

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

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

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

Time-reversal symmetry leads to the following relationships:

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

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. The last term in the second line of (2) is dealt with as follows,

$$\begin{aligned} \frac{D}{2(\omega_{nm}^S)^2} \frac{1}{(\omega_{nm}^S - \omega)^2} &= \frac{f_{mn}}{2} \frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{(\omega_{nm}^S)^2} \frac{\Delta_{nm}^c}{(\omega_{nm}^S - \omega)^2} = \frac{f_{mn}}{2} \frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{(\omega_{nm}^S)^2} \left( \frac{1}{\omega_{nm}^S - \omega} \right)_{;k^c} \\ &= -\frac{f_{mn}}{2} \left( \frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{(\omega_{nm}^S)^2} \right)_{;k^c} \frac{1}{\omega_{nm}^S - \omega}. \quad (3) \end{aligned}$$

We use the fact that

$$(\omega_{nm}^S)_{;k^c} = (\omega_{nm}^{\text{LDA}})_{;k^c} = \frac{p_{nn}^c - p_{mm}^c}{m_e} \equiv \Delta_{nm}^c, \quad (4) \quad \boxed{\text{wk}}$$

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

$$\left( \frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{(\omega_{nm}^S)^2} \right)_{;k^c} = \frac{r_{nm}^b}{(\omega_{nm}^S)^2} (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c} + \frac{\mathcal{V}_{mn}^{\Sigma,a}}{(\omega_{nm}^S)^2} (r_{nm}^b)_{;k^c} - \frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{2(\omega_{nm}^S)^3} (\omega_{nm}^S)_{;k^c}. \quad (5) \quad \boxed{\text{chn}}$$

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

$$(\omega_{nm}^S)_{;k^c} = \Delta_{nm}^{\text{LDA},c}, \quad (6) \quad \boxed{\text{eli.1}}$$

and,

$$(r_{nm}^b)_{;k^a} \approx \frac{r_{nm}^a \Delta_{mn}^{\text{LDA},b} + r_{nm}^b \Delta_{mn}^{\text{LDA},a}}{\omega_{nm}^{\text{LDA}}} + \frac{i}{\omega_{nm}^{\text{LDA}}} \sum_{\ell} \left( \omega_{\ell m}^{\text{LDA}} r_{n\ell}^a r_{\ell m}^b - \omega_{n\ell}^{\text{LDA}} r_{n\ell}^b r_{\ell m}^a \right). \quad (7) \quad \boxed{\text{eli.2}}$$

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

$$(\mathcal{V}_{nm}^{\Sigma,a,\ell})_{;k^b} = (\mathcal{V}_{nm}^{\text{LDA},a,\ell})_{;k^b} + (\mathcal{V}_{nm}^{S,a,\ell})_{;k^b}, \quad (8)$$

where,

$$\begin{aligned} (\mathcal{V}_{nm}^{\text{LDA},a})_{;k^b} &= \frac{1}{2} \sum_q \left( v_{nq}^{\text{LDA},a} \mathcal{F}_{qm}^{\ell} + \mathcal{F}_{nq}^{\ell} v_{qm}^{\text{LDA},a} \right)_{;k^b} \\ &= \frac{1}{2} \sum_q \left( (v_{nq}^{\text{LDA},a})_{;k^b} \mathcal{F}_{qm}^{\ell} + v_{nq}^{\text{LDA},a} (\mathcal{F}_{qm}^{\ell})_{;k^b} + (\mathcal{F}_{nq}^{\ell})_{;k^b} v_{qm}^{\text{LDA},a} + \mathcal{F}_{nq}^{\ell} (v_{qm}^{\text{LDA},a})_{;k^b} \right) \end{aligned} \quad (9)$$

(10)

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

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

Lastly,

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

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

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