Orbital magnetization

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We shall discuss the *modern theory of orbital magnetization*. This constitutes the first step for understanding the very interesting *topological magneto-electric effect with axion electrodynamics* (not discussing here, though).

I. SURFACE CHARGE AND SURFACE CURRENT [1, 2]

A. Surface charge and electric polarization

From the charge conservation, we have

$$\begin{aligned}\int\left(\frac{d\rho(\boldsymbol{r})}{dt}\right)dv &= \int\left(\boldsymbol{\nabla}\cdot\boldsymbol{J}\right)dv \\ &= \int\left(\boldsymbol{J}\cdot\boldsymbol{n}\right)dA. \end{aligned} \tag{1}$$

When we consider the surface region with the surface charge density σ_{surf} we have the total charge of

$$\int\left(\frac{d\rho(\boldsymbol{r})}{dt}\right)dv = \int\left(\frac{d\sigma_{\mathrm{surf}}}{dt}\right)dA. \tag{2}$$

This leads to

$$\frac{d\sigma_{\mathrm{surf}}}{dt} = \underbrace{\boldsymbol{J}}_{\frac{d\boldsymbol{P}}{dt}}\cdot\boldsymbol{n}, \tag{3}$$

and thus

$$\sigma_{\mathrm{surf}} = \boldsymbol{P}\cdot\boldsymbol{n}. \tag{4}$$

This *surface* charge density σ_{surf} is thus related to the *bulk* electric polarization $\boldsymbol{P}$. Here, the electric polarization $\boldsymbol{P}$ is given by the Zak phase

$$\phi_n(\lambda) = \int_0^{\frac{2\pi}{a}} dk\left(i\left\langle u_{k,n}(\lambda)\middle|\frac{\partial}{\partial k}\middle|u_{k,n}(\lambda)\right\rangle\right). \tag{5}$$

or the Wannier center

$$\bar{x}_n(\lambda) = \frac{a}{2\pi}\phi_n(\lambda) = \frac{a}{2\pi}\int_0^{\frac{2\pi}{a}} dk\left(i\left\langle u_{k,n}(\lambda)\middle|\frac{\partial}{\partial k}\middle|u_{k,n}(\lambda)\right\rangle\right). \tag{6}$$

for the filled band n; namely

$$\boldsymbol{P}(\lambda) = -\frac{e}{2\pi}\sum_n\phi_n(\lambda) = -\frac{e}{a}\sum_n\bar{x}_n(\lambda). \tag{7}$$

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B. Surface current and orbital magnetization

Let us seek the similar *surface-bulk* relation for the surface *bound* current $\boldsymbol{K}_{\text{surf}}$. The classical electromagnetism tells us that the magnetic dipole moment along z-axis m that is produced by a current I flowing in a loop on xy-plane is given by

$$m = IA \tag{8}$$

where A is the area of the loop. The relation can be rewritten in terms of vectors as

$$\underbrace{\begin{bmatrix} 0 \\ 0 \\ m \end{bmatrix}}_{\boldsymbol{m}} = A \underbrace{\begin{bmatrix} -I\sin\phi \\ I\cos\phi \\ 0 \end{bmatrix}}_{\boldsymbol{I}} \times \underbrace{\begin{bmatrix} \cos\phi \\ \sin\phi \\ 0 \end{bmatrix}}_{\boldsymbol{n}}, \tag{9}$$

where $\boldsymbol{n} = \begin{bmatrix} \cos\phi \\ \sin\phi \\ 0 \end{bmatrix}$ is the unit vector normal to the loop. The relation can be inverted to give

$$\underbrace{\begin{bmatrix} -I\sin\phi \\ I\cos\phi \\ 0 \end{bmatrix}}_{\boldsymbol{I}} = \frac{1}{A} \underbrace{\begin{bmatrix} 0 \\ 0 \\ m \end{bmatrix}}_{\boldsymbol{m}} \times \underbrace{\begin{bmatrix} \cos\phi \\ \sin\phi \\ 0 \end{bmatrix}}_{\boldsymbol{n}}. \tag{10}$$

Let us then consider a ribbon with width of d that is made by extruding the loop we have just considered. The relation (10) does not change. Dividing both sides of Eq.(10) by d we have

$$\boldsymbol{K}_{\text{surf}} = \frac{1}{Ad}\boldsymbol{m} \times \boldsymbol{n}. \tag{11}$$

where $\boldsymbol{K}_{\text{surf}} = \frac{\boldsymbol{I}}{d}$ is the *surface* current.

Now consider a magnetized cylinder with thickness of d having the top and bottom surfaces of area A. In this case, we can expect that

$$\boldsymbol{K}_{\text{surf}} = \boldsymbol{M} \times \boldsymbol{n} \tag{12}$$

hold. Here

$$\boldsymbol{M} = \frac{\boldsymbol{m}}{Ad}, \tag{13}$$

is the *magnetization*, that is, the magnetic moment per unit volume. As for the typical magnetized materials, namely, the ferromagnets, the magnetization $\boldsymbol{M}$ is predominantly emerged as a result that macroscopic number of the localized electron spins align in particular direction (here z-axis). Putting this *spin* magnetization $\boldsymbol{M}_{\text{spin}}$ aside we shall investigate the *orbital* magnetization $\boldsymbol{M}_{\text{orb}}$. The orbital magnetization $\boldsymbol{M}_{\text{orb}}$ emerged due to the the orbital degree of freedom of the Bloch electrons has attracted much attentions these days. The *surface-bulk* relation can thus be written as

$$\boldsymbol{K}_{\text{surf}} = \boldsymbol{M}_{\text{orb}} \times \boldsymbol{n}. \tag{14}$$

The next question is how can the orbital magnetization $\boldsymbol{M}_{\text{orb}}$ be expressed in terms of microscopic bulk quantities.

II. ORBITAL MAGNETIZATION [2, 3]

A. Real-space expression

Given an isolated atom, the ratio between the orbital magnetic dipole moment $\boldsymbol{m}_{\text{orb}}$ and the orbital angular momentum $\boldsymbol{L} = \boldsymbol{x} \times \boldsymbol{p}$ is called the gyromagnetic ratio, γ_s. The magnetic moment can thus be given in terms of $\boldsymbol{x}$

and $\boldsymbol{p}$ as

$$\boldsymbol{m}_{\rm orb} = \underbrace{\gamma_s}_{-\frac{e}{2m_e}} \left(\boldsymbol{x} \times \underbrace{\boldsymbol{p}}_{m_e \boldsymbol{v}}\right) = -\frac{e}{2}\left(\boldsymbol{x} \times \boldsymbol{v}\right). \tag{15}$$

Let us imagine that we have a 2D *topologically trivial* insulating crystal [3] with the area of A, wherein these isolated atoms form a 2D array. The orbital magnetization $\boldsymbol{M}_{\rm orb}$ would then be written as

$$\boldsymbol{M}_{\rm orb} = -\frac{e}{2A}\sum_i \left(\boldsymbol{x}_i \times \boldsymbol{v}_i\right). \tag{16}$$

The corresponding quantum-mechanical expression is given by

$$\boldsymbol{M}_{\rm orb} = -\frac{e}{2A}\sum_i \left\langle \phi_i \right| (\boldsymbol{x} \times \boldsymbol{v}) \left| \phi_i \right\rangle, \tag{17}$$

where $|\phi_i\rangle$ is the so-called *Wannier function* that is defined by Fourier-transforming the Bloch function $\left|\psi_{\boldsymbol{k}}\right\rangle = e^{i\boldsymbol{k}\cdot\boldsymbol{x}}\left|u_{\boldsymbol{k}}\right\rangle$, that is,

$$\begin{aligned} |\phi_i\rangle &= \frac{A_0}{(2\pi)^2}\int_{\rm BZ} d^2k e^{-i\boldsymbol{k}\cdot\boldsymbol{x}_i}\left|\psi_{\boldsymbol{k}}\right\rangle \\ &= \frac{A_0}{(2\pi)^2}\int_{\rm BZ} d^2k e^{-i\boldsymbol{k}\cdot(\boldsymbol{x}_i-\boldsymbol{x})}\left|u_{\boldsymbol{k}}\right\rangle \end{aligned} \tag{18}$$

which is localized at $\boldsymbol{x}_i$, where A_0 is the unit-cell area and the integration is performed within the Brillouin zone. Conversely, the Bloch function $\left|\psi_{\boldsymbol{k}}\right\rangle$ can be expressed by the Wannier functions $|\phi_i\rangle$ as

$$\left|\psi_{\boldsymbol{k}}\right\rangle = \sum_i e^{i\boldsymbol{k}\cdot\boldsymbol{x}_i}\left|\phi_i\right\rangle. \tag{19}$$

Here, we have a dangerous position operator $\boldsymbol{x}$ in Eq. (17). Following Thonhauser *et al.* [2, 3], let us split $\boldsymbol{M}_{\rm orb}$ in Eq. (17) into two contributions:

$$\boldsymbol{M}_{\rm orb} = \underbrace{\left(-\frac{e}{2A}\sum_i \left\langle\phi_i\right|((\boldsymbol{x}-\bar{\boldsymbol{x}}_i)\times\boldsymbol{v})\left|\phi_i\right\rangle\right)}_{\boldsymbol{M}_{LC}} + \underbrace{\left(-\frac{e}{2A}\sum_i \left\langle\phi_i\right|(\bar{\boldsymbol{x}}_i\times\boldsymbol{v})\left|\phi_i\right\rangle\right)}_{\boldsymbol{M}_{IC}}, \tag{20}$$

where

$$\bar{\boldsymbol{x}}_i = \left\langle\phi_i\right|\boldsymbol{x}\left|\phi_i\right\rangle = \frac{A_0}{(2\pi)^2}\int_{\rm BZ} d^2k\left(i\left\langle u_{\boldsymbol{k}}\left|\frac{\partial}{\partial\boldsymbol{k}}\right|u_{\boldsymbol{k}}\right\rangle\right) \tag{21}$$

is the 2D analog of the Wannier center. Here, $\boldsymbol{M}_{LC}$ can be interpreted as the magnetization arising due to the *local circulation* of the electrons within the unit-cell area A_0 in the interior bulk region. Using the translational symmetry, we have

$$\begin{aligned} \boldsymbol{M}_{LC} &= -\frac{e}{2\left(NA_0\right)}\sum_i \left\langle\phi_i\right|((\boldsymbol{x}-\bar{\boldsymbol{x}}_i)\times\boldsymbol{v})\left|\phi_i\right\rangle \\ &= -\frac{e}{2A_0}\left\langle\phi_0\right|((\boldsymbol{x}-\bar{\boldsymbol{x}}_0)\times\boldsymbol{v})\left|\phi_0\right\rangle \\ &= -\frac{e}{2A_0}\left\langle\phi_0\right|(\boldsymbol{x}\times\boldsymbol{v})\left|\phi_0\right\rangle \end{aligned} \tag{22}$$

since

$$\bar{\boldsymbol{v}}_0 = \left\langle\phi_0\right|\boldsymbol{v}\left|\phi_0\right\rangle = 0. \tag{23}$$

We can manipulate Eq. (22) a bit further to obtain

$$\begin{aligned}\boldsymbol{M}_{LC} &= -\frac{e}{2A_0}\left\langle \phi_0 \middle| \left(\boldsymbol{x} \times \left(-\frac{i}{\hbar}\left[\boldsymbol{x}, H\right] \right) \right) \middle| \phi_0 \right\rangle \\ &= \frac{ie}{2A_0\hbar} \underbrace{\langle \phi_0 | (\boldsymbol{x} \times \boldsymbol{x} H) | \phi_0 \rangle}_{0} - \frac{ie}{2A_0\hbar} \langle \phi_0 | (\boldsymbol{x} \times H\boldsymbol{x}) | \phi_0 \rangle \\ &= -\frac{ie}{2A_0\hbar} \langle \phi_0 | (\boldsymbol{x} \times H\boldsymbol{x}) | \phi_0 \rangle \end{aligned} \tag{24}$$

$\boldsymbol{M}_{IC}$ is, on the other hand, interpreted as the magnetization arising due to the *itinerant circulation* of the electrons only at the *surface* region. While in the interior region,

$$\boldsymbol{M}_{IC} = -\frac{e}{2A_0}\left(\bar{\boldsymbol{x}}_0 \times \underbrace{\bar{\boldsymbol{v}}_0}_{0} \right) = 0, \tag{25}$$

in the surface region, since

$$\bar{\boldsymbol{v}}_s = \langle \phi_s | \boldsymbol{v} | \phi_s \rangle \tag{26}$$

may not necessarily vanish at the surface, we have

$$\boldsymbol{M}_{IC} = -\frac{e}{2A}\sum_s \left(\bar{\boldsymbol{x}}_s \times \bar{\boldsymbol{v}}_s \right), \tag{27}$$

where the sum s runs only over the sites in the surface region.

B. Reciprocal-space expression

Back in the Bloch basis, after some algebra, Eq. (24) becomes

$$\boldsymbol{M}_{LC} = -\frac{e}{2\hbar}\mathrm{Im}\int \frac{d^2k}{(2\pi)^2}\left\langle \frac{\partial u_{\boldsymbol{k}}}{\partial \boldsymbol{k}} \middle| \times H_{\boldsymbol{k}} \middle| \frac{\partial u_{\boldsymbol{k}}}{\partial \boldsymbol{k}} \right\rangle, \tag{28}$$

where $H_{\boldsymbol{k}} = e^{-i\boldsymbol{k}\cdot\boldsymbol{x}} H e^{i\boldsymbol{k}\cdot\boldsymbol{x}}$ and Eq. (27) becomes

$$\boldsymbol{M}_{IC} = -\frac{e}{2\hbar}\mathrm{Im}\int \frac{d^2k}{(2\pi)^2} E_{\boldsymbol{k}} \underbrace{\left\langle \frac{\partial u_{\boldsymbol{k}}}{\partial \boldsymbol{k}} \middle| \times \middle| \frac{\partial u_{\boldsymbol{k}}}{\partial \boldsymbol{k}} \right\rangle}_{\boldsymbol{\Omega}_{\boldsymbol{k}}:\ \text{Berry curvature}}, \tag{29}$$

where $E_{\boldsymbol{k}} = \langle u_{\boldsymbol{k}} | H | u_{\boldsymbol{k}} \rangle$ is the band energy. We thus finally arrive at

$$\boldsymbol{M}_{\mathrm{orb}} = \boldsymbol{M}_{LC} + \boldsymbol{M}_{\boldsymbol{IC}} = -\frac{e}{2\hbar}\mathrm{Im}\int \frac{d^2k}{(2\pi)^2}\left\langle \frac{\partial u_{\boldsymbol{k}}}{\partial \boldsymbol{k}} \middle| \times \left(H_{\boldsymbol{k}} + E_{\boldsymbol{k}} \right) \middle| \frac{\partial u_{\boldsymbol{k}}}{\partial \boldsymbol{k}} \right\rangle, \tag{30}$$

an expression of $\boldsymbol{M}_{\mathrm{orb}}$ that contains only bulk quantities!

Unlike the electric polarization $\boldsymbol{P}$ in Eq. (7) which defined modulo e, however, the orbital magnetization $\boldsymbol{M}_{\mathrm{orb}}$ in Eq. (30) is well-defined [2].

[1] E. M. Purcell, *Electricity and Magnetism*, 2nd ed. (Cambridge University Press, Cambridge, 2011)

[2] D. Vanderbilt, *Berry Phases in Electronic Structure Theory: Electric Polarization, Orbital Magnetization and Topological Insulators*, (Cambridge University Press, Cambridge, 2018).

[3] T. Thonhauser, D. Ceresoli, D. Vanderbilt, and R. Resta, Phys. Rev. Lett. **95**, 137205 (2005).