Derivation of Optical Response Coefficients

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Introduction

In this paper, we will examine how the results from the paper "Second-Order Optical Response in Semiconductors" (1999) by J. E. Sipe and A. I. Shkrebtii. In this paper we will analytically derive expressions, using nonlinear susceptibility $\chi_2(-\omega_\Sigma; \omega_\beta, \omega_\Gamma)$, for injection currents (called "circular photocurrent") and shift currents.

Own Notes

The Dipole Hamiltonian

We start with the many-body Hamiltonian:

$$H(t) = \int \psi^\dagger(\mathbf{x}, t)\mathcal{H}(t)\psi(\boldsymbol{x}, t)d\mathbf{x} + H_{\text{rest}} \tag{1}$$

Where $\psi(\boldsymbol{x}, t)$ is the electron field operator from a Heisenberg description (i.e. $\{\psi(\boldsymbol{x}, t), \psi^\dagger(\boldsymbol{x'}, t)\} = \delta(\mathbf{x} - \mathbf{x}')$ is satisfied), and we define $\mathcal{H}(t) = \frac{1}{2m}[\frac{\hbar}{i}\nabla - \frac{e}{c}\mathbf{A}]^2 + V(\mathbf{x})$. Here $V(\mathbf{x})$ is the periodic potential.

We now define a new operator, $\widetilde{\psi}(\mathbf{x}, t) = \psi(\mathbf{x}, t)e^{-i\mathbf{K}(t)\cdot\mathbf{x}}$. In defining this quantity, we find that:

$$\begin{aligned}
&\implies i\hbar\frac{d}{dt}\psi(\mathbf{x}, t) = [\psi(\mathbf{x}, t), H(t)] \\
&\implies i\hbar\frac{d}{dt}\left(\widetilde{\psi}(\mathbf{x}, t)e^{i\mathbf{K}(t)\cdot\mathbf{x}}\right) = [\widetilde{\psi}(\mathbf{x}, t)e^{i\mathbf{K}(t)\cdot\mathbf{x}}, H(t)] \\
&\implies i\hbar\frac{d}{dt}\left(\widetilde{\psi}(\mathbf{x}, t)\right) = [\widetilde{\psi}(\mathbf{x}, t), H_{\text{eff}}(t)]
\end{aligned}$$

In the last line, we made use of the fact that $i\hbar\psi\frac{d}{dt}e^{i\mathbf{k}(t)\cdot\mathbf{x}} = i\psi e^{i\mathbf{k}(t)\cdot\mathbf{x}}(i\mathbf{x}\cdot\mathbf{F}) = -e\psi e^{i\mathbf{k}(t)\cdot\mathbf{x}}\mathbf{x}\cdot\mathbf{E}$. After making use of this fact, we can go and relate that $H_{\text{eff}}(t) = \int \widetilde{\psi}^\dagger(\mathbf{x}, t)(\mathcal{H}_0(t) - e\mathbf{x}\cdot\mathbf{E}(t))\widetilde{\psi}(\boldsymbol{x}, t)d\mathbf{x} + H_{\text{rest}}$ where $\mathcal{H}_0(t) = [\frac{\hbar}{i}\nabla]^2 + V(\mathbf{x})$.

Here we will be using the notation that $A(k)$ is the Berry connection, so $A(\mathbf{k}) = \psi^*(\mathbf{k})\nabla_\mathbf{k}\psi(\mathbf{k})$.

Current Density Operator

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The current density operator is defined via

$$\mathbf{J}(t) = \frac{e}{m}\int \frac{d\mathbf{x}}{\Omega}\psi^{\dagger}[\frac{\hbar}{i}\nabla - \frac{e}{c}\mathbf{A}]\psi \tag{2}$$

However, we evoke $\widetilde{\psi}(\mathbf{x},t) = \psi(\mathbf{x},t)e^{-i\mathbf{K}(t)\cdot\mathbf{x}}$ to find that the $\mathbf{A}$ term cancels out when we recognize that $\hbar\frac{d\mathbf{k}}{dt} = e\mathbf{E} = e\frac{d\mathbf{A}}{dt}$. We now have:

$$\mathbf{J}(t) = \frac{e}{m}\int \frac{d\mathbf{x}}{\Omega}\widetilde{\psi}^{\dagger}[\frac{\hbar}{i}\nabla]\widetilde{\psi} \tag{3}$$

$$= e\sum_{n,m}\int \frac{d\mathbf{k}}{\Omega}\mathbf{v}_{nm}a_n^{\dagger}(k)a_m(k) \tag{4}$$

We are now in a position to introduce the polarization vectors:

$$\mathbf{P}_{\text{inter}}(t) = e\int \frac{d\mathbf{k}}{\Omega}\sum_{n,m}\mathbf{r}_{nm}(\mathbf{k})a_n^{\dagger}(\mathbf{k})a_m(\mathbf{k}) \tag{5}$$

$$\mathbf{P}_{\text{intra}}(t) = ie\int \frac{d\mathbf{k}}{\Omega}\sum_{n}[a_n^{\dagger}(\mathbf{k})\partial a_n(\mathbf{k})] \tag{6}$$

Such that it is now true that our Hamiltonian can be written as $H(t) = H_0 - \Omega(\mathbf{P}_{\text{inter}}(t) + \mathbf{P}_{\text{intra}}(t)) \cdot \mathbf{E} + H_{\text{rest}}$. We next declare that $\mathbf{P}(t) = \mathbf{P}_{\text{inter}}(t) + \mathbf{P}_{\text{intra}}(t)$ so that we know $\mathbf{J}(t) = \frac{d\mathbf{P}(t)}{dt}$

1 Derivations of Material in Notes

Perturbation on the Correlation Function

We start with the correlation function with time evolution (which I have derived up to a sign):

$$\frac{\partial c_{mn}}{\partial t} + i\omega_{mn}c_{mn} = -\frac{eE^b(t)}{\hbar}c_{mn;b} + \frac{ieE^b(t)}{\hbar}\sum_p\left(r^b_{mp}c_{pn} - c_{mp}r^b_{pn}\right) \tag{7}$$

. To derive the n-th orders of $C^{(n)}_{mn}$, we use a perturbative approach. We start with the 0-th order, $C^{(0)}_{mn} = \delta_{mn}f_n$

To derive the next order, according to Sipe, we take the RHS of Equation (7) at $C^{(0)}_m n$ and solve the differential to find $C^{(1)}_m n$. Please see work below:

Derivation of $C^{(1)}_{mn}$

$$\frac{\partial}{\partial t} C^{(1)}_{mn} + i\omega_{mn} C^{(1)}_{mn} = -\frac{eE^b_\beta}{\hbar} e^{-i\omega_\beta t}\Big(\underbrace{\cancel{\frac{\partial C^{(0)}_{mn}}{\partial k^b}}}_{\boxed{\text{Q: Why is this 0?}}}{}^{\to 0} - i(A^b_{mm} - A^b_{nn}) C^{(0)}_{mn}$$

$$+ \frac{ie}{\hbar} E^b_\beta e^{-i\omega_\beta t} \sum_p \left(\Gamma^b_{mp} C^{(0)}_{pn} - C^{(0)}_{mp}\Gamma^b_{pn}\right)\Big)$$

Plug in $C^{(0)}_{mn} = f_n \delta_{mn}$

$$\frac{\partial}{\partial t} C^{(1)}_{mn} + i\omega_{mn} C^{(1)}_{mn} = \frac{-eE^b_\beta e^{-i\omega_\beta t}}{\hbar}\Big[\cancel{-i(A^b_{mm} - A^b_{nn}) f_n \delta_{mn}}^{\to 0}$$

$$- i \sum_p \left(\Gamma^b_{mp} f_n \delta_{pn} - f_p \delta_{mp} \Gamma^b_{pn}\right)\Big]$$

$$= \frac{-eE^b_\beta}{\hbar} e^{-i\omega_\beta t}\left[-i\left(\Gamma^b_{mn} f_n - f_m \Gamma^b_{mn}\right)\right]$$

$$\frac{\partial}{\partial t} C^{(1)}_{mn} + i\omega_{mn} C^{(1)}_{mn} = \frac{ie}{\hbar}\left[\Gamma^b_{mn} f_{nm}\right] E^b_\beta e^{-i\omega_\beta t}$$

Ansatz: $C^{(1)}_{mn} = A e^{-i\omega_\beta t} E^b_\beta$

$$A(-i\omega_\beta) + (i\omega_{mn})A = \frac{ie}{\hbar}\left[\Gamma^b_{mn} f_{nm}\right]$$

$$A = \frac{e\Gamma^b_{mn} f_{nm}}{\hbar(\omega_{mn} - \omega_\beta)}$$

$$\therefore \quad C^{(1)}_{mn} = \frac{e\Gamma^b_{mn} f_{nm}}{\hbar(\omega_{mn} - \omega_\beta)} E^b_\beta e^{-i\omega_\beta t}$$

We then rederive the result of Sipe.

$$c_{mn}^{(1)} = \mathcal{B}_{mn}^{b} E_{\beta}^{b} e^{-i\omega_{\beta} t} \tag{8}$$

where $\mathcal{B}_{mn}^{b} \equiv \frac{e f_{nm} r_{mn}^{b}}{\hbar(\omega_{mn} - \omega_{\beta})}$

We preform an identical treatment to find $C_{mn}^{(2)}$, whereupon we leave $C_{mn}^{(2)}$ on the LHS and plug in $C_{mn}^{(1)}$ on the RHS. We also introduce a new Electric field $E^{c}(t) = E_{\gamma}^{c} e^{-i\omega_{\gamma} t}$. The derivation is shown below:

Derivation of $C^{(2)}_{mn}$

$$\frac{\partial}{\partial t} C^{(2)}_{mn} + i\omega_{mn} C^{(2)}_{mn} = -\frac{eE^c_\gamma}{\hbar} e^{-i\omega_\gamma t}\left(\frac{\partial C^{(1)}_{mn}}{\partial k^c} - i\left(A^c_{mm}-A^c_{nn}\right)C^{(1)}_{mn}\right)$$

$$+\frac{ie}{\hbar}E^c_\gamma e^{-i\omega_\gamma t}\sum_p\left(\Gamma^c_{mp} C^{(1)}_{pn} - C^{(1)}_{mp}\Gamma^c_{pn}\right)$$

If we define $B^b_{mn} = \dfrac{e f_{nm} r^b_{mn}}{\hbar(\omega_{mn}-\omega_\beta)}$ so that

$$C^{(1)} = B^b_{mn} E^b_\beta e^{-i\omega_\beta t}$$

$$\frac{\partial}{\partial t} C^{(2)}_{mn} + i\omega_{mn} C^{(2)}_{mn} = -\frac{eE^c_\gamma}{\hbar} e^{-i\omega_\gamma t}\Big[\frac{\partial}{\partial k^c}\left(B^b_{mn} E^b_\beta e^{-i\omega_\beta t}\right)$$

$$-i\left(A^c_{mm}-A^c_{nn}\right)B^b_{mn} E^b_\beta e^{-i\omega_\beta t}$$

$$-i\sum_p \Gamma^c_{mp} B^b_{pn} E^b_\beta e^{-i\omega_\beta t} - B^b_{mp} E^b_\beta e^{-i\omega_\beta t}\Gamma^c_{pn}\Big]$$

$$= -\frac{e}{\hbar}E^c_\gamma E^b_\beta e^{-i(\omega_\gamma+\omega_\beta)t}\Big[B^b_{mn;c} - i\sum_p \Gamma^c_{mp} B^b_{pn} - B^b_{mp}\Gamma^c_{pn}\Big]$$

Ansatz: let $C^{(2)}_{mn} = A e^{-i(\omega_\gamma+\omega_\beta)t}\left(\frac{e}{\hbar}\right)E^c_\gamma E^b_\beta$

$$\Rightarrow A\left(-i(\omega_\gamma+\omega_\beta)\right) + i\omega_{mn}A = B^b_{mn;c} - i\sum_p \Gamma^c_{mp} B^b_{pn} - B^b_{mp}\Gamma^c_{pn}$$

$$\Rightarrow A = \frac{1}{i(\omega_{mn}-\omega_\gamma-\omega_\beta)}\left(B^b_{mn;c} - i\sum_p\left(\Gamma^c_{mp} B^b_{pn} - B^b_{mp}\Gamma^c_{pn}\right)\right)$$

So
$$C^{(2)}_{mn} = \frac{-e}{i\hbar(\omega_{mn}-\omega_\gamma-\omega_\beta)}E^c_\gamma E^b_\beta e^{-i(\omega_\gamma+\omega_\beta)t}\left(B^b_{mn;c} - i\sum_p\left(\Gamma^c_{mp} B^b_{pn} - B^b_{mp}\Gamma^c_{pn}\right)\right)$$

This again agrees with Sipe.

$$c_{mn}^{(2)} = \frac{e}{i\hbar\left(\omega_{mn} - \omega_{\Sigma}\right)}[-\mathcal{B}_{mn;c}^{b} + i\sum_{p}\left(r_{mp}^{c}\mathcal{B}_{pn}^{b} - \mathcal{B}_{mp}^{b}r_{pn}^{c}\right)]E_{\beta}^{b}E_{\gamma}^{c}e^{-i\omega_{\Sigma}t} \tag{9}$$

1.1.1 First Order Response

We can use the expression $\langle \mathbf{J}(t)\rangle^{(1)} = \frac{d\langle \mathbf{P}_{inter}(t)\rangle^{(1)}}{dt}$ since $\langle \mathbf{J}_{intra}(t)\rangle^{(1)} = 0$. Therefore, we find Equation (5) that $\langle \mathbf{P}_{inter}(t)\rangle^{(1)} = e\int \frac{d\mathbf{k}}{(2\pi)^3}\sum_{n,m} c_{mn}^{(1)}\mathbf{r}_{nm}$.

We now define a susceptibility such that $\langle P_{\text{inter}}^{a}(t)\rangle^{(1)} = \chi_1^{ab}\left(-\omega_\beta;\omega_\beta\right)E_\beta^b e^{-i\omega_\beta t}$. Then it follows that $\chi_1^{ab}(-\omega;\omega) = \frac{e^2}{\hbar}\int\frac{d\mathbf{k}}{8\pi^3}\sum_{n,m}\frac{f_{nm}r_{nm}^{a}r_{mn}^{b}}{(\omega_{mn}-\omega-i\eta)}$. Here, η is a small frequency which can be taken to 0 at the end of the day. We can simplify this even further with the aid of a principal value. The principal value is defined such that $\lim_{\eta\to 0^+}\int_{-\infty}^{\infty}\frac{f(x)}{x-x_0+i\eta}dx = P\int_{-\infty}^{\infty}\frac{f(x)}{x-x_0}dx - i\pi f\left(x_0\right)$. So for $\chi_1^{ab}(-\omega;\omega)$, we choose our function to be a Dirac Delta function, $\delta\left(\omega_{mn}-\omega\right)$. The advantage of this distinction is we can now separate $\chi_1^{ab}(-\omega;\omega)$ into its real and imaginary components,

$$\mathrm{Re}\left[\chi_1^{ab}(-\omega;\omega)\right] = \frac{e^2}{\hbar}\int\frac{d\mathbf{k}}{8\pi^3}\sum_{n,m}\mathcal{P}\frac{f_{nm}r_{nm}^{a}r_{mn}^{b}}{(\omega_{mn}-\omega)} \tag{10}$$

$$\mathrm{Im}\left[\chi_1^{ab}(-\omega;\omega)\right] = \frac{e^2\pi}{\hbar}\int\frac{d\mathbf{k}}{8\pi^3}\sum_{n,m}f_{nm}r_{nm}^{a}r_{mn}^{b}\delta\left(\omega_{mn}-\omega\right) \tag{11}$$

1.1.2 Second Order Response

For the sake of clarity, we will now break up intraband into the first order response from the correlation function and the second order response from the correlation function, $\langle \mathbf{J}_{intra}(t)\rangle^{(2)} = \langle \mathbf{J}_{intra}(t)\rangle^{(I)} + \langle \mathbf{J}_{intra}(t)\rangle^{(II)}$. We will use Equation (6) to aid in this, as well as Equations (8) and (9). The process for finding the expression of $\langle \mathbf{J}_{intra}(t)\rangle^{(I)}$ and $\langle \mathbf{J}_{intra}(t)\rangle^{(II)}$ is shown below:

$$\langle P_{intra}(t)\rangle = \left\langle ie\int \frac{d\vec{k}}{\Omega}\sum_n \frac{1}{2}\left[a_n^{\dagger}\frac{\partial a_n(\vec{k})}{\partial\vec{k}} - \frac{\partial a_n^{\dagger}}{\partial\vec{k}}a_n(\vec{k})\right] - i A_{nn}\vec{a}_n^{\dagger}a_n\right\rangle$$

$$\langle\vec{J}(t)\rangle^{(2)} = \frac{d\langle P_{intra}^{(2)}(t)\rangle}{dt} = e\int\frac{d\vec{k}}{\Omega}\sum_{nm}\overline{V}_{nm}(\vec{k},t)\,a_n^{\dagger}(\vec{k})\,a_m(\vec{k})$$

$$\& \quad \overline{V}^a_{nm} = V^a_{nn}(\vec{k})\,\delta_{nm} - \frac{e\Gamma^b_{nm;a}E^b}{\hbar}$$

$$\langle J(t)\rangle^{(2)} = e\int\frac{d\vec{k}}{(2\pi)^3}\sum_{nm}\overline{V}_{nm}(\vec{k},t)\,C_{nm}$$

$$= e\int\frac{d\vec{k}}{(2\pi)^3}\sum_{nm}\left(V_{nn}\delta_{nm} - \frac{e\Gamma^b_{nm;a}}{\hbar}E^b\right)C_{nm}$$

we see that $V^a_{nn}\delta_{nm}C^{(1)}_{nm} = 0$ b/c $f_{nm}\delta_{nm} = 0$, so this term vanishes. Also since $C^{(2)}_{nm} \propto E^2$, we let the term $\frac{e\Gamma^b_{nm;a}}{\hbar}E^b C^{(2)}_{nm} \sim 0$. Then, we find:

$$\langle J(t)\rangle^{(2)} = e\int\frac{d\vec{k}}{(2\pi)^3}\Bigg(\underbrace{\sum_n V^a_{nn}C^{(2)}_{nn}}_{\langle J\rangle^{(II)}} + \underbrace{\sum_{n,m}\frac{e\Gamma^b_{nm;a}}{\hbar}E^b C^{(1)}_{nm}}_{\langle J\rangle^{(I)}}\Bigg)$$

Make sure the argument for dropping the $E^b c_{mn}$ holds up. After adjusting for the minus sign I inexplicably dropped at the end, the result tells us that we should define:

$$\langle J^a_{intra}(t)\rangle^{(I)} = -\frac{e^2}{\hbar}\int\frac{d\mathbf{k}}{(2\pi)^3}\sum_{n,m}c^{(1)}_{mn}r^b_{nm;a}E^b(t) \tag{12}$$

.

$$\langle J^a_{intra}(t)\rangle^{(II)} = e\int \frac{d\mathbf{k}}{8\pi^3}\sum_n c^{(2)}_{nn} v^a_{nn} \tag{13}$$

We evaluate Equation (13). We plug in the result of $C^{(2)}_{nn}$ according to Equation (9). The derivation is shown below:

$$\langle J^a_{intra}(t)\rangle^{(II)} = e\int \frac{d\vec{k}}{(2\pi)^3}\sum_n C^{(2)}_{nn}\, V^a_{nn}$$

$$= \frac{e^2}{i\hbar(\omega_{nn}-\omega_\beta-\omega_\gamma)}\sum_n\Big[-\mathcal{B}^b_{nn;c} + i\sum_p\big(r^c_{np}\mathcal{B}^b_{pn} - \mathcal{B}^b_{np}\, r^c_{pn}\big)\Big] V^a_{nn} E^b_\beta E^c_\gamma e^{-i(\omega_\beta+\omega_\gamma)t}$$

since $\mathcal{B}^b_{mn} \equiv \dfrac{e\, f_{nm} r^b_{mn}}{\hbar(\omega_{mn}-\omega_\beta)}$ at $m=n$, $f_{nn}=0$ so
$\mathcal{B}^b_{nn}=0$

$$\langle J^a_{intra}(t)\rangle^{(II)} = \frac{-e^2\,\cancel{i}}{\cancel{i}\hbar(\omega_\beta+\omega_\gamma)}\sum_n\sum_p\big(r^c_{np}\mathcal{B}^b_{pn} - \mathcal{B}^b_{np}\, r^c_{pn}\big) V^a_{nn} E^b_\beta E^c_\gamma e^{''}$$

define $\Delta_{mn}(\vec{k}) \equiv \vec{V}_{mm}(\vec{k}) - \vec{V}_{nn}(\vec{k})$

$$\langle J^a_{intra}(t)\rangle^{(II)} = \frac{-e^2}{\hbar(\omega_\beta+\omega_\gamma)}\sum_{n,p}\underbrace{\big(V_{nn}\, r^c_{np}\mathcal{B}^b_{pn} - \mathcal{B}^b_{np}\, r^c_{pn} V_{nn}\big)}_{\text{since indices are summed up, switch the dummy index on the last term } n\leftrightarrow p} E^b_\beta E^c_\gamma$$

$$\langle J^a_{intra}(t)\rangle^{(II)} = \frac{-e^2}{\hbar(\omega_\beta+\omega_\gamma)}\sum_{n,p}\big(V_{nn}\, r^c_{np}\mathcal{B}^b_{pn} - V_{pp}\,\mathcal{B}^b_{pn}\, r^c_{np}\big) E^b_\beta E^c_\gamma e^{''}$$

$$\langle J^a_{intra}(t)\rangle^{(II)} = \frac{e^2}{\hbar(\omega_\beta+\omega_\gamma)}\sum_{n,p}\big(\Delta_{pn}\, r^c_{np}\mathcal{B}^b_{pn}\big) E^b_\beta E^c_\gamma e^{-i(\omega_\beta+\omega_\gamma)t}$$

This agrees with Sipe. We define the sum according to $K^{abc} = \frac{e^2}{\hbar\omega_\Sigma}\left[\int \frac{d\mathbf{k}}{8\pi^3}\sum_{n,m}\Delta^a_{mn} r^c_{nm}\mathcal{B}^b_{mn}\right]$. So our end result:

$$< J^a_{\text{intra}}(t) >^{(II)} = K^{abc} E^b_\beta E^c_\gamma e^{-i\omega_\Sigma t} \tag{14}$$

2 Calculations

2.0.1 Effective Hamiltonian Without Separating Between Inter and Intra Bands

In the Sipe literature, we are given the following Hamiltonian:

$$H_{eff}(t) = \int \widetilde{\psi}^{\dagger}(\mathbf{x},t) \left[\mathcal{H}_o - e\mathbf{x}\cdot\mathbf{E}(t)\right] \widetilde{\psi}(\mathbf{x},t) d\mathbf{x} + H_{rest} \tag{15}$$

We now considering transforming $\widetilde{\psi}(\mathbf{x},t)$ into fermionic opterators in momentum space such that $\widetilde{\psi}(\mathbf{x},t) = \sum_n \int d\mathbf{k} a_n(\mathbf{k})\psi_n(\mathbf{k};\mathbf{x})$. We will also make use of the relation from Blount:

$$\langle n\mathbf{k}|\mathbf{x}|m\mathbf{k}'\rangle = \delta\left(\mathbf{k}-\mathbf{k}'\right) A_{nm}(\mathbf{k}) + \delta_{nm}\left[+i\frac{\partial}{\partial \mathbf{k}}\delta\left(\mathbf{k}-\mathbf{k}'\right)\right] \tag{16}$$

and

$$\mathcal{H}_o\psi_n(\mathbf{k};\mathbf{x}) = \hbar\omega_n(\mathbf{k})\psi_n(\mathbf{k};\mathbf{x}) \tag{17}$$

The derivation is given in:

$$H_{eff} = \int \left\{ \sum_{n,m} \int d\vec{k} \left[\underbrace{\psi_n^{\dagger} a_n^{\dagger} \left(\omega_m a_m \psi_m\right)}_{\int \psi_n^{\dagger}\psi_m d\vec{x} = \delta_{nm}} - eE_j \sum_{n,m} \int d\vec{k}\, \psi_n^{\dagger} a_n^{\dagger} x_j a_m \psi_m \right\} d\vec{x}$$

From above, we have:

$$H_{eff} = \int d\vec{k} \left\{ \sum_n \hbar\omega_n a_n^{\dagger} a_n - eE_j \sum_{n,m} a_n^{\dagger} \left(\delta(\vec{k}-\vec{k}')\vec{A}^j_{nm}(\vec{k}) + i\frac{\partial}{\partial k^j}\delta(\vec{k}-\vec{k}')\right) a_m \right\}$$

$$H_{eff} = \int d\vec{k} \left\{ \hbar\omega_n a_n^{\dagger} a_n - eE_j \sum_{n,m} A^j_{nm} a_n^{\dagger} a_m + i a_n^{\dagger} \frac{\partial}{\partial k^j} a_m \right\}$$

$$H_{eff} = \int d\vec{k} \left\{ \sum_n \hbar\omega_n a_n^{\dagger} a_n - eE_j \sum_{n,m} A^j_{nm} a_n^{\dagger} a_m + \frac{i}{2}\left(a_n^{\dagger} \frac{\partial}{\partial k^j} a_n - \left(\frac{\partial}{\partial k^j} a_n^{\dagger}\right) a_n \right) \right\}$$

$$H_{eff} = \int d\vec{k} \left\{ \sum_n \left[\hbar\omega_n a^{\dagger} a_n + \frac{i}{2}\left(a_n^{\dagger} \frac{\partial}{\partial k^j} a_n - \left(\frac{\partial}{\partial k^j} a_n^{\dagger}\right) a_n \right)\right] - eE_j \sum_{n,m} \left[A^j_{nm} a_n^{\dagger} a_m \right] \right\}$$

The end result is:

$$\hat{H}_{\text{eff}} = \int d\mathbf{k}\Big\{ \sum_n \Big[\hbar\omega_n a_n^\dagger(\mathbf{k})a_n(\mathbf{k}) - \frac{ieE_j}{2}(a_n^\dagger(\mathbf{k})\frac{\partial a_n(\mathbf{k})}{\partial k^j} - \frac{\partial a_n^\dagger(\mathbf{k})}{\partial k^j}a_n(\mathbf{k})\Big] - eE_j \sum_{n,m} A_{nm}^j a_n^\dagger(\mathrm{k})a_m(\mathbf{k})\Big\} \quad (18)$$

The Correlation Function

We now consider the correlation function without consideration to inter and intra band separation. We now define a polarization vector $\mathbf{P}(t)$ such that: $H_{eff}(t) = H_o - \Omega\mathbf{P}(t)\cdot\mathbf{E}(t) + H_{rest}$ and Ω is a reference volume. We can now consider the correlation function $c_{mn}(\mathbf{k}) = \frac{\Omega}{(2\pi)^2} < a_n^\dagger(\mathbf{k})a_m(\mathbf{k}) >$. We now consider the evolution of the correlation function through the Heisenberg Equation of Motion:

$\frac{\Omega}{(2\pi)^2}\frac{d}{dt} < a_n^\dagger(\mathbf{k})a_m(\mathbf{k}) >=< [H_{\text{eff}},\, \frac{\Omega}{(2\pi)^2}a_n^\dagger(\mathbf{k})a_m(\mathbf{k})] >$.