Orbital Magnetization in Condensed Matter: Part 1

Raffaele Resta

Trieste, 2020

Outline

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Back to basics: Macroscopics

$$\begin{aligned} \mathbf{B} &= \mathbf{H} + 4\pi\mathbf{M} \\ \mathbf{M} &= \mathbf{M}_{\text{spin}} + \mathbf{M}_{\text{orbital}} \end{aligned}$$

- $\mathbf{M}_{\text{spin}}$ and $\mathbf{M}_{\text{orbital}}$ separately measurable **(really?)**
- Spontaneous $\mathbf{M}$ (in $\mathbf{B} = 0$) in ferromagnetic materials, orbital & spin, due to spin-orbit interaction.
- Induced $\mathbf{M}$ by a time-reversal-symmetry breaking perturbation (e.g. a macroscopic $\mathbf{B}$ field).
  $\mathbf{M}$ is purely orbital in a nonmagnetic insulator.

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Electric-magnetic analogies in continuous media

$$\mathbf{B} = \mathbf{H} + 4\pi\mathbf{M} \qquad \mathbf{E} = \mathbf{D} - 4\pi\mathbf{P}$$

$$\nabla \times \mathbf{M} = \mathbf{j}/c \qquad \nabla \cdot \mathbf{P} = -\rho$$

- A dissipationless current circulates at the surface of a uniformly magnetized sample:

$$\mathbf{K}_{\text{surface}} = c\mathbf{M} \times \mathbf{n}$$

- A surface charge piles up at the surface of a uniformly polarized sample:

$$\sigma_{\text{surface}} = -\mathbf{P} \cdot \mathbf{n}$$

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(for both crystalline and disordered systems)

- The system has **no surface** by construction.
- Any quantity defined or computed within PBC is by definition "bulk".
- However... The position operator **r** is **incompatible** with Born-von Kármán PBCs.
- The matrix elements of **r** over Bloch orbitals are **ill defined**.
- Because of this, the problem of macroscopic electric polarization remained **unsolved** until the early 1990s.
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Heuristically, by analogy with the electrical case

- For an insulator, in absence of inversion symmetry, in zero **E** field, we have

$$\mathbf{P}_{\text{electronic}} = -\frac{2e}{V_{\text{cell}}} \sum_{n \in \text{occupied}} \langle w_n | \mathbf{r} | w_n \rangle$$

- By analogy, in absence of time-reversal symmetry, in zero **B** field, it is tempting to write:

$$\mathbf{M} = -\frac{2e}{2cV_{\text{cell}}} \sum_{n \in \text{occupied}} \langle w_n | \mathbf{r} \times \mathbf{v} | w_n \rangle$$

- **Question:** Is this the correct formula for the bulk magnetization
- **Answer:** No!
  There is an additional term, having no electrical analogue.

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From Wannier back to Bloch

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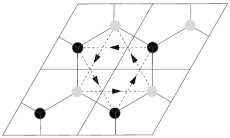
The Haldanium model material
(Haldane, PRL 1988)



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- Insulator at half-filling (only the lowest band occupied).
- Zero flux through the unit cell:
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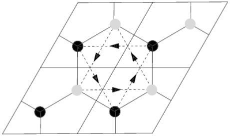

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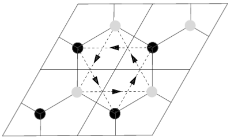

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(2d, single occupancy, single band, atomic units)

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- **(B)** Finite system of area L^2 cut from the bulk (so-called "open" boundary conditions)

$$M = \frac{1}{2cL^2} \int d\mathbf{r} \; \mathbf{r} \times \mathbf{j}(\mathbf{r}) = \frac{1}{2cL^2} \sum_{n \in \text{occupied}} \langle \varphi_n | \mathbf{r} \times \mathbf{v} | \varphi_n \rangle$$

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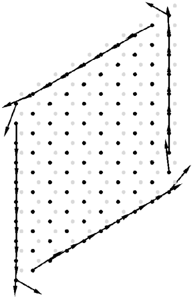
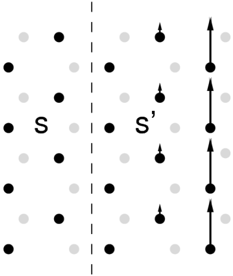
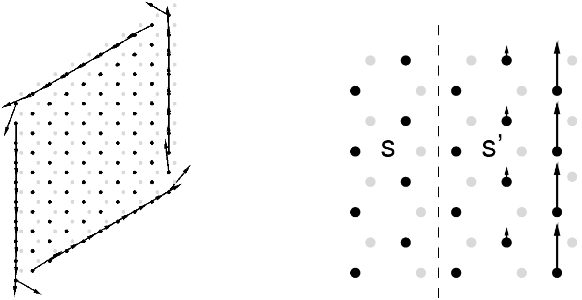
Chasing the missing term:
Localized-orbital (Boys/Wannier) analysis





- The Boys/Wannier localized orbitals at the sample edge carry a **net** current and contribute to **M**.

Chasing the missing term: Localized-orbital (Boys/Wannier) analysis



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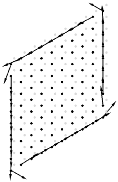
The historical derivation



- Even the additional edge contribution can be computed using Bloch states and PBCs, where the system has no edge.

- This is possible **only** in trivial insulators:
  no Chern insulators, no metals

- Formulated in this way in Vanderbilt's textbook

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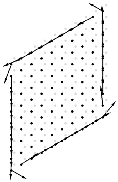

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The very first calculation for real materials
(D. Ceresoli, U. Gertsmann, A.P. Seitsonen, & F. Mauri, 2010)

Metal	e	Expt.	FLAPW		This method	
			LDA	PBE	LDA	PBE
bcc-Fe	[001]	0.081	0.053	0.051	0.0640	0.0658
bcc-Fe	[111]	–	–	–	0.0633	0.0660
hcp-Co	[001]	0.133	0.069	0.073	0.0924	0.0957
hcp-Co	[100]	–	–	–	0.0837	0.0867
fcc-Ni	[111]	0.053	0.038	0.037	0.0315	0.0519
fcc-Ni	[001]	–	–	–	0.0308	0.0556

TABLE III: Orbital magnetization $\mathbf{M}(e)$ in μ_B per atom of ferromagnetic metals parallel to the spin, for different spin orientations e. The easy axis for Fe, Co and Ni are, respectively, [001], [001] and [111]. Experimental results from Ref. 24; FLAPW results from Ref. 5.

Caveat: Role of the core electrons in a pseudopotential framework

Outline

The textbook formulæ

$$\mathbf{M} = \frac{\mathbf{m}}{V} = \frac{1}{2cV}\int d\mathbf{r}\, \mathbf{r} \times \mathbf{j}(\mathbf{r})$$

$$\mathbf{P} = \frac{\mathbf{d}}{V} = \frac{1}{V}\int d\mathbf{r}\, \mathbf{r}\, \rho(\mathbf{r})$$

The textbook formulæ

$$\mathbf{M} = \frac{\mathbf{m}}{V} = \frac{1}{2cV} \int d\mathbf{r} \, \textcolor{red}{\mathbf{r}} \times \mathbf{j}(\mathbf{r})$$

$$\mathbf{P} = \frac{\mathbf{d}}{V} = \frac{1}{V} \int d\mathbf{r} \, \textcolor{red}{\mathbf{r}} \, \rho(\mathbf{r})$$

P and **M** as reciprocal-space integrals

- 1992: **Polarization** (insulators)

$$P_\alpha = -\frac{2ie}{(2\pi)^d} \sum_{j=1}^{n_b} \int_{\mathrm{BZ}} d\mathbf{k} \, \langle u_{j\mathbf{k}} | \partial_\alpha u_{j\mathbf{k}} \rangle + P_\alpha^{(\mathrm{nuclei})}$$

- 2005-06: **Orbital magnetization** (including metals)

$$M_\gamma = -\frac{ie}{2\hbar c} \varepsilon_{\gamma\alpha\beta} \sum_j \int_{\epsilon_{j\mathbf{k}} \leq \mu} \frac{d\mathbf{k}}{(2\pi)^d} \langle \partial_\alpha u_{j\mathbf{k}} | \left(H_\mathbf{k} + \epsilon_{j\mathbf{k}} - 2\mu \right) | \partial_\beta u_{j\mathbf{k}} \rangle$$

- Do they have anything in common?

P and **M** as reciprocal-space integrals

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They could not be more different!

$$P_\alpha^{(\mathrm{el})} = -2ie \sum_{j=1}^{n_b} \int_{\mathrm{BZ}} \frac{d\mathbf{k}}{(2\pi)^d} \langle u_{j\mathbf{k}} | \partial_{k_\alpha} u_{j\mathbf{k}} \rangle = -2e \int_{\mathrm{BZ}} \frac{d\mathbf{k}}{(2\pi)^d} \mathcal{A}_\alpha(\mathbf{k})$$

$$M_\gamma = -\frac{ie}{2\hbar c} \varepsilon_{\gamma\alpha\beta} \sum_{\varepsilon_{j\mathbf{k}}<\mu} \int_{\mathrm{BZ}} \frac{d\mathbf{k}}{(2\pi)^d} \langle \partial_{k_\alpha} u_{j\mathbf{k}} | (H_\mathbf{k} + \epsilon_{j\mathbf{k}} - 2\mu) | \partial_{k_\beta} u_{j\mathbf{k}} \rangle$$

- **Polarization**
  - Insulators only
  - Gauge-dependent integrand
  - Integral of a 1-form
  - At bare bones, **P** is 1-dimensional
  - Bulk **P** multiple valued
  - Tinkering with the boundaries can alter **P**
- **Orbital Magnetization**
  - Insulators and metals
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- **Orbital Magnetization:** **prototype of class II observables**
  - Insulators and metals
  - Gauge-invariant integrand
  - Integral of a 2-form
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Insulators, metals, and more

- Magnetization in a "normal" (zero-Chern-number) insulator proof obtained via WFs:

$$\mathbf{M} = -\frac{ie}{\hbar c(2\pi)^3} \sum_{n \in \text{occupied}} \int_{\text{BZ}} d\mathbf{k} \, \langle \partial_{\mathbf{k}} u_{n\mathbf{k}} | \times [H(\mathbf{k}) + \varepsilon(\mathbf{k}) \,] | \partial_{\mathbf{k}} u_{n\mathbf{k}} \rangle$$

- Magnetization in metals & Chern insulators:

$$\mathbf{M} = -\frac{ie}{\hbar c(2\pi)^3} \sum_{n} \int_{\varepsilon_n(\mathbf{k}) < \mu} d\mathbf{k} \, \langle \partial_{\mathbf{k}} u_{n\mathbf{k}} | \times [H(\mathbf{k}) + \varepsilon(\mathbf{k}) - 2\mu] \, | \partial_{\mathbf{k}} u_{n\mathbf{k}} \rangle$$

Relationship to Hall conductivity

- Magnetization in a metal & in a Chern insulator

$$\mathbf{M} = -\frac{ie}{\hbar c(2\pi)^3} \sum_n \int_{\varepsilon_n(\mathbf{k})<\mu} d\mathbf{k}\, \langle \partial_{\mathbf{k}} u_{n\mathbf{k}} | \times [H(\mathbf{k}) + \varepsilon(\mathbf{k}) - 2\mu]\, | \partial_{\mathbf{k}} u_{n\mathbf{k}} \rangle$$

$$\frac{d\mathbf{M}}{d\mu} \propto \sum_n \int_{\varepsilon_n(\mathbf{k})<\mu} d\mathbf{k}\; i \langle \partial_{\mathbf{k}} u_{n\mathbf{k}} | \times | \partial_{\mathbf{k}} u_{n\mathbf{k}} \rangle$$

- This is the intrinsic part of the AHE (in metals)
- In 2d, integrated over the BZ, it gives the quantized Hall conductivity in a Chern insulator
- What has $d\mathbf{M}/d\mu$ to do with the Hall conductivity?

Relationship to Hall conductivity

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