

Theory of Orbital Magnetization in Solids

(Modern Theory of Orbital Magnetization)

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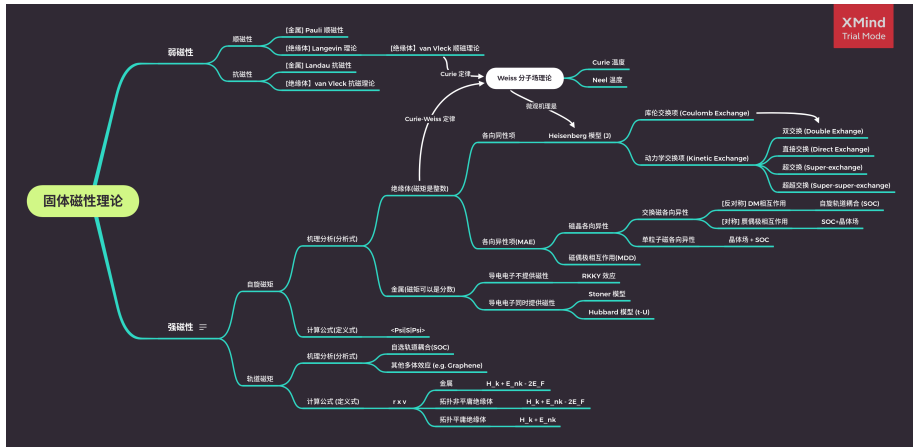
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- 2 Attempt for Solving Orbital Magnetization
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Tree of magnetic theory in solids



$$\mathbf{M}_{\text{total}} = \mathbf{M}_{\text{spin}} + \mathbf{M}_{\text{orb}}$$

Why we survived without M_{orb} ?

Someone may tell you

In many common materials of everyday interest, the orbital contribution **is small** compared to the spin contribution. The orbital magnetic moment **is usually weak or even quenched completely** due to time-reversal (or momentum-reversal) symmetry.

Table: Magnetization of some materials¹ (in μ_B)

Material	M_{spin}	$M_{\text{orb}}^{\text{Exp}}$	$M_{\text{orb}}^{\text{DFT}}$
<i>bcc</i> - Fe	2.083	0.081	0.066
<i>fcc</i> - Co	1.523	0.120	0.076
<i>fcc</i> - Ni	0.518	0.053	0.052

Question still there

Why orbital magnetic moment is **usually weak**?

¹T. Thonhauser, Int. J. Mod. Phys. B **25**, 1429 (2011).

Importance of the M_{orb}

- In some cases, the orbital magnetization is simply **cannot be ignored**.
- **A wealth of applications** are directly related to the orbital magnetization, which include but not limit to:
 - Nuclear magnetic resonance (NMR) in solid states^a
 - Electron paramagnetic resonance (EPR) g-tensor ^b
 - Magnetic susceptibility
 - Orbital magnetoelectric coupling and response^c
 - Spin Hall conductivity^d
 - Non-abelian quantum Hall states^e
- The **modern theory of orbital magnetization** is further important because of its close connection to the **modern theory of electric polarization** in solids.

^aT. Thonhauser *et al.*, J. Chem. Phys. **131**, 101101 (2009).

^bD. Ceresoli *et al.*, Phys. Rev. B **81**, 060409(R) (2010).

^cA.M. Essin *et al.*, Phys. Rev. Lett. **102**, 146805 (2009).

^dS. Murakami, Phys. Rev. Lett. **97**, 236805 (2006).

^eN. R. Cooper *et al.*, Phys. Rev. Lett. **102**, 176807 (2009).

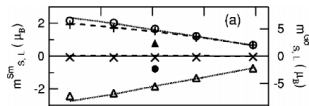
Some cases that cannot ignore the M_{orb}

- H. J. Gotsis *et al.*, Phys. Rev. B **68**, 224427 (2003).

U (Ry)	0.0	0/ $J=0$	0.2	0.4	0.5	0.6
$m_{\text{total}} (\mu_B)$	11.50	11.57	11.42	11.44	10.89	10.99
$m_{\text{orb}} (\mu_B)$	-2.57	-2.37	-3.80	-4.28	-4.78	-4.85
$m_{\text{spin}} (\mu_B)$	5.27	5.34	5.29	5.15	4.94	4.92
n_{+1}	0.65	0.78	0.95	0.97	0.98	0.98
n_{+2}	0.58	0.67	0.49	0.29	0.03	0.01
n_{+3}	0.41	0.32	0.02	0.01	0.02	0.01

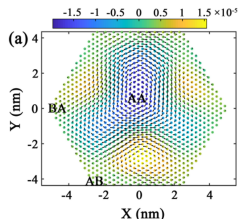
In this article, the DFT calculation result shows, the M_{orb} and M_{spin} of Sm 4*f* electrons in SmAl₂ is comparable.

- S. Qiao *et al.*, Phys. Rev. B **70**, 134418 (2004).



In this case, the x-ray experimental result shows that, the $M_{\text{spin}}(\bigcirc)$ and $M_{\text{orb}}(\triangle)$ of Sm in Sm_{0.982}Gd_{0.018}Al₂ is equally contribute to the total magnetization, and therefore canceled with each other.

- Si-Yu Li *et al.*, Phys. Rev. B **102**, 121406(R) (2020).



In twisted bilayer graphene, it is said that, the magnetization mainly comes from the M_{orb} , which is about 10.7 μ_B per moiré supercell.

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From classical model to the quantum system

The classical definition of the magnetic moment is,

$$\mathbf{m}_{\text{orb}} = \frac{1}{2c} \int d^3r \, \mathbf{r} \times \mathbf{J} = \frac{1}{2c} \int d^3r \, \mathbf{r} \times \rho \mathbf{v} \quad (1)$$

In the quantum system, the total orbital moment (\mathbf{m}_{orb}) is:

$$\mathbf{m}_{\text{orb}} = -\frac{e}{2c} \sum_n f_n \langle \psi_n | \hat{\mathbf{r}} \times \hat{\mathbf{v}} | \psi_n \rangle \quad (2a)$$

$$\mathbf{v} = -\frac{i}{\hbar} [\hat{\mathbf{r}}, \hat{H}] \quad (2b)$$

Then, the orbital magnetization (\mathbf{M}_{orb}) can be defined as the magnetic moment per unit volume,

$$\mathbf{M}_{\text{orb}} = \frac{\mathbf{m}_{\text{orb}}}{V_{\text{all}}} = -\frac{e}{2cV_{\text{all}}} \sum_n f_n \langle \psi_n | \hat{\mathbf{r}} \times \hat{\mathbf{v}} | \psi_n \rangle \quad (3)$$

where the V_{all} is the total volume of the target system.

The ill-defined $\hat{\mathbf{r}}$ under Bloch states

In the solids, the eigenstates $|\psi_n\rangle$ become Bloch states $|\psi_{n\mathbf{k}}\rangle$ with the Bloch wave vector \mathbf{k} ,

$$\mathbf{M}_{\text{orb}} = -\frac{e}{2cV_{\text{all}}} \sum_{n,\mathbf{k}} f_{n\mathbf{k}} \langle \psi_{n\mathbf{k}} | \hat{\mathbf{r}} \times \hat{\mathbf{v}} | \psi_{n\mathbf{k}} \rangle \quad (4a)$$

$$= -\frac{e}{2c} \sum_n \int_{\text{BZ}} \frac{d^d k}{(2\pi)^d} f_{n\mathbf{k}} \langle \psi_{n\mathbf{k}} | \hat{\mathbf{r}} \times \hat{\mathbf{v}} | \psi_{n\mathbf{k}} \rangle \quad (4b)$$

As we all know, the $\hat{\mathbf{r}}$ operator is ill-defined under Bloch states^a,

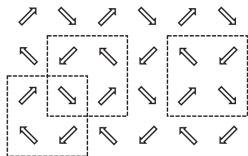
$$\begin{aligned} \langle \psi_{n\mathbf{k}} | \hat{\mathbf{r}} | \psi_{m\mathbf{k}'} \rangle &= (1 - \delta_{nm}) \delta(\mathbf{k} - \mathbf{k}') \mathbf{A}_{nm}(\mathbf{k}) \\ &\quad + \delta_{nm} \left[\delta(\mathbf{k} - \mathbf{k}') \mathbf{A}_{nn} + i \frac{\partial}{\partial \mathbf{k}} \delta(\mathbf{k} - \mathbf{k}') \right] \end{aligned} \quad (5)$$

where, \mathbf{A} is Berry connection, which is gauge dependent. Thus, $\langle \psi_{n\mathbf{k}} | \hat{\mathbf{r}} | \psi_{n\mathbf{k}} \rangle$ is divergent.

^aE.I. Blount, Solid State Phys. **13**, 305 (1962).

Using local bulk current?

One may hope to solve the problem from the point of view of local bulk current density $\mathbf{J}(\mathbf{r})$.



But in fact, the knowledge of $\mathbf{J}(\mathbf{r})$, in principle, is insufficient to calculate the \mathcal{M}_{orb} . Just like what happen when we try to using the electron density $\rho(\mathbf{r})$ to get the electric polarization \mathbf{P} in solids.

To see this, we define the magnetization $\mathcal{M}_{\text{orb}}(\mathbf{r})$ via $\mathbf{J}(\mathbf{r})$,

$$c\nabla \times \mathcal{M}_{\text{orb}}(\mathbf{r}) = \mathbf{J}(\mathbf{r}) \quad (6)$$

However, $\mathcal{M}_{\text{orb}}(\mathbf{r})$ can simply be repalced by another $\mathcal{M}'_{\text{orb}}(\mathbf{r})$, which corresponding to the same $\mathbf{J}(\mathbf{r})$,

$$\mathcal{M}_{\text{orb}}(\mathbf{r}) \rightarrow \mathcal{M}'_{\text{orb}}(\mathbf{r}) = \mathcal{M}_{\text{orb}}(\mathbf{r}) + \mathcal{M}_{\text{orb}}^0 + \nabla \xi(\mathbf{r}) \quad (7)$$

Muffin-tin approximation



Non-overlapping muffin-tin spheres center around the atoms in solids. Within these spheres, which are finite systems, the moment can be calculated according to,

$$\mathbf{m}_{\text{orb}} = -\frac{e}{2c} \sum_{n\mathbf{k}} f_n \langle \psi_{n\mathbf{k}} | \hat{\mathbf{r}} \times \hat{\mathbf{v}} | \psi_{n\mathbf{k}} \rangle \quad (8)$$

Often, the orbital magnetization indeed originates from regions near the atom cores, making this approximation acceptable.

Table: Orbital magnetization using muffin-tin approximation² (in μ_B)

Material	$M_{\text{orb}}^{\text{Exp}}$	$M_{\text{orb}}^{\text{DFT}}$	muffin-tin	interstitial
<i>bcc</i> - Fe	0.081	0.066	0.043	0.023
<i>fcc</i> - Co	0.120	0.076	0.063	0.013
<i>fcc</i> - Ni	0.053	0.052	0.051	0.001

²T. Thonhauser, Int. J. Mod. Phys. B **25**, 1429 (2011).

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Derivation in the Wannier representation

For now, let us focus on a simplest model: a **2D finite but large enough** solid, with **one topological trivial insulating band**, described by **spinless** Hamiltonian with **broken time-reversal symmetry**. As we show above, its orbital magnetization can be expressed as,

$$\mathbf{M}_{\text{orb}} = -\frac{e}{2cV_{\text{all}}} \sum_{\mathbf{k}} \langle \psi_{\mathbf{k}} | \hat{\mathbf{r}} \times \hat{\mathbf{v}} | \psi_{\mathbf{k}} \rangle \quad (9)$$

Using the Wannier representation,

$$\mathbf{M}_{\text{orb}} = -\frac{e}{2cV_{\text{all}}} \sum_{\mathbf{R}} \langle \omega_{\mathbf{R}} | \hat{\mathbf{r}} \times \hat{\mathbf{v}} | \omega_{\mathbf{R}} \rangle \quad (10)$$

where **\mathbf{R}** traverses all primitive cells in the solid.

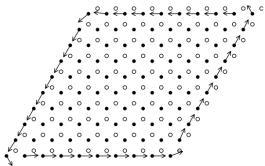
Key Point

It must be noted that, the Wannier function in such a solid system can be split into two regions: “**in the bulk**” and “**on the surface**”. Those two regions behave totally differently when it comes to Eq. (10).

Two terms in orbital magnetization

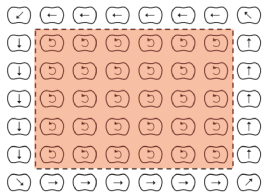
$$\begin{aligned}
 \mathbf{M}_{\text{orb}} &= -\frac{e}{2cV_{\text{all}}} \sum_{\mathbf{R}}^{\text{all}} \langle \omega_{\mathbf{R}} | \hat{\mathbf{r}} \times \hat{\mathbf{v}} | \omega_{\mathbf{R}} \rangle \\
 &= -\frac{e}{2cV_{\text{all}}} \sum_{\mathbf{R}}^{\text{bulk}} \langle \omega_{\mathbf{R}} | \hat{\mathbf{r}} \times \hat{\mathbf{v}} | \omega_{\mathbf{R}} \rangle - \frac{e}{2cV_{\text{all}}} \sum_{\mathbf{R}}^{\text{surf}} \langle \omega_{\mathbf{R}} | \hat{\mathbf{r}} \times \hat{\mathbf{v}} | \omega_{\mathbf{R}} \rangle \\
 &= -\frac{e}{2cV_{\text{all}}} \sum_{\mathbf{R}}^{\text{all}} \langle \omega_{\mathbf{R}} | (\hat{\mathbf{r}} - \mathbf{r}_{\mathbf{R}}) \times \hat{\mathbf{v}} | \omega_{\mathbf{R}} \rangle - \frac{e}{2cV_{\text{all}}} \sum_{\mathbf{R}}^{\text{all}} \langle \omega_{\mathbf{R}} | \mathbf{r}_{\mathbf{R}} \times \hat{\mathbf{v}} | \omega_{\mathbf{R}} \rangle \\
 &= \mathbf{M}_{\text{orb}}^{\text{LC}} + \mathbf{M}_{\text{orb}}^{\text{IC}}
 \end{aligned} \tag{11}$$

where $\mathcal{O}_s = \langle \omega_s | \hat{\mathcal{O}} | \omega_s \rangle$.



The 1st term is what we called “**local circulation(LC)**” term, the 2nd is “**itinerant circulation(IC)**”. And, as what we show in second step, the \mathbf{R} summation contains the **bulk** and **surface** regions.

Local circulation term $M_{\text{orb}}^{\text{LC}}$

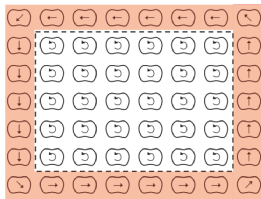


The “LC” term corresponding to the magnetization mainly generated by the bulk electrons.

$$\begin{aligned}
 M_{\text{orb}}^{\text{LC}} &= -\frac{e}{2cV_{\text{all}}} \sum_{\mathbf{R}}^{\text{all}} \langle \omega_{\mathbf{R}} | (\hat{\mathbf{r}} - \mathbf{r}_{\mathbf{R}}) \times \hat{\mathbf{v}} | \omega_{\mathbf{R}} \rangle \\
 &\approx -\frac{e}{2cV_{\text{all}}} \sum_{\mathbf{R}}^{\text{bulk}} \langle \omega_{\mathbf{R}} | (\hat{\mathbf{r}} - \mathbf{r}_{\mathbf{R}}) \times \hat{\mathbf{v}} | \omega_{\mathbf{R}} \rangle \\
 &= -\frac{e}{2cV_{\text{all}}} (N - n) \langle \omega_0 | (\hat{\mathbf{r}} - \mathbf{r}_0) \times \hat{\mathbf{v}} | \omega_0 \rangle \\
 &\approx -\frac{eN}{2cV_{\text{all}}} \langle \omega_0 | (\hat{\mathbf{r}} - \mathbf{r}_0) \times \hat{\mathbf{v}} | \omega_0 \rangle \\
 &= -\frac{e}{2cV_{\text{cell}}} \langle \omega_0 | \hat{\mathbf{r}} \times \hat{\mathbf{v}} | \omega_0 \rangle
 \end{aligned} \tag{12}$$

where the n and N is the Wannier functions' quantity on the surface and in the whole system, while $N \sim n^2$. ω_0 is one Wannier function in the bulk, V_{cell} is the volume of a primitive cell.

Itinerant circulation term $M_{\text{orb}}^{\text{IC}}$



The “IC” term corresponding to the magnetization mainly generated by the surface electrons.

$$\begin{aligned}
 M_{\text{orb}}^{\text{IC}} &= -\frac{e}{2cV_{\text{all}}} \sum_{\mathbf{R}}^{\text{all}} \langle \omega_{\mathbf{R}} | \mathbf{r}_{\mathbf{R}} \times \hat{\mathbf{v}} | \omega_{\mathbf{R}} \rangle \\
 &= -\frac{e}{2cV_{\text{all}}} \sum_{\mathbf{R}}^{\text{all}} \mathbf{r}_{\mathbf{R}} \times \mathbf{v}_{\mathbf{R}} \\
 &= -\frac{e}{2cV_{\text{all}}} \sum_{\mathbf{R}}^{\text{surf}} \mathbf{r}_{\mathbf{R}} \times \mathbf{v}_{\mathbf{R}} \\
 &\approx -\frac{e}{2cV_{\text{cell}}} \frac{n}{N} \mathbf{r}_{\text{surf}} \times \mathbf{v}_{\text{surf}}
 \end{aligned} \tag{13}$$

where, \mathbf{r}_{surf} and \mathbf{v}_{surf} is the absolute position and velocity of the surface wannier functions.

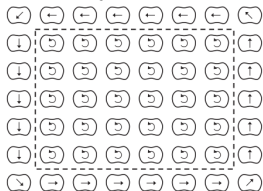
Express $M_{\text{orb}}^{\text{IC}}$ using bulk Wannier functions (2D system)

With some tricky derivation³, one can express the $M_{\text{orb}}^{\text{IC}}$ with bulk Wannier functions.

$$M_{\text{orb}}^{\text{IC},z} = -\frac{e}{2cV_{\text{all}}} \sum_{\mathbf{R}}^{\text{surf}} \mathbf{r}_{\mathbf{R}} \times \mathbf{v}_{\mathbf{R}}$$

$$= \frac{e}{2\hbar c V_{\text{cell}}} \text{Im} \left\{ \sum_{\mathbf{R}} \langle \omega_0 | \hat{H} | \omega_{\mathbf{R}} \rangle (R_x \langle \omega_{\mathbf{R}} | \hat{y} | \omega_0 \rangle - R_y \langle \omega_{\mathbf{R}} | \hat{x} | \omega_0 \rangle) \right\} \quad (14)$$

where, $\hat{\mathbf{r}} = (\hat{x}, \hat{y}, \hat{z})$, $\mathbf{R} = (R_x, R_y, R_z)$, $M_{\text{orb}}^{\text{IC},z}$ represent the magnetization in z direction, for this is a 2D system.



BE CAREFUL, the relation above is valid only when the edge states **can merge into the bulk adiabatically**.

The surface term can express with the bulk wannier functions.

³T. Thonhauser *et al.*, Phys. Rev. Lett. **95**, 137205 (2005).

Back to Bloch representation (2D system)

The final step is transform the expressions for $M_{\text{orb}}^{\text{LC}}$ and $M_{\text{orb}}^{\text{IC}}$ back to the Bloch representation.

$$|\omega_{\mathbf{R}}\rangle = \frac{V_{\text{cell}}}{(2\pi)^d} \int d^d k \, e^{-i\mathbf{k}\cdot(\mathbf{r}-\mathbf{R})} |u_{\mathbf{k}}\rangle \quad (15a)$$

$$|u_{\mathbf{k}}\rangle = e^{-i\mathbf{k}\cdot\mathbf{r}} |\psi_{\mathbf{k}}\rangle \quad (15b)$$

Little surprisingly, we get quite a concise formula,

$$M_{\text{orb}}^{\text{LC},z} = \frac{e}{\hbar c} \text{Im} \int_{\text{BZ}} \frac{d^2 k}{(2\pi)^2} \langle \partial_x u_{\mathbf{k}} | \hat{H}_{\mathbf{k}} | \partial_y u_{\mathbf{k}} \rangle \quad (16a)$$

$$M_{\text{orb}}^{\text{IC},z} = \frac{e}{\hbar c} \text{Im} \int_{\text{BZ}} \frac{d^2 k}{(2\pi)^2} \langle \partial_x u_{\mathbf{k}} | E_{\mathbf{k}} | \partial_y u_{\mathbf{k}} \rangle \quad (16b)$$

where, $\partial_i \equiv \partial/\partial k_i$, $\hat{H}_{\mathbf{k}} \equiv e^{-i\mathbf{k}\cdot\mathbf{r}} \hat{H} e^{i\mathbf{k}\cdot\mathbf{r}}$, the $E_{\mathbf{k}}$ denote the corresponding eigenvalues.

Extended to 3D

The orbital magnetization formula can be easily extended to 3D,

$$M_{\text{orb}}^{\text{LC}} = \frac{e}{2\hbar c} \text{Im} \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} \langle \nabla_{\mathbf{k}} u_{\mathbf{k}} | \times \hat{H}_{\mathbf{k}} | \nabla_{\mathbf{k}} u_{\mathbf{k}} \rangle \quad (17a)$$

$$M_{\text{orb}}^{\text{IC}} = \frac{e}{2\hbar c} \text{Im} \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} \langle \nabla_{\mathbf{k}} u_{\mathbf{k}} | \times E_{\mathbf{k}} | \nabla_{\mathbf{k}} u_{\mathbf{k}} \rangle \quad (17b)$$

You may already notice that, the “IC” term can further be written with the Berry curvature $\Omega_{\mathbf{k}}$,

$$M_{\text{orb}}^{\text{IC}} = -\frac{e}{2\hbar c} \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} E_{\mathbf{k}} \Omega_{\mathbf{k}} \quad (18)$$

Anyway, for now, we get the orbital magnetization for a one band spinless trivial insulating system,

$$M_{\text{orb}} = \frac{e}{2\hbar c} \text{Im} \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} \langle \nabla_{\mathbf{k}} u_{\mathbf{k}} | \times (\hat{H}_{\mathbf{k}} + E_{\mathbf{k}}) | \nabla_{\mathbf{k}} u_{\mathbf{k}} \rangle \quad (19)$$

A paradox about the energy shift

As we all know, an energy shift on the Hamiltonian does not change its physical behaviors, but

$$\begin{aligned} M_{\text{orb}} \rightarrow M'_{\text{orb}} &= \frac{e}{2\hbar c} \text{Im} \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} \langle \nabla_{\mathbf{k}} u_{\mathbf{k}} | \times (\hat{H}_{\mathbf{k}} + \varepsilon + E_{\mathbf{k}} + \varepsilon) | \nabla_{\mathbf{k}} u_{\mathbf{k}} \rangle \\ \Delta M_{\text{orb}} &= M'_{\text{orb}} - M_{\text{orb}} = \frac{e\varepsilon}{\hbar c (2\pi)^2} \mathbf{C} \end{aligned} \quad (20)$$

where, $\mathbf{C} = (C_x, C_y, C_z)$ is the Chern number in three directions, ε is the energy shift of the Hamiltonian.

Paradox

That means, in a system whose Chern number not equal to zero (e.g., metal or topological non-trivial insulator), there will be something weird.

The explanation for this “paradox” can be revealed from the pre-condition that the following equation satisfied.

$$M_{\text{orb}}^{\text{IC}} = \frac{e}{2\hbar c} \text{Im} \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} \langle \nabla_{\mathbf{k}} u_{\mathbf{k}} | \times E_{\mathbf{k}} | \nabla_{\mathbf{k}} u_{\mathbf{k}} \rangle$$

Orbital magnetization for non-zero Chern number system

$$M_{\text{orb}}^{\text{IC}} \doteq \frac{e}{2\hbar c} \text{Im} \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} \langle \nabla_{\mathbf{k}} u_{\mathbf{k}} | \times E_{\mathbf{k}} | \nabla_{\mathbf{k}} u_{\mathbf{k}} \rangle$$

As we mention before, there is a pre-condition (○) when we using the bulk wannier function represent “IC” term, that is, **the edge states can merge into the bulk adiabatically**. But for the edge states protected by the topology, obviously, that pre-condition cannot be satisfied.

Topological edge states term

In other words, the M_{orb} must add a 3rd term corresponding to the topological edge state. Let us show it first and explain later.

$$M_{\text{orb}}^{\text{TP}} = \frac{e}{2\hbar c} \text{Im} \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} \langle \nabla_{\mathbf{k}} u_{\mathbf{k}} | \times (-2\mu_0) | \nabla_{\mathbf{k}} u_{\mathbf{k}} \rangle \quad (21)$$

where, μ_0 is the chemical potential under zero temperature, which is exactly the Fermi energy, $\mu_0 = E_{\text{F}}$.

Topological edge states term

Now, let us consider a 2D system,

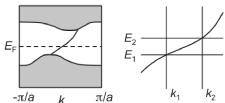
$$M_{\text{orb}}^{\text{TP},z} = \frac{e}{\hbar c} \text{Im} \int_{\text{BZ}} \frac{d^2 k}{(2\pi)^2} \langle \partial_x u_{\mathbf{k}} | (-2\mu_0) | \partial_y u_{\mathbf{k}} \rangle \quad (22)$$

$$\frac{dM_{\text{orb}}^z}{d\mu_0} = \frac{dM_{\text{orb}}^{\text{TP},z}}{d\mu_0} = -\frac{e}{2\pi\hbar c} C \quad (23)$$

Owing to the main equation $c\nabla \times \mathbf{M} = \mathbf{j}$, a macroscopic current of intensity $I = cM_{\text{orb}}^z$, together with Eq. (23),

$$\frac{dI}{d\mu_0} = -\frac{eC}{2\pi\hbar} \quad (24)$$

On the other hand, raising the chemical potential by $d\mu_0$ fills $dk/2\pi$ states per unit length on one band, which means,



$$dI = -env \frac{dk}{2\pi} = -en \frac{1}{\hbar} \frac{dE_{\text{total}}}{dk} \frac{dk}{2\pi} \approx -\frac{en}{2\pi\hbar} d\mu_0 \quad (25)$$

where, v is the electrons' group velocity, n is the bands' quantity that participate in conducting.

Compare Eq. (24) and Eq. (25), we can conclude that, the Chern number C is exactly the number of chiral edge channels n .

Final formula for orbital magnetization in solids

Orbital Magnetization

$$\mathbf{M}_{\text{orb}} = \frac{e}{2\hbar c} \text{Im} \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} \langle \nabla_{\mathbf{k}} u_{\mathbf{k}} | \times (\hat{H}_{\mathbf{k}} + E_{\mathbf{k}} - 2\mu_0) | \nabla_{\mathbf{k}} u_{\mathbf{k}} \rangle \quad (26a)$$

$$= \frac{e}{2\hbar c} \text{Im} \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} \langle \nabla_{\mathbf{k}} u_{\mathbf{k}} | \times (\hat{H}_{\mathbf{k}} + E_{\mathbf{k}} - 2E_F) | \nabla_{\mathbf{k}} u_{\mathbf{k}} \rangle \quad (26b)$$

Orbital Magnetization for Multiple Bands

For the multiple band case, we just need to include the summation for bands,

Orbital Magnetization for Multiple Bands

$$M_{\text{orb}} = \frac{e}{2\hbar c} \text{Im} \sum_n \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} f_{n\mathbf{k}} \langle \nabla_{\mathbf{k}} u_{n\mathbf{k}} | \times (\hat{H}_{\mathbf{k}} + E_{n\mathbf{k}} - 2\mu_0) | \nabla_{\mathbf{k}} u_{n\mathbf{k}} \rangle \quad (27a)$$

$$= \frac{e}{2\hbar c} \text{Im} \sum_n \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} f_{n\mathbf{k}} \langle \nabla_{\mathbf{k}} u_{n\mathbf{k}} | \times (\hat{H}_{\mathbf{k}} + E_{n\mathbf{k}} - 2E_F) | \nabla_{\mathbf{k}} u_{n\mathbf{k}} \rangle \quad (27b)$$

where, $f_{n\mathbf{k}}$ is the electrons occupation function.

The semi-classical derivation for M_{orb} (Part I)

PRL **95**, 137204 (2005)

PHYSICAL REVIEW LETTERS

week ending
23 SEPTEMBER 2005

Berry Phase Correction to Electron Density of States in Solids

Di Xiao, Junren Shi, and Qian Niu

Department of Physics, The University of Texas, Austin, Texas 78712-0264, USA

(Received 14 February 2005; published 22 September 2005; corrected 5 October 2005)

A very different derivation of M_{orb} was given by Di Xiao *et al.* based on the semiclassical theory,

$$\dot{\mathbf{r}} = \frac{1}{\hbar} \partial_{\mathbf{k}} E_n(\mathbf{k}) - \dot{\mathbf{k}} \times \boldsymbol{\Omega}_n(\mathbf{k}) \quad (28a)$$

$$\dot{\mathbf{k}} = -\frac{e}{\hbar} \mathbf{F}(\mathbf{r}) - \frac{e}{\hbar} \dot{\mathbf{r}} \times \mathbf{B}(\mathbf{r}) \quad (28b)$$

$$n_e = \frac{N_e}{V_{\text{all}}} = \int^{\mu(\mathbf{B})} \frac{d^d \mathbf{k}}{(2\pi)^d} \left(1 + \frac{e \mathbf{B} \cdot \boldsymbol{\Omega}}{\hbar} \right) \quad (28c)$$

In 1999, Sundaram and Niu observed that, the orbital magnetic moment of a wave packet centered at \mathbf{k} in band n is,⁴

$$\mathbf{m}_{n\mathbf{k}} = \frac{-ie}{2\hbar c} \langle \nabla_{\mathbf{k}} u_{n\mathbf{k}} | \times (\hat{H}_{\mathbf{k}} - E_{n\mathbf{k}}) | \nabla_{\mathbf{k}} u_{n\mathbf{k}} \rangle \quad (29)$$

⁴G. Sundaram *et al.*, Phys. Rev. B **59**, 14915 (1999).

The semi-classical derivation for M_{orb} (Part II)

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Berry Phase Correction to Electron Density of States in Solids

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Combine the Eq. (28) and Eq. (29), they find the total energy of the whole system can be written as,

$$E_{\text{total}} = \sum_n \int^{\mu(B)} \frac{d^3k}{(2\pi)^3} f_{n\mathbf{k}} \left(1 + \frac{e}{\hbar c} \mathbf{B} \cdot \boldsymbol{\Omega}_{n\mathbf{k}} \right) (E_{n\mathbf{k}} - \mathbf{m}_{n\mathbf{k}} \cdot \mathbf{B}) \quad (30)$$

Then the orbital magnetization can be directly calculated as,

$$M_{\text{orb}} = \nabla_B E_{\text{total}}|_{B=0} = M_{\text{orb}}^{\text{MOM}} + M_{\text{orb}}^{\text{DOS}} \quad (31a)$$

$$M_{\text{orb}}^{\text{MOM}} = \frac{e}{2\hbar c} \text{Im} \sum_n \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} f_{n\mathbf{k}} \langle \nabla_{\mathbf{k}} u_{n\mathbf{k}} | \times (\hat{H}_{\mathbf{k}} - E_{n\mathbf{k}}) | \nabla_{\mathbf{k}} u_{n\mathbf{k}} \rangle \quad (31b)$$

$$M_{\text{orb}}^{\text{DOS}} = \frac{e}{2\hbar c} \text{Im} \sum_n \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} f_{n\mathbf{k}} \langle \nabla_{\mathbf{k}} u_{n\mathbf{k}} | \times 2(E_{n\mathbf{k}} - \mu_0) | \nabla_{\mathbf{k}} u_{n\mathbf{k}} \rangle \quad (31c)$$

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Compare orbital magnetization M_{orb} and polarization P

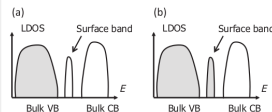
Both of the M_{orb} and P comes from the ill-defined operator \hat{r} , and because of which, have a close relation with the **Berry connection**.

In the M_{orb} , we use $\langle \psi_{\mathbf{k}} | \mathbf{r} \times \mathbf{v} | \psi_{\mathbf{k}} \rangle \rightarrow \langle \omega_0 | \mathbf{r} \times \mathbf{v} | \omega_0 \rangle$ to sneak by the problem, while, in the P , $\langle \psi_{\mathbf{k}} | \mathbf{r} | \psi_{\mathbf{k}} \rangle \rightarrow \langle \omega_0 | \mathbf{r} | \omega_0 \rangle$ do the same.

Unlike P , which only involved the Bloch functions $|u_{n\mathbf{k}}\rangle$, the M_{orb} also **need the Hamiltonian**. E.g., if \hat{H} is scaled by a multiplicative factor, M_{orb} gets the same scaled, while P remain invariant.

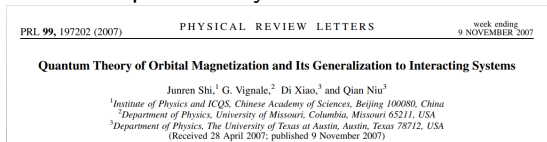
Unlike P , which is a “valued lattice”, the M_{orb} is **single valued**. It can be understand from two perspectives,

- Solids allow the static charge on the surface, but do not allow surface charge accumulation.
- In nature, there are electric charges but no magnetic monopoles.

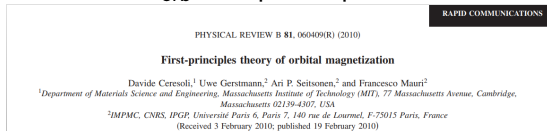


Theory beyond “modern theory”

- Finite-temperature system.



- Calculate M_{orb} in a pseudopotential context.



- Many-particle version M_{orb} .



Total magnetization in solids

Total magnetization in solids

The total magnetization of the system can be written as follow,

$$M_{\text{total}} = M_{\text{spin}} + M_{\text{orb}} \quad (32a)$$

$$M_{\text{spin}} = \mu_B \frac{-g_s}{\hbar} \sum_n \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} f_{n\mathbf{k}} \langle \psi_{n\mathbf{k}} | \hat{\mathbf{S}} | \psi_{n\mathbf{k}} \rangle \quad (32b)$$

$$M_{\text{orb}} = \mu_B \frac{m_e}{\hbar^2} \sum_n \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} f_{n\mathbf{k}} \text{Im} \langle \nabla_{\mathbf{k}} u_{n\mathbf{k}} | \times (\hat{H}_{\mathbf{k}} + E_{n\mathbf{k}} - 2\mu_0) | \nabla_{\mathbf{k}} u_{n\mathbf{k}} \rangle \quad (32c)$$

An interesting question

Why the M_{spin} is often far large than (10 to 100 times) M_{orb} in solids?

Why M_{spin} is often large than M_{orb} ?

The effective Hamiltonian near the ground state:

$$\hat{H}_{\text{eff}} = \begin{pmatrix} \hat{H}_{\uparrow} & \hat{W}_{\text{SOC}} \\ \hat{W}_{\text{SOC}} & \hat{H}_{\downarrow} \end{pmatrix}, \quad \hat{W}_{\text{SOC}} \approx \lambda \hat{\mathbf{L}} \cdot \hat{\mathbf{S}} \quad (33)$$

Some personal arguments

- In solid, the time-reversal symmetry (TRS) breaking in spin channels is common, for it is a requirement of fermions symmetry.
- The TRS breaking in orbital space is not as common as spin. Usually, orbits “borrows” the TRS breaking from spin space with the help of spin-orbit coupling (SOC).
- The orbital magnetization introduced via SOC may have the same magnitude order as SOC.
- There **are** ways to break the orbital TRS without SOC. In those cases, theoretically, the orbital magnetic moment can have a comparable value as spin's. (That is another long story...)

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Summary

- $M_{\text{total}} = M_{\text{spin}} + M_{\text{orb}}$. Often, the M_{spin} accounts for a large proportion.
- The modern theory of orbital magnetization gives,

$$M_{\text{orb}} = \frac{e}{2\hbar c} \text{Im} \sum_n \int_{\text{BZ}} \frac{d^3k}{(2\pi)^3} f_{n\mathbf{k}} \langle \nabla_{\mathbf{k}} u_{n\mathbf{k}} | \times (\hat{H}_{\mathbf{k}} + E_{n\mathbf{k}} - 2\mu_0) | \nabla_{\mathbf{k}} u_{n\mathbf{k}} \rangle$$

which is a single particle equation valid for both insulators and metals.

- Besides the trivial edge state, the topological edge state also gives a term in the orbital magnetization.
- The same equation can also gain from the semi-classical theory.
- The modern theory of orbital magnetization have a close relation with the modern theory of polarization.