Motivation

In the presence of time-reversal symmetry (TRS) breaking, a differential absorption (and optical conductivity) of left handed and right handed circularly polarized light can occur. In this note we show how to calculate this circular dichroism in the Kubo formalism when only on-shell (energy conserving) transitions are allowed.

A Diversion About Delta Functions: The Poisson Kernel

If $f(x)$ has integral 1 on $\mathbb{R}$, then the function

$$f_\eta(x) = \frac{1}{\eta} f(\frac{x}{\eta})$$

also has integral 1 on $\mathbb{R}$ and

$$\lim_{\eta \to 0} f_\eta(x) = \delta(x)$$

A simple contour integral shows that

$$\int_{-\infty}^{+\infty} dx \, \frac{1}{x^2 + 1} = \pi$$

Therefore we can write

$$\pi \, \delta(x) = \lim_{\eta \to 0} \frac{1}{\eta} \frac{1}{(x/\eta)^2 + 1} = \lim_{\eta \to 0} \frac{\eta}{x^2 + \eta^2}$$

Now we contrive the "Poisson kernel"

$$\lim_{\eta \to 0} \frac{1}{x + i\eta} = \lim_{\eta \to 0} \frac{x}{x^2 + \eta^2} - \frac{i\eta}{x^2 + \eta^2} = (x/\eta)\pi\delta(x) - i\pi\delta(x) = -i\pi\delta(x)$$

Which we will find useful in what follows.

Circular Dichroism via the Kubo Formula

Now we note that the absorbent part of the optical conductivity for circularly polarized light is (proof in the subsequent section)

$$\mathrm{Re}(\sigma_\pm) = \frac{1}{2}\big(\mathrm{Re}(\sigma_{xx}) + \mathrm{Re}(\sigma_{yy}) \pm 2\,\mathrm{Im}(\sigma_{xy})\big),$$

where the optical conductivities are a function of photon energy. To find the circular dichroism, we will want the imaginary part of the Hall conductivity (which for a TRS preserving model can only be non-zero in subsets of the Brillouin zone).

Now, the Kubo formula for the first-order direct interband contributions to the Hall conductivity, σ_{xy}, is a sum over all momentum conserving transitions

$$\sigma_{\mu\nu} = i\frac{e^2}{\hbar} \sum_{s,s'} \int \frac{d\vec{k}}{(2\pi)^{\mathrm{dim}}} \frac{f(\epsilon_s) - f(\epsilon_{s'})}{\epsilon_{s'} - \epsilon_s} \frac{\langle s|\hbar v_\mu|s'\rangle\langle s'|\hbar v_\nu|s\rangle}{\hbar\omega - (\epsilon_{s'} - \epsilon_s) + i\eta}$$

where s are valence states, s' are conduction states, f are the Fermi distribution function, $\hbar\omega$ is the energy of the incident light, and v_μ are the velocity operators given by

$$v_\mu = \frac{1}{\hbar}\frac{\partial H}{\partial k_\mu}$$

Inserting the Poisson kernel representation of the delta function (in the limit of $\eta \to 0$, i.e. no off-shell transitions)

$$\sigma_{\mu\nu} = \pi \frac{e^2}{\hbar} \sum_{s,s'} \int \frac{d\vec{k}}{(2\pi)^{\dim}} \frac{f(\epsilon_s) - f(\epsilon_{s'})}{\epsilon_{s'} - \epsilon_s} \langle s|\hbar v_\mu|s'\rangle\langle s'|\hbar v_\nu|s\rangle \delta(\hbar\omega - (\epsilon_{s'} - \epsilon_s))$$

so that

$$\mathrm{Im}(\sigma_{xy}) = \pi \frac{e^2}{\hbar} \sum_{s,s'} \int \frac{d\vec{k}}{(2\pi)^{\dim}} \frac{f(\epsilon_s) - f(\epsilon_{s'})}{\epsilon_{s'} - \epsilon_s} \delta(\hbar\omega - (\epsilon_{s'} - \epsilon_s)) \mathrm{Im}[\langle s|\hbar v_x|s'\rangle\langle s'|\hbar v_y|s\rangle]$$

So we see that the key to evaluating the Hall conductivity is to determine the matrix elements $\langle s|\hbar v_x|s'\rangle\langle s'|\hbar v_y|s\rangle$. All that is needed to find $\mathrm{Im}(\sigma_{xy})$ and hence the circular dichroism is to (1) diagonalize $H(k)$ to find the valence and conduction Bloch states, (2) determine the velocity operators v_μ by differentiating $H(k)$, and (3) evaluate a few inner products.

Circular Dichroism Formula

As we saw above, in the Kubo formula, any real or imaginary part originates with the matrix elements $\langle s|\hbar v_x|s'\rangle\langle s'|\hbar v_y|s\rangle$. Let $M_\mu = \langle s|\hbar v_\mu|s'\rangle$ and $M_\mu^* = \langle s'|\hbar v_\mu|s\rangle$, then we have

$$M_\pm = \frac{1}{\sqrt{2}}(M_x \pm iM_y),$$

so that

$$\begin{aligned}
|M_\pm|^2 &= \frac{1}{2}(M_x \pm iM_y)(M_x^* \mp iM_y^*) \\
&= \frac{1}{2}(M_x^R + iM_x^I \pm iM_y^R \mp M_y^I)(M_x^R - iM_x^I \mp iM_y^R \mp M_y^I) \\
&= \frac{1}{2}\big((M_x^R \mp M_y^I)^2 + (M_x^I \pm M_y^R)^2\big) \\
&= \frac{1}{2}(M_x^{R,2} + M_y^{I,2} \mp 2M_x^R M_y^I + M_x^{I,2} + M_y^{R,2} \pm M_x^I M_y^R).
\end{aligned}$$

where the superscripts R and I refer to the real and imaginary parts respectively.
We have

$$|M_i|^2 = M_i^{R,2} + M_i^{I,2},$$

and

$$M_x M_y^* = (M_x^R + iM_x^I)(M_y^R - iM_y^I) = (M_x^R M_y^R + M_x^I M_y^I) + i(M_x^I M_y^R - M_x^R M_y^I),$$

from which it immediately follows that

$$|M_\pm|^2 = \frac{1}{2}(|M_x|^2 + |M_y|^2 \pm 2\,\mathrm{Im}(M_x M_y^*))$$