We start with the expression for the susceptibility for the intraband transtitions,

$$\chi^{s,\ell}_{i,\mathrm{abc}} = -\frac{e^3}{\Omega\hbar^2\omega_3}\sum_{mn\mathbf{k}} \frac{\mathcal{V}^{\Sigma,\mathrm{a},\ell}_{mn}}{\omega^S_{nm}-\omega_3}\left(\frac{f_{mn}r^{\mathrm{b}}_{nm}}{\omega^S_{nm}-\omega_\beta}\right)_{;k^{\mathrm{c}}}, \tag{1}$$

chii

where s denotes *surface* and S refers to the *scissors* correction. This expression diverges as $\omega_3 \to 0$. To eliminate this divergence we take the partial fraction expansion,

$$\begin{aligned} I &= C\left[-\frac{1}{2(\omega^S_{nm})^2}\frac{1}{\omega^S_{nm}-\omega} + \frac{2}{(\omega^S_{nm})^2}\frac{1}{\omega^S_{nm}-2\omega} + \frac{1}{2(\omega^S_{nm})^2}\frac{1}{\omega}\right] \\ &- D\left[-\frac{3}{2(\omega^S_{nm})^2}\frac{1}{\omega^S_{nm}-\omega} + \frac{4}{(\omega^S_{nm})^3}\frac{1}{\omega^S_{nm}-2\omega} + \frac{1}{2(\omega^S_{nm})^3}\frac{1}{\omega} - \frac{1}{2(\omega^S_{nm})^2}\frac{1}{(\omega^S_{nm}-\omega)^2}\right], \end{aligned} \tag{2}$$

where $C = f_{mn}\mathcal{V}^{\Sigma,\mathrm{a}}_{mn}(r^{\mathrm{LDA,b}}_{nm})_{;k^{\mathrm{c}}}$, and $D = f_{mn}\mathcal{V}^{\Sigma,\mathrm{a}}_{mn}r^{\mathrm{b}}_{nm}\Delta^{\mathrm{c}}_{nm}$.

Time-reversal symmetry leads to the following relationships:

$$\begin{aligned} \mathbf{r}_{mn}(\mathbf{k}) &= \mathbf{r}_{nm}(-\mathbf{k}), \\ \mathbf{r}_{mn;\mathbf{k}}(\mathbf{k}) &= -\mathbf{r}_{nm;\mathbf{k}}(-\mathbf{k}), \\ \mathcal{V}^{\Sigma,\mathrm{a}}_{mn}(-\mathbf{k}) &= -\mathcal{V}^{\Sigma,\mathrm{a}}_{nm}(\mathbf{k}), \\ \omega^S_{mn}(-\mathbf{k}) &= \omega^S_{mn}(\mathbf{k}), \\ \Delta^a_{nm}(-\mathbf{k}) &= -\Delta^a_{nm}(\mathbf{k}). \end{aligned}$$

For a clean cold semiconductor, $f_n = 1$ for an occupied or valence ($n = v$) band, and $f_n = 0$ for an empty or conduction ($n = c$) band independent of $\mathbf{k}$, and $f_{nm} = -f_{mn}$.

The $\frac{1}{\omega}$ terms cancel each other out. The last term in the second line of (pfi 2) is dealt with as follows,

$$\begin{aligned} \frac{D}{2(\omega^S_{nm})^2}\frac{1}{(\omega^S_{nm}-\omega)^2} &= \frac{f_{mn}}{2}\frac{\mathcal{V}^{\Sigma,\mathrm{a}}_{mn}r^{\mathrm{b}}_{nm}}{(\omega^S_{nm})^2}\frac{\Delta^{\mathrm{c}}_{nm}}{(\omega^S_{nm}-\omega)^2} = \frac{f_{mn}}{2}\frac{\mathcal{V}^{\Sigma,\mathrm{a}}_{mn}r^{\mathrm{b}}_{nm}}{(\omega^S_{nm})^2}\left(\frac{1}{\omega^S_{nm}-\omega}\right)_{;k^{\mathrm{c}}} \\ &= -\frac{f_{mn}}{2}\left(\frac{\mathcal{V}^{\Sigma,\mathrm{a}}_{mn}r^{\mathrm{b}}_{nm}}{(\omega^S_{nm})^2}\right)_{;k^{\mathrm{c}}}\frac{1}{\omega^S_{nm}-\omega}. \end{aligned} \tag{3}$$

We use the fact that

$$(\omega^S_{nm})_{;k^{\mathrm{c}}} = (\omega^{\mathrm{LDA}}_{nm})_{;k^{\mathrm{c}}} = \frac{p^{\mathrm{c}}_{nn}-p^{\mathrm{c}}_{mm}}{m_e} \equiv \Delta^{\mathrm{c}}_{nm}, \tag{4}$$

wk

and for the last line, we performed an integration by parts over the Brillouin zone, where the contribution from the edges vanishes.

1 Generalized Derivative

Using the chain rule we obtain

$$\left(\frac{\mathcal{V}^{\Sigma,\mathrm{a}}_{mn} r^{\mathrm{b}}_{nm}}{(\omega^S_{nm})^2}\right)_{;k^{\mathrm{c}}} = \frac{r^{\mathrm{b}}_{nm}}{(\omega^S_{nm})^2}\left(\mathcal{V}^{\Sigma,\mathrm{a}}_{mn}\right)_{;k^{\mathrm{c}}} + \frac{\mathcal{V}^{\Sigma,\mathrm{a}}_{mn}}{(\omega^S_{nm})^2}\left(r^{\mathrm{b}}_{nm}\right)_{;k^{\mathrm{c}}} - \frac{\mathcal{V}^{\Sigma,\mathrm{a}}_{mn} r^{\mathrm{b}}_{nm}}{2(\omega^S_{nm})^3}\left(\omega^S_{nm}\right)_{;k^{\mathrm{c}}} . \qquad (5) \quad \boxed{\texttt{chrn}}$$

The individual terms for this expression can be expanded as follows. First,

$$\left(\omega^S_{nm}\right)_{;k^{\mathrm{c}}} = \Delta^{\mathrm{LDA,c}}_{nm}, \qquad (6) \quad \boxed{\texttt{eli.1}}$$

and,

$$(r^{\mathrm{b}}_{nm})_{;k^{\mathrm{a}}} \approx \frac{r^{\mathrm{a}}_{nm}\Delta^{\mathrm{LDA,b}}_{mn} + r^{\mathrm{b}}_{nm}\Delta^{\mathrm{LDA,a}}_{mn}}{\omega^{\mathrm{LDA}}_{nm}} + \frac{i}{\omega^{\mathrm{LDA}}_{nm}}\sum_{\ell}\left(\omega^{\mathrm{LDA}}_{\ell m} r^{\mathrm{a}}_{n\ell} r^{\mathrm{b}}_{\ell m} - \omega^{\mathrm{LDA}}_{n\ell} r^{\mathrm{b}}_{n\ell} r^{\mathrm{a}}_{\ell m}\right). \qquad (7) \quad \boxed{\texttt{eli.2}}$$

1.1 Generalized derivative for $\mathcal{V}^{\Sigma,\mathbf{a},\ell}_{nm}$

We must include the generalized derivative for $\mathcal{V}^{\Sigma,\mathrm{a},\ell}_{nm}$. We can seperate the expression into its components,

$$\left(\mathcal{V}^{\Sigma,\mathrm{a},\ell}_{nm}\right)_{;k^{\mathrm{b}}} = \left(\mathcal{V}^{\mathrm{LDA,a},\ell}_{nm}\right)_{;k^{\mathrm{b}}} + \left(\mathcal{V}^{S,\mathrm{a},\ell}_{nm}\right)_{;k^{\mathrm{b}}}, \qquad (8)$$

where,

$$\left(\mathcal{V}^{\mathrm{LDA,a}}_{nm}\right)_{;k^{\mathrm{b}}} = \frac{1}{2}\sum_{q}\left((v^{\mathrm{LDA,a}}_{nq})_{;k^{\mathrm{b}}}\mathcal{F}^{\ell}_{qm} + v^{\mathrm{LDA,a}}_{nq}(\mathcal{F}^{\ell}_{qm})_{;k^{\mathrm{b}}} + (\mathcal{F}^{\ell}_{nq})_{;k^{\mathrm{b}}} v^{\mathrm{LDA,a}}_{qm} + \mathcal{F}^{\ell}_{nq}(v^{\mathrm{LDA,a}}_{qm})_{;k^{\mathrm{b}}}\right), \qquad (9) \quad \boxed{\texttt{a.2}}$$

and $\left(v^{\mathrm{LDA,a}}_{nn}\right)_{;k^{\mathrm{b}}}$ is given by

$$\left(v^{\mathrm{LDA,a}}_{nn}\right)_{;k^{\mathrm{b}}} = \frac{\hbar}{m_e}\delta_{\mathrm{ab}} - \sum_{\ell\neq n}\omega^{\mathrm{LDA}}_{\ell n}\left(r^{\mathrm{a}}_{n\ell} r^{\mathrm{b}}_{\ell n} + r^{\mathrm{b}}_{n\ell} r^{\mathrm{a}}_{\ell n}\right). \qquad (10)$$

Lastly,

$$\mathcal{V}^{S,\mathrm{a},\ell}_{nm} = \frac{1}{2}\sum_{q}\left((v^{S,\mathrm{a}}_{nq})_{;k^{\mathrm{b}}}\mathcal{F}_{qm} + v^{S,\mathrm{a}}_{nq}(\mathcal{F}_{qm})_{;k^{\mathrm{b}}} + (\mathcal{F}_{nq})_{;k^{\mathrm{b}}} v^{S,\mathrm{a}}_{qm} + \mathcal{F}_{nq}(v^{S,\mathrm{a}}_{qm})_{;k^{\mathrm{b}}}\right), \qquad (11) \quad \boxed{\texttt{a.3b}}$$

where $\left(v_{nm}^{S,\mathrm{a}}\right)_{;k^{\mathrm{b}}}$ is given by

$$\left(v_{nm}^{S,\mathrm{a}}\right)_{;k^{\mathrm{b}}} = i\Delta f_{mn}(r_{nm}^{\mathrm{a}})_{;k^{\mathrm{b}}}. \tag{12}$$

For the I term of (2), we notice that the energy denominators are invariant under $\mathbf{k} \to -\mathbf{k}$, and then we only look at the numerators, then

$$\begin{aligned}
C \to f_{mn}\mathcal{V}_{mn}^{\Sigma,\mathrm{a}}(r_{nm}^{\mathrm{b}})_{;k^{\mathrm{c}}}|_{\mathbf{k}} + f_{mn}\mathcal{V}_{mn}^{\Sigma,\mathrm{a}}(r_{nm}^{\mathrm{b}})_{;k^{\mathrm{c}}}|_{-\mathbf{k}} &= f_{mn}\left[\mathcal{V}_{mn}^{\Sigma,\mathrm{a}}(r_{nm}^{\mathrm{b}})_{;k^{\mathrm{c}}}|_{\mathbf{k}} + (-\mathcal{V}_{nm}^{\Sigma,\mathrm{a}})(-(r_{mn}^{\mathrm{b}})_{;k^{\mathrm{c}}})|_{\mathbf{k}}\right] \\
&= f_{mn}\left[\mathcal{V}_{mn}^{\Sigma,\mathrm{a}}(r_{nm}^{\mathrm{b}})_{;k^{\mathrm{c}}} + \mathcal{V}_{nm}^{\Sigma,\mathrm{a}}(r_{mn}^{\mathrm{b}})_{;k^{\mathrm{c}}}\right] \\
&= f_{mn}\left[\mathcal{V}_{mn}^{\Sigma,\mathrm{a}}(r_{nm}^{\mathrm{b}})_{;k^{\mathrm{c}}} + (\mathcal{V}_{mn}^{\Sigma,\mathrm{a}}(r_{nm}^{\mathrm{b}})_{;k^{\mathrm{c}}})^*\right] \\
&= m_e f_{mn}\omega_{mn}\left[i\mathcal{R}_{mn}^{\mathrm{a}}(r_{nm}^{\mathrm{b}})_{;k^{\mathrm{c}}} + (i\mathcal{R}_{mn}^{\mathrm{a}}(r_{nm}^{\mathrm{b}})_{;k^{\mathrm{c}}})^*\right] \\
&= i m_e f_{mn}\omega_{mn}\left[\mathcal{R}_{mn}^{\mathrm{a}}(r_{nm}^{\mathrm{b}})_{;k^{\mathrm{c}}} - (\mathcal{R}_{mn}^{\mathrm{a}}(r_{nm}^{\mathrm{b}})_{;k^{\mathrm{c}}})^*\right] \\
&= -2m_e f_{mn}\omega_{mn}\mathrm{Im}[\mathcal{R}_{mn}^{\mathrm{a}}(r_{nm}^{\mathrm{b}})_{;k^{\mathrm{c}}}],
\end{aligned} \tag{13}$$