

I. EXPRESSIONS FOR χ_{abc}^S IN TERMS OF $\mathcal{V}_{mn}^{\sigma,a,\ell}$

As can be seen from the prefactor of Eqs. (??) and (??), they diverge as $\tilde{\omega} \rightarrow 0$. To remove this apparent divergence of χ^S , we perform a partial fraction expansion in $\tilde{\omega}$.

II. INTRABAND CONTRIBUTIONS

For the intraband term of Eq. (??) we obtain

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

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:

$$\begin{aligned} \mathbf{r}_{mn}(\mathbf{k})|_{-\mathbf{k}} &= \mathbf{r}_{nm}(\mathbf{k})|_{\mathbf{k}}, \\ (\mathbf{r}_{mn})_{;\mathbf{k}}(\mathbf{k})|_{-\mathbf{k}} &= (-\mathbf{r}_{nm})_{;\mathbf{k}}(\mathbf{k})|_{\mathbf{k}}, \\ \mathcal{V}_{mn}^{\sigma,a,\ell}(\mathbf{k})|_{-\mathbf{k}} &= -\mathcal{V}_{nm}^{\sigma,a,\ell}(\mathbf{k})|_{\mathbf{k}}, \\ (\mathcal{V}_{mn}^{\sigma,a,\ell})_{;\mathbf{k}}(\mathbf{k})|_{-\mathbf{k}} &= (\mathcal{V}_{nm}^{\sigma,a,\ell})_{;\mathbf{k}}(\mathbf{k})|_{\mathbf{k}}, \\ \omega_{mn}^\sigma(\mathbf{k})|_{-\mathbf{k}} &= \omega_{nm}^\sigma(\mathbf{k})|_{\mathbf{k}}, \\ \Delta_{nm}^a(\mathbf{k})|_{-\mathbf{k}} &= -\Delta_{nm}^a(\mathbf{k})|_{\mathbf{k}}. \end{aligned} \quad (2)$$

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 $f_{nm} = -f_{mn}$. Using above relationships, we can show that the $1/\omega$ terms cancel each other out. Therefore, all the remaining non-zero terms in expressions (1) are simple ω and 2ω resonant denominators well behaved at zero frequency.

To apply time-reversal invariance, we notice that the energy denominators are invariant under $\mathbf{k} \rightarrow -\mathbf{k}$, and then we only look at the numerators, then

$$\begin{aligned} C &\rightarrow f_{mn} \mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA},b})_{;k^c} |_{\mathbf{k}} + f_{mn} \mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA},b})_{;k^c} |_{-\mathbf{k}} \\ &= f_{mn} \left[\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA},b})_{;k^c} |_{\mathbf{k}} + (-\mathcal{V}_{nm}^{\sigma,a,\ell}) (-r_{mn}^{\text{LDA},b})_{;k^c} |_{\mathbf{k}} \right] \\ &= f_{mn} \left[\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA},b})_{;k^c} + \mathcal{V}_{nm}^{\sigma,a,\ell} (r_{mn}^{\text{LDA},b})_{;k^c} \right] \\ &= f_{mn} \left[\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA},b})_{;k^c} + \left(\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA},b})_{;k^c} \right)^* \right] \\ &= 2f_{mn} \text{Re} \left[\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA},b})_{;k^c} \right], \end{aligned} \quad (3)$$

and likewise,

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

The last term in the second line of Eq. (1) is dealt with as follows.

$$\begin{aligned} \frac{D}{2(\omega_{nm}^\sigma)^2} \frac{1}{(\omega_{nm}^\sigma - \tilde{\omega})^2} &= \frac{f_{mn}}{2} \frac{\mathcal{V}_{mn}^{\sigma,a} r_{nm}^b}{(\omega_{nm}^\sigma)^2} \frac{\Delta_{nm}^c}{(\omega_{nm}^\sigma - \tilde{\omega})^2} = -\frac{f_{mn}}{2} \frac{\mathcal{V}_{mn}^{\sigma,a} r_{nm}^b}{(\omega_{nm}^\sigma)^2} \left(\frac{1}{\omega_{nm}^\sigma - \tilde{\omega}} \right)_{;k^c} \\ &= \frac{f_{mn}}{2} \left(\frac{\mathcal{V}_{mn}^{\sigma,a} r_{nm}^b}{(\omega_{nm}^\sigma)^2} \right)_{;k^c} \frac{1}{\omega_{nm}^\sigma - \tilde{\omega}}, \end{aligned} \quad (5)$$

where we used Eqs. (??) and for the last line, we performed an integration by parts over the Brillouin zone, where the contribution from the edges vanishes.[1] Now, we apply the chain rule, to get

$$\left(\frac{\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^2} \right)_{;k^c} = \frac{r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^2} (\mathcal{V}_{mn}^{\sigma,a,\ell})_{;k^c} + \frac{\mathcal{V}_{mn}^{\sigma,a,\ell}}{(\omega_{nm}^\sigma)^2} (r_{nm}^{\text{LDA,b}})_{;k^c} - \frac{2\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^3} (\omega_{nm}^\sigma)_{;k^c}, \quad (6)$$

and work the time-reversal on each term. The first term is reduced to

$$\begin{aligned} \frac{r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^2} (\mathcal{V}_{mn}^{\sigma,a,\ell})_{;k^c} | \mathbf{k} + \frac{r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^2} (\mathcal{V}_{mn}^{\sigma,a,\ell})_{;k^c} | -\mathbf{k} &= \frac{r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^2} (\mathcal{V}_{mn}^{\sigma,a,\ell})_{;k^c} | \mathbf{k} + \frac{r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^2} (\mathcal{V}_{nm}^{\sigma,a,\ell})_{;k^c} | \mathbf{k} \\ &= \frac{1}{(\omega_{nm}^\sigma)^2} \left[r_{nm}^{\text{LDA,b}} (\mathcal{V}_{mn}^{\sigma,a,\ell})_{;k^c} + \left(r_{nm}^{\text{LDA,b}} (\mathcal{V}_{mn}^{\sigma,a,\ell})_{;k^c} \right)^* \right] \\ &= \frac{2}{(\omega_{nm}^\sigma)^2} \text{Re} \left[r_{nm}^{\text{LDA,b}} (\mathcal{V}_{mn}^{\sigma,a,\ell})_{;k^c} \right], \end{aligned} \quad (7)$$

the second term is reduced to

$$\begin{aligned} \frac{\mathcal{V}_{mn}^{\sigma,a,\ell}}{(\omega_{nm}^\sigma)^2} (r_{nm}^{\text{LDA,b}})_{;k^c} | \mathbf{k} + \frac{\mathcal{V}_{mn}^{\sigma,a,\ell}}{(\omega_{nm}^\sigma)^2} (r_{nm}^{\text{LDA,b}})_{;k^c} | -\mathbf{k} &= \frac{\mathcal{V}_{mn}^{\sigma,a,\ell}}{(\omega_{nm}^\sigma)^2} (r_{nm}^{\text{LDA,b}})_{;k^c} | \mathbf{k} + \frac{\mathcal{V}_{nm}^{\sigma,a,\ell}}{(\omega_{nm}^\sigma)^2} (r_{nm}^{\text{LDA,b}})_{;k^c} | \mathbf{k} \\ &= \frac{1}{(\omega_{nm}^\sigma)^2} \left[\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA,b}})_{;k^c} + \left(\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA,b}})_{;k^c} \right)^* \right] \\ &= \frac{2}{(\omega_{nm}^\sigma)^2} \text{Re} \left[\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA,b}})_{;k^c} \right], \end{aligned} \quad (8)$$

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

$$\begin{aligned} \frac{2\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^3} (\omega_{nm}^\sigma)_{;k^c} | \mathbf{k} + \frac{2\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^3} (\omega_{nm}^\sigma)_{;k^c} | -\mathbf{k} &= \frac{2\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^3} \Delta_{nm}^c | \mathbf{k} + \frac{2\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^3} \Delta_{nm}^c | -\mathbf{k} \\ &= \frac{2\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^3} \Delta_{nm}^c | \mathbf{k} + \frac{2\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^3} \Delta_{nm}^c | \mathbf{k} \\ &= \frac{2}{(\omega_{nm}^\sigma)^3} \left[\mathcal{V}_{nm}^{\sigma,a,\ell} r_{mn}^{\text{LDA,b}} + \left(\mathcal{V}_{nm}^{\sigma,a,\ell} r_{mn}^{\text{LDA,b}} \right)^* \right] \Delta_{nm}^c \\ &= \frac{4}{(\omega_{nm}^\sigma)^3} \text{Re} \left[\mathcal{V}_{nm}^{\sigma,a,\ell} r_{mn}^{\text{LDA,b}} \right] \Delta_{nm}^c. \end{aligned} \quad (9)$$

Combining the results from (7), (8), and (9) into (6),

$$\begin{aligned} \frac{f_{mn}}{2} \left[\left(\frac{\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^2} \right)_{;k^c} | \mathbf{k} + \left(\frac{\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^\sigma)^2} \right)_{;k^c} | -\mathbf{k} \right] \frac{1}{\omega_{nm}^\sigma - \tilde{\omega}} &= \\ \left(2 \text{Re} \left[r_{nm}^{\text{LDA,b}} (\mathcal{V}_{mn}^{\sigma,a,\ell})_{;k^c} \right] + 2 \text{Re} \left[\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA,b}})_{;k^c} \right] - \frac{4}{\omega_{nm}^\sigma} \text{Re} \left[\mathcal{V}_{nm}^{\sigma,a,\ell} r_{mn}^{\text{LDA,b}} \right] \Delta_{nm}^c \right) \frac{f_{mn}}{2(\omega_{nm}^\sigma)^2} \frac{1}{\omega_{nm}^\sigma - \tilde{\omega}}. \end{aligned} \quad (10)$$

We substitute (3), (4), and (10) in (1),

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

If we simplify,

$$\begin{aligned}
I = & -\frac{2f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA,b}})_{;k^c}]}{2(\omega_{nm}^\sigma)^2} \frac{1}{\omega_{nm}^\sigma - \tilde{\omega}} + \frac{4f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA,b}})_{;k^c}]}{(\omega_{nm}^\sigma)^2} \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}} \\
& + \frac{6f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}] \Delta_{nm}^c}{2(\omega_{nm}^\sigma)^3} \frac{1}{\omega_{nm}^\sigma - \tilde{\omega}} - \frac{8f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}] \Delta_{nm}^c}{(\omega_{nm}^\sigma)^3} \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}} \\
& + \frac{2f_{mn} \operatorname{Re} [r_{nm}^{\text{LDA,b}} (\mathcal{V}_{mn}^{\sigma,a,\ell})_{;k^c}]}{2(\omega_{nm}^\sigma)^2} \frac{1}{\omega_{nm}^\sigma - \tilde{\omega}} \\
& + \frac{2f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA,b}})_{;k^c}]}{2(\omega_{nm}^\sigma)^2} \frac{1}{\omega_{nm}^\sigma - \tilde{\omega}} \\
& - \frac{4f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} r_{mn}^{\text{LDA,b}}] \Delta_{nm}^c}{2(\omega_{nm}^\sigma)^3} \frac{1}{\omega_{nm}^\sigma - \tilde{\omega}}, \tag{11}
\end{aligned}$$

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 above equation

$$\begin{aligned}
I_{2\omega} = & -\frac{e^3}{\hbar^2} \sum_{mn\mathbf{k}} \left[\frac{4f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA,b}})_{;k^c}]}{(\omega_{nm}^\sigma)^2} - \frac{8f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}] \Delta_{nm}^c}{(\omega_{nm}^\sigma)^3} \right] \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}} \\
= & -\frac{e^3}{\hbar^2} \sum_{mn\mathbf{k}} \frac{4f_{mn}}{(\omega_{nm}^\sigma)^2} \left[\operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA,b}})_{;k^c}] - \frac{2 \operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}] \Delta_{nm}^c}{\omega_{nm}^\sigma} \right] \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}}. \tag{12}
\end{aligned}$$

We can express the energies in terms of transitions between bands. Therefore, $\omega_{nm}^\sigma = \omega_{cv}^\sigma$ for transitions between conduction and valence bands. To take the limit $\eta \rightarrow 0$, we use

$$\lim_{\eta \rightarrow 0} \frac{1}{x \pm i\eta} = P \frac{1}{x} \mp i\pi\delta(x), \tag{13}$$

and can finally rewrite (12) in the desired form,

$$\operatorname{Im}[\chi_{i,a,\ell bc,2\omega}^{s,\ell}] = -\frac{\pi|e|^3}{2\hbar^2} \sum_{v\mathbf{c}\mathbf{k}} \frac{4}{(\omega_{cv}^\sigma)^2} \left(\operatorname{Re} [\mathcal{V}_{vc}^{\sigma,a,\ell} (r_{cv}^{\text{LDA,b}})_{;k^c}] - \frac{2 \operatorname{Re} [\mathcal{V}_{vc}^{\sigma,a,\ell} r_{cv}^{\text{LDA,b}}] \Delta_{cv}^c}{\omega_{cv}^\sigma} \right) \delta(\omega_{cv}^\sigma - 2\omega). \tag{14}$$

where we added a 1/2 from the sum over $\mathbf{k} \rightarrow -\mathbf{k}$. We do the same for the $\tilde{\omega}$ terms in (11) to obtain

$$\begin{aligned}
I_\omega = & -\frac{e^3}{2\hbar^2} \sum_{nm\mathbf{k}} \left[-\frac{2f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA,b}})_{;k^c}]}{(\omega_{nm}^\sigma)^2} + \frac{6f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}] \Delta_{nm}^c}{(\omega_{nm}^\sigma)^3} \right. \\
& + \frac{2f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} (r_{nm}^{\text{LDA,b}})_{;k^c}]}{(\omega_{nm}^\sigma)^2} - \frac{4f_{mn} \operatorname{Re} [\mathcal{V}_{nm}^{\sigma,a,\ell} r_{mn}^{\text{LDA,b}}] \Delta_{nm}^c}{(\omega_{nm}^\sigma)^3} \\
& \left. + \frac{2f_{mn} \operatorname{Re} [r_{nm}^{\text{LDA,b}} (\mathcal{V}_{mn}^{\sigma,a,\ell})_{;k^c}]}{(\omega_{nm}^\sigma)^2} \right] \frac{1}{\omega_{nm}^\sigma - \tilde{\omega}}. \tag{15}
\end{aligned}$$

We reduce in the same way as (12),

$$I_\omega = -\frac{e^3}{2\hbar^2} \sum_{nm\mathbf{k}} \frac{f_{mn}}{(\omega_{nm}^\sigma)^2} \left[2 \operatorname{Re} [r_{nm}^{\text{LDA,b}} (\mathcal{V}_{mn}^{\sigma,a,\ell})_{;k^c}] + \frac{2 \operatorname{Re} [\mathcal{V}_{mn}^{\sigma,a,\ell} r_{nm}^{\text{LDA,b}}] \Delta_{nm}^c}{\omega_{nm}^\sigma} \right] \frac{1}{\omega_{nm}^\sigma - \tilde{\omega}}, \tag{16}$$

and using (13) we obtain our final form,

$$\operatorname{Im}[\chi_{i,a,\ell bc,\omega}^{s,\ell}] = -\frac{\pi|e|^3}{2\hbar^2} \sum_{cv} \frac{1}{(\omega_{cv}^\sigma)^2} \left(\operatorname{Re} [r_{cv}^{\text{LDA,b}} (\mathcal{V}_{vc}^{\sigma,a,\ell})_{;k^c}] + \frac{\operatorname{Re} [\mathcal{V}_{vc}^{\sigma,a,\ell} r_{cv}^{\text{LDA,b}}] \Delta_{cv}^c}{\omega_{cv}^\sigma} \right) \delta(\omega_{cv}^\sigma - \omega), \tag{17}$$

where again we added a 1/2 from the sum over $\mathbf{k} \rightarrow -\mathbf{k}$.

III. INTERBAND CONTRIBUTIONS

We follow an equivalent procedure for the interband contribution. From Eq. (??) we have

$$E = A \left[-\frac{1}{2\omega_{lm}^\sigma(2\omega_{lm}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{lm}^\sigma - \tilde{\omega}} + \frac{2}{\omega_{nm}^\sigma(2\omega_{lm}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}} + \frac{1}{2\omega_{lm}^\sigma\omega_{nm}^\sigma} \frac{1}{\tilde{\omega}} \right] \\ - B \left[-\frac{1}{2\omega_{nl}^\sigma(2\omega_{nl}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{nl}^\sigma - \tilde{\omega}} + \frac{2}{\omega_{nm}^\sigma(2\omega_{nl}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}} + \frac{1}{2\omega_{nl}^\sigma\omega_{nm}^\sigma} \frac{1}{\tilde{\omega}} \right], \quad (18)$$

where $A = f_{ml}\mathcal{V}_{mn}^{\sigma,a}r_{nl}^c r_{lm}^b$ and $B = f_{ln}\mathcal{V}_{mn}^{\sigma,a}r_{nl}^c r_{lm}^b$.

Just as above, the $\frac{1}{\tilde{\omega}}$ terms cancel out. We multiply out the A and B terms,

$$E = \left[-\frac{A}{2\omega_{lm}^\sigma(2\omega_{lm}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{lm}^\sigma - \tilde{\omega}} + \frac{2A}{\omega_{nm}^\sigma(2\omega_{lm}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}} \right] \\ + \left[\frac{B}{2\omega_{nl}^\sigma(2\omega_{nl}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{nl}^\sigma - \tilde{\omega}} - \frac{2B}{\omega_{nm}^\sigma(2\omega_{nl}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}} \right]. \quad (19)$$

As before, we notice that the energy denominators are invariant under $\mathbf{k} \rightarrow -\mathbf{k}$ so we need only look at the numerators. Starting with A ,

$$A \rightarrow f_{ml}\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^c r_{lm}^b|_{\mathbf{k}} + f_{ml}\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^c r_{lm}^b|_{-\mathbf{k}} \\ = f_{ml} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^c r_{lm}^b|_{\mathbf{k}} + (-\mathcal{V}_{nm}^{\sigma,a,\ell}) r_{ln}^c r_{ml}^b|_{\mathbf{k}}] \\ = f_{ml} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^c r_{lm}^b - \mathcal{V}_{nm}^{\sigma,a,\ell}r_{ln}^c r_{ml}^b] \\ = f_{ml} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^c r_{lm}^b - (\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^c r_{lm}^b)^*] \\ = -2f_{ml} \text{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^c r_{lm}^b],$$

then B ,

$$B \rightarrow f_{ln}\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^b r_{lm}^c|_{\mathbf{k}} + f_{ln}\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^b r_{lm}^c|_{-\mathbf{k}} \\ = f_{ln} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^b r_{lm}^c|_{\mathbf{k}} + (-\mathcal{V}_{nm}^{\sigma,a,\ell}) r_{ln}^b r_{ml}^c|_{\mathbf{k}}] \\ = f_{ln} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^b r_{lm}^c - \mathcal{V}_{nm}^{\sigma,a,\ell}r_{ln}^b r_{ml}^c] \\ = f_{ln} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^b r_{lm}^c - (\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^b r_{lm}^c)^*] \\ = -2f_{ln} \text{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^b r_{lm}^c].$$

We then substitute in (19),

$$E = \left[\frac{2f_{ml} \text{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^c r_{lm}^b]}{2\omega_{lm}^\sigma(2\omega_{lm}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{lm}^\sigma - \tilde{\omega}} - \frac{4f_{ml} \text{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^c r_{lm}^b]}{\omega_{nm}^\sigma(2\omega_{lm}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}} \right] \\ - \left[\frac{2f_{ln} \text{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^b r_{lm}^c]}{2\omega_{nl}^\sigma(2\omega_{nl}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{nl}^\sigma - \tilde{\omega}} + \frac{4f_{ln} \text{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^b r_{lm}^c]}{\omega_{nm}^\sigma(2\omega_{nl}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}} \right].$$

We manipulate indices and simplify,

$$E = \left[\frac{f_{ml} \text{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^c r_{lm}^b]}{\omega_{lm}^\sigma(2\omega_{lm}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{lm}^\sigma - \tilde{\omega}} - \frac{f_{ln} \text{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^b r_{lm}^c]}{\omega_{nl}^\sigma(2\omega_{nl}^\sigma - \omega_{nm}^\sigma)} \frac{1}{\omega_{nl}^\sigma - \tilde{\omega}} \right] \\ + \left[\frac{f_{ln} \text{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^b r_{lm}^c]}{2\omega_{nl}^\sigma - \omega_{nm}^\sigma} - \frac{f_{ml} \text{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^c r_{lm}^b]}{2\omega_{lm}^\sigma - \omega_{nm}^\sigma} \right] \frac{4}{\omega_{nm}^\sigma} \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}} \\ = \left[\frac{f_{mn} \text{Im} [\mathcal{V}_{ml}^{\sigma,a,\ell}r_{ln}^c r_{nm}^b]}{2\omega_{nm}^\sigma - \omega_{lm}^\sigma} - \frac{f_{mn} \text{Im} [\mathcal{V}_{ln}^{\sigma,a,\ell}r_{nm}^b r_{ml}^c]}{2\omega_{nm}^\sigma - \omega_{nl}^\sigma} \right] \frac{1}{\omega_{nm}^\sigma} \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}} \\ + \left[\frac{f_{ln} \text{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^b r_{lm}^c]}{2\omega_{nl}^\sigma - \omega_{nm}^\sigma} - \frac{f_{ml} \text{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell}r_{nl}^c r_{lm}^b]}{2\omega_{lm}^\sigma - \omega_{nm}^\sigma} \right] \frac{4}{\omega_{nm}^\sigma} \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}},$$

and substitute in (??),

$$I = -\frac{e^3}{2\hbar^2} \sum_{nm} \frac{1}{\omega_{nm}^\sigma} \left[\frac{f_{mn} \operatorname{Im} [\mathcal{V}_{ml}^{\sigma,a,\ell} \{r_{ln}^c r_{nm}^b\}]}{2\omega_{nm}^\sigma - \omega_{lm}^\sigma} - \frac{f_{mn} \operatorname{Im} [\mathcal{V}_{ln}^{\sigma,a,\ell} \{r_{nm}^b r_{ml}^c\}]}{2\omega_{nm}^\sigma - \omega_{nl}^\sigma} \right] \frac{1}{\omega_{nm}^\sigma - \tilde{\omega}} \\ + 4 \left[\frac{f_{ln} \operatorname{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell} \{r_{nl}^b r_{lm}^c\}]}{2\omega_{nl}^\sigma - \omega_{nm}^\sigma} - \frac{f_{ml} \operatorname{Im} [\mathcal{V}_{mn}^{\sigma,a,\ell} \{r_{nl}^c r_{lm}^b\}]}{2\omega_{lm}^\sigma - \omega_{nm}^\sigma} \right] \frac{1}{\omega_{nm}^\sigma - 2\tilde{\omega}}.$$

Finally, we take $n = c$, $m = v$, and $l = q$ and substitute,

$$I = -\frac{e^3}{2\hbar^2} \sum_{cv} \frac{1}{\omega_{cv}^\sigma} \left(\left[\frac{f_{vc} \operatorname{Im} [\mathcal{V}_{vq}^{\sigma,a,\ell} \{r_{qc}^b r_{cv}^c\}]}{2\omega_{cv}^\sigma - \omega_{qv}^\sigma} - \frac{f_{vc} \operatorname{Im} [\mathcal{V}_{qc}^{\sigma,a,\ell} \{r_{cv}^b r_{vq}^c\}]}{2\omega_{cv}^\sigma - \omega_{cq}^\sigma} \right] \frac{1}{\omega_{cv}^\sigma - \tilde{\omega}} \right. \\ \left. + 4 \left[\frac{f_{qc} \operatorname{Im} [\mathcal{V}_{vc}^{\sigma,a,\ell} \{r_{cq}^b r_{qv}^c\}]}{2\omega_{cq}^\sigma - \omega_{cv}^\sigma} - \frac{f_{vq} \operatorname{Im} [\mathcal{V}_{vc}^{\sigma,a,\ell} \{r_{cq}^c r_{qv}^b\}]}{2\omega_{qv}^\sigma - \omega_{cv}^\sigma} \right] \frac{1}{\omega_{cv}^\sigma - 2\tilde{\omega}} \right) \\ = \frac{e^3}{2\hbar^2} \sum_{cv} \frac{1}{\omega_{cv}^\sigma} \left(\left[\frac{\operatorname{Im} [\mathcal{V}_{qc}^{\sigma,a,\ell} \{r_{cv}^b r_{vq}^c\}]}{2\omega_{cv}^\sigma - \omega_{cq}^\sigma} - \frac{\operatorname{Im} [\mathcal{V}_{vq}^{\sigma,a,\ell} \{r_{qc}^c r_{cv}^b\}]}{2\omega_{cv}^\sigma - \omega_{qv}^\sigma} \right] \frac{1}{\omega_{cv}^\sigma - \tilde{\omega}} \right. \\ \left. - 4 \left[\frac{f_{qc} \operatorname{Im} [\mathcal{V}_{vc}^{\sigma,a,\ell} \{r_{cq}^b r_{qv}^c\}]}{2\omega_{cq}^\sigma - \omega_{cv}^\sigma} - \frac{f_{vq} \operatorname{Im} [\mathcal{V}_{vc}^{\sigma,a,\ell} \{r_{cq}^c r_{qv}^b\}]}{2\omega_{qv}^\sigma - \omega_{cv}^\sigma} \right] \frac{1}{\omega_{cv}^\sigma - 2\tilde{\omega}} \right).$$

We use (13),

$$I = \frac{\pi|e^3|}{2\hbar^2} \sum_{cv} \frac{1}{\omega_{cv}^\sigma} \left(\left[\frac{\operatorname{Im} [\mathcal{V}_{qc}^{\sigma,a,\ell} \{r_{cv}^b r_{vq}^c\}]}{2\omega_{cv}^\sigma - \omega_{cq}^\sigma} - \frac{\operatorname{Im} [\mathcal{V}_{vq}^{\sigma,a,\ell} \{r_{qc}^c r_{cv}^b\}]}{2\omega_{cv}^\sigma - \omega_{qv}^\sigma} \right] \delta(\omega_{cv}^\sigma - \omega) \right. \\ \left. - 4 \left[\frac{f_{qc} \operatorname{Im} [\mathcal{V}_{vc}^{\sigma,a,\ell} \{r_{cq}^b r_{qv}^c\}]}{2\omega_{cq}^\sigma - \omega_{cv}^\sigma} - \frac{f_{vq} \operatorname{Im} [\mathcal{V}_{vc}^{\sigma,a,\ell} \{r_{cq}^c r_{qv}^b\}]}{2\omega_{qv}^\sigma - \omega_{cv}^\sigma} \right] \delta(\omega_{cv}^\sigma - 2\omega) \right),$$

and recognize that for the 1ω terms, $q \neq (v, c)$, and for the 2ω q can have two distinct values such that,

$$I = \frac{\pi|e^3|}{2\hbar^2} \sum_{cv} \frac{1}{\omega_{cv}^\sigma} \left(\sum_{q \neq (v,c)} \left[\frac{\operatorname{Im} [\mathcal{V}_{qc}^{\sigma,a,\ell} \{r_{cv}^b r_{vq}^c\}]}{2\omega_{cv}^\sigma - \omega_{cq}^\sigma} - \frac{\operatorname{Im} [\mathcal{V}_{vq}^{\sigma,a,\ell} \{r_{qc}^c r_{cv}^b\}]}{2\omega_{cv}^\sigma - \omega_{qv}^\sigma} \right] \delta(\omega_{cv}^\sigma - \omega) \right. \\ \left. - 4 \left[\sum_{v' \neq v} \frac{\operatorname{Im} [\mathcal{V}_{vc}^{\sigma,a,\ell} \{r_{cv'}^b r_{v'v}^c\}]}{2\omega_{cv'}^\sigma - \omega_{cv}^\sigma} - \sum_{c' \neq c} \frac{\operatorname{Im} [\mathcal{V}_{vc}^{\sigma,a,\ell} \{r_{cc'}^c r_{c'v}^b\}]}{2\omega_{c'v}^\sigma - \omega_{cv}^\sigma} \right] \delta(\omega_{cv}^\sigma - 2\omega) \right).$$

[1] N. W. Ashcroft and N. D. Mermin. *Solid State Physics*. Brooks Cole, Saunders College, Philadelphia, 1976.