

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,

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

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:

$$\begin{aligned} \mathbf{r}_{mn}(\mathbf{k})|_{-\mathbf{k}} &= \mathbf{r}_{nm}(\mathbf{k})|_{\mathbf{k}}, \\ (\mathbf{r}_{mn})_{;\mathbf{k}}(\mathbf{k})|_{-\mathbf{k}} &= (-\mathbf{r}_{nm})_{;\mathbf{k}}(\mathbf{k})|_{\mathbf{k}}, \\ \mathcal{V}_{mn}^{\Sigma,a}(\mathbf{k})|_{-\mathbf{k}} &= -\mathcal{V}_{nm}^{\Sigma,a}(\mathbf{k})|_{\mathbf{k}}, \\ (\mathcal{V}_{mn}^{\Sigma,a})_{;\mathbf{k}}(\mathbf{k})|_{-\mathbf{k}} &= (\mathcal{V}_{nm}^{\Sigma,a})_{;\mathbf{k}}(\mathbf{k})|_{\mathbf{k}}, \\ \omega_{mn}^S(\mathbf{k})|_{-\mathbf{k}} &= \omega_{nm}^S(\mathbf{k})|_{\mathbf{k}}, \\ \Delta_{nm}^a(\mathbf{k})|_{-\mathbf{k}} &= -\Delta_{nm}^a(\mathbf{k})|_{\mathbf{k}}. \end{aligned} \quad (3)$$

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

$$\begin{aligned} C &\rightarrow f_{mn}\mathcal{V}_{mn}^{\Sigma,a}(r_{nm}^{\text{LDA},b})_{;k^c}|_{\mathbf{k}} + f_{mn}\mathcal{V}_{mn}^{\Sigma,a}(r_{nm}^{\text{LDA},b})_{;k^c}|_{-\mathbf{k}} \\ &= f_{mn} \left[\mathcal{V}_{mn}^{\Sigma,a}(r_{nm}^{\text{LDA},b})_{;k^c}|_{\mathbf{k}} + (-\mathcal{V}_{nm}^{\Sigma,a})(-r_{mn}^{\text{LDA},b})_{;k^c}|_{\mathbf{k}} \right] \\ &= f_{mn} \left[\mathcal{V}_{mn}^{\Sigma,a}(r_{nm}^{\text{LDA},b})_{;k^c} + \mathcal{V}_{nm}^{\Sigma,a}(r_{mn}^{\text{LDA},b})_{;k^c} \right] \\ &= f_{mn} \left[\mathcal{V}_{mn}^{\Sigma,a}(r_{nm}^{\text{LDA},b})_{;k^c} + \left(\mathcal{V}_{mn}^{\Sigma,a}(r_{nm}^{\text{LDA},b})_{;k^c} \right)^* \right] \\ &= 2f_{mn} \text{Re} \left[\mathcal{V}_{mn}^{\Sigma,a}(r_{nm}^{\text{LDA},b})_{;k^c} \right], \end{aligned} \quad (4)$$

and likewise,

$$\begin{aligned}
D &\rightarrow f_{mn} \mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b \Delta_{nm}^c |_{\mathbf{k}} + f_{mn} \mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b \Delta_{nm}^c |_{-\mathbf{k}} \\
&= f_{mn} \left[\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b \Delta_{nm}^c |_{\mathbf{k}} + (-\mathcal{V}_{nm}^{\Sigma,a}) r_{mn}^b (-\Delta_{nm}^c) |_{\mathbf{k}} \right] \\
&= f_{mn} \left[\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b + \mathcal{V}_{nm}^{\Sigma,a} r_{mn}^b \right] \Delta_{nm}^c \\
&= f_{mn} \left[\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b + \left(\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b \right)^* \right] \Delta_{nm}^c \\
&= 2f_{mn} \text{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b \right] \Delta_{nm}^c.
\end{aligned} \tag{5}$$

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

$$\begin{aligned}
\frac{D}{2(\omega_{nm}^S)^2} \frac{1}{(\omega_{nm}^S - \omega)^2} &= \frac{f_{mn}}{2} \frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{(\omega_{nm}^S)^2} \frac{\Delta_{nm}^c}{(\omega_{nm}^S - \omega)^2} = \frac{f_{mn}}{2} \frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{(\omega_{nm}^S)^2} \left(\frac{1}{\omega_{nm}^S - \omega} \right)_{;k^c} \\
&= -\frac{f_{mn}}{2} \left(\frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{(\omega_{nm}^S)^2} \right)_{;k^c} \frac{1}{\omega_{nm}^S - \omega}.
\end{aligned} \tag{6}$$

We use the fact that

$$(\omega_{nm}^S)_{;k^c} = (\omega_{nm}^{\text{LDA}})_{;k^c} = \frac{p_{nn}^c - p_{mm}^c}{m_e} \equiv \Delta_{nm}^c, \tag{7}$$

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

$$\left(\frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{(\omega_{nm}^S)^2} \right)_{;k^c} = \frac{r_{nm}^b}{(\omega_{nm}^S)^2} (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c} + \frac{\mathcal{V}_{mn}^{\Sigma,a}}{(\omega_{nm}^S)^2} (r_{nm}^b)_{;k^c} - \frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{2(\omega_{nm}^S)^3} (\omega_{nm}^S)_{;k^c}. \tag{8}$$

We will check each term of (8) over $\mathbf{k} \rightarrow -\mathbf{k}$ using the relations in (3). The first term is reduced to

$$\begin{aligned}
\frac{r_{nm}^b}{(\omega_{nm}^S)^2} (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c} |_{\mathbf{k}} + \frac{r_{nm}^b}{(\omega_{nm}^S)^2} (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c} |_{-\mathbf{k}} &= \frac{r_{nm}^b}{(\omega_{nm}^S)^2} (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c} |_{\mathbf{k}} + \frac{r_{mn}^b}{(\omega_{nm}^S)^2} (\mathcal{V}_{nm}^{\Sigma,a})_{;k^c} |_{\mathbf{k}} \\
&= \frac{1}{(\omega_{nm}^S)^2} \left[r_{nm}^b (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c} + \left(r_{nm}^b (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c} \right)^* \right] \\
&= \frac{2}{(\omega_{nm}^S)^2} \text{Re} \left[r_{nm}^b (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c} \right],
\end{aligned} \tag{9}$$

the second term is reduced to

$$\begin{aligned}
\frac{\mathcal{V}_{mn}^{\Sigma,a}}{(\omega_{nm}^S)^2} \left(r_{nm}^b \right)_{;k^c} | \mathbf{k} + \frac{\mathcal{V}_{mn}^{\Sigma,a}}{(\omega_{nm}^S)^2} \left(r_{nm}^b \right)_{;k^c} | -\mathbf{k} &= \frac{\mathcal{V}_{mn}^{\Sigma,a}}{(\omega_{nm}^S)^2} \left(r_{nm}^b \right)_{;k^c} | \mathbf{k} + \frac{\mathcal{V}_{nm}^{\Sigma,a}}{(\omega_{nm}^S)^2} \left(r_{mn}^b \right)_{;k^c} | \mathbf{k} \\
&= \frac{1}{(\omega_{nm}^S)^2} \left[\mathcal{V}_{mn}^{\Sigma,a} \left(r_{nm}^b \right)_{;k^c} + \left(\mathcal{V}_{mn}^{\Sigma,a} \left(r_{nm}^b \right)_{;k^c} \right)^* \right] \\
&= \frac{2}{(\omega_{nm}^S)^2} \text{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} \left(r_{nm}^b \right)_{;k^c} \right], \tag{10}
\end{aligned}$$

and by using (7), the third term is reduced to

$$\begin{aligned}
\frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{2(\omega_{nm}^S)^3} (\omega_{nm}^S)_{;k^c} | \mathbf{k} + \frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{2(\omega_{nm}^S)^3} (\omega_{nm}^S)_{;k^c} | -\mathbf{k} &= \frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{2(\omega_{nm}^S)^3} \Delta_{nm}^c | \mathbf{k} + \frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{2(\omega_{nm}^S)^3} \Delta_{nm}^c | -\mathbf{k} \\
&= \frac{\mathcal{V}_{nm}^{\Sigma,a} r_{mn}^b}{2(\omega_{nm}^S)^3} \Delta_{nm}^c | \mathbf{k} + \frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{2(\omega_{nm}^S)^3} \Delta_{nm}^c | \mathbf{k} \\
&= \frac{1}{2(\omega_{nm}^S)^3} \left[\mathcal{V}_{nm}^{\Sigma,a} r_{mn}^b + \left(\mathcal{V}_{nm}^{\Sigma,a} r_{mn}^b \right)^* \right] \Delta_{nm}^c \\
&= \frac{1}{(\omega_{nm}^S)^3} \text{Re} \left[\mathcal{V}_{nm}^{\Sigma,a} r_{mn}^b \right] \Delta_{nm}^c. \tag{11}
\end{aligned}$$

Combining the results from (9), (10), and (11) into (8),

$$\begin{aligned}
& - \frac{f_{mn}}{2} \left[\left(\frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{(\omega_{nm}^S)^2} \right)_{;k^c} | \mathbf{k} + \left(\frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b}{(\omega_{nm}^S)^2} \right)_{;k^c} | -\mathbf{k} \right] \frac{1}{\omega_{nm}^S - \omega} = \\
& - f_{mn} \left(2 \text{Re} \left[r_{nm}^b (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c} \right] + 2 \text{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} (r_{nm}^b)_{;k^c} \right] - \frac{1}{\omega_{nm}^S} \text{Re} \left[\mathcal{V}_{nm}^{\Sigma,a} r_{mn}^b \right] \Delta_{nm}^c \right) \frac{1}{2(\omega_{nm}^S)^2} \frac{1}{\omega_{nm}^S - \omega}. \tag{12}
\end{aligned}$$

We have all the elements to be substituted in (2). We substitute (4), (5), and (12) in (2),

$$\begin{aligned}
I &= \left[- \frac{2f_{mn} \text{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} \left(r_{nm}^{\text{LDA},b} \right)_{;k^c} \right]}{2(\omega_{nm}^S)^2} \frac{1}{\omega_{nm}^S - \omega} + \frac{4f_{mn} \text{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} \left(r_{nm}^{\text{LDA},b} \right)_{;k^c} \right]}{(\omega_{nm}^S)^2} \frac{1}{\omega_{nm}^S - 2\omega} \right] \\
& - \left[- \frac{6f_{mn} \text{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b \right] \Delta_{nm}^c}{2(\omega_{nm}^S)^2} \frac{1}{\omega_{nm}^S - \omega} + \frac{8f_{mn} \text{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b \right] \Delta_{nm}^c}{(\omega_{nm}^S)^3} \frac{1}{\omega_{nm}^S - 2\omega} \right. \\
& \left. + \frac{f_{mn} \left(2 \text{Re} \left[r_{nm}^b (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c} \right] + 2 \text{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} (r_{nm}^b)_{;k^c} \right] - \frac{1}{\omega_{nm}^S} \text{Re} \left[\mathcal{V}_{nm}^{\Sigma,a} r_{mn}^b \right] \Delta_{nm}^c \right)}{2(\omega_{nm}^S)^2} \frac{1}{\omega_{nm}^S - \omega} \right].
\end{aligned}$$

If we simplify and homogenize,

$$\begin{aligned}
I = & -\frac{2f_{mn} \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} \left(r_{nm}^{\text{LDA},b} \right)_{;k^c} \right]}{2(\omega_{nm}^S)^2} \frac{1}{\omega_{nm}^S - \omega} + \frac{4f_{mn} \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} \left(r_{nm}^{\text{LDA},b} \right)_{;k^c} \right]}{(\omega_{nm}^S)^2} \frac{1}{\omega_{nm}^S - 2\omega} \\
& + \frac{6f_{mn} \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b \right] \Delta_{nm}^c}{2(\omega_{nm}^S)^2} \frac{1}{\omega_{nm}^S - \omega} - \frac{8f_{mn} \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b \right] \Delta_{nm}^c}{(\omega_{nm}^S)^3} \frac{1}{\omega_{nm}^S - 2\omega} \\
& - \frac{2f_{mn} \operatorname{Re} \left[r_{nm}^b \left(\mathcal{V}_{mn}^{\Sigma,a} \right)_{;k^c} \right]}{2(\omega_{nm}^S)^2} \frac{1}{\omega_{nm}^S - \omega} \\
& - \frac{2f_{mn} \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} \left(r_{nm}^b \right)_{;k^c} \right]}{2(\omega_{nm}^S)^2} \frac{1}{\omega_{nm}^S - \omega} \\
& + \frac{f_{mn} \operatorname{Re} \left[\mathcal{V}_{nm}^{\Sigma,a} r_{mn}^b \right] \Delta_{nm}^c}{2(\omega_{nm}^S)^3} \frac{1}{\omega_{nm}^S - \omega}, \tag{13}
\end{aligned}$$

we conveniently collect the terms in columns of ω and 2ω . We can now express the susceptibility in terms of ω and 2ω . Separating the 2ω terms and substituting in (1),

$$\begin{aligned}
I_{2\omega} = & -\frac{e^3}{\hbar^2} \sum_{mn\mathbf{k}} \left[\frac{4f_{mn} \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} \left(r_{nm}^{\text{LDA},b} \right)_{;k^c} \right]}{(\omega_{nm}^S)^2} - \frac{8f_{mn} \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b \right] \Delta_{nm}^c}{(\omega_{nm}^S)^3} \right] \frac{1}{\omega_{nm}^S - 2\omega} \\
= & \frac{e^3}{\hbar^2} \sum_{mn\mathbf{k}} \frac{4f_{mn}}{(\omega_{nm}^S)^2} \left[-\operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} \left(r_{nm}^{\text{LDA},b} \right)_{;k^c} \right] + \frac{2 \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b \right] \Delta_{nm}^c}{(\omega_{nm}^S)} \right] \frac{1}{\omega_{nm}^S - 2\omega} \tag{14}
\end{aligned}$$

Work in progress: I need to understand more about $\eta = 0$ and the switch to ω_{cv} in order to get equation (90) and (A16).

We do the same for the ω terms in (13) and substitute in (1),

$$\begin{aligned}
I_{\omega} = & -\frac{e^3}{2\hbar^2} \sum_{mn\mathbf{k}} \left[-\frac{2f_{mn} \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} \left(r_{nm}^{\text{LDA},b} \right)_{;k^c} \right]}{(\omega_{nm}^S)^2} + \frac{6f_{mn} \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^b \right] \Delta_{nm}^c}{(\omega_{nm}^S)^2} \right. \\
& - \frac{2f_{mn} \operatorname{Re} \left[r_{nm}^b \left(\mathcal{V}_{mn}^{\Sigma,a} \right)_{;k^c} \right]}{(\omega_{nm}^S)^2} - \frac{2f_{mn} \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} \left(r_{nm}^b \right)_{;k^c} \right]}{(\omega_{nm}^S)^2} \\
& \left. + \frac{f_{mn} \operatorname{Re} \left[\mathcal{V}_{nm}^{\Sigma,a} r_{mn}^b \right] \Delta_{nm}^c}{(\omega_{nm}^S)^3} \right] \frac{1}{\omega_{nm}^S - \omega}, \tag{15}
\end{aligned}$$

and we reduce in the same way as (14),

$$\begin{aligned}
I_\omega = \frac{e^3}{2\hbar^2} \sum_{mn\mathbf{k}} \frac{f_{mn}}{(\omega_{nm}^S)^2} & \left[2 \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,\mathbf{a}} \left(r_{nm}^{\text{LDA},\mathbf{b}} \right)_{;k^c} \right] \right. \\
& - 6 \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,\mathbf{a}} r_{nm}^{\mathbf{b}} \right] \Delta_{nm}^c + 2 \operatorname{Re} \left[r_{nm}^{\mathbf{b}} \left(\mathcal{V}_{mn}^{\Sigma,\mathbf{a}} \right)_{;k^c} \right] \\
& \left. + 2 \operatorname{Re} \left[\mathcal{V}_{mn}^{\Sigma,\mathbf{a}} \left(r_{nm}^{\mathbf{b}} \right)_{;k^c} \right] - \frac{\operatorname{Re} \left[\mathcal{V}_{nm}^{\Sigma,\mathbf{a}} r_{mn}^{\mathbf{b}} \right] \Delta_{nm}^c}{(\omega_{nm}^S)} \right] \frac{1}{\omega_{nm}^S - \omega} \quad (16)
\end{aligned}$$