_{\mathrm{F}})}{\partial \rho_j^\beta} = 0 \tag{K.54}$$

Which results in

$$\begin{aligned}\sum_i \frac{\partial n_i(\{n_i, \mathbf{m}_i\}, E_{\mathrm{F}})}{\partial \rho_j^\beta} = &-\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_i\int_{-\infty}^{\infty} dE f\left(E - E_{\mathrm{F}}(\{n_i, \mathbf{m}_i\})\right) \operatorname{Im}\operatorname{Tr}\frac{\partial G_{ii}(\mathbf{k}, E + i\eta; \{n_i, \mathbf{m}_i\})}{\partial \rho_j^\beta} \\ &-\frac{1}{\pi}\frac{1}{N}\sum_{\mathbf{k}}\sum_i\int_{-\infty}^{\infty} dE \frac{\partial f\left(E - E_{\mathrm{F}}(\{n_i, \mathbf{m}_i\})\right)}{\partial \rho_j^\beta} \operatorname{Im}\operatorname{Tr} G_{ii}(\mathbf{k}, E + i\eta; \{n_i, \mathbf{m}_i\}) = 0 \ .\end{aligned} \tag{K.55}$$

Appendix L

Units

Here we are going to list the units of all the quantities calculated in this work. We will use the following notation for the units:

$$\begin{aligned} E &= \text{energy} \\ T &= \text{time} \\ S &= \text{distance} \\ M &= \text{mass} \\ C &= \text{charge} \\ V &= \text{voltage} \\ B &= \text{magnetic field} \end{aligned} \quad . \tag{L.1}$$

For example: $E = MS^2T^{-2}$.

Time-dependent monoeletronic Green function (Eq. (2.1):

$$[G(t)] = [\hbar^{-1}] = E^{-1}T^{-1} \ . \tag{L.2}$$

Energy-dependent monoeletronic Green function (Eq. (2.10)):

$$[G(\omega)] = [(\hbar\omega)^{-1}] = E^{-1} \ . \tag{L.3}$$

Time-dependent susceptibility (Eq. (3.1)):

$$[\chi(t)] = [\hbar^{-1}] = E^{-1}T^{-1} \ . \tag{L.4}$$

Note that $c(t)$ is adimensional and we are not including $\hbar$ in the spin operator.

Energy-dependent susceptibility (e.g., Eq. (3.89)):

$$[\chi(\omega)] = [\hbar\omega][G(\omega)][G(\omega)] = E^{-1} \ . \tag{L.5}$$

The results above are consistent with the Fourier transform defined in Eq. (5.113).

Hoppings in reciprocal and real space (Eq. (5.80)):

$$[t_{ij}^{\mu\nu}(\mathbf{k}_\parallel)] = [t_{ij}^{\mu\nu}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)] = E \tag{L.6}$$

Derivative of the $\mathbf{k}_\parallel$-space hopping with respect to $\mathbf{k}_\parallel$ (Eq. (A.14)):

$$[\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t_{ij}^{\mu\nu}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{e}}^\alpha] = [(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)][t_{ij}^{\mu\nu}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)] = SE \tag{L.7}$$

Electric field:

$$[eE_0] = ES^{-1} \tag{L.8}$$

Spin disturbance due to an electric field (Eq. (5.321)):

$$\begin{aligned}[\delta\langle \hat{S}_{\ell_1}^m(\mathbf{q}_\parallel, t)\rangle] =&[e][E_0][(\hbar\omega)^{-1}][\mathcal{D}_{\ell_1}^m(\omega)] \\ =&[eE_0][(\hbar\omega)^{-1}][\chi_{\ell_1\ell_1\ell\ell'}^{m\sigma\;\mu\mu\gamma\xi}(\mathbf{k}_\parallel,\omega)][\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t_{\ell\ell'}^{\gamma\xi}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E] \\ =&(CVS^{-1})E^{-1}E^{-1}SE = (ES^{-1})E^{-1}E^{-1}SE \\ =&1\end{aligned} \tag{L.9}$$

Since we are not including $\hbar$ in the spin operator, the spin disturbance is adimensional. If we plot in units of eE_0, we have

$$\left[\frac{\delta\langle \hat{S}_{\ell_1}^m(\mathbf{q}_\parallel, t)\rangle}{eE_0}\right] = E^{-1}S \tag{L.10}$$

In our calculation, this unit corresponds to Å/ Ry. If we want to write in Hartree atomic units, we can write $0.529\text{Å} = a_0$, where a_0 is the Bohr radius, and $1\text{Ry} = 13.6\text{eV} = 0.5\text{H}$ (H - Hartree). Therefore, $1\text{Å}/\text{Ry} = \frac{2}{0.529}$ a.u. $= 3.78$ a.u..

If we multiply this by $\hbar\omega$, we have

$$\left[\hbar\omega\frac{\delta\langle \hat{S}_{\ell_1}^m(\mathbf{q}_\parallel, t)\rangle}{eE_0}\right] = [\mathcal{D}_{\ell_1}^m(\omega)] = E^{-1}SE = S \tag{L.11}$$

This is calculated in Å, so to write in a.u. units, we just need to divide by 0.529.

The magnetic moment is obtained by multiplying the spin by $g\mu_B = 2\mu_B$ (which is 1 in atomic units).

To calculate the units of currents, we need

$$\begin{aligned}[\mathcal{J}_{\ell_1}^m(\omega)] =& \left[\frac{\hbar}{e}\frac{1}{eE_0}\hbar\omega\langle \hat{I}_{\ell_1}^{\mathrm{C}}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel, t)\rangle\right] \\ =&[t_{ij}^{\mu\nu}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel)][\chi_{\ell_1\ell_1\ell\ell'}^{m\sigma\;\mu\mu\gamma\xi}(\mathbf{k}_\parallel,\omega)][\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t_{\ell\ell'}^{\gamma\xi}(\mathbf{k}_\parallel) \cdot \hat{\mathbf{u}}_E] \\ =&EE^{-1}SE \\ =&SE\end{aligned} \tag{L.12}$$

This is the quantity calculated in the program, in units of Ry.Å. On the program, we calculate

$$\begin{aligned}\left[\frac{1}{\hbar\omega}\mathcal{J}^{m}_{\ell_1}(\omega)\right] &= \left[\frac{\hbar}{e}\frac{1}{eE_0}\langle\hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel,t)\rangle\right]\\ &=S\end{aligned} \tag{L.13}$$

Charge current due to an electric field (Eq. (5.251)):

$$\begin{aligned}[\langle\hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel,t)\rangle] =&[e^2][E_0][\hbar^{-1}][(\hbar\omega)^{-1}][\mathcal{J}^{m}_{\ell_1}(\omega)]\\ =&[e\hbar^{-1}][eE_0][(\hbar\omega)^{-1}][\mathcal{J}^{m}_{\ell_1}(\omega)]\\ =&CE^{-1}T^{-1}(CVS^{-1})E^{-1}SE = CE^{-1}T^{-1}(ES^{-1})E^{-1}SE\\ =&CT^{-1}\end{aligned} \tag{L.14}$$

In units of eE_0 (conductance),

$$\begin{aligned}\left[\frac{\langle\hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel,t)\rangle}{eE_0}\right] =&[e][\hbar^{-1}][(\hbar\omega)^{-1}][\mathcal{J}^{m}_{\ell_1}(\omega)]\\ =&[e\hbar^{-1}][(\hbar\omega)^{-1}][t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)][\chi^{m\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)][\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E]\\ =&CE^{-1}T^{-1}E^{-1}EE^{-1}SE\\ =&T^{-1}V^{-1}S = CT^{-1}E^{-1}S\end{aligned} \tag{L.15}$$

Conductance in units of $e/\hbar$,

$$\begin{aligned}\left[\frac{\langle\hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel,t)\rangle}{eE_0}\right] =&[e\hbar^{-1}][(\hbar\omega)^{-1}][t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)][\chi^{m\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)][\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E]\\ =&[e\hbar^{-1}]E^{-1}EE^{-1}SE = S[e\hbar^{-1}]\end{aligned} \tag{L.16}$$

Multiplying by $\hbar\omega$

$$\begin{aligned}\left[\hbar\omega\frac{\langle\hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel,t)\rangle}{eE_0}\right] =&[e][\hbar^{-1}][\mathcal{J}^{m}_{\ell_1}(\omega)]\\ =&[e\hbar^{-1}][t^{\mu\nu}_{ij}(\mathbf{R}'_\parallel-\mathbf{R}_\parallel)][\chi^{m\sigma\,\mu\mu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)][\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E]\\ =&CE^{-1}T^{-1}EE^{-1}SE\\ =&T^{-1}V^{-1}SE = CT^{-1}S\end{aligned} \tag{L.17}$$

Taking into account a transversal cross section of the material, A, the conductivity of a film with N_ℓ layers is given by

$$\sigma^{\mathrm{C}} = \frac{1}{A}\sum_{\mathbf{R}'}\sum_{\ell_1}\frac{\langle\hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel,\mathbf{R}'_\parallel,t)\rangle}{E_0} \tag{L.18}$$

where the sum over $\mathbf{R}'$ covers sites only in one direction (to where the conductivity is to be calculated). For a bcc(110) slab with N layers, for example, where the area of the unit cell in each layer is $\frac{a^2}{\sqrt{2}}$ (a is the lattice parameter),

$$\sigma^{\mathrm{C}} = \frac{\sqrt{2}}{Na^2}\sum_{\mathbf{R}'}\sum_{\ell_1}\frac{\langle \hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel, t)\rangle}{E_0} \tag{L.19}$$

In the program, we calculate

$$\begin{aligned}\frac{\hbar}{e^2}\hbar\omega\sigma^{\mathrm{C}} =&\frac{\sqrt{2}}{Na^2}\sum_{\mathbf{R}'}\sum_{\ell_1}\frac{\hbar}{e}\frac{1}{eE_0}\hbar\omega\langle \hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel, t)\rangle\\ =&\frac{\sqrt{2}}{Na^2}\sum_{\mathbf{R}'}\sum_{\ell_1}\mathcal{J}^m_{\ell_1}(\omega)\end{aligned} \tag{L.20}$$

which gives the conductivity multiplied by the energy (frequency) in units of $\frac{e^2}{\hbar}$ The units are given by

$$\begin{aligned}\left[\frac{\hbar}{e^2}\hbar\omega\sigma^{\mathrm{C}}\right] =&[a^{-2}][\mathcal{J}^m_{\ell_1}(\omega)]\\ =&S^{-2}S.E = S^{-1}E\end{aligned} \tag{L.21}$$

This result is in Ry.Å^{-1}. Multiplying by 13.6 we have the result in eV.Å^{-1}. We can also use once again the conversion 1Å/Ry $= \frac{2}{0.529}$ a.u. $= 3.78$ a.u. to obtain the result in atomic units.

If we calculate the ratio $\frac{\delta\langle \hat{S}^m_{\ell_1}(\mathbf{q}_\parallel, t)\rangle}{\langle \hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel, t)\rangle}$, we have

$$\begin{aligned}\left[\frac{\delta\langle \hat{S}^m_{\ell_1}(\mathbf{q}_\parallel, t)\rangle}{\langle \hat{I}^{\mathrm{C}}_{\ell_1}(\mathbf{R}_\parallel, \mathbf{R}'_\parallel, t)\rangle}\right] =& \left[\frac{\hbar}{e}\right]\left[\frac{|\mathcal{D}^m_{\ell_1}(\omega)|}{|\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega)|}\right]\\ =&ETC^{-1}E^{-1}\\ =&C^{-1}T\end{aligned} \tag{L.22}$$

This already removes the dependency on E_0 and the divergence with ω^{-1}.

If instead we define the coefficient between the amplitude of oscillation and the charge current density,

$$\begin{aligned}\left[\frac{\delta\langle \hat{S}^m_{\ell_1}(\mathbf{q}_\parallel, t)\rangle}{j^{\mathrm{C}}}\right] =& \left[\frac{\delta\langle \hat{S}^m_{\ell_1}(\mathbf{q}_\parallel, t)\rangle}{\sigma^{\mathrm{C}}\cdot E_0}\right]\\ =& \left[\frac{\hbar}{e}\right]\left[\hbar\omega\frac{\delta\langle \hat{S}^m_{\ell_1}(\mathbf{q}_\parallel, t)\rangle}{eE_0}\right]\left[\frac{e^2}{\hbar}\frac{1}{\hbar\omega\sigma^{\mathrm{C}}\cdot}\right]\\ =&ETC^{-1}SSE^{-1}\\ =&TC^{-1}S^2\end{aligned} \tag{L.23}$$

Effective field (Eq. (5.347)): the effective field calculated in the program is given by

$$\begin{aligned}\left[g_S\mu_B\frac{\hbar\omega}{eE_0}\mathcal{B}(\omega)\right] =&[\chi^{-1}(\omega)][\mathcal{D}(\omega)] \\ =&ES\end{aligned}\,, \tag{L.24}$$

where we have used the units of χ and $\mathcal{D}$ given by Eqs. L.5 and L.11. Multiplying by all the prefactors, we have

$$\begin{aligned}[\mathcal{B}(\omega)] =&[(g_S\mu_B)^{-1}][eE_0][(\hbar\omega)^{-1}][\chi^{-1}(\omega)][\mathcal{D}(\omega)] \\ =&E^{-1}B(ES^{-1})E^{-1}ES \\ =&B\end{aligned}\,, \tag{L.25}$$

as it should be.

Spin-Orbit Torques (Eq. (5.354)): the units of the torque caused by the spin-orbit interaction calculated in the program is

$$\begin{aligned}\frac{\delta\langle\hat{\tau}^m_{\ell_1}(t)\rangle}{eE_0} &= \left[\frac{\lambda_{\ell_1}}{\hbar\omega}\left|\mathcal{T}^m_{\ell_1}(\omega)\right|\right] \\ &= [\lambda_{\ell_1}]\left[(\hbar\omega)^{-1}\right]\left[L'^m_{\mu\nu}\chi^{\sigma\sigma'\ \mu\nu\gamma\xi}_{\ell_1\ell_1\ell\ell'}(\mathbf{k}_\parallel,\omega)\boldsymbol{\nabla}_{\mathbf{k}_\parallel}t^{\gamma\xi}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\right] \\ &=E.E^{-1}.E^{-1}.SE = S\end{aligned}\,, \tag{L.26}$$

Appendix M

Magnetic interactions from the force theorem

We start rewriting our ground state hamiltonian given by Eq. (1.37) as

$$\hat{H}_{ij}^{\mu\nu} = H_{ij}^{0\,\mu\nu}\sigma^0 + \boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_i B_i^{\mathrm{xc}\,\mu\nu}\delta_{ij} + \boldsymbol{\sigma}\cdot\mathbf{B}_i^{\mathrm{soc}\,\mu\nu}\delta_{ij}\ . \tag{M.1}$$

where $B_i^{\mathrm{xc}\,\mu\nu} = -\delta_{\mu\nu}\frac{U^i}{2}m_i$, with μ and ν restricted to the d orbitals (where $U^i \neq 0$). Here, $\hat{\mathbf{e}}_i = \frac{\mathbf{m}_i}{m_i}$ is the direction of the magnetization.

M.1 Heisenberg model

We can map our hamiltonian into a generalized Heisenberg model, described by

$$E = -\frac{1}{2}\sum_{ij}\hat{\mathbf{e}}_i\cdot\overline{\mathcal{J}}_{ij}\cdot\hat{\mathbf{e}}_j - \sum_i \mathbf{m}_i\cdot\mathbf{B}_i^{\mathrm{ext}}\ , \tag{M.2}$$

where $\hat{\mathbf{e}}_i = \frac{\mathbf{m}_i}{m_i}$. To simplify the following calculations, we can rewrite it assuming a constant magnetization length as

$$E = -\frac{1}{2}\sum_{ij}\mathbf{m}_i\cdot\mathcal{J}_{ij}\cdot\mathbf{m}_j - \sum_i \mathbf{m}_i\cdot\mathbf{B}_i^{\mathrm{ext}}\ , \tag{M.3}$$

where $\overline{\mathcal{J}}_{ij} = m_i m_j \mathcal{J}_{ij}$ are 3×3 matrices in real space (x, y, z). From Eq. (M.5), the exchange interaction tensor can be obtained by

$$\begin{aligned}
\mathcal{J}_{ij} =& -\frac{\partial^2 E}{\partial\mathbf{m}_j\partial\mathbf{m}_i}\\
=&\frac{1}{2}\begin{pmatrix} J_{ij}^{xx}+J_{ji}^{xx} & J_{ij}^{xy}+J_{ji}^{yx} & J_{ij}^{xz}+J_{ji}^{zx}\\ J_{ij}^{yx}+J_{ji}^{xy} & J_{ij}^{yy}+J_{ji}^{yy} & J_{ij}^{yz}+J_{ji}^{zy}\\ J_{ij}^{zy}+J_{ji}^{yz} & J_{ij}^{zy}+J_{ji}^{yz} & J_{ij}^{zz}+J_{ji}^{zz}\end{pmatrix}\\
=&\begin{pmatrix} J_{ij}^{xx} & J_{ij}^{xy} & J_{ij}^{xz}\\ J_{ij}^{yx} & J_{ij}^{yy} & J_{ij}^{yz}\\ J_{ij}^{zy} & J_{ij}^{zy} & J_{ij}^{zz}\end{pmatrix}\ ,
\end{aligned} \tag{M.4}$$

where we have used the fact that the tensor can be chosen such as $\mathcal{J}_{ij} = \mathcal{J}^t_{ji}$.

The anisotropy term can also be obtained from the on-site part of the tensor $\overline{\mathcal{J}}$ as $\mathbf{K}_i = \overline{\mathcal{J}}_{ii}/2$, with the convention

$$E = -\sum_i \hat{\mathbf{e}}_i \cdot \mathbf{K}_i \cdot \hat{\mathbf{e}}_i \ . \tag{M.5}$$

With this convention, $K_i^{\alpha\alpha} > 0$ reduces the energy and the axis α is an easy axis.

M.2 Landau-Lifshitz-Gilbert equation

We can describe the magnetization precession in the Heisenberg model using Landau-Lifshitz-Gilbert phenomenological equation,

$$\dot{\mathbf{m}}_i = -\gamma \mathbf{m}_i \times \mathbf{B}_i^{\text{eff}} + \alpha \mathbf{m}_i \times \dot{\mathbf{m}}_i. \tag{M.6}$$

The effective field can be obtained as

$$\begin{aligned} \mathbf{B}_k^{\text{eff}} &= -\frac{\partial H}{\partial \mathbf{m}_k} \\ &= -\frac{\partial H}{\partial m_k^x}\hat{\mathbf{x}} - \frac{\partial H}{\partial m_k^y}\hat{\mathbf{y}} - \frac{\partial H}{\partial m_k^z}\hat{\mathbf{z}} \\ &= \sum_j \mathcal{J}_{kj}\mathbf{m}_j + \mathbf{B}_k^{\text{ext}} \ . \end{aligned} \tag{M.7}$$

Now we choose a local spin coordinate system such that, in a collinear system (where $\mathbf{B}_i^{\text{ext}} = B_i \hat{\mathbf{z}}$),

$$\begin{aligned} \mathbf{m}_i(t) =& M_i\hat{\mathbf{z}} + m_i^x(t)\hat{\mathbf{x}} + m_i^y(t)\hat{\mathbf{y}} \\ \mathbf{B}_i^{\text{ext}}(t) =& B_i\hat{\mathbf{z}} + b_i^x(t)\hat{\mathbf{x}} + b_i^y(t)\hat{\mathbf{y}} \ . \end{aligned} \tag{M.8}$$

In the ground state,

$$\begin{aligned} M_i\hat{\mathbf{z}} \times \mathbf{B}_i^{\text{eff}} = 0 \\ -M_i B_i^{\text{eff}^y}\hat{\mathbf{x}} + M_i B_i^{\text{eff}^x}\hat{\mathbf{y}} = 0 \end{aligned} \tag{M.9}$$

In linear response theory, $\dot{M}_i = 0$ and we have

$$\begin{aligned} \dot{m}_i^x =& -\gamma\left[m_i^y(t)B_i - M_i b_i^y(t)\right] - \alpha M_i \dot{m}_i^y(t) \\ \dot{m}_i^y =& -\gamma\left[M_i b_i^x(t) - m_i^x(t)B_i\right] + \alpha M_i \dot{m}_i^x(t) \ . \end{aligned} \tag{M.10}$$

M.3 Liechtenstein Formula

From Eq. (M.4), we see that to obtain the exchange interaction tensor we need to calculate how the energy changes when we rotate the magnetization. The band energy of our system, for $T = 0K$, can be calculated as

$$\begin{aligned} E &= \int_{-\infty}^{E_\mathrm{F}} dE (E - E_\mathrm{F}) \rho(E) \\ &= -\frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int_{-\infty}^{E_\mathrm{F}} dE (E - E_\mathrm{F}) \sum_m G_{mm}(E). \end{aligned} \tag{M.11}$$

Taking derivatives of the energy with respect to the magnetization,

$$\begin{aligned} \frac{\partial E}{\partial \mathbf{m}_i} &= -\frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int_{-\infty}^{E_\mathrm{F}} dE (E - E_\mathrm{F}) \sum_m \frac{\partial G_{mm}(E)}{\partial \mathbf{m}_i} \\ &= -\frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int_{-\infty}^{E_\mathrm{F}} dE (E - E_\mathrm{F}) \sum_{mj} G_{mj}(E) \left[\frac{\partial H}{\partial \mathbf{m}_i} \right]_{jj} G_{jm}(E) \\ &= -\frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int_{-\infty}^{E_\mathrm{F}} dE (E - E_\mathrm{F}) \sum_{mj} G_{jm}(E) G_{mj}(E) \left[\frac{\partial H}{\partial \mathbf{m}_i} \right]_{ii} \delta_{ij} \\ &= -\frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int_{-\infty}^{E_\mathrm{F}} dE (E - E_\mathrm{F}) \left[-\frac{\partial G_{ii}(E)}{\partial E} \right] \left[B_i^{\mathrm{xc}} \otimes \frac{\partial \left(\boldsymbol{\sigma} \cdot \hat{\mathbf{e}}_i \right)}{\partial \mathbf{m}_i} \right]. \end{aligned} \tag{M.12}$$

Integrating by parts,

$$\begin{aligned} \frac{\partial E}{\partial \mathbf{m}_i} &= \frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \left\{ (E - E_\mathrm{F}) G_{ii}(E) \Big|_{-\infty}^{E_\mathrm{F}} \left[B_i^{\mathrm{xc}} \otimes \frac{\partial \left(\boldsymbol{\sigma} \cdot \hat{\mathbf{e}}_i \right)}{\partial \mathbf{m}_i} \right] \right. \\ &\qquad \left. - \int_{-\infty}^{E_\mathrm{F}} dE G_{ii}(E) \left[B_i^{\mathrm{xc}} \otimes \frac{\partial \left(\boldsymbol{\sigma} \cdot \hat{\mathbf{e}}_i \right)}{\partial \mathbf{m}_i} \right] \right\} \\ &= -\frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int_{-\infty}^{E_\mathrm{F}} dE G_{ii}(E) \left[B_i^{\mathrm{xc}} \otimes \frac{\partial \left(\boldsymbol{\sigma} \cdot \hat{\mathbf{e}}_i \right)}{\partial \mathbf{m}_i} \right]. \end{aligned} \tag{M.13}$$

The second derivative of the energy with respect to the magnetization is taken relative to site j (that may be the same as i, which gives rise to the anisotropy term),

$$\begin{aligned} \frac{\partial^2 E}{\partial \mathbf{m}_j \partial \mathbf{m}_i} &= -\frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int_{-\infty}^{E_\mathrm{F}} dE \frac{\partial}{\partial \mathbf{m}_j} \left\{ G_{ii}(E) \left[B_i^{\mathrm{xc}} \otimes \frac{\partial \left(\boldsymbol{\sigma} \cdot \hat{\mathbf{e}}_i \right)}{\partial \mathbf{m}_i} \right] \right\} \\ &= -\frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int_{-\infty}^{E_\mathrm{F}} dE \left\{ \frac{\partial G_{ii}(E)}{\partial \mathbf{m}_j} \left[B_i^{\mathrm{xc}} \otimes \frac{\partial \left(\boldsymbol{\sigma} \cdot \hat{\mathbf{e}}_i \right)}{\partial \mathbf{m}_i} \right] + G_{ii}(E) \left[B_i^{\mathrm{xc}} \otimes \frac{\partial^2 \left(\boldsymbol{\sigma} \cdot \hat{\mathbf{e}}_i \right)}{\partial \mathbf{m}_j \partial \mathbf{m}_i} \right] \right\} \\ &= -\frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int_{-\infty}^{E_\mathrm{F}} dE \left\{ G_{ij}(E) \left[B_j^{\mathrm{xc}} \otimes \frac{\partial \left(\boldsymbol{\sigma} \cdot \hat{\mathbf{e}}_j \right)}{\partial \mathbf{m}_j} \right] G_{ji}(E) \left[B_i^{\mathrm{xc}} \otimes \frac{\partial \left(\boldsymbol{\sigma} \cdot \hat{\mathbf{e}}_i \right)}{\partial \mathbf{m}_i} \right] \right. \\ &\qquad \left. + G_{ii}(E) \left[B_i^{\mathrm{xc}} \otimes \frac{\partial^2 \left(\boldsymbol{\sigma} \cdot \hat{\mathbf{e}}_i \right)}{\partial \mathbf{m}_i^2} \right] \delta_{ij} \right\} \\ &= -\frac{1}{\pi} \operatorname{Im} \operatorname{Tr} \int_{-\infty}^{E_\mathrm{F}} dE \left\{ \left[B_i^{\mathrm{xc}} \otimes \frac{\partial \left(\boldsymbol{\sigma} \cdot \hat{\mathbf{e}}_i \right)}{\partial \mathbf{m}_i} \right] G_{ij}(E) \left[B_j^{\mathrm{xc}} \otimes \frac{\partial \left(\boldsymbol{\sigma} \cdot \hat{\mathbf{e}}_j \right)}{\partial \mathbf{m}_j} \right] G_{ji}(E) \right. \\ &\qquad \left. + G_{ii}(E) \left[B_i^{\mathrm{xc}} \otimes \frac{\partial^2 \left(\boldsymbol{\sigma} \cdot \hat{\mathbf{e}}_i \right)}{\partial \mathbf{m}_i^2} \right] \delta_{ij} \right\}. \end{aligned} \tag{M.14}$$

where we have used the cyclic property of the trace and the identity

$$
\begin{aligned}
(E-H)G =& 1 \\
\frac{\partial\left[(E-H)G\right]}{\partial \mathbf{m}_j} =& 0 \\
-\frac{\partial H}{\partial \mathbf{m}_j}G + (E-H)\frac{\partial G}{\partial \mathbf{m}_j} =& 0 \\
\frac{\partial G}{\partial \mathbf{m}_j} =& G\frac{\partial H}{\partial \mathbf{m}_j}G \\
\frac{\partial G_{ii}}{\partial \mathbf{m}_j} =& \sum_m G_{im}\left[\frac{\partial H}{\partial \mathbf{m}_j}\right]_{mm} G_{mi} \\
\frac{\partial G_{ii}}{\partial \mathbf{m}_j} =& G_{ij}\left[\frac{\partial H}{\partial \mathbf{m}_j}\right]_{jj} G_{ji}
\end{aligned}
\tag{M.15}
$$

Therefore, the exchange interaction tensor can be calculated as

$$
\begin{aligned}
\mathcal{J}_{ij} = \frac{1}{\pi}\,\mathrm{Im}\,\mathrm{Tr}\int_{-\infty}^{E_\mathrm{F}} dE \Bigg\{ &\left[B_i^{\mathrm{xc}} \otimes \frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_i\right)}{\partial \mathbf{m}_i}\right] G_{ij}(E) \left[B_j^{\mathrm{xc}} \otimes \frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_j\right)}{\partial \mathbf{m}_j}\right] G_{ji}(E) \\
&+ G_{ii}(E)\left[B_i^{\mathrm{xc}} \otimes \frac{\partial^2\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_i\right)}{\partial \mathbf{m}_i^2}\right]\delta_{ij}\Bigg\}.
\end{aligned}
\tag{M.16}
$$

Note that, since B^{xc} is diagonal over orbitals, the first term can be rewritten by expliciting the orbital trace (but keeping the spin trace) as

$$
\begin{aligned}
J_{ij}^{\alpha\beta} =& \frac{U_iU_j\langle m_i\rangle\langle m_j\rangle}{4\pi}\,\mathrm{Im}\sum_{\mu\nu}\int_{-\infty}^{E_\mathrm{F}} dE\,\mathrm{tr}\left\{\left[\frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_i\right)}{\partial \mathbf{m}_i}\right]_\alpha G_{ij}^{\mu\nu}(E)\left[\frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_j\right)}{\partial \mathbf{m}_j}\right]_\beta G_{ji}^{\nu\mu}(E)\right\} \\
=& -U_iU_j\sum_{\mu\nu}\chi_{(0)ij}^{\alpha\beta\,\mu\nu}(\omega=0),
\end{aligned}
\tag{M.17}
$$

where we have used the Hartree-Fock susceptibility at $\omega = 0$ defined in Eq. (C.43), and α and β are the transverse directions defined by $\hat{\mathbf{e}}_i$ (see Eqs. M.28 and M.30 below).

Substituting Eq. (M.28),

$$
\begin{aligned}
J_{ij}^{\alpha\beta} =& \frac{U_iU_j}{4\pi}\,\mathrm{Im}\sum_{\mu\nu}\int_{-\infty}^{E_\mathrm{F}} dE\,\mathrm{tr}\left\{\left[\boldsymbol{\sigma} - \left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)_i\hat{\mathbf{e}}_i\right]_\alpha G_{ij}^{\mu\nu}(E)\left[\boldsymbol{\sigma} - \left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_j\right)\hat{\mathbf{e}}_j\right]_\beta G_{ji}^{\nu\mu}(E)\right\} \\
=& -U_iU_j\sum_{\mu\nu}\chi_{(0)ij}^{\alpha\beta\,\mu\nu}(\omega=0),
\end{aligned}
\tag{M.18}
$$

For the second term, we have

$$\begin{aligned}\mathcal{J}_i =& \frac{1}{\pi}\,\mathrm{Im}\,\mathrm{Tr}\int_{-\infty}^{E_{\mathrm{F}}} dE G_{ii}(E)\left[B_i^{\mathrm{xc}}\otimes\frac{\partial^2\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_i\right)}{\partial\mathbf{m}_i^2}\right] \\ =& \frac{1}{\pi}\,\mathrm{Im}\,\mathrm{tr}\sum_{\mu\nu}\int_{-\infty}^{E_{\mathrm{F}}} dE G_{ii}^{\mu\nu}(E)B_i^{\mathrm{xc}\,\nu\mu}\left[\frac{\partial^2\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_i\right)}{\partial\mathbf{m}_i^2}\right] \\ =& -\frac{U_i\langle m_i\rangle}{2\pi}\,\mathrm{Im}\,\mathrm{tr}\sum_{\mu}\int_{-\infty}^{E_{\mathrm{F}}} dE G_{ii}^{\mu\mu}(E)\left[\frac{\partial^2\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_i\right)}{\partial\mathbf{m}_i^2}\right]\ .\end{aligned} \tag{M.19}$$

where tr represents the trace in spin space. The second derivative tensor will be calculated below (Eq. (M.29)), and each of its components is related to a Pauli matrix (or zero). In general, using Eq. (M.29), we can write

$$\begin{aligned}\mathcal{J}_i^{\alpha\beta} =& -\frac{U_i\langle m_i\rangle}{2}\frac{1}{\langle m_i\rangle^2}\left[\hat{\mathbf{e}}_{\mathbf{i}}\otimes\mathbf{m}_i+\mathbf{m}_i\otimes\hat{\mathbf{e}}_{\mathbf{i}}+\left(\mathbf{m}_i\cdot\hat{\mathbf{e}}_{\mathbf{i}}\right)\mathbf{1}-3\left(\mathbf{m}_i\cdot\hat{\mathbf{e}}_{\mathbf{i}}\right)\hat{\mathbf{e}}_{\mathbf{i}}\otimes\hat{\mathbf{e}}_{\mathbf{i}}\right] \\ =& -\frac{U_i}{2\langle m_i\rangle^2}\left[\mathbf{m}_i\otimes\mathbf{m}_i+\mathbf{m}_i\otimes\mathbf{m}_i+\left(\mathbf{m}_i\cdot\mathbf{m}_i\right)\mathbf{1}-3\left(\mathbf{m}_i\cdot\mathbf{m}_i\right)\mathbf{m}_i\otimes\mathbf{m}_i\right]\ .\end{aligned} \tag{M.20}$$

Extending the integration over the upper complex plane, $E\to z=E+iy$, we can use the usual contour on the imaginary axis $z=E_{\mathrm{F}}+iy\Rightarrow dz=idy$ to obtain

$$\begin{aligned}\mathcal{J}_{ij} = -\frac{1}{\pi}\,\mathrm{Re}\,\mathrm{Tr}\int_{\eta}^{\infty} dy \Bigg\{&\left[B_i^{\mathrm{xc}}\otimes\frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_i\right)}{\partial\mathbf{m}_i}\right]G_{ij}(E_{\mathrm{F}}+iy)\left[B_j^{\mathrm{xc}}\otimes\frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_j\right)}{\partial\mathbf{m}_j}\right]G_{ji}(E_{\mathrm{F}}+iy) \\ &+G_{ii}(E_{\mathrm{F}}+iy)\left[B_i^{\mathrm{xc}}\otimes\frac{\partial^2\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_i\right)}{\partial\mathbf{m}_i^2}\right]\delta_{ij}\Bigg\}.\end{aligned} \tag{M.21}$$

The contour over $R\to\infty$ does not contribute because the first term involves a product of 2 Green functions and the second term has a trace of Pauli matrices.

For multilayers, we can take into account the symmetry and calculate $\mathcal{J}_{\ell\ell'}(\mathbf{q})$ as

$$\begin{aligned}\mathcal{J}_{\ell\ell'}(\mathbf{q}) =& \sum_{\mathbf{R}} e^{i\mathbf{q}\cdot\mathbf{R}}\mathcal{J}_{\ell\ell'}(\mathbf{R}) \\ =& -\frac{1}{\pi}\sum_{\mathbf{R}} e^{i\mathbf{q}\cdot\mathbf{R}}\,\mathrm{Re}(A_{\ell\ell'}) \\ =& -\frac{1}{\pi}\sum_{\mathbf{R}} e^{i\mathbf{q}\cdot\mathbf{R}}\frac{1}{2}\left(A_{\ell\ell'}+A_{\ell\ell'}^{\dagger}\right) \\ =& -\frac{1}{2\pi}\sum_{\mathbf{R}}\left\{e^{i\mathbf{q}\cdot\mathbf{R}}A_{\ell\ell'}+(e^{-i\mathbf{q}\cdot\mathbf{R}}A_{\ell\ell'})^*\right\}\end{aligned} \tag{M.22}$$

where A_{ij} is given by the terms inside the real part in Eq. (M.21). The first term of the equation above can be written as:

$$
\begin{aligned}
\mathcal{J}^{(1)}_{\ell\ell'}(\mathbf{q}) = -\frac{1}{2\pi}\,\mathrm{Tr}\int_{\eta}^{\infty} dy \sum_{\mathbf{R}} e^{i\mathbf{q}\cdot\mathbf{R}} \Bigg\{ & \left[B^{\mathrm{xc}}_{\ell} \otimes \frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_{\ell}\right)}{\partial \mathbf{m}_{\ell}}\right] G_{\ell\ell'}(\mathbf{R}; E_{\mathrm{F}}+iy) \\
& \times \left[B^{\mathrm{xc}}_{\ell'} \otimes \frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_{\ell'}\right)}{\partial \mathbf{m}_{\ell'}}\right] G_{\ell'\ell}(-\mathbf{R}; E_{\mathrm{F}}+iy) \\
& + G_{\ell\ell}(0; E_{\mathrm{F}}+iy)\left[B^{\mathrm{xc}}_{\ell} \otimes \frac{\partial^2\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_{\ell}\right)}{\partial \mathbf{m}^2_{\ell}}\right]\delta_{\ell\ell'}\delta(\mathbf{R})\Bigg\} \\
= -\frac{1}{2\pi}\,\mathrm{Tr}\int_{\eta}^{\infty} dy \sum_{\mathbf{R}} e^{i\mathbf{q}\cdot\mathbf{R}} \Bigg\{ & \left[B^{\mathrm{xc}}_{\ell} \otimes \frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_{\ell}\right)}{\partial \mathbf{m}_{\ell}}\right] \frac{1}{N}\sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot\mathbf{R}} G_{\ell\ell'}(\mathbf{k}; E_{\mathrm{F}}+iy) \\
& \times \left[B^{\mathrm{xc}}_{\ell'} \otimes \frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_{\ell'}\right)}{\partial \mathbf{m}_{\ell'}}\right] \frac{1}{N}\sum_{\mathbf{k}'} e^{i\mathbf{k}'\cdot\mathbf{R}} G_{\ell'\ell}(\mathbf{k}'; E_{\mathrm{F}}+iy) \\
& + \frac{1}{N}\sum_{\mathbf{k}} G_{\ell\ell}(\mathbf{k}; E_{\mathrm{F}}+iy)\left[B^{\mathrm{xc}}_{\ell} \otimes \frac{\partial^2\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_{\ell}\right)}{\partial \mathbf{m}^2_{\ell}}\right]\delta_{\ell\ell'}\delta(\mathbf{R})\Bigg\} \ .
\end{aligned}
\tag{M.23}
$$

Since $\frac{1}{N}\sum_{\mathbf{R}} e^{i(\mathbf{q}-\mathbf{k}+\mathbf{k}')\cdot\mathbf{R}} = \delta(\mathbf{q}-\mathbf{k}+\mathbf{k}')$,

$$
\begin{aligned}
\mathcal{J}^{(1)}_{\ell\ell'}(\mathbf{q}) = -\frac{1}{2\pi}\,\mathrm{Tr}\int_{\eta}^{\infty} dy \frac{1}{N}\sum_{\mathbf{k}} \Bigg\{ & \left[B^{\mathrm{xc}}_{\ell} \otimes \frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_{\ell}\right)}{\partial \mathbf{m}_{\ell}}\right] G_{\ell\ell'}(\mathbf{k}; E_{\mathrm{F}}+iy) \\
& \times \left[B^{\mathrm{xc}}_{\ell'} \otimes \frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_{\ell'}\right)}{\partial \mathbf{m}_{\ell'}}\right] G_{\ell'\ell}(\mathbf{k}-\mathbf{q}; E_{\mathrm{F}}+iy) \\
& + G_{\ell\ell}(\mathbf{k}; E_{\mathrm{F}}+iy)\left[B^{\mathrm{xc}}_{\ell} \otimes \frac{\partial^2\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}_{\ell}\right)}{\partial \mathbf{m}^2_{\ell}}\right]\delta_{\ell\ell'}\Bigg\} \ .
\end{aligned}
\tag{M.24}
$$

The second term can be obtained from the same term, by changing $\mathbf{q} \to -\mathbf{q}$ and taking the complex conjugate, such that the final result is given by

$$
\mathcal{J}_{\ell\ell'}(\mathbf{q}) = \mathcal{J}^{(1)}_{\ell\ell'}(\mathbf{q}) + [\mathcal{J}^{(1)}_{\ell\ell'}(-\mathbf{q})]^* \tag{M.25}
$$

The derivatives of $\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}$ can be calculated taking into account that

$$
\begin{aligned}
\hat{\mathbf{e}} =& \frac{\mathbf{m}}{m} \\
=& \frac{m_x\hat{\mathbf{x}} + m_y\hat{\mathbf{y}} + m_z\hat{\mathbf{z}}}{\sqrt{m_x^2+m_y^2+m_z^2}} \ ,
\end{aligned}
\tag{M.26}
$$

where we have omitted the site index for simplicity. Its derivative with respect to the magnetization vector can be written as

$$
\begin{aligned}
\frac{\partial \hat{\mathbf{e}}}{\partial \mathbf{m}} =& \frac{\partial \hat{\mathbf{e}}}{\partial m_x}\hat{\mathbf{x}} + \frac{\partial \hat{\mathbf{e}}}{\partial m_y}\hat{\mathbf{y}} + \frac{\partial \hat{\mathbf{e}}}{\partial m_z}\hat{\mathbf{z}} \\
=& \left[\frac{\hat{\mathbf{x}}}{m} - \hat{\mathbf{e}}\frac{m_x}{m^2}\right]\hat{\mathbf{x}} + \left[\frac{\hat{\mathbf{y}}}{m} - \hat{\mathbf{e}}\frac{m_y}{m^2}\right]\hat{\mathbf{y}} + \left[\frac{\hat{\mathbf{z}}}{m} - \hat{\mathbf{e}}\frac{m_z}{m^2}\right]\hat{\mathbf{z}} \\
=& \frac{1}{m}\left[\hat{\mathbf{x}}\otimes\hat{\mathbf{x}} + \hat{\mathbf{y}}\otimes\hat{\mathbf{y}} + \hat{\mathbf{z}}\otimes\hat{\mathbf{z}} - \hat{\mathbf{e}}\otimes\hat{\mathbf{e}}\right] \\
=& \frac{1}{m}\left[\mathbf{1} - \hat{\mathbf{e}}\otimes\hat{\mathbf{e}}\right] \ .
\end{aligned}
\tag{M.27}
$$

Therefore,

$$
\begin{aligned}
\frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)}{\partial \mathbf{m}} =& \boldsymbol{\sigma}\cdot\frac{\partial \hat{\mathbf{e}}}{\partial \mathbf{m}} \\
=& \frac{1}{m}\left[\boldsymbol{\sigma} - \left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)\hat{\mathbf{e}}\right].
\end{aligned}
\tag{M.28}
$$

The second derivative, needed for the anisotropy term, is given by

$$
\begin{aligned}
\frac{\partial^2\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)}{\partial \mathbf{m}^2} =& \frac{\partial}{\partial \mathbf{m}}\left(\frac{1}{m}\right)\otimes\left[\boldsymbol{\sigma} - \left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)\hat{\mathbf{e}}\right] - \frac{1}{m}\left[\boldsymbol{\sigma}\cdot\left(\frac{\partial \hat{\mathbf{e}}}{\partial \mathbf{m}}\right)\right]\otimes\hat{\mathbf{e}} - \frac{1}{m}\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)\frac{\partial \hat{\mathbf{e}}}{\partial \mathbf{m}} \\
=& -\frac{\mathbf{m}}{m^3}\otimes\left[\boldsymbol{\sigma} - \left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)\hat{\mathbf{e}}\right] - \frac{1}{m^2}\left[\boldsymbol{\sigma}\otimes\hat{\mathbf{e}} - (\boldsymbol{\sigma}\cdot\hat{\mathbf{e}})\hat{\mathbf{e}}\otimes\hat{\mathbf{e}}\right] - \frac{1}{m^2}\left[\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)\mathbf{1} - \left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)\hat{\mathbf{e}}\otimes\hat{\mathbf{e}}\right] \\
=& -\frac{1}{m^2}\left[\hat{\mathbf{e}}\otimes\boldsymbol{\sigma} - \left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)\hat{\mathbf{e}}\otimes\hat{\mathbf{e}} + \boldsymbol{\sigma}\otimes\hat{\mathbf{e}} - (\boldsymbol{\sigma}\cdot\hat{\mathbf{e}})\hat{\mathbf{e}}\otimes\hat{\mathbf{e}} + \left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)\mathbf{1} - \left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)\hat{\mathbf{e}}\otimes\hat{\mathbf{e}}\right] \\
=& -\frac{1}{m^2}\left[\hat{\mathbf{e}}\otimes\boldsymbol{\sigma} + \boldsymbol{\sigma}\otimes\hat{\mathbf{e}} + \left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)\mathbf{1} - 3\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{e}}\right)\hat{\mathbf{e}}\otimes\hat{\mathbf{e}}\right] \ .
\end{aligned}
\tag{M.29}
$$

where we have used Eq. (M.27).

If we consider a ferromagnetic ground state where the magnetization points along the $\hat{\mathbf{z}}$ axis on all magnetic sites, i.e., $\hat{\mathbf{e}}_i = \hat{\mathbf{z}}$, we have

$$
\begin{aligned}
\frac{\partial\left(\boldsymbol{\sigma}\cdot\hat{\mathbf{z}}\right)}{\partial \mathbf{m}} =& \frac{1}{m}\left[\sigma^x\hat{\mathbf{x}} + \sigma^y\hat{\mathbf{y}}\right] \\
=& \frac{1}{m}\begin{pmatrix} \sigma^x \\ \sigma^y \\ 0 \end{pmatrix} \ ,
\end{aligned}
\tag{M.30}
$$

and

$$\frac{\partial^2 (\boldsymbol{\sigma} \cdot \hat{\mathbf{e}})}{\partial \mathbf{m}^2} = -\frac{1}{m^2} \left[\hat{\mathbf{z}} \otimes \boldsymbol{\sigma} + \boldsymbol{\sigma} \otimes \hat{\mathbf{z}} + \sigma^z \mathbf{1} - 3\sigma^z \hat{\mathbf{z}} \otimes \hat{\mathbf{z}} \right]$$
$$= -\frac{1}{m^2} \left[\begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ \sigma^x & \sigma^y & \sigma^z \end{pmatrix} + \begin{pmatrix} 0 & 0 & \sigma^x \\ 0 & 0 & \sigma^y \\ 0 & 0 & \sigma^z \end{pmatrix} + \begin{pmatrix} \sigma^z & 0 & 0 \\ 0 & \sigma^z & 0 \\ 0 & 0 & \sigma^z \end{pmatrix} - \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 3\sigma^z \end{pmatrix} \right]$$
$$= -\frac{1}{m^2} \begin{pmatrix} \sigma^z & 0 & \sigma^x \\ 0 & \sigma^z & \sigma^y \\ \sigma^x & \sigma^y & 0 \end{pmatrix} . \tag{M.31}$$

The on-site tensor written in Eq. (M.20) becomes

$$\mathcal{J}_i = -\frac{U_i}{2m_i} \begin{pmatrix} m_i^z & 0 & m_i^x \\ 0 & m_i^z & m_i^y \\ m_i^x & m_i^y & 0 \end{pmatrix} . \tag{M.32}$$

Appendix N

Landau-Lifshitz-Gilbert equation

The basic Landau-Lifshitz-Equation can be written as

$$\dot{\mathbf{m}}_i = -\gamma \mathbf{m}_i \times \mathbf{B}_i^{\text{eff}} + \alpha \mathbf{m}_i \times \dot{\mathbf{m}}_i. \tag{N.1}$$

where $\mathbf{m}_i(t)$ is a unit vector pointing along the magnetization direction of site i, $\mathbf{B}_i^{\text{eff}}$ is the effective magnetic field applied on the magnetization (that may include external fields and internal fields), γ is the gyromagnetic ratio ($\gamma = \frac{g\mu_B}{\hbar}$ for the electron), and α is the (Gilbert-)damping constant. (T. L. Gilbert, Magnetics, IEEE Transactions on 40, 3443 (2004).)

In terms of the components, we can write (omitting the site index for simplicity):

$$\begin{aligned}
\dot{m}^x &= -\gamma[m^y B_z^{\text{eff}} - m^z B_y^{\text{eff}}] + \alpha\,[m^y \dot{m}^z - m^z \dot{m}^y] \\
\dot{m}^y &= -\gamma[m^z B_x^{\text{eff}} - m^x B_z^{\text{eff}}] + \alpha\,[m^z \dot{m}^x - m^x \dot{m}^z]\ . \\
\dot{m}^z &= -\gamma[m^x B_y^{\text{eff}} - m^y B_x^{\text{eff}}] + \alpha\,[m^x \dot{m}^y - m^y \dot{m}^x]
\end{aligned} \tag{N.2}$$

Multiplying Eq. (N.1) from the left by $\mathbf{m}_i\times$,

$$\mathbf{m}_i \times \dot{\mathbf{m}}_i = -\gamma \mathbf{m}_i \times (\mathbf{m}_i \times \mathbf{B}_i^{\text{eff}}) + \alpha \mathbf{m}_i \times (\mathbf{m}_i \times \dot{\mathbf{m}}_i). \tag{N.3}$$

Using the identity $\mathbf{a} \times (\mathbf{b} \times \mathbf{c}) = \mathbf{b}(\mathbf{a} \cdot \mathbf{c}) - \mathbf{c}(\mathbf{a} \cdot \mathbf{b})$ on the second term of the RHS,

$$\begin{aligned}
\alpha \mathbf{m}_i \times (\mathbf{m}_i \times \dot{\mathbf{m}}_i) =& \alpha \mathbf{m}_i(\mathbf{m}_i \cdot \dot{\mathbf{m}}_i) - \alpha \dot{\mathbf{m}}_i(\mathbf{m}_i \cdot \mathbf{m}_i) \\
=& -\alpha \dot{\mathbf{m}}_i\ ,
\end{aligned} \tag{N.4}$$

since $\mathbf{m}_i \cdot \dot{\mathbf{m}}_i = 0$. Substituting back into Eq. (N.3), we obtain

$$\mathbf{m}_i \times \dot{\mathbf{m}}_i = -\gamma \mathbf{m}_i \times (\mathbf{m}_i \times \mathbf{B}_i^{\text{eff}}) - \alpha \dot{\mathbf{m}}_i\ . \tag{N.5}$$

Using this result for the Gilbert term in Eq. (N.1), we can write

$$\dot{\mathbf{m}}_i = -\gamma \mathbf{m}_i \times \mathbf{B}_i^{\text{eff}} - \alpha\gamma \mathbf{m}_i \times (\mathbf{m}_i \times \mathbf{B}_i^{\text{eff}}) - \alpha^2 \dot{\mathbf{m}}_i\ . \tag{N.6}$$

Rearranging the terms,

$$(1+\alpha^2)\dot{\mathbf{m}}_i = -\gamma \mathbf{m}_i \times \mathbf{B}_i^{\text{eff}} - \alpha\gamma \mathbf{m}_i \times (\mathbf{m}_i \times \mathbf{B}_i^{\text{eff}}) \ . \tag{N.7}$$

And finally

$$\dot{\mathbf{m}}_i = -\frac{\gamma}{(1+\alpha^2)} \mathbf{m}_i \times \mathbf{B}_i^{\text{eff}} - \frac{\alpha\gamma}{(1+\alpha^2)} \mathbf{m}_i \times (\mathbf{m}_i \times \mathbf{B}_i^{\text{eff}}) \ , \tag{N.8}$$

which is a renormalized Landau-Lifshitz equation (Landau-Lifshitz equation in the Gilbert form). This form of the equation gives the proper behavior for large dampings ($\dot{\mathbf{m}}_i \to 0$ when $\alpha \to \infty$).

N.1 Linear response

We can linearize LLG equation assuming that the magnetization oscillates around its equilibrium position $\hat{\mathbf{z}}$ with a small cone angle such that $\dot{m}^z \simeq 0$.

$$\begin{aligned}
\dot{m}^x &= -\gamma[m^y B_z^{\text{eff}} - m^z B_y^{\text{eff}}] - \alpha m^z \dot{m}^y \\
\dot{m}^y &= -\gamma[m^z B_x^{\text{eff}} - m^x B_z^{\text{eff}}] + \alpha m^z \dot{m}^x \\
0 &= -\gamma[m^x B_y^{\text{eff}} - m^y B_x^{\text{eff}}] + \alpha \left[m^x \dot{m}^y - m^y \dot{m}^x\right]
\end{aligned} \ . \tag{N.9}$$

From the first two equations above, we can write

$$\begin{aligned}
\dot{m}^x + i\dot{m}^y &= -\gamma\left[m^y B_z^{\text{eff}} - i m^x B_z^{\text{eff}} - m^z B_y^{\text{eff}} + i m^z B_x^{\text{eff}}\right] - \alpha m^z \dot{m}^y + i\alpha m^z \dot{m}^x \\
\dot{m}^x + i\dot{m}^y &= -\gamma\left[\left(m^y - i m^x\right) B_z^{\text{eff}} - m^z \left(B_y^{\text{eff}} - i B_x^{\text{eff}}\right)\right] - \alpha m^z \left(\dot{m}^y - i\dot{m}^x\right) \\
\dot{m}^x + i\dot{m}^y &= -\gamma\left[-i\left(i m^y + m^x\right) B_z^{\text{eff}} + i m^z \left(i B_y^{\text{eff}} + B_x^{\text{eff}}\right)\right] + i\alpha m^z \left(i\dot{m}^y + \dot{m}^x\right) \ , \\
\dot{m}^+ &= -\gamma\left[-i B_z^{\text{eff}} m^+ + i m^z B_+^{\text{eff}}\right] + i\alpha m^z \dot{m}^+ \\
\left(1 - i\alpha m^z\right) \dot{m}^+ &= -\gamma\left[-i B_z^{\text{eff}} m^+ + i m^z B_+^{\text{eff}}\right]
\end{aligned} \tag{N.10}$$

and finally, we obtain

$$\left(1 - i\alpha m^z\right) \dot{m}^+(t) - i\gamma B_z^{\text{eff}} m^+(t) = -i\gamma m^z B_+^{\text{eff}} \ . \tag{N.11}$$

N.2 Connection to the Heisenberg model

The effective field can be obtained as $\mathbf{B}_k^{\text{eff}} = -\frac{\partial H}{\partial \mathbf{m}_k}$. We can use the Heisenberg model written in Eq. (M.2) or Eq. (M.5) (the extra terms that appear on the former is parallel to the magnetization, which vanish on the cross product of the LLG equation) to obtain

$$
\begin{aligned}
\mathbf{B}_k^{\mathrm{eff}} &= -\frac{\partial H}{\partial \mathbf{m}_k} \\
&= -\frac{\partial H}{\partial m_k^x}\hat{\mathbf{x}} - \frac{\partial H}{\partial m_k^y}\hat{\mathbf{y}} - \frac{\partial H}{\partial m_k^z}\hat{\mathbf{z}} \\
&= \sum_j \mathcal{J}_{kj}\mathbf{m}_j + \mathbf{B}_k^{\mathrm{ext}} \ .
\end{aligned}
\tag{N.12}
$$

Now, let's consider a magnetic field composed by a static part on the $\hat{\mathbf{z}}$ direction and a time-dependent transverse part, that is,

$$
\mathbf{B}_{\mathrm{eff}}(t) = b_x(t)\hat{\mathbf{x}} + b_y(t)\hat{\mathbf{y}} + B_0\hat{\mathbf{z}} \ . \tag{N.13}
$$

Substituting in Eq. (N.11), we get

$$
\left(1 - i\alpha m^z\right)\dot{m}^+(t) - i\gamma B_0 m^+(t) = -i\gamma m^z B_+^{\mathrm{eff}}(t) \ . \tag{N.14}
$$

Using the Fourier transform defined by

$$
f(t) = \frac{1}{2\pi}\int e^{-i\omega t}F(\omega)d\omega \ , \tag{N.15}
$$

we can write

$$
\begin{aligned}
-i\omega\left(1 - i\alpha m^z\right)m^+(\omega) - i\gamma B_0 m^+(\omega) &= -i\gamma m^z B_+^{\mathrm{eff}}(\omega) \\
\left[\omega\left(1 - i\alpha m^z\right) + \gamma B_0\right]m_0^+ &= \gamma m^z B_+^{\mathrm{eff}}(\omega)
\end{aligned} \ .
\tag{N.16}
$$

and finally,

$$
m_0^+(\omega) = \frac{\gamma m^z}{\left[\omega\left(1 - i\alpha m^z\right) + \gamma B_0\right]}B_+^{\mathrm{eff}}(\omega) \ . \tag{N.17}
$$

From the linear response formalism, the proportionality is given by the susceptibility χ as

$$
m^+(\omega) = \chi^{+-}(\omega)B_+^{\mathrm{eff}}(\omega) \ , \tag{N.18}
$$

where the response function $\chi^{+-}(\omega)$ can be identified as

$$
\chi^{+-}(\omega) = \frac{\gamma m^z}{\omega\left(1 - i\alpha m^z\right) + \gamma B_0} \ . \tag{N.19}
$$

It's convenient to write it as

$$
\begin{aligned}
\chi^{+-}(\omega) &= \frac{\gamma m^z}{\omega+\gamma B_0 - i\omega\alpha m^z}\frac{\omega+\gamma B_0 + i\omega\alpha m^z}{\omega+\gamma B_0 + i\omega\alpha m^z} \\
&= \frac{\gamma m^z\left(\omega+\gamma B_0 + i\omega\alpha m^z\right)}{\left(\omega+\gamma B_0\right)^2+\left(\omega\alpha m^z\right)^2} \\
&= \gamma m^z \frac{\left(\omega+\gamma B_0 + i\omega\alpha m^z\right)}{\left(1+\alpha^2 m^{z2}\right)\omega^2 + 2\omega\gamma B_0 + \left(\gamma B_0\right)^2} \\
&= \frac{\gamma m^z}{\left(1+\alpha^2 m^{z2}\right)}\frac{\left(\omega+\gamma B_0 + i\omega\alpha m^z\right)}{\omega^2+\frac{2\omega\gamma B_0}{(1+\alpha^2 m^{z2})}+\frac{(\gamma B_0)^2}{(1+\alpha^2 m^{z2})}} \\
&= \frac{\gamma m^z}{\left(1+\alpha^2 m^{z2}\right)}\frac{\left(\omega+\gamma B_0 + i\omega\alpha m^z\right)}{\left[\omega+\frac{(\gamma B_0)}{(1+\alpha^2 m^{z2})}\right]^2-\frac{(\gamma B_0)^2}{(1+\alpha^2 m^{z2})^2}+\frac{(\gamma B_0)^2(1+\alpha^2 m^{z2})}{(1+\alpha^2 m^{z2})^2}} \\
&= \frac{\gamma m^z}{\left(1+\alpha^2 m^{z2}\right)}\frac{\left(\omega+\gamma B_0 + i\omega\alpha m^z\right)}{\left[\omega+\frac{(\gamma B_0)}{(1+\alpha^2 m^{z2})}\right]^2+\frac{(\gamma B_0)^2\alpha^2 m^{z2}}{(1+\alpha^2 m^{z2})^2}}
\end{aligned} \quad . \tag{N.20}
$$

If we write $\eta = \alpha m^z$ and $\omega_0 = \frac{\gamma B_0}{(1+\eta^2)}$, we can rewrite the expression above as

$$
\begin{aligned}
\chi^{+-}(\omega) &= \frac{\gamma m^z}{(1+\eta^2)}\frac{\omega+\left(1+\eta^2\right)\omega_0+i\omega\eta}{\left(\omega+\omega_0\right)^2+\left(\eta\omega_0\right)^2} \\
&= \frac{m^z\omega_0}{B_0}\frac{\omega+\left(1+\eta^2\right)\omega_0+i\omega\eta}{\left(\omega+\omega_0\right)^2+\left(\eta\omega_0\right)^2}
\end{aligned} \quad . \tag{N.21}
$$

$$
\begin{aligned}
\chi^{+-}(\omega) &= \frac{\gamma m^z}{\omega+\gamma B_0 - i\omega\alpha m^z}\frac{\omega+\gamma B_0 + i\omega\alpha m^z}{\omega+\gamma B_0 + i\omega\alpha m^z} \\
&= \frac{\gamma m^z\left(\omega+\gamma B_0 + i\omega\alpha m^z\right)}{\left(\omega+\gamma B_0\right)^2+\left(\omega\alpha m^z\right)^2} \\
&= \frac{\gamma m^z\left(\omega+\gamma B_0\right)}{\left(\omega+\gamma B_0\right)^2+\left(\omega\alpha m^z\right)^2} + i\frac{\omega\alpha\gamma(m^z)^2}{\left(\omega+\gamma B_0\right)^2+\left(\omega\alpha m^z\right)^2} \\
&= \mathrm{Re}\left[\chi^{+-}(\omega)\right] + i\,\mathrm{Im}\left[\chi^{+-}(\omega)\right]
\end{aligned} \quad . \tag{N.22}
$$

$$
\chi^{+-}(\omega) = \frac{\gamma m^z\left(\omega+\gamma B_0\right)}{\left(\omega+\gamma B_0\right)^2+\left(\omega\alpha m^z\right)^2} + i\frac{\omega\alpha\gamma(m^z)^2}{\left(\omega+\gamma B_0\right)^2+\left(\omega\alpha m^z\right)^2} \quad . \tag{N.23}
$$

N.3 Dimer

When we have two magnetic units, all the equations become site-dependent and we need to take into account the interaction between them. We can rewrite Eq. (N.11) as

$$
\left(1 - i\alpha m_i^z\right)\dot{m}_i^+(t) - i\gamma B_i^z m_i^+(t) = -i\gamma m_i^z B_i^+ \ . \tag{N.24}
$$

where, for a dimer, $i = 1, 2$ and $\mathbf{B}_i = \mathbf{B}_i^{\mathrm{eff}}$ is the total effective field that acts on site i. It can be obtained by

$$\mathbf{B}_i = -\frac{\partial E}{\partial \mathbf{m}_1} \ . \tag{N.25}$$

Here, E is the total energy of the system. This is the main quantity where we can introduce the different effects to be taken into account: exchange interaction, external field, anisotropies, Dzyaloshinskii-Moriya interaction, currents, and so on.

Let's consider a dimer described by the following total energy:

$$\begin{split} E(\mathbf{m}_1, \mathbf{m}_2) = &-\frac{1}{2} J_{12} \mathbf{m}_1 \cdot \mathbf{m}_2 - \frac{1}{2} J_{21} \mathbf{m}_2 \cdot \mathbf{m}_1 - \frac{1}{2} \mathbf{D}_{12} \cdot \mathbf{m}_1 \times \mathbf{m}_2 - \frac{1}{2} \mathbf{D}_{21} \cdot \mathbf{m}_2 \times \mathbf{m}_1 \\ &- \mathbf{B}_1^{\mathrm{ext}} \cdot \mathbf{m}_1 - \mathbf{B}_2^{\mathrm{ext}} \cdot \mathbf{m}_2 \pm \mathbf{B}_1^{\mathrm{an}} \cdot \mathbf{m}_1 \pm \mathbf{B}_2^{\mathrm{an}} \cdot \mathbf{m}_2 \end{split} \ . \tag{N.26}$$

We are representing the anisotropy as a field that we will later choose to be in the direction of the magnetization.

Appendix O

Connection with D. Edwards Kernel

To connect our U matrix (given by Eqs. 3.182-3.185) with the one obtained by David Edwards (arxiv.org:1506.05622), we consider that $v(\mathbf{q}) = 0$ from his side, and $U_2 = 0$ (given by Eq. (3.185)) from our side. Then, since $[U]^{m}_{\alpha\beta\gamma\xi} = [U_1^m]\delta_{\alpha\beta}\delta_{\gamma\xi}$, Eq. (3.187) becomes

$$[\chi]_{ijkl}^{MN\,\mu\nu\gamma\xi}(\omega) = [\chi_{(0)}]_{ijkl}^{MN\,\mu\nu\gamma\xi}(\omega) - [\chi_{(0)}]_{ijmm}^{MP\,\mu\nu\alpha\alpha}(\omega)[U_1^m]^{PL}[\chi]_{mmkl}^{LN\,\beta\beta\gamma\xi}(\omega) \ . \tag{O.1}$$

Particularizing the equation above for the usual magnetic susceptibility, given by $\chi_{ij}^{MN} = \sum_{\mu\nu}\chi_{ij}^{MN\,\mu\nu} = \sum_{\mu\nu}\chi_{iijj}^{MN\,\mu\mu\nu\nu}$ (where the sum is over the d orbitals), and making the sum over orbitals explicit, we have

$$\begin{aligned}
\sum_{\mu\nu}[\chi]_{iijj}^{MN\,\mu\mu\nu\nu}(\omega) &= \sum_{\mu\nu}[\chi_{(0)}]_{iijj}^{MN\,\mu\mu\nu\nu}(\omega) - \sum_{\mu\nu\alpha\beta}[\chi_{(0)}]_{iimm}^{MP\,\mu\mu\alpha\alpha}(\omega)[U_1^m]^{PL}[\chi]_{mmjj}^{LN\,\beta\beta\nu\nu}(\omega) \\
\sum_{\mu\nu}[\chi]_{ij}^{MN\,\mu\nu}(\omega) &= \sum_{\mu\nu}[\chi_{(0)}]_{ij}^{MN\,\mu\nu}(\omega) - \sum_{\mu\nu\alpha\beta}[\chi_{(0)}]_{im}^{MP\,\mu\alpha}(\omega)[U_1^m]^{PL}[\chi]_{mj}^{LN\,\beta\nu}(\omega) \\
[\chi]_{ij}^{MN}(\omega) &= [\chi_{(0)}]_{ij}^{MN}(\omega) - [\chi_{(0)}]_{im}^{MP}(\omega)[U_1^m]^{PL}[\chi]_{mj}^{LN}(\omega) \ .
\end{aligned} \tag{O.2}$$

Appendix P

Rectifield currents and voltages (second order)

When we apply an ac electric field, a charge current given by Eq. (5.251) is generated in the system. The resistance depends on the magnetization angle—and since it is precessing, the resistance is also oscillating. The measured voltage is then given by

$$V(t) = R(t)I(t) \ . \tag{P.1}$$

If we consider that an uniform electric field is applied on the system, the generated current can be written as

$$\begin{aligned} I(t) =& G'(t)V(t) \\ =& G(t)E(t) \ . \end{aligned} \tag{P.2}$$

where G' is the conductance and G is an "integrated conductance". For an oscillatory electric field, the current is given by Eq. (5.251).

P.1 Currents

P.1.1 Longitudinal

The longitudinal measurement is affected by the anisotropic magnetoresistance (AMR) and by the spin Hall magnetoresistance (SMR). In this case, the conductance can be written as

$$\begin{aligned} G =& G_0 + \Delta G_1 \sin^2\theta\cos^2\phi + \Delta G_2 \sin^2\theta\sin^2\phi \\ =& G_0 + \Delta G_1 \frac{\langle m^x\rangle^2}{|\mathbf{m}|^2} + \Delta G_2 \frac{\langle m^y\rangle^2}{|\mathbf{m}|^2} \ . \end{aligned} \tag{P.3}$$

where ΔG_1 and ΔG_2 are the AMR and SMR conductance variations, respectively, and θ, ϕ are the spherical angles of the magnetization. The x and y components of the magnetization (in the lattice frame of reference) may have static and dynamical components, i.e., $\langle m^\alpha\rangle(t) = m^\alpha + \delta\langle m^\alpha\rangle(t)$. The time-dependent part of the magnetization is given by Eq. (5.321). Since $\delta\langle m^\alpha\rangle(t) \ll 1$, we can write

$$\begin{aligned}
G(t) =& G_0 + \Delta G_1 \frac{[\langle m^x\rangle(t)]^2}{|\mathbf{m}|^2} + \Delta G_2 \frac{[\langle m^y\rangle(t)]^2}{|\mathbf{m}|^2} \\
\approx& G_0 + \Delta G_1 \frac{(m^x)^2 + 2m^x\delta\langle m^x\rangle(t)}{|\mathbf{m}|^2} + \Delta G_2 \frac{(m^y)^2 + 2m^y\delta\langle m^y\rangle(t)}{|\mathbf{m}|^2} \\
=& G_0 + \Delta G_1 \frac{(m^x)^2}{|\mathbf{m}|^2} + \Delta G_2 \frac{(m^y)^2}{|\mathbf{m}|^2} + \Delta G_1 \frac{2m^x\delta\langle m^x\rangle(t)}{|\mathbf{m}|^2} + \Delta G_2 \frac{2m^y\delta\langle m^y\rangle(t)}{|\mathbf{m}|^2} \\
=& G + \Delta G_1 \frac{2m^x\delta\langle m^x\rangle(t)}{|\mathbf{m}|^2} + \Delta G_2 \frac{2m^y\delta\langle m^y\rangle(t)}{|\mathbf{m}|^2} \ .
\end{aligned} \tag{P.4}$$

P.1.2 Transverse

P.2 Voltages

P.2.1 Longitudinal

The longitudinal measurement is affected by the anisotropic magnetoresistance (AMR) and by the spin Hall magnetoresistance (SMR). In this case, the resistance is given by

$$\begin{aligned}
R =& R_0 + \Delta R_1 \sin^2\theta \cos^2\phi + \Delta R_2 \sin^2\theta \sin^2\phi \\
=& R_0 + \Delta R_1 \frac{\langle m^x\rangle^2}{|\mathbf{m}|^2} + \Delta R_2 \frac{\langle m^y\rangle^2}{|\mathbf{m}|^2} \ .
\end{aligned} \tag{P.5}$$

where ΔR_1 and ΔR_2 are the AMR and SMR coefficients, respectively, and θ, ϕ are the spherical angles of the magnetization. The x and y components of the magnetization (in the lattice frame of reference) may have static and dynamical components, i.e., $\langle m^\alpha\rangle(t) = m^\alpha + \delta\langle m^\alpha\rangle(t)$. The time-dependent part of the magnetization is given by Eq. (5.321). Since $\delta\langle m^x\rangle(t) \ll |\mathbf{m}|$, the absolute value of the magnetization is $|\mathbf{m}|$, and we can write for each layer

$$\begin{aligned}
R(t) =& R_\parallel - (R_\parallel - R_\perp) \frac{[\langle m^x\rangle(t)]^2}{|\mathbf{m}|^2} \\
\approx& R_\parallel - (R_\parallel - R_\perp) \frac{(m^x)^2 + 2m^x\delta\langle m^x\rangle(t)}{|\mathbf{m}|^2} \ .
\end{aligned} \tag{P.6}$$

Therefore, using Eqs. 5.251, 5.321 and P.6, the voltage measured in the experiments is given by

$$\begin{aligned}
V_{\parallel,\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, t) =& -\frac{e^2 E_0}{\hbar^2\omega}\left[R_\parallel - (R_\parallel - R_\perp)\frac{(m^x_{\ell_1})^2 + 2m^x_{\ell_1}\delta\langle m^x_{\ell_1}\rangle(t)}{|\mathbf{m}|^2}\right]\left|\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega)\right|\cos[\omega t - \phi^{\mathrm{C}}(\omega)] \\
=& -\frac{e^2 E_0}{\hbar^2\omega}\left[R_\parallel - (R_\parallel - R_\perp)\frac{(m^x_{\ell_1})^2}{|\mathbf{m}|^2}\right]\left|\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega)\right|\cos[\omega t - \phi^{\mathrm{C}}(\omega)] \\
&+ \frac{e^3 E_0^2}{\hbar^3\omega^2}(R_\parallel - R_\perp)\frac{2m^x_{\ell_1}}{|\mathbf{m}|^2}\left|\mathcal{D}^x_{\ell_1}(\omega)\right|\sin[\omega t - \phi^x(\omega)]\left|\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega)\right|\cos[\omega t - \phi^{\mathrm{C}}(\omega)] \; .
\end{aligned} \tag{P.7}$$

where $\mathbf{R}'_\parallel - \mathbf{R}_\parallel$ is in the direction of the electric field (i.e., longitudinal current), and $\mathbf{q} = 0$. Using the relation $\sin\alpha\cos\beta = \frac{1}{2}\left[\sin(\alpha+\beta) + \sin(\alpha-\beta)\right]$, we have

$$\begin{aligned}
V_{\parallel,\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, t) =& -\frac{e^2 E_0}{\hbar^2\omega}\left[R_\parallel - (R_\parallel - R_\perp)\frac{(m^x_{\ell_1})^2}{|\mathbf{m}|^2}\right]\left|\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega)\right|\cos[\omega t - \phi^{\mathrm{C}}(\omega)] \\
&+ \frac{e^3 E_0^2}{\hbar^3\omega^2}(R_\parallel - R_\perp)\frac{m^x_{\ell_1}}{|\mathbf{m}|^2}\left|\mathcal{D}^x_{\ell_1}(\omega)\right|\left|\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega)\right|\sin[2\omega t - \phi^x(\omega) - \phi^{\mathrm{C}}(\omega)] \\
&+ \frac{e^3 E_0^2}{\hbar^3\omega^2}(R_\parallel - R_\perp)\frac{m^x_{\ell_1}}{|\mathbf{m}|^2}\left|\mathcal{D}^x_{\ell_1}(\omega)\right|\left|\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega)\right|\sin[\phi^{\mathrm{C}}(\omega) - \phi^x(\omega)] \; .
\end{aligned} \tag{P.8}$$

Note that we have two second order terms: one with doubled frequency 2ω and another one which is dc. The latter is the one used in the experiments, and it can be written as

$$V^{\mathrm{dc}}_{\parallel,\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel) = \frac{e^3 E_0^2}{\hbar^3\omega^2}(R_\parallel - R_\perp)\frac{m^x_{\ell_1}}{|\mathbf{m}|^2}\left|\mathcal{D}^x_{\ell_1}(\omega)\right|\left|\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel,\omega)\right|\sin[\phi^{\mathrm{C}}(\omega) - \phi^x(\omega)] \; . \tag{P.9}$$

This is the longitudinal dc voltage that appears when an ac-charge current is flowing on magnetic system and the magnetization is set into precession.

P.2.2 Transverse (Hall) voltage

We now focus our attention on the transverse voltage. In this case, the current is also given by Eq. (5.251) but now $\mathbf{R}'_\parallel - \mathbf{R}_\parallel$ is in the transverse direction. The transverse resistance have contributions from the Anomalous and the Planar Hall effects,

$$\begin{aligned}
R =& R_{\mathrm{AHE}}\cos\theta + R_{\mathrm{PHE}}\sin^2\theta\cos\phi\sin\phi \\
=& R_{\mathrm{AHE}}\frac{\langle m^z\rangle}{|\mathbf{m}|} + R_{\mathrm{PHE}}\frac{\langle m^x\rangle}{|\mathbf{m}|}\frac{\langle m^y\rangle}{|\mathbf{m}|} \; .
\end{aligned} \tag{P.10}$$

When the magnetization is precessing, we can separate the static and the dynamical part of the magnetization to obtain

$$
\begin{aligned}
R(t) =& R_{\mathrm{AHE}} \frac{m^z + \delta\langle m^z\rangle(t)}{|\mathbf{m}|} + R_{\mathrm{PHE}} \left[\frac{m^x + \delta\langle m^x\rangle(t)}{|\mathbf{m}|}\right] \left[\frac{m^y + \delta\langle m^y\rangle(t)}{|\mathbf{m}|}\right] \\
\approx& R_{\mathrm{AHE}} \frac{m^z + \delta\langle m^z\rangle(t)}{|\mathbf{m}|} + R_{\mathrm{PHE}} \frac{m^x m^y + m^y \delta\langle m^x\rangle(t) + m^x \delta\langle m^y\rangle(t)}{|\mathbf{m}|^2} \ .
\end{aligned}
\tag{P.11}
$$

Using Eqs. 5.251, 5.321 and P.11, the voltage measured in the experiments is given by

$$
\begin{aligned}
V_\perp(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, t) =& -\frac{e^2 E_0}{\hbar^2 \omega} \left[R_{\mathrm{AHE}} \frac{m^z + \delta\langle m^z\rangle(t)}{|\mathbf{m}|} + R_{\mathrm{PHE}} \frac{m^x m^y + m^y \delta\langle m^x\rangle(t) + m^x \delta\langle m^y\rangle(t)}{|\mathbf{m}|^2}\right] \\
& \times \left|\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega)\right| \cos[\omega t - \phi^{\mathrm{C}}(\omega)] \\
=& -\frac{e^2 E_0}{\hbar^2 \omega} \left[R_{\mathrm{AHE}} \frac{m^z}{|\mathbf{m}|} + R_{\mathrm{PHE}} \frac{m^x m^y}{|\mathbf{m}|^2}\right] \left|\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega)\right| \cos[\omega t - \phi^{\mathrm{C}}(\omega)] \\
& - \frac{e^3 E_0^2}{\hbar^3 \omega^2} \left|\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega)\right| \cos[\omega t - \phi^{\mathrm{C}}(\omega)] \left\{ \frac{R_{\mathrm{AHE}}}{|\mathbf{m}|} \left|\mathcal{D}^z_{\ell_1}(\omega)\right| \sin[\omega t - \phi^z(\omega)] \right. \\
& \left. + \frac{R_{\mathrm{PHE}}}{|\mathbf{m}|^2} \left[m^y \left|\mathcal{D}^x_{\ell_1}(\omega)\right| \sin[\omega t - \phi^x(\omega)] + m^x \left|\mathcal{D}^y_{\ell_1}(\omega)\right| \sin[\omega t - \phi^y(\omega)]\right] \right\} \ .
\end{aligned}
\tag{P.12}
$$

where now $\mathbf{R}'_\parallel - \mathbf{R}_\parallel$ is in the direction perpendicular to the electric field (i.e., transverse current). Once again, the last term (second order) will give rise to an oscillatory voltage with double frequency, 2ω and a dc term given by

$$
\begin{aligned}
V^{\mathrm{dc}}_\perp(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, t) =& -\frac{e^3 E_0^2}{\hbar^3 \omega^2} \left|\mathcal{J}^{\mathrm{C}}_{\ell_1}(\mathbf{R}'_\parallel - \mathbf{R}_\parallel, \omega)\right| \left\{ \frac{R_{\mathrm{AHE}}}{|\mathbf{m}|} \left|\mathcal{D}^z_{\ell_1}(\omega)\right| \sin[\phi^{\mathrm{C}}(\omega) - \phi^z(\omega)] \right. \\
& \left. + \frac{R_{\mathrm{PHE}}}{|\mathbf{m}|^2} \left[m^y \left|\mathcal{D}^x_{\ell_1}(\omega)\right| \sin[\phi^{\mathrm{C}}(\omega) - \phi^x(\omega)] + m^x \left|\mathcal{D}^y_{\ell_1}(\omega)\right| \sin[\phi^{\mathrm{C}}(\omega) - \phi^y(\omega)]\right] \right\} \ .
\end{aligned}
\tag{P.13}
$$

Appendix Q

DC component of spin pumping

The semi-classical expression for the spin current pumped out of a magnetic unit is given by

$$\begin{aligned}\mathbf{I}_S^{\text{pump}} =& \frac{\hbar}{4\pi}\left(A_r \mathbf{m}\times\frac{d\mathbf{m}}{dt} - A_i\frac{d\mathbf{m}}{dt}\right)\\ =& \frac{\hbar}{4\pi}\left(\operatorname{Re} g^{\uparrow\downarrow}\mathbf{m}\times\frac{d\mathbf{m}}{dt} + \operatorname{Im} g^{\uparrow\downarrow}\frac{d\mathbf{m}}{dt}\right) \ ,\end{aligned} \tag{Q.1}$$

where $g^{\uparrow\downarrow}$ is the spin mixing conductance. When the magnetization precesses with a small cone angle around the equilibrium position $\mathbf{m}_0$, we can write $\mathbf{m} = \mathbf{m}_0 + \delta\mathbf{m}(t)$. Therefore, the pumped spin current is given by

$$\mathbf{I}_S^{\text{pump}} = \frac{\hbar}{4\pi}\left[\operatorname{Re} g^{\uparrow\downarrow}(\mathbf{m}_0+\delta\mathbf{m}(t))\times\frac{d\delta\mathbf{m}(t)}{dt} + \operatorname{Im} g^{\uparrow\downarrow}\frac{d\delta\mathbf{m}(t)}{dt}\right] \ . \tag{Q.2}$$

We have then an ac component

$$\mathbf{I}_S^{\text{ac}} = \frac{\hbar}{4\pi}\left[\operatorname{Re} g^{\uparrow\downarrow}\mathbf{m}_0\times\frac{d\delta\mathbf{m}(t)}{dt} + \operatorname{Im} g^{\uparrow\downarrow}\frac{d\delta\mathbf{m}(t)}{dt}\right] \ , \tag{Q.3}$$

and a dc one

$$\mathbf{I}_S^{\text{dc}} = \frac{\hbar}{4\pi}\operatorname{Re} g^{\uparrow\downarrow}\delta\mathbf{m}(t)\times\frac{d\delta\mathbf{m}(t)}{dt} \ . \tag{Q.4}$$

Using Eq. (5.321) for the spin accumulation, the dc component of the spin accumulation can be written as

$$I_S^{\text{dc},m} = \frac{e^2E_0^2}{4\pi\hbar\omega}\operatorname{Re} g^{\uparrow\downarrow}\epsilon_{mnk}|\mathcal{D}_{\ell_1}^n(\omega)||\mathcal{D}_{\ell_1}^k(\omega)|\sin[\omega t-\phi^n(\omega)]\cos[\omega t-\phi^k(\omega)] \ . \tag{Q.5}$$

Using the relation $\sin\alpha\cos\beta = \frac{1}{2}\left[\sin(\alpha+\beta)+\sin(\alpha-\beta)\right]$, we end up with

$$I_S^{\text{dc},m} = \frac{e^2E_0^2}{2\pi\hbar\omega}\operatorname{Re} g^{\uparrow\downarrow}\epsilon_{mnk}|\mathcal{D}_{\ell_1}^n(\omega)||\mathcal{D}_{\ell_1}^k(\omega)|\left\{\sin[2\omega t-\phi^n(\omega)-\phi^k(\omega)]+\sin[\phi^k(\omega)-\phi^n(\omega)]\right\} \ . \tag{Q.6}$$

Appendix R

Relations

Hopping in real space (real numbers)

$$\begin{aligned} t_{ij}^{\mu\nu} &= t_{ji}^{\nu\mu} \\ [t_{ij}^{\mu\nu}]^* &= t_{ij}^{\mu\nu} \end{aligned} \tag{R.1}$$

Hopping in mixed space (Eq. (5.80))

$$\begin{aligned} \left[t_{\ell\ell'}^{\mu\nu}(\mathbf{k}_\parallel)\right]^* &= \sum_{\mathbf{R}''_\parallel} e^{-i\mathbf{k}_\parallel\cdot\mathbf{R}''_\parallel} t_{\ell\ell'}^{\mu\nu}(\mathbf{R}''_\parallel) = t_{\ell\ell'}^{\mu\nu}(-\mathbf{k}_\parallel) \\ &= \sum_{\mathbf{R}''_\parallel} e^{i\mathbf{k}_\parallel\cdot\mathbf{R}''_\parallel} t_{\ell\ell'}^{\mu\nu}(-\mathbf{R}''_\parallel) \\ &= \sum_{\mathbf{R}''_\parallel} e^{i\mathbf{k}_\parallel\cdot\mathbf{R}''_\parallel} t_{\ell'\ell}^{\nu\mu}(\mathbf{R}''_\parallel) = t_{\ell'\ell}^{\nu\mu}(\mathbf{k}_\parallel) \end{aligned} \tag{R.2}$$

Derivative of the hopping (Eq. (A.14))

$$\begin{aligned} \left[\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t_{\ell\ell'}^{\mu\nu}(\mathbf{k}_\parallel)\right]^* &= -\sum_{\mathbf{R}''_\parallel} i\mathbf{R}''_\parallel e^{-i\mathbf{k}_\parallel\cdot\mathbf{R}''_\parallel} t_{\ell\ell'}^{\mu\nu}(\mathbf{R}''_\parallel) = -\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t_{\ell\ell'}^{\mu\nu}(-\mathbf{k}_\parallel) \\ &= \sum_{\mathbf{R}''_\parallel} i\mathbf{R}''_\parallel e^{i\mathbf{k}_\parallel\cdot\mathbf{R}''_\parallel} t_{\ell\ell'}^{\mu\nu}(-\mathbf{R}''_\parallel) \\ &= \sum_{\mathbf{R}''_\parallel} i\mathbf{R}''_\parallel e^{i\mathbf{k}_\parallel\cdot\mathbf{R}''_\parallel} t_{\ell'\ell}^{\nu\mu}(\mathbf{R}''_\parallel) = \boldsymbol{\nabla}_{\mathbf{k}_\parallel} t_{\ell'\ell}^{\nu\mu}(\mathbf{k}_\parallel) \end{aligned} \tag{R.3}$$

which means that

$$\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t_{\ell'\ell}^{\nu\mu}(\mathbf{k}_\parallel) = -\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t_{\ell\ell'}^{\mu\nu}(-\mathbf{k}_\parallel) \tag{R.4}$$

Momentum components in real space (Eq. (5.71)):

$$\begin{aligned}\left[\mathbf{p}^{\mu\nu}_{\ell\ell'}(\mathbf{R}_j-\mathbf{R}_i)\right]^* =&\frac{m}{\hbar}\sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)}\left[\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E\right]^* \\ =&\frac{m}{\hbar}\sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\nu\mu}_{\ell'\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E=\mathbf{p}^{\nu\mu}_{\ell'\ell}(\mathbf{R}_i-\mathbf{R}_j)\end{aligned} \tag{R.5}$$

$$\begin{aligned}\mathbf{p}^{\nu\mu}_{\ell'\ell}(\mathbf{R}_i-\mathbf{R}_j) =&\frac{m}{\hbar}\sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\nu\mu}_{\ell'\ell}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E \\ =&\frac{m}{\hbar}\sum_{\mathbf{k}_\parallel} e^{-i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\mu\nu}_{\ell\ell'}(-\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E \\ =&-\frac{m}{\hbar}\sum_{\mathbf{k}_\parallel} e^{i\mathbf{k}_\parallel\cdot(\mathbf{R}_i-\mathbf{R}_j)}\boldsymbol{\nabla}_{\mathbf{k}_\parallel} t^{\mu\nu}_{\ell\ell'}(\mathbf{k}_\parallel)\cdot\hat{\mathbf{u}}_E=-\mathbf{p}^{\mu\nu}_{\ell\ell'}(\mathbf{R}_j-\mathbf{R}_i)\end{aligned} \tag{R.6}$$

Therefore,

$$\left[\mathbf{p}^{\mu\nu}_{\ell\ell'}(\mathbf{R}_j-\mathbf{R}_i)\right]^* = -\mathbf{p}^{\mu\nu}_{\ell\ell'}(\mathbf{R}_j-\mathbf{R}_i) \tag{R.7}$$

Green function in real space (Eq. (4.4)):

$$\left[G^{\sigma\sigma'\,\mu\nu}_{ij}(E+i\eta)\right]^* = \sum_\alpha \langle j\nu\sigma'\,|\alpha\rangle\,(E-\epsilon_\alpha-i\eta)^{-1}\,\langle\alpha|\,i\mu\sigma\rangle = G^{\sigma'\sigma\,\nu\mu}_{ji}(E-i\eta) \tag{R.8}$$

Appendix S

Transformation of basis

The susceptibility written in $\{+, \uparrow, \downarrow, -\}$ basis can be transformed to the $\{0, x, y, z\}$ basis considering that

$$\begin{aligned}
\hat{\rho}_{ij}^{\mu\nu} = \hat{S}_{ij}^{0,\mu\nu} =& \sum_{\alpha\beta} c_{i\mu\alpha}^{\dagger} \sigma_{\alpha\beta}^{0} c_{j\nu\beta} = \hat{S}_{ij}^{\uparrow,\mu\nu} + \hat{S}_{ij}^{\downarrow,\mu\nu} \\
\hat{S}_{ij}^{x,\mu\nu} =& \frac{1}{2} \sum_{\alpha\beta} c_{i\mu\alpha}^{\dagger} \sigma_{\alpha\beta}^{x} c_{j\nu\beta} = \frac{1}{2} \left(\hat{S}_{ij}^{+,\mu\nu} + \hat{S}_{ij}^{-,\mu\nu} \right) \\
\hat{S}_{ij}^{y,\mu\nu} =& \frac{1}{2} \sum_{\alpha\beta} c_{i\mu\alpha}^{\dagger} \sigma_{\alpha\beta}^{y} c_{j\nu\beta} = \frac{1}{2i} \left(\hat{S}_{ij}^{+,\mu\nu} - \hat{S}_{ij}^{-,\mu\nu} \right) \\
\hat{S}_{ij}^{z,\mu\nu} =& \frac{1}{2} \sum_{\alpha\beta} c_{i\mu\alpha}^{\dagger} \sigma_{\alpha\beta}^{z} c_{j\nu\beta} = \frac{1}{2} \left(\hat{S}_{ij}^{\uparrow,\mu\nu} - \hat{S}_{ij}^{\downarrow,\mu\nu} \right) \ ,
\end{aligned} \tag{S.1}$$

or in matrix form,

$$\begin{pmatrix} \hat{\rho}_{ij}^{\mu\nu} \\ \hat{S}_{ij}^{x,\mu\nu} \\ \hat{S}_{ij}^{y,\mu\nu} \\ \hat{S}_{ij}^{z,\mu\nu} \end{pmatrix} = \begin{pmatrix} 0 & 1 & 1 & 0 \\ \frac{1}{2} & 0 & 0 & \frac{1}{2} \\ \frac{1}{2i} & 0 & 0 & -\frac{1}{2i} \\ 0 & \frac{1}{2} & -\frac{1}{2} & 0 \end{pmatrix} \begin{pmatrix} \hat{S}_{ij}^{+,\mu\nu} \\ \hat{S}_{ij}^{\uparrow,\mu\nu} \\ \hat{S}_{ij}^{\downarrow,\mu\nu} \\ \hat{S}_{ij}^{-,\mu\nu} \end{pmatrix} . \tag{S.2}$$

The inverse transformation is given by

$$\begin{aligned}
\hat{S}_{ij}^{+,\mu\nu} &= c_{i\mu\uparrow}^{\dagger} c_{j\nu\downarrow} = \frac{1}{2} \sum_{\alpha\beta} c_{i\mu\alpha}^{\dagger} \sigma_{\alpha\beta}^{+} c_{j\nu\beta} = \hat{S}_{ij}^{x,\mu\nu} + i\hat{S}_{ij}^{y,\mu\nu} \\
\hat{S}_{ij}^{\uparrow,\mu\nu} &= c_{i\mu\uparrow}^{\dagger} c_{j\nu\uparrow} = \frac{1}{2} \sum_{\alpha\beta} c_{i\mu\alpha}^{\dagger} \sigma_{\alpha\beta}^{\uparrow} c_{j\nu\beta} = \frac{1}{2}\hat{S}_{ij}^{0,\mu\nu} + \hat{S}_{ij}^{z,\mu\nu} \\
\hat{S}_{ij}^{\downarrow,\mu\nu} &= c_{i\mu\downarrow}^{\dagger} c_{j\nu\downarrow} = \frac{1}{2} \sum_{\alpha\beta} c_{i\mu\alpha}^{\dagger} \sigma_{\alpha\beta}^{\downarrow} c_{j\nu\beta} = \frac{1}{2}\hat{S}_{ij}^{0,\mu\nu} - \hat{S}_{ij}^{z,\mu\nu} \\
\hat{S}_{ij}^{-,\mu\nu} &= c_{i\mu\downarrow}^{\dagger} c_{j\nu\uparrow} = \frac{1}{2} \sum_{\alpha\beta} c_{i\mu\alpha}^{\dagger} \sigma_{\alpha\beta}^{-} c_{j\nu\beta} = \hat{S}_{ij}^{x,\mu\nu} - i\hat{S}_{ij}^{y,\mu\nu} \ .
\end{aligned} \tag{S.3}$$

In matrix form,

$$\begin{pmatrix} \hat{S}_{ij}^{+,\mu\nu} \\ \hat{S}_{ij}^{\uparrow,\mu\nu} \\ \hat{S}_{ij}^{\downarrow,\mu\nu} \\ \hat{S}_{ij}^{-,\mu\nu} \end{pmatrix} = \begin{pmatrix} 0 & 1 & i & 0 \\ \frac{1}{2} & 0 & 0 & 1 \\ \frac{1}{2} & 0 & 0 & -1 \\ 0 & 1 & -i & 0 \end{pmatrix} \begin{pmatrix} \hat{\rho}_{ij}^{\mu\nu} \\ \hat{S}_{ij}^{x,\mu\nu} \\ \hat{S}_{ij}^{y,\mu\nu} \\ \hat{S}_{ij}^{z,\mu\nu} \end{pmatrix} . \tag{S.4}$$

Therefore, if we want to obtain the magnetic response in the cartesian basis,

$$\begin{aligned}
&\begin{pmatrix} \hat{S}^{+} \\ \hat{S}^{\uparrow} \\ \hat{S}^{\downarrow} \\ \hat{S}^{-} \end{pmatrix} = \begin{pmatrix} \chi^{+-} & \chi^{+\uparrow} & \chi^{+\downarrow} & \chi^{++} \\ \chi^{\uparrow-} & \chi^{\uparrow\uparrow} & \chi^{\uparrow\downarrow} & \chi^{\uparrow+} \\ \chi^{\downarrow-} & \chi^{\downarrow\uparrow} & \chi^{\downarrow\downarrow} & \chi^{\downarrow+} \\ \chi^{--} & \chi^{-\uparrow} & \chi^{-\downarrow} & \chi^{-+} \end{pmatrix} \begin{pmatrix} \delta B^{+} \\ \delta B^{\uparrow} \\ \delta B^{\downarrow} \\ \delta B^{-} \end{pmatrix} \\
&\begin{pmatrix} 0 & 1 & i & 0 \\ \frac{1}{2} & 0 & 0 & 1 \\ \frac{1}{2} & 0 & 0 & -1 \\ 0 & 1 & -i & 0 \end{pmatrix} \begin{pmatrix} \hat{\rho} \\ \hat{S}^{x} \\ \hat{S}^{y} \\ \hat{S}^{z} \end{pmatrix} = \begin{pmatrix} \chi^{+-} & \chi^{+\uparrow} & \chi^{+\downarrow} & \chi^{++} \\ \chi^{\uparrow-} & \chi^{\uparrow\uparrow} & \chi^{\uparrow\downarrow} & \chi^{\uparrow+} \\ \chi^{\downarrow-} & \chi^{\downarrow\uparrow} & \chi^{\downarrow\downarrow} & \chi^{\downarrow+} \\ \chi^{--} & \chi^{-\uparrow} & \chi^{-\downarrow} & \chi^{-+} \end{pmatrix} \begin{pmatrix} 0 & 1 & i & 0 \\ \frac{1}{2} & 0 & 0 & 1 \\ \frac{1}{2} & 0 & 0 & -1 \\ 0 & 1 & -i & 0 \end{pmatrix} \begin{pmatrix} \delta V \\ \delta B^{x} \\ \delta B^{y} \\ \delta B^{z} \end{pmatrix} \\
&\begin{pmatrix} \hat{\rho} \\ \hat{S}^{x} \\ \hat{S}^{y} \\ \hat{S}^{z} \end{pmatrix} = \begin{pmatrix} 0 & 1 & 1 & 0 \\ \frac{1}{2} & 0 & 0 & \frac{1}{2} \\ \frac{1}{2i} & 0 & 0 & -\frac{1}{2i} \\ 0 & \frac{1}{2} & -\frac{1}{2} & 0 \end{pmatrix} \begin{pmatrix} \chi^{+-} & \chi^{+\uparrow} & \chi^{+\downarrow} & \chi^{++} \\ \chi^{\uparrow-} & \chi^{\uparrow\uparrow} & \chi^{\uparrow\downarrow} & \chi^{\uparrow+} \\ \chi^{\downarrow-} & \chi^{\downarrow\uparrow} & \chi^{\downarrow\downarrow} & \chi^{\downarrow+} \\ \chi^{--} & \chi^{-\uparrow} & \chi^{-\downarrow} & \chi^{-+} \end{pmatrix} \begin{pmatrix} 0 & 1 & i & 0 \\ \frac{1}{2} & 0 & 0 & 1 \\ \frac{1}{2} & 0 & 0 & -1 \\ 0 & 1 & -i & 0 \end{pmatrix} \begin{pmatrix} \delta V \\ \delta B^{x} \\ \delta B^{y} \\ \delta B^{z} \end{pmatrix} .
\end{aligned} \tag{S.5}$$

Comparing to

$$\begin{pmatrix} \hat{\rho} \\ \hat{S}^{x} \\ \hat{S}^{y} \\ \hat{S}^{z} \end{pmatrix} = \begin{pmatrix} \chi^{00} & \chi^{0x} & \chi^{0y} & \chi^{0z} \\ \chi^{x0} & \chi^{xx} & \chi^{xy} & \chi^{xz} \\ \chi^{y0} & \chi^{yx} & \chi^{yy} & \chi^{yz} \\ \chi^{z0} & \chi^{zx} & \chi^{zy} & \chi^{zz} \end{pmatrix} \begin{pmatrix} \delta V \\ \delta B^{x} \\ \delta B^{y} \\ \delta B^{z} \end{pmatrix} , \tag{S.6}$$

we have

$$\begin{pmatrix} \chi^{00} & \chi^{0x} & \chi^{0y} & \chi^{0z} \\ \chi^{x0} & \chi^{xx} & \chi^{xy} & \chi^{xz} \\ \chi^{y0} & \chi^{yx} & \chi^{yy} & \chi^{yz} \\ \chi^{z0} & \chi^{zx} & \chi^{zy} & \chi^{zz} \end{pmatrix} = \begin{pmatrix} 0 & 1 & 1 & 0 \\ \frac{1}{2} & 0 & 0 & \frac{1}{2} \\ \frac{1}{2i} & 0 & 0 & -\frac{1}{2i} \\ 0 & \frac{1}{2} & -\frac{1}{2} & 0 \end{pmatrix} \begin{pmatrix} \chi^{+-} & \chi^{+\uparrow} & \chi^{+\downarrow} & \chi^{++} \\ \chi^{\uparrow-} & \chi^{\uparrow\uparrow} & \chi^{\uparrow\downarrow} & \chi^{\uparrow+} \\ \chi^{\downarrow-} & \chi^{\downarrow\uparrow} & \chi^{\downarrow\downarrow} & \chi^{\downarrow+} \\ \chi^{--} & \chi^{-\uparrow} & \chi^{-\downarrow} & \chi^{-+} \end{pmatrix} \begin{pmatrix} 0 & 1 & i & 0 \\ \frac{1}{2} & 0 & 0 & 1 \\ \frac{1}{2} & 0 & 0 & -1 \\ 0 & 1 & -i & 0 \end{pmatrix} . \tag{S.7}$$

Appendix T

Rotation of the susceptibilities

The susceptibility can be written in the $\{+,\uparrow,\downarrow,-\}$ basis as

$$\begin{pmatrix} \chi^{+-} & \chi^{+\uparrow} & \chi^{+\downarrow} & \chi^{++} \\ \chi^{\uparrow -} & \chi^{\uparrow\uparrow} & \chi^{\uparrow\downarrow} & \chi^{\uparrow +} \\ \chi^{\downarrow -} & \chi^{\downarrow\uparrow} & \chi^{\downarrow\downarrow} & \chi^{\downarrow +} \\ \chi^{--} & \chi^{-\uparrow} & \chi^{-\downarrow} & \chi^{-+} \end{pmatrix} . \tag{T.1}$$

Note that this form of the susceptibility has its largest values on the diagonal. The $\alpha-$component of the spin operator is given by

$$S_i^\alpha = \frac{1}{2}\sum_{\mu s s'} c^\dagger_{i\mu s}\sigma^\alpha_{ss'}c_{i\mu s'} \ , \tag{T.2}$$

where

$$\sigma^x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \quad \sigma^y = \begin{pmatrix} 0 & -\mathrm{i} \\ \mathrm{i} & 0 \end{pmatrix} \quad \sigma^z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \quad \sigma^0 = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} , \tag{T.3}$$

and

$$\sigma^+ = \begin{pmatrix} 0 & 2 \\ 0 & 0 \end{pmatrix} \quad \sigma^\uparrow = \begin{pmatrix} 2 & 0 \\ 0 & 0 \end{pmatrix} \quad \sigma^\downarrow = \begin{pmatrix} 0 & 0 \\ 0 & 2 \end{pmatrix} \quad \sigma^- = \begin{pmatrix} 0 & 0 \\ 2 & 0 \end{pmatrix} . \tag{T.4}$$

With this definition, the number of particles is given by $n = 2S^0$.

The components of this basis are related to the cartesian ones by

$$\begin{cases} S^+ = & S^x + \mathrm{i}S^y \\ S^\uparrow = & \frac{n}{2} + S^z \\ S^\downarrow = & \frac{n}{2} - S^z \\ S^- = & S^x - \mathrm{i}S^y \end{cases} \Rightarrow \begin{cases} S^x = & \frac{S^+ + S^-}{2} \\ S^y = & \frac{S^+ - S^-}{2\mathrm{i}} \\ S^z = & \frac{S^\uparrow - S^\downarrow}{2} \\ n = & S^\uparrow + S^\downarrow \end{cases} \tag{T.5}$$

or, in matrix form,

$$\begin{pmatrix} S^+ \\ S^\uparrow \\ S^\downarrow \\ S^- \end{pmatrix} = \begin{pmatrix} 1 & \mathrm{i} & 0 & 0 \\ 0 & 0 & 1 & \frac{1}{2} \\ 0 & 0 & -1 & \frac{1}{2} \\ 1 & -\mathrm{i} & 0 & 0 \end{pmatrix} \begin{pmatrix} S^x \\ S^y \\ S^z \\ n \end{pmatrix} \Rightarrow \begin{pmatrix} S^x \\ S^y \\ S^z \\ n \end{pmatrix} = \begin{pmatrix} \frac{1}{2} & 0 & 0 & \frac{1}{2} \\ \frac{1}{2\mathrm{i}} & 0 & 0 & -\frac{1}{2\mathrm{i}} \\ 0 & \frac{1}{2} & -\frac{1}{2} & 0 \\ 0 & 1 & 1 & 0 \end{pmatrix} \begin{pmatrix} S^+ \\ S^\uparrow \\ S^\downarrow \\ S^- \end{pmatrix} . \tag{T.6}$$

In spherical coordinates, the cartesian components of the spin are

$$\begin{aligned} S^x &= S \sin\theta \cos\phi \\ S^y &= S \sin\theta \sin\phi \\ S^z &= S \cos\theta \end{aligned} \tag{T.7}$$

The occupation n is independent of the angle. Considering now a primed system of reference that points along the magnetization, the components of $\mathbf{S}'$ (i.e., in the local frame) can be obtained from the coordinates $\mathbf{S}$ (in the global frame) first by rotating it by ϕ around $\hat{\mathbf{z}}$ ($x \to y$) and then around $\hat{\mathbf{y}}'$ (the intermediate axis $z \to x$). The rotation matrices (including the density, which is not affected by the rotation) are given by

$$R_z(\phi) = \begin{pmatrix} \cos\phi & -\sin\phi & 0 & 0 \\ \sin\phi & \cos\phi & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \quad \text{, and} \quad R_y(\theta) = \begin{pmatrix} \cos\theta & 0 & \sin\theta & 0 \\ 0 & 1 & 0 & 0 \\ -\sin\theta & 0 & \cos\theta & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \tag{T.8}$$

Alternatively, the vector $\mathbf{S}$ can be rotated by $-\phi$ around $\hat{\mathbf{z}}$, and then by $-\theta$ around $\hat{\mathbf{y}}$, such that

$$\mathbf{S} = R_y(-\theta) R_z(-\phi) \mathbf{S}' \tag{T.9}$$

In matrix form

$$\begin{aligned} \begin{pmatrix} S'^x \\ S'^y \\ S'^z \\ S'^0 \end{pmatrix} &= \begin{pmatrix} \cos\theta & 0 & -\sin\theta & 0 \\ 0 & 1 & 0 & 0 \\ \sin\theta & 0 & \cos\theta & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} \cos\phi & \sin\phi & 0 & 0 \\ -\sin\phi & \cos\phi & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} S^x \\ S^y \\ S^z \\ S^0 \end{pmatrix} \\ &= \begin{pmatrix} \cos\theta\cos\phi & \cos\theta\sin\phi & -\sin\theta & 0 \\ -\sin\phi & \cos\phi & 0 & 0 \\ \sin\theta\cos\phi & \sin\theta\sin\phi & \cos\theta & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} S^x \\ S^y \\ S^z \\ S^0 \end{pmatrix} . \end{aligned} \tag{T.10}$$

This way, the spin vector in the global frame of reference $\mathbf{S} = \{S\sin\theta\cos\phi, S\sin\theta\sin\phi, S\cos\theta\}$ becomes $\mathbf{S}' = \{0, 0, S\}$ in the local frame.

Transforming to the circular coordinates,

$$\begin{pmatrix} \frac{1}{2} & 0 & 0 & \frac{1}{2} \\ \frac{1}{2\mathrm{i}} & 0 & 0 & -\frac{1}{2\mathrm{i}} \\ 0 & \frac{1}{2} & -\frac{1}{2} & 0 \\ 0 & 1 & 1 & 0 \end{pmatrix} \begin{pmatrix} S'^{+} \\ S'^{\uparrow} \\ S'^{\downarrow} \\ S'^{-} \end{pmatrix} = \begin{pmatrix} \cos\theta\cos\phi & \cos\theta\sin\phi & -\sin\theta & 0 \\ -\sin\phi & \cos\phi & 0 & 0 \\ \sin\theta\cos\phi & \sin\theta\sin\phi & \cos\theta & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} \frac{1}{2} & 0 & 0 & \frac{1}{2} \\ \frac{1}{2\mathrm{i}} & 0 & 0 & -\frac{1}{2\mathrm{i}} \\ 0 & \frac{1}{2} & -\frac{1}{2} & 0 \\ 0 & 1 & 1 & 0 \end{pmatrix} \begin{pmatrix} S^{+} \\ S^{\uparrow} \\ S^{\downarrow} \\ S^{-} \end{pmatrix} \tag{T.11}$$

or

$$\begin{aligned} \begin{pmatrix} S'^{+} \\ S'^{\uparrow} \\ S'^{\downarrow} \\ S'^{-} \end{pmatrix} &= \begin{pmatrix} 1 & \mathrm{i} & 0 & 0 \\ 0 & 0 & 1 & \frac{1}{2} \\ 0 & 0 & -1 & \frac{1}{2} \\ 1 & -\mathrm{i} & 0 & 0 \end{pmatrix} \begin{pmatrix} \cos\theta\cos\phi & \cos\theta\sin\phi & -\sin\theta & 0 \\ -\sin\phi & \cos\phi & 0 & 0 \\ \sin\theta\cos\phi & \sin\theta\sin\phi & \cos\theta & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix} \begin{pmatrix} \frac{1}{2} & 0 & 0 & \frac{1}{2} \\ \frac{1}{2\mathrm{i}} & 0 & 0 & -\frac{1}{2\mathrm{i}} \\ 0 & \frac{1}{2} & -\frac{1}{2} & 0 \\ 0 & 1 & 1 & 0 \end{pmatrix} \begin{pmatrix} S^{+} \\ S^{\uparrow} \\ S^{\downarrow} \\ S^{-} \end{pmatrix} \\ &= \begin{pmatrix} 1 & \mathrm{i} & 0 & 0 \\ 0 & 0 & 1 & \frac{1}{2} \\ 0 & 0 & -1 & \frac{1}{2} \\ 1 & -\mathrm{i} & 0 & 0 \end{pmatrix} \begin{pmatrix} \frac{1}{2}\cos\theta e^{-\mathrm{i}\phi} & -\frac{1}{2}\sin\theta & \frac{1}{2}\sin\theta & \frac{1}{2}\cos\theta e^{\mathrm{i}\phi} \\ -\frac{\mathrm{i}}{2}e^{-\mathrm{i}\phi} & 0 & 0 & \frac{\mathrm{i}}{2}e^{\mathrm{i}\phi} \\ \frac{1}{2}\sin\theta e^{-\mathrm{i}\phi} & \frac{1}{2}\cos\theta & -\frac{1}{2}\cos\theta & \frac{1}{2}\sin\theta e^{\mathrm{i}\phi} \\ 0 & 1 & 1 & 0 \end{pmatrix} \begin{pmatrix} S^{+} \\ S^{\uparrow} \\ S^{\downarrow} \\ S^{-} \end{pmatrix} \\ &= \begin{pmatrix} \frac{1}{2}(1+\cos\theta)e^{-\mathrm{i}\phi} & -\frac{1}{2}\sin\theta & \frac{1}{2}\sin\theta & -\frac{1}{2}(1-\cos\theta)e^{\mathrm{i}\phi} \\ \frac{1}{2}\sin\theta e^{-\mathrm{i}\phi} & \frac{1}{2}(1+\cos\theta) & \frac{1}{2}(1-\cos\theta) & \frac{1}{2}\sin\theta e^{\mathrm{i}\phi} \\ -\frac{1}{2}\sin\theta e^{-\mathrm{i}\phi} & \frac{1}{2}(1-\cos\theta) & \frac{1}{2}(1+\cos\theta) & -\frac{1}{2}\sin\theta e^{\mathrm{i}\phi} \\ -\frac{1}{2}(1-\cos\theta)e^{-\mathrm{i}\phi} & -\frac{1}{2}\sin\theta & \frac{1}{2}\sin\theta & \frac{1}{2}(1+\cos\theta)e^{\mathrm{i}\phi} \end{pmatrix} \begin{pmatrix} S^{+} \\ S^{\uparrow} \\ S^{\downarrow} \\ S^{-} \end{pmatrix} . \end{aligned} \tag{T.12}$$

Transposing and conjugating Eq. (T.12),

$$\begin{pmatrix} S'^{-} & S'^{\uparrow} & S'^{\downarrow} & S'^{+} \end{pmatrix} = \begin{pmatrix} S^{-} & S^{\uparrow} & S^{\downarrow} & S^{+} \end{pmatrix} \begin{pmatrix} \frac{1}{2}(1+\cos\theta)e^{\mathrm{i}\phi} & \frac{1}{2}\sin\theta e^{\mathrm{i}\phi} & -\frac{1}{2}\sin\theta e^{\mathrm{i}\phi} & -\frac{1}{2}(1 - \\ -\frac{1}{2}\sin\theta & \frac{1}{2}(1+\cos\theta) & \frac{1}{2}(1-\cos\theta) & -\frac{1}{2} \\ \frac{1}{2}\sin\theta & \frac{1}{2}(1-\cos\theta) & \frac{1}{2}(1+\cos\theta) & \frac{1}{2}\,\mathrm{s} \\ -\frac{1}{2}(1-\cos\theta)e^{-\mathrm{i}\phi} & \frac{1}{2}\sin\theta e^{-\mathrm{i}\phi} & -\frac{1}{2}\sin\theta e^{-\mathrm{i}\phi} & \frac{1}{2}(1+\mathrm{c} \end{pmatrix} \tag{T.13}$$

The susceptibility given in Eq. (T.1) can be written as

$$\left\langle\!\left\langle \begin{pmatrix} S^{+} \\ S^{\uparrow} \\ S^{\downarrow} \\ S^{-} \end{pmatrix}, \begin{pmatrix} S^{-} & S^{\uparrow} & S^{\downarrow} & S^{+} \end{pmatrix} \right\rangle\!\right\rangle = \begin{pmatrix} \chi^{+-} & \chi^{+\uparrow} & \chi^{+\downarrow} & \chi^{++} \\ \chi^{\uparrow-} & \chi^{\uparrow\uparrow} & \chi^{\uparrow\downarrow} & \chi^{\uparrow+} \\ \chi^{\downarrow-} & \chi^{\downarrow\uparrow} & \chi^{\downarrow\downarrow} & \chi^{\downarrow+} \\ \chi^{--} & \chi^{-\uparrow} & \chi^{-\downarrow} & \chi^{-+} \end{pmatrix} . \tag{T.14}$$

Therefore, the susceptibility can be calculated in the local frame as

$$
\begin{aligned}
&\left\langle\left\langle \begin{pmatrix} S'^{+} \\ S'^{\uparrow} \\ S'^{\downarrow} \\ S'^{-} \end{pmatrix}, \begin{pmatrix} S'^{-} & S'^{\uparrow} & S'^{\downarrow} & S'^{+} \end{pmatrix} \right\rangle\right\rangle \\
&= \left\langle\left\langle \begin{pmatrix} \frac{1}{2}(1+\cos\theta)e^{-\mathrm{i}\phi} & -\frac{1}{2}\sin\theta & \frac{1}{2}\sin\theta & -\frac{1}{2}(1-\cos\theta)e^{\mathrm{i}\phi} \\ \frac{1}{2}\sin\theta e^{-\mathrm{i}\phi} & \frac{1}{2}(1+\cos\theta) & \frac{1}{2}(1-\cos\theta) & \frac{1}{2}\sin\theta e^{\mathrm{i}\phi} \\ -\frac{1}{2}\sin\theta e^{-\mathrm{i}\phi} & \frac{1}{2}(1-\cos\theta) & \frac{1}{2}(1+\cos\theta) & -\frac{1}{2}\sin\theta e^{\mathrm{i}\phi} \\ -\frac{1}{2}(1-\cos\theta)e^{-\mathrm{i}\phi} & -\frac{1}{2}\sin\theta & \frac{1}{2}\sin\theta & \frac{1}{2}(1+\cos\theta)e^{\mathrm{i}\phi} \end{pmatrix} \begin{pmatrix} S^{+} \\ S^{\uparrow} \\ S^{\downarrow} \\ S^{-} \end{pmatrix}, \right. \right. \\
&\left. \left. \begin{pmatrix} S^{-} & S^{\uparrow} & S^{\downarrow} & S^{+} \end{pmatrix} \begin{pmatrix} \frac{1}{2}(1+\cos\theta)e^{\mathrm{i}\phi} & \frac{1}{2}\sin\theta e^{\mathrm{i}\phi} & -\frac{1}{2}\sin\theta e^{\mathrm{i}\phi} & -\frac{1}{2}(1-\cos\theta)e^{\mathrm{i}\phi} \\ -\frac{1}{2}\sin\theta & \frac{1}{2}(1+\cos\theta) & \frac{1}{2}(1-\cos\theta) & -\frac{1}{2}\sin\theta \\ \frac{1}{2}\sin\theta & \frac{1}{2}(1-\cos\theta) & \frac{1}{2}(1+\cos\theta) & \frac{1}{2}\sin\theta \\ -\frac{1}{2}(1-\cos\theta)e^{-\mathrm{i}\phi} & \frac{1}{2}\sin\theta e^{-\mathrm{i}\phi} & -\frac{1}{2}\sin\theta e^{-\mathrm{i}\phi} & \frac{1}{2}(1+\cos\theta)e^{-\mathrm{i}\phi} \end{pmatrix} \right\rangle\right\rangle \\
&= \begin{pmatrix} \frac{1}{2}(1+\cos\theta)e^{-\mathrm{i}\phi} & -\frac{1}{2}\sin\theta & \frac{1}{2}\sin\theta & -\frac{1}{2}(1-\cos\theta)e^{\mathrm{i}\phi} \\ \frac{1}{2}\sin\theta e^{-\mathrm{i}\phi} & \frac{1}{2}(1+\cos\theta) & \frac{1}{2}(1-\cos\theta) & \frac{1}{2}\sin\theta e^{\mathrm{i}\phi} \\ -\frac{1}{2}\sin\theta e^{-\mathrm{i}\phi} & \frac{1}{2}(1-\cos\theta) & \frac{1}{2}(1+\cos\theta) & -\frac{1}{2}\sin\theta e^{\mathrm{i}\phi} \\ -\frac{1}{2}(1-\cos\theta)e^{-\mathrm{i}\phi} & -\frac{1}{2}\sin\theta & \frac{1}{2}\sin\theta & \frac{1}{2}(1+\cos\theta)e^{\mathrm{i}\phi} \end{pmatrix} \begin{pmatrix} \chi^{+-} & \chi^{+\uparrow} & \chi^{+\downarrow} & \chi^{++} \\ \chi^{\uparrow-} & \chi^{\uparrow\uparrow} & \chi^{\uparrow\downarrow} & \chi^{\uparrow+} \\ \chi^{\downarrow-} & \chi^{\downarrow\uparrow} & \chi^{\downarrow\downarrow} & \chi^{\downarrow+} \\ \chi^{--} & \chi^{-\uparrow} & \chi^{-\downarrow} & \chi^{-+} \end{pmatrix} \\
&\quad \begin{pmatrix} \frac{1}{2}(1+\cos\theta)e^{\mathrm{i}\phi} & \frac{1}{2}\sin\theta e^{\mathrm{i}\phi} & -\frac{1}{2}\sin\theta e^{\mathrm{i}\phi} & -\frac{1}{2}(1-\cos\theta)e^{\mathrm{i}\phi} \\ -\frac{1}{2}\sin\theta & \frac{1}{2}(1+\cos\theta) & \frac{1}{2}(1-\cos\theta) & -\frac{1}{2}\sin\theta \\ \frac{1}{2}\sin\theta & \frac{1}{2}(1-\cos\theta) & \frac{1}{2}(1+\cos\theta) & \frac{1}{2}\sin\theta \\ -\frac{1}{2}(1-\cos\theta)e^{-\mathrm{i}\phi} & \frac{1}{2}\sin\theta e^{-\mathrm{i}\phi} & -\frac{1}{2}\sin\theta e^{-\mathrm{i}\phi} & \frac{1}{2}(1+\cos\theta)e^{-\mathrm{i}\phi} \end{pmatrix}.
\end{aligned}
\tag{T.15}
$$

Since these matrices are transforming the susceptibility to the local frame, they are site dependent, i.e.,

$$
\chi'_{ij} = T^{\dagger}(\theta_i, \phi_i)\chi_{ij}T(\theta_j, \phi_j) \tag{T.16}
$$

where $\mathbf{S}_i = (S_i \sin\theta_i \cos\phi_i, S_i \sin\theta_i \sin\phi_i, S_i \cos\theta_i)$.

Appendix U

Total Energy

$$
\begin{aligned}
V_{\mathrm{xc}}^{\mu} &= U_n(\rho^{\mu} - \rho_0^{\mu}) - \frac{1}{2}U_n \sum_{\nu}(\rho^{\nu} - \rho_0^{\nu}) = \frac{\delta E_{\mathrm{xc}}^{n}}{\delta \rho^{\mu}} \\
E_{\mathrm{xc}}^{n} &= \frac{1}{2}U_n \sum_{\mu}(\rho^{\mu} - \rho_0^{\mu})^2 - \frac{1}{4}U_n \left(\sum_{\mu}(\rho^{\mu} - \rho_0^{\mu})\right)^2 \\
&= \frac{1}{2}U_n \sum_{\mu}(\rho^{\mu} - \rho_0^{\mu})^2 - \frac{1}{4}U_n \left(\rho^{\mathrm{tot}} - \rho_0^{\mathrm{tot}}\right)^2 \\
E_{\mathrm{pot}}^{n} &= \sum_{\mu} \rho^{\mu} V^{\mu} \\
&= \left[\sum_{\mu} U_n \rho^{\mu}(\rho^{\mu} - \rho_0^{\mu}) - \frac{1}{2}U_n \rho^{\mu} \sum_{\nu}(\rho^{\nu} - \rho_0^{\nu})\right] \\
&= \sum_{\mu} \left[U_n \rho^{\mu}(\rho^{\mu} - \rho_0^{\mu}) - \frac{1}{2}U_n \rho^{\mu}(\rho^{\mathrm{tot}} - \rho_0^{\mathrm{tot}})\right] \\
&= \sum_{\mu} \left[U_n \rho^{\mu}(\rho^{\mu} - \rho_0^{\mu})\right] - \frac{1}{2}U_n \rho^{\mathrm{tot}}(\rho^{\mathrm{tot}} - \rho_0^{\mathrm{tot}}) \\
\mathbf{B}_{\mathrm{xc}} &= -\frac{1}{2}U_m \mathbf{m} = \frac{\delta E_{\mathrm{xc}}^{m}}{\delta \mathbf{m}} \\
E_{\mathrm{xc}}^{m} &= -\frac{1}{4}U_m m^2 \\
E_{\mathrm{pot}}^{m} &= \mathbf{m} \cdot \mathbf{B} = -\frac{1}{2}U_m m^2 \\
E_{\mathrm{tot}} &= E_{\mathrm{band}} + E_{\mathrm{xc}}^{n} - E_{\mathrm{pot}}^{n} + E_{\mathrm{xc}}^{m} - E_{\mathrm{pot}}^{m} \\
&= E_{\mathrm{band}} + \frac{1}{2}U_n \sum_{\mu}(\rho^{\mu} - \rho_0^{\mu})^2 - \frac{1}{4}U_n \left(\rho^{\mathrm{tot}} - \rho_0^{\mathrm{tot}}\right)^2 - \sum_{\mu} \left[U_n \rho^{\mu}(\rho^{\mu} - \rho_0^{\mu})\right] + \frac{1}{2}U_n \rho^{\mathrm{tot}}(\rho^{\mathrm{tot}} - \rho_0^{\mathrm{tot}}) + \frac{1}{4}U \\
&= E_{\mathrm{band}} - \frac{1}{2}U_n \sum_{\mu} \left[(\rho^{\mu})^2 - (\rho_0^{\mu})^2\right] + \frac{1}{4}U_n \left[(\rho^{\mathrm{tot}})^2 - (\rho_0^{\mathrm{tot}})^2\right] + \frac{1}{4}U_m m^2
\end{aligned}
\tag{U.1}
$$

Appendix V

Derivatives of the Green function

Eq. M.15:

$$
\begin{aligned}
(E-H)G =& 1 \\
\frac{\partial\left[(E-H)G\right]}{\partial \mathbf{m}} =& 0 \\
-\frac{\partial H}{\partial \mathbf{m}_j}G+(E-H)\frac{\partial G}{\partial \mathbf{m}_j} =& 0 \\
\frac{\partial G}{\partial \mathbf{m}_j} =& G\frac{\partial H}{\partial \mathbf{m}_j}G \\
\frac{\partial G_{ii}}{\partial \mathbf{m}_j} =& \sum_m G_{im}\left[\frac{\partial H}{\partial \mathbf{m}_j}\right]_{mm}G_{mi} \\
\frac{\partial G_{ii}}{\partial \mathbf{m}_j} =& G_{ij}\left[\frac{\partial H}{\partial \mathbf{m}_j}\right]_{jj}G_{ji}
\end{aligned}
\tag{V.1}
$$

$$
\begin{aligned}
(E-H)G =& 1 \\
\frac{\partial\left[(E-H)G\right]}{\partial E} =& 0 \\
G+(E-H)\frac{\partial G}{\partial E} =& 0 \\
(E-H)\frac{\partial G}{\partial E} =& -G \\
\frac{\partial G}{\partial E} =& -G^2
\end{aligned}
\tag{V.2}
$$