

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

$$\chi_{i,\text{abc}}^{s,\ell} = -\frac{e^3}{\Omega\hbar^2\omega_3} \sum_{mn\mathbf{k}} \frac{\mathcal{V}_{mn}^{\Sigma,\text{a},\ell}}{\omega_{nm}^S - \omega_3} \left(\frac{f_{mn}r_{nm}^b}{\omega_{nm}^S - \omega_\beta} \right)_{;k^c}, \quad (1) \quad \boxed{\text{chi i}}$$

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

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

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

Time-reversal symmetry leads to the following relationships:

$$\begin{aligned} \mathbf{r}_{mn}(\mathbf{k}) &= \mathbf{r}_{nm}(-\mathbf{k}), \\ \mathbf{r}_{mn;\mathbf{k}}(\mathbf{k}) &= -\mathbf{r}_{nm;\mathbf{k}}(-\mathbf{k}), \\ \mathcal{V}_{mn}^{\Sigma,\text{a}}(-\mathbf{k}) &= -\mathcal{V}_{nm}^{\Sigma,\text{a}}(\mathbf{k}), \\ \omega_{mn}^S(-\mathbf{k}) &= \omega_{nm}^S(\mathbf{k}), \\ \Delta_{nm}^a(-\mathbf{k}) &= -\Delta_{nm}^a(\mathbf{k}). \end{aligned}$$

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. We notice that the energy denominators are invariant under $\mathbf{k} \rightarrow -\mathbf{k}$, and then we only look at the numerators, then

$$\begin{aligned} C \rightarrow f_{mn} \mathcal{V}_{mn}^{\Sigma,\text{a}}(r_{nm}^b)_{;k^c} |_{\mathbf{k}} + f_{mn} \mathcal{V}_{mn}^{\Sigma,\text{a}}(r_{nm}^b)_{;k^c} |_{-\mathbf{k}} &= f_{mn} \left[\mathcal{V}_{mn}^{\Sigma,\text{a}}(r_{nm}^b)_{;k^c} |_{\mathbf{k}} + (-\mathcal{V}_{nm}^{\Sigma,\text{a}})(-(r_{mn}^b)_{;k^c}) |_{\mathbf{k}} \right] \\ &= f_{mn} \left[\mathcal{V}_{mn}^{\Sigma,\text{a}}(r_{nm}^b)_{;k^c} + \mathcal{V}_{nm}^{\Sigma,\text{a}}(r_{mn}^b)_{;k^c} \right] \\ &= f_{mn} \left[\mathcal{V}_{mn}^{\Sigma,\text{a}}(r_{nm}^b)_{;k^c} + (\mathcal{V}_{mn}^{\Sigma,\text{a}}(r_{nm}^b)_{;k^c})^* \right] \end{aligned} \quad (3)$$

The last term in the second line of [\(2\)](#) is dealt with as follows,

$$\begin{aligned} \frac{D}{2(\omega_{nm}^S)^2} \frac{1}{(\omega_{nm}^S - \omega)^2} &= \frac{f_{mn}}{2} \frac{\mathcal{V}_{mn}^{\Sigma,\text{a}} r_{nm}^b}{(\omega_{nm}^S)^2} \frac{\Delta_{nm}^c}{(\omega_{nm}^S - \omega)^2} = \frac{f_{mn}}{2} \frac{\mathcal{V}_{mn}^{\Sigma,\text{a}} r_{nm}^b}{(\omega_{nm}^S)^2} \left(\frac{1}{\omega_{nm}^S - \omega} \right)_{;k^c} \\ &= -\frac{f_{mn}}{2} \left(\frac{\mathcal{V}_{mn}^{\Sigma,\text{a}} r_{nm}^b}{(\omega_{nm}^S)^2} \right)_{;k^c} \frac{1}{\omega_{nm}^S - \omega}. \end{aligned} \quad (4)$$

We use the fact that

$$(\omega_{nm}^S)_{;k^c} = (\omega_{nm}^{\text{LDA}})_{;k^c} = \frac{p_{nn}^c - p_{mm}^c}{m_e} \equiv \Delta_{nm}^c, \quad (5) \quad \boxed{\text{wk}}$$

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

$$\left(\frac{\mathcal{V}_{mn}^{\Sigma,a,b} r_{nm}^b}{(\omega_{nm}^S)^2} \right)_{;k^c} = \frac{r_{nm}^b}{(\omega_{nm}^S)^2} (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c} + \frac{\mathcal{V}_{mn}^{\Sigma,a}}{(\omega_{nm}^S)^2} (r_{nm}^b)_{;k^c} - \frac{\mathcal{V}_{mn}^{\Sigma,a,b} r_{nm}^b}{2(\omega_{nm}^S)^3} (\omega_{nm}^S)_{;k^c}. \quad (6) \quad \boxed{\text{chn}}$$

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

$$(\omega_{nm}^S)_{;k^c} = \Delta_{nm}^{\text{LDA},c}, \quad (7) \quad \boxed{\text{eli.1}}$$

and,

$$(r_{nm}^b)_{;k^a} \approx \frac{r_{nm}^a \Delta_{mn}^{\text{LDA},b} + r_{nm}^b \Delta_{mn}^{\text{LDA},a}}{\omega_{nm}^{\text{LDA}}} + \frac{i}{\omega_{nm}^{\text{LDA}}} \sum_{\ell} \left(\omega_{\ell m}^{\text{LDA}} r_{n\ell}^a r_{\ell m}^b - \omega_{n\ell}^{\text{LDA}} r_{n\ell}^b r_{\ell m}^a \right). \quad (8) \quad \boxed{\text{eli.2}}$$

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

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

$$\left(\mathcal{V}_{nm}^{\Sigma,a,\ell} \right)_{;k^b} = \left(\mathcal{V}_{nm}^{\text{LDA},a,\ell} \right)_{;k^b} + \left(\mathcal{V}_{nm}^{S,a,\ell} \right)_{;k^b}, \quad (9)$$

where,

$$(\mathcal{V}_{nm}^{\text{LDA},a})_{;k^b} = \frac{1}{2} \sum_q \left((v_{nq}^{\text{LDA},a})_{;k^b} \mathcal{F}_{qm}^{\ell} + v_{nq}^{\text{LDA},a} (\mathcal{F}_{qm}^{\ell})_{;k^b} + (\mathcal{F}_{nq}^{\ell})_{;k^b} v_{qm}^{\text{LDA},a} + \mathcal{F}_{nq}^{\ell} (v_{qm}^{\text{LDA},a})_{;k^b} \right), \quad (10) \quad \boxed{\text{a.2}}$$

and $(v_{nn}^{\text{LDA},a})_{;k^b}$ is given by

$$(v_{nn}^{\text{LDA},\text{a}})_{;k^{\text{b}}} = \frac{\hbar}{m_e} \delta_{\text{ab}} - \sum_{\ell \neq n} \omega_{\ell n}^{\text{LDA}} \left(r_{n\ell}^{\text{a}} r_{\ell n}^{\text{b}} + r_{n\ell}^{\text{b}} r_{\ell n}^{\text{a}} \right). \quad (11)$$

Lastly,

$$\mathcal{V}_{nm}^{S,\text{a},\ell} = \frac{1}{2} \sum_q \left((v_{nq}^{S,\text{a}})_{;k^{\text{b}}} \mathcal{F}_{qm} + v_{nq}^{S,\text{a}} (\mathcal{F}_{qm})_{;k^{\text{b}}} + (\mathcal{F}_{nq})_{;k^{\text{b}}} v_{qm}^{S,\text{a}} + \mathcal{F}_{nq} (v_{qm}^{S,\text{a}})_{;k^{\text{b}}} \right), \quad (12) \quad \boxed{\text{a.3b}}$$

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

$$(v_{nm}^{S,\text{a}})_{;k^{\text{b}}} = i \Delta f_{mn} (r_{nm}^{\text{a}})_{;k^{\text{b}}}. \quad (13)$$