

1 Interband Contributions

Hello world!

2 Intraband Contributions

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

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

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,

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

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

Time-reversal symmetry leads to the following relationships:

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

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

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

and likewise,

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

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

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

We use the fact that

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

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

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

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

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

the second term is reduced to

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

and by using $\frac{\partial \mathbf{k}}{\partial \omega}$, the third term is reduced to

$$\begin{aligned}
\frac{2\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^S)^3} (\omega_{nm}^S)_{;k^c} | \mathbf{k} + \frac{2\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^S)^3} (\omega_{nm}^S)_{;k^c} | -\mathbf{k} &= \frac{2\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^S)^3} \Delta_{nm}^c | \mathbf{k} + \frac{2\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^S)^3} \Delta_{nm}^c | -\mathbf{k} \\
&= \frac{2\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^S)^3} \Delta_{nm}^c | \mathbf{k} + \frac{2\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^S)^3} \Delta_{nm}^c | \mathbf{k} \\
&= \frac{2}{(\omega_{nm}^S)^3} \left[\mathcal{V}_{nm}^{\Sigma,a} r_{mn}^{\text{LDA,b}} + (\mathcal{V}_{nm}^{\Sigma,a} r_{mn}^{\text{LDA,b}})^* \right] \Delta_{nm}^c \\
&= \frac{4}{(\omega_{nm}^S)^3} \text{Re} [\mathcal{V}_{nm}^{\Sigma,a} r_{mn}^{\text{LDA,b}}] \Delta_{nm}^c.
\end{aligned} \tag{11}$$

third_term_g

Combining the results from $\frac{\text{first term and derivative}}{\text{first term and derivative}}$, (9), (10), and (11) into (8),

$$\begin{aligned}
\frac{f_{mn}}{2} \left[\left(\frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^S)^2} \right)_{;k^c} | \mathbf{k} + \left(\frac{\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^{\text{LDA,b}}}{(\omega_{nm}^S)^2} \right)_{;k^c} | -\mathbf{k} \right] \frac{1}{\omega_{nm}^S - \omega} = \\
f_{mn} \left(2 \text{Re} \left[r_{nm}^{\text{LDA,b}} (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c} \right] + 2 \text{Re} \left[\mathcal{V}_{mn}^{\Sigma,a} (r_{nm}^{\text{LDA,b}})_{;k^c} \right] - \frac{4}{\omega_{nm}^S} \text{Re} [\mathcal{V}_{nm}^{\Sigma,a} r_{mn}^{\text{LDA,b}}] \Delta_{nm}^c \right) \frac{1}{2(\omega_{nm}^S)^2} \frac{1}{\omega_{nm}^S - \omega}.
\end{aligned} \tag{12}$$

derivative_u

We have all the elements to be substituted in $\frac{\text{pfi}}{\text{pfi}}$. We substitute $\frac{\text{ct}}{\text{ct}}$, $\frac{\text{dt}}{\text{dt}}$, and $\frac{\text{derivative under k}}{\text{derivative under k}}$ in (2),

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

If we simplify,

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

simplified_i

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 [\(I1\)](#),

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

We can express the energies in terms of transitions between bands. Therefore, $\omega_{nm}^S = \omega_{cv}^S$ for transitions between conduction and valence bands. We analyze the limit,

$$\lim_{\eta \rightarrow 0} \frac{1}{x \pm i\eta} = P \frac{1}{x} \mp i\pi \delta(x), \quad (15)$$

and can finally rewrite [\(I4\)](#) in the desired form,

$$\operatorname{Im}[\chi_{i,\text{abc},2\omega}^{s,\ell}] = -\frac{\pi e^3}{\hbar^2} \sum_{mn\mathbf{k}} \frac{4}{(\omega_{nm}^S)^2} \left(\operatorname{Re} [\mathcal{V}_{mn}^{\Sigma,a} (r_{nm}^{\text{LDA,b}})_{;k^c}] - \frac{2 \operatorname{Re} [\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^{\text{LDA,b}}] \Delta_{nm}^c}{\omega_{nm}^S} \right) \delta(\omega_{nm}^S - 2\omega). \quad (16) \quad \boxed{\text{imchi2w}}$$

We do the same for the ω terms in [\(I3\)](#) and substitute in [\(I1\)](#),

$$\begin{aligned} I_{\omega} &= -\frac{e^3}{2\hbar^2} \sum_{mn\mathbf{k}} \left[-\frac{2f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\Sigma,a} (r_{nm}^{\text{LDA,b}})_{;k^c}]}{(\omega_{nm}^S)^2} + \frac{6f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^{\text{LDA,b}}] \Delta_{nm}^c}{(\omega_{nm}^S)^3} \right. \\ &\quad \left. + \frac{2f_{mn} \operatorname{Re} [\mathcal{V}_{mn}^{\Sigma,a} (r_{nm}^{\text{LDA,b}})_{;k^c}]}{(\omega_{nm}^S)^2} - \frac{4f_{mn} \operatorname{Re} [\mathcal{V}_{nm}^{\Sigma,a} r_{mn}^{\text{LDA,b}}] \Delta_{nm}^c}{(\omega_{nm}^S)^3} \right. \\ &\quad \left. + \frac{2f_{mn} \operatorname{Re} [r_{nm}^{\text{LDA,b}} (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c}]}{(\omega_{nm}^S)^2} \right] \frac{1}{\omega_{nm}^S - \omega}, \end{aligned} \quad (17) \quad \boxed{\text{wchii}}$$

and we reduce in the same way as [\(I4\)](#),

$$I_{\omega} = -\frac{e^3}{2\hbar^2} \sum_{mn\mathbf{k}} \frac{f_{mn}}{(\omega_{nm}^S)^2} \left[\frac{2 \operatorname{Re} [\mathcal{V}_{mn}^{\Sigma,a} r_{nm}^{\text{LDA,b}}] \Delta_{nm}^c}{\omega_{nm}^S} + 2 \operatorname{Re} [r_{nm}^{\text{LDA,b}} (\mathcal{V}_{mn}^{\Sigma,a})_{;k^c}] \right] \frac{1}{\omega_{nm}^S - \omega} \quad (18) \quad \boxed{\text{wchii_simpli}}$$