

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

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

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

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

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 allows us to write,  $\mathbf{r}_{mn}(\mathbf{k}) = \mathbf{r}_{nm}(-\mathbf{k})$ ,  $\mathbf{r}_{mn;\mathbf{k}}(\mathbf{k}) = -\mathbf{r}_{nm;\mathbf{k}}(-\mathbf{k})$ ,  $\mathcal{V}_{mn}^{\Sigma,a}(-\mathbf{k}) = -\mathcal{V}_{nm}^{\Sigma,a}(\mathbf{k})$ ,  $\omega_{mn}^S(-\mathbf{k}) = \omega_{nm}^S(\mathbf{k})$ , and  $\Delta_{nm}^a(-\mathbf{k}) = -\Delta_{nm}^a(\mathbf{k})$ . Also, 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 last term in the second line of Eq. (2) is dealt with as follows.

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

where we used Eqs. (??) and (??), 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

$$\left( \frac{\mathcal{V}_{mn}^{\Sigma,a}r_{nm}^b}{(\omega_{nm}^S)^2} \right)_{;k^c} = \frac{r_{nm}^b}{(\omega_{nm}^S)^2} (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c} + \frac{\mathcal{V}_{mn}^{\Sigma,a}}{(\omega_{nm}^S)^2} (r_{nm}^b)_{;k^c} - \frac{\mathcal{V}_{mn}^{\Sigma,a}r_{nm}^b}{2(\omega_{nm}^S)^3} (\omega_{nm}^S)_{;k^c}, \quad (4)$$

where in the appendix ?? we show how to calculate  $(\mathcal{V}_{nm}^{\Sigma,a})_{;k^b}$ . For  $(\omega_{nm}^S)_{;k^c}$  we simply use Eq. (??), that using Eq. (??) gives

$$(\omega_{nm}^S)_{;k^c} = \Delta_{nm}^{\text{LDA},c}, \quad (5)$$

and from Eq. (??)

$$(r_{nm}^b)_{;k^a} \approx \frac{r_{nm}^a\Delta_{mn}^{\text{LDA},b} + r_{nm}^b\Delta_{mn}^{\text{LDA},a}}{\omega_{nm}^{\text{LDA}}} + \frac{i}{\omega_{nm}^{\text{LDA}}} \sum_{\ell} \left( \omega_{\ell m}^{\text{LDA}} r_{n\ell}^a r_{\ell m}^b - \omega_{n\ell}^{\text{LDA}} r_{n\ell}^b r_{\ell m}^a \right), \quad (6)$$