Non-relativistic velocity operators on the triclinic lattice

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Optical transitions between energy bands in crystals are governed by the matrix elements of the appropriate operators between these bands. In the electric dipole approximation, this operator is a velocity operator. Here we provide a pedagogical overview of the formulation of non-relativistic velocity operators on the general triclinic lattice.

1 Context

The frequency-dependent optical conductivity $\sigma_{\mu\nu}(\omega)$ is an experimentally measurable response property of a material to illumination by light. One way to calculate this (in the equilibrium limit) is the Kubo formula. Now, the Kubo formula for the first-order direct interband electric-dipole moment mediated optical transitions is a sum over all momentum conserving transitions

$$\sigma_{\mu\nu}(\omega) = i\frac{e^2}{\hbar}\sum_{s,s'}\int_{\mathrm{BZ}}\frac{d\boldsymbol{k}}{(2\pi)^{\mathrm{dim}}}\frac{f(\epsilon_s)-f(\epsilon_{s'})}{\epsilon_{s'}-\epsilon_s}\frac{\langle s|\hbar\hat{v}_\mu|s'\rangle\langle s'|\hbar\hat{v}_\nu|s\rangle}{\hbar\omega-(\epsilon_{s'}-\epsilon_s)+i\eta}$$

where s are valence states, s' are conduction states, $f(\epsilon)=1/(e^{\epsilon/kT}+1)$ are the Fermi distribution function, $\hbar\omega$ is the energy of the incident light, η is the dephasing (extent to which off-shell transitions are allowed), and $\hat{v}_\mu$ are the velocity operators.

2 Velocity Operators

The covariant current operator is defined as

$$\hat{j}_i = e\hat{v}_i, \tag{1}$$

and the contravariant velocity operators are defined by the gradient of the Bloch Hamiltonian with respect to momentum

$$\begin{pmatrix}\hat{v}^a\\ \hat{v}^b\\ \hat{v}^c\end{pmatrix} = \boldsymbol{\nabla}_p H = \begin{pmatrix}\dfrac{\partial H}{\partial\hat{p}^a}\\ \dfrac{\partial H}{\partial\hat{p}^b}\\ \dfrac{\partial H}{\partial\hat{p}^c}\end{pmatrix}. \tag{2}$$

To obtain the covariant velocity operators, we use $\boldsymbol{e}^i = \sum_j g^{ij}\boldsymbol{e}_j$ to express the contravariant unit vectors $\boldsymbol{e}^i$ in terms of the covariant unit vectors $\boldsymbol{e}_i$.

Now, the metric tensor of the triclinic lattice is

$$[g_{ij}] = \begin{pmatrix}|a|^2 & |a||b|\cos(\gamma) & |a||c|\cos(\beta)\\ |a||b|\cos(\gamma) & |b|^2 & |b||c|\cos(\alpha)\\ |a||c|\cos(\beta) & |b||c|\cos(\alpha) & |c|^2\end{pmatrix}. \tag{3}$$

However we want the metric tensor of the triclinic *coordinate system*. To see why, note that we are not so much interested in measuring distances in unit cells or Brillouin zones, but rather the contribution of the non-orthogonality of the axes to the optical conductivity. Note that the unit cell is much smaller than the wavelength of the light for the case where these velocity operators hold. Now, the metric tensor for the triclinic coordinate system is

$$[g_{ij}] = \begin{pmatrix} 1 & \cos(\gamma) & \cos(\beta) \\ \cos(\gamma) & 1 & \cos(\alpha) \\ \cos(\beta) & \cos(\alpha) & 1 \end{pmatrix}, \tag{4}$$

and matrix inversion yields

$$[g^{ij}] = \frac{1}{\det([g_{ij}])} \begin{pmatrix} 1-\cos^2(\alpha) & \cos(\alpha)\cos(\beta)-\cos(\gamma) & \cos(\alpha)\cos(\gamma)-\cos(\beta) \\ \cos(\alpha)\cos(\beta)-\cos(\gamma) & 1-\cos^2(\beta) & \cos(\beta)\cos(\gamma)-\cos(\alpha) \\ \cos(\alpha)\cos(\gamma)-\cos(\beta) & \cos(\beta)\cos(\gamma)-\cos(\alpha) & 1-\cos^2(\gamma) \end{pmatrix} \tag{5}$$

where

$$\frac{1}{\det([g_{ij}])} = \frac{1}{1-\cos^2(\alpha)-\cos^2(\beta)-\cos^2(\gamma)+2\cos(\alpha)\cos(\beta)\cos(\gamma)}. \tag{6}$$

We then have

$$\hat{v}_a = \frac{1-\cos^2(\alpha)}{\det([g_{ij}])}\frac{\partial H}{\partial \hat{p}^a} + \frac{\cos(\alpha)\cos(\beta)-\cos(\gamma)}{\det([g_{ij}])}\frac{\partial H}{\partial \hat{p}^b} + \frac{\cos(\alpha)\cos(\gamma)-\cos(\beta)}{\det([g_{ij}])}\frac{\partial H}{\partial \hat{p}^c} \tag{7a}$$

$$\hat{v}_b = \frac{\cos(\alpha)\cos(\beta)-\cos(\gamma)}{\det([g_{ij}])}\frac{\partial H}{\partial \hat{p}^a} + \frac{1-\cos^2(\beta)}{\det([g_{ij}])}\frac{\partial H}{\partial \hat{p}^b} + \frac{\cos(\beta)\cos(\gamma)-\cos(\alpha)}{\det([g_{ij}])}\frac{\partial H}{\partial \hat{p}^c} \tag{7b}$$

$$\hat{v}_c = \frac{\cos(\alpha)\cos(\gamma)-\cos(\beta)}{\det([g_{ij}])}\frac{\partial H}{\partial \hat{p}^a} + \frac{\cos(\beta)\cos(\gamma)-\cos(\alpha)}{\det([g_{ij}])}\frac{\partial H}{\partial \hat{p}^b} + \frac{1-\cos^2(\gamma)}{\det([g_{ij}])}\frac{\partial H}{\partial \hat{p}^c} \tag{7c}$$

where we obtained the $\boldsymbol{e}^i$ in terms of the $\boldsymbol{e}_i$ and took $\hat{v}_i$ to be all the components in the $\boldsymbol{e}_i$ direction.

Note that the derivates of the Hamiltonian with respect to momentum are straightforward to evaluate in k-space, where we recall the relation

$$\hat{p}^i = \hbar \hat{k}^i. \tag{8}$$

3 Example: Bernal Graphene

Bernal graphene has a hexagonal unit cell where $|a| = |b| = 2.46$ Å and $|c| = 6.70$ Å, and $\alpha = \beta = \pi/2$ and $\gamma = 2\pi/3$. So $\cos(\alpha) = \cos(\beta) = 0$ and $\cos(\gamma) = -1/2$, so that

$$\frac{1}{\det([g_{ij}])} = \frac{1}{1-1/4} = \frac{4}{3} \tag{9}$$

and we see

$$\hat{v}_a = \frac{4}{3}\frac{\partial H}{\partial \hat{p}^a} + \frac{2}{3}\frac{\partial H}{\partial \hat{p}^b} \tag{10a}$$

$$\hat{v}_b = \frac{2}{3}\frac{\partial H}{\partial \hat{p}^a} + \frac{4}{3}\frac{\partial H}{\partial \hat{p}^b} \tag{10b}$$

$$\hat{v}_c = \frac{\partial H}{\partial \hat{p}^c} \tag{10c}$$

One can then evaluate the momentum derivatives, for example by using the Slonczewski-Weiss-McClure model, or the Charlier-Michenaud-Gonze-Vigneron model for Bernal graphene.