Orbital magnetization in insulators with broken time-reversal symmetry

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THE STATE UNIVERSITY OF NEW JERSEY
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Collaboration

- **Collaboration**
  - Timo Thonhauser (Rutgers)
  - David Vanderbilt (Rutgers)
  - Davide Ceresoli (SISSA, Trieste, Italy, and Rutgers)
  - Raffaele Resta (SISSA, Trieste, Italy)
- **Preprints**
  - *"Orbital magnetization in extended systems," accepted for publication in ChemPhysChem.*
  - *"Orbital magnetization in periodic insulators," http://arXiv.org/abs/cond-mat/0505518.*

Motivations: Orbital Magnetization

Much current interest in "spintronics"

- Magnetic semiconductors
- Half metallic magnets
- Spin injection
- Anomalous Hall conductivity
- Spin Hall effect
- Etc.

Back to basics:

$$\mathbf{H} = \mathbf{B} - 4\pi\,\mathbf{M}$$

$$\mathbf{K}_{surf} = \mathbf{M} \times \mathbf{n}$$

$$\mathbf{M} = \mathbf{M}_{spin} + \mathbf{M}_{orbital}\ \textbf{?}$$

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Motivations: Orbital Magnetization

Condensed Matter, abstract
cond-mat/0502340

From: Di Xiao [view email]
Date: Mon, 14 Feb 2005 18:05:30 GMT (10kb)

Berry phase correction to electron density of states in solids

Authors: Di Xiao, Junren Shi, Qian Niu
Comments: submitted to PRL
Subj-class: Mesoscopic Systems and Quantum Hall Effect

Liouville's theorem on the conservation of phase space volume is violated by Berry phase in the semiclassical dynamics of Bloch electrons. This leads to a modification of the phase space density of states, whose significance is discussed in a number of examples: field modification of the Fermi-sea volume, connection to the anomalous Hall effect, and a general formula for orbital magnetization. The effective quantum mechanics of Bloch electrons is also sketched, where the modified density of states plays an essential role.

$$\boldsymbol{M} = \frac{e}{2\hbar} \int^{\mu_0} \frac{\mathrm{d}\boldsymbol{k}}{(2\pi)^d} \mathrm{i} \left\langle \frac{\partial u}{\partial \boldsymbol{k}} \right| \times [2\mu_0 - \varepsilon_0(\boldsymbol{k}) - \hat{H}_0] \left| \frac{\partial u}{\partial \boldsymbol{k}} \right\rangle$$

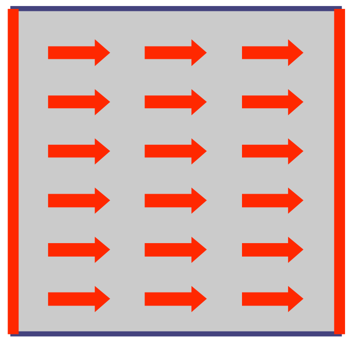
Semiclassical argument only!

Motivations: Orbital Magnetization

Our approach:

- Fully quantum
- Based on Wannier representation
- Analogous to Berry-phase theory of polarization

Analogy: Electric Polarization



- ***P*** is a bulk property

$$\Rightarrow \quad \sigma = \mathbf{P} \cdot \mathbf{n}$$

- σ is *only apparently* a surface property

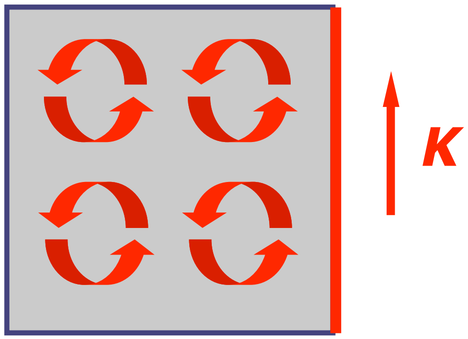
<u>References</u>

- King-Smith and Vanderbilt, PRB, 1993.
- Vanderbilt and King-Smith, PRB, 1993.
- Resta, RMP, 1994.

<u>Conditions and caveats:</u>

- Bulk is periodic insulator
- Surface is
  - Unreconstructed (1x1)
  - Insulating, in gap common to bulk and surface
- σ only determined up to "quantum" e/A_{surf}

Orbital Magnetization



$\boldsymbol{K} = \boldsymbol{M} \times \boldsymbol{n}$

Is ***M*** a *bulk property?*

Is ***K*** *only apparently* a surface property?

Definition:

If ***K*** is predetermined at all surfaces in such a way that $\boldsymbol{K} = \boldsymbol{M} \times \boldsymbol{n}$ for some vector ***M***, then ***M*** is the bulk magnetization.

Orbital Magnetization

Clarification:

- Microscopic $\boldsymbol{M}(\mathbf{r})$ defined via $\nabla \times \boldsymbol{M}(\mathbf{r}) = \boldsymbol{J}(\mathbf{r})$
- $\boldsymbol{M}(\mathbf{r})$ ill-defined: $\boldsymbol{M}(\mathbf{r}) \rightarrow \boldsymbol{M}(\mathbf{r}) + \boldsymbol{M}_0 + \nabla\phi$
- Therefore, cannot define $\boldsymbol{M}$ as cell average of $\boldsymbol{M}(\mathbf{r})$

Conclusion: $\boldsymbol{M}$ is not, even in principle, a functional of the bulk current distribution $\boldsymbol{J}(\mathbf{r})$

(Hirst, RMP, 1997)

Just as: $\boldsymbol{P}$ is not, even in principle, a functional of the bulk charge density distribution $\rho(\mathbf{r})$

Strong reasons to expect bulk ***M***

- Nearsightedness:

  Surface current depends only on local environment

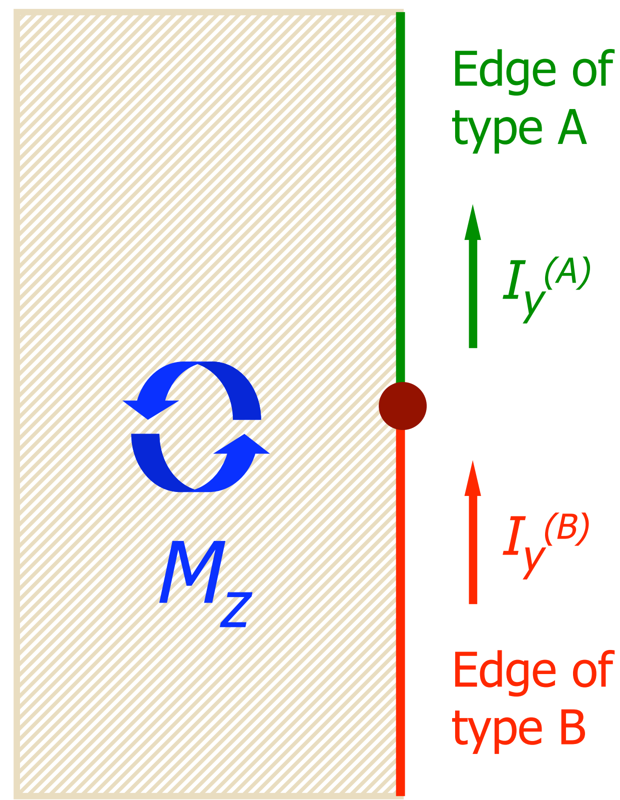
- Stationary quantum state:

  $d\rho/dt = 0$

- Conservation of charge:

  $\nabla\cdot\boldsymbol{J} = 0$

So: $I_y^{(A)} = I_y^{(B)} = M_z$



Central Claims of This Work

- Orbital magnetization is a bulk property
- Expandable in terms of bulk band-structure properties
- Closely related to Berry phases and Berry curvature
- Sum of two distinct contributions
- Suitable for calculation using standard band-structure codes

Theoretical Context

- One-particle Hamiltonian $[H, TR] \neq 0$
- $B_{macro} = 0$ (or commensurate)

} 1-particle states labeled by ***k***

- Insulator
- Chern number $C = 0$

} Wannier representable

- Spinless electrons
- 2D
- Isolated occupied band

} For simplicity of presentation

- Tight-binding models

} For tests

Vocabulary (One band in 2D)

Derivatives act in 2D k-space

$$k_\alpha \,, \qquad \alpha = \{x, y\}$$

Berry connection

$$A_\alpha(\mathbf{k}) = i\,\langle u_\mathbf{k}|\,\frac{\partial}{\partial k_\alpha}\,|u_\mathbf{k}\rangle$$

Berry curvature

$$\Omega(\mathbf{k}) = \nabla \times \mathbf{A} = \frac{\partial A_y}{\partial x} - \frac{\partial A_x}{\partial y}$$

Chern number

$$C = \frac{1}{2\pi}\int_{\mathrm{BZ}} \Omega(\mathbf{k})\, d^2k = \frac{1}{2\pi}\oint_{\mathrm{BZ}} \mathbf{A}(\mathbf{k}) \cdot \mathbf{dk}$$

Anomalous Hall conductivity

$$\sigma_{xy} = \frac{q^2}{(2\pi)^2\hbar}\int_{\mathrm{BZ}} \Omega(\mathbf{k})\, f(E_\mathbf{k} - \mu)\, d^2k$$

Electric polarizaton

$$P_\alpha = \frac{q}{(2\pi)^2}\int_{\mathrm{BZ}} A_\alpha(\mathbf{k})\, d^2k$$

Bloch function $\quad \psi_\mathbf{k}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}}\, u_\mathbf{k}(\mathbf{r})$

Derivation of Electric Polarization

Dipole moment of finite sample

$$d_x = q \sum_j \langle \psi_j | x | \psi_j \rangle \qquad |\psi_j\rangle = \text{Eigenstate}$$

$$= q \sum_m \langle w_m | x | w_m \rangle \qquad |w_m\rangle = \text{Loc. molec. orb.}$$

Thermodynamic limit

$$|w_m\rangle \to |\mathbf{R}\rangle \qquad |\mathbf{R}\rangle = \text{Bulk Wannier func.}$$

$$P_x = \frac{d_x}{A_{\text{sample}}} = \frac{q}{A_0} \langle \mathbf{0} | x | \mathbf{0} \rangle$$

Derivation of Electric Polarization

Transform to k-space

$$|\mathbf{R}\rangle = \frac{A_0}{(2\pi)^2} \int_{\mathrm{BZ}} d^2k \, e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{R})} \, |u_{\mathbf{k}}\rangle$$

$$x\,|\mathbf{R}\rangle = i\,\frac{A_0}{(2\pi)^2} \int_{\mathrm{BZ}} d^2k \, e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{R})} \, |\frac{\partial u_{\mathbf{k}}}{\partial k_x}\rangle$$

$$y\,|\mathbf{R}\rangle = i\,\frac{A_0}{(2\pi)^2} \int_{\mathrm{BZ}} d^2k \, e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{R})} \, |\frac{\partial u_{\mathbf{k}}}{\partial k_y}\rangle$$

$$P_\alpha = \frac{iq}{(2\pi)^2} \int_{\mathrm{BZ}} d^2k \, \langle u_{\mathbf{k}}| \frac{\partial}{\partial k_\alpha} |u_{\mathbf{k}}\rangle$$

Derivation of Orbital Magnetization?

Magnetization of finite sample

$$M = \frac{q}{2Ac}\sum_j \langle \psi_j | x v_y - y v_x | \psi_j \rangle$$

$$= \frac{-iq}{2\hbar A c}\sum_m \langle w_m |\, x[y,H] - y[x,H]\, | w_m \rangle$$

$$= \frac{-q}{\hbar A c}\,\mathrm{Im}\sum_m \langle w_m |\, xHy\, | w_m \rangle$$

Magnetization in thermodynamic limit

$$M_{\mathrm{LC}} = \frac{-q}{\hbar c A_0}\,\mathrm{Im}\,\langle \mathbf{0} | xHy | \mathbf{0} \rangle$$

$M = M_{LC}$?

Derivation of Orbital Magnetization?

$$M_{\rm LC} = \frac{-q}{\hbar c A_0}\,{\rm Im}\,\langle \mathbf{0}|xHy|\mathbf{0}\rangle$$

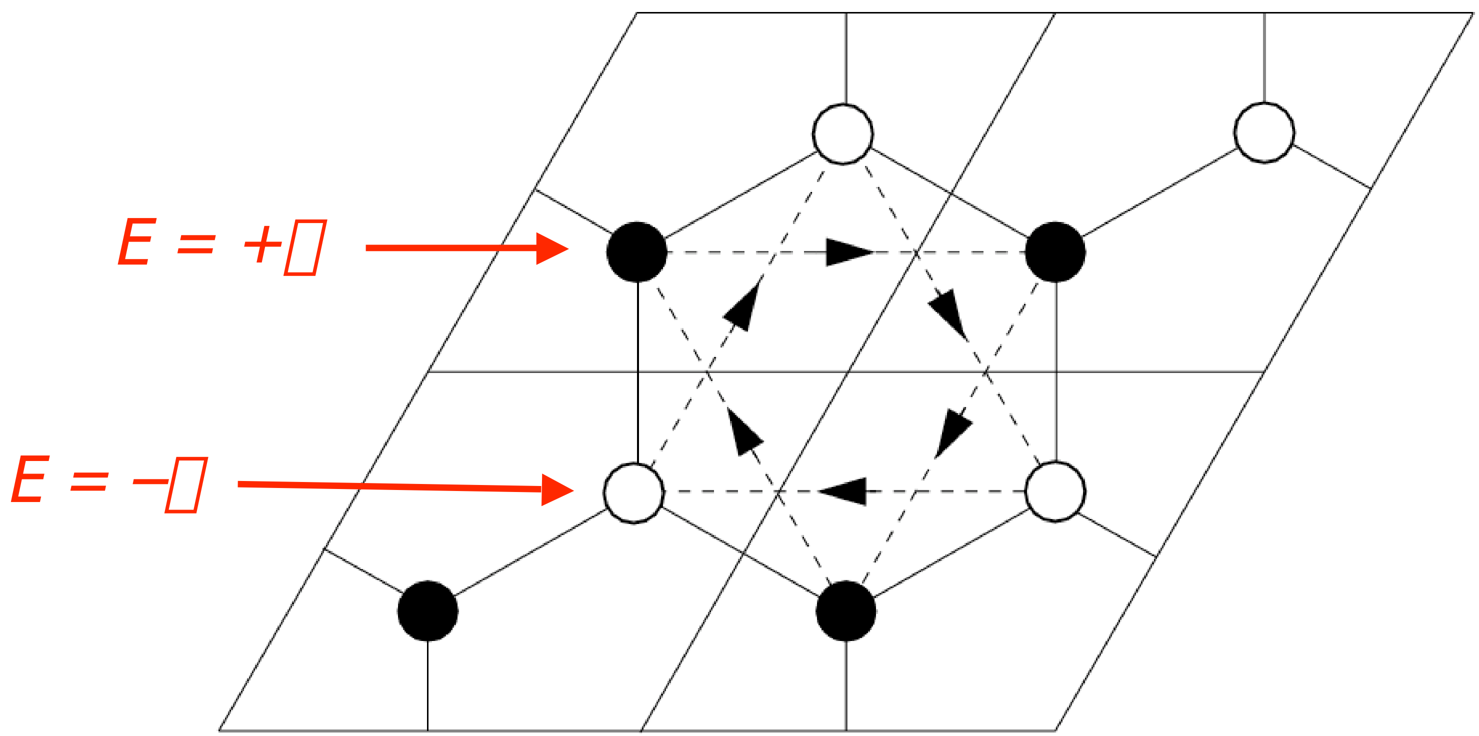
Transform to k-space

$$M_{\rm LC} = \frac{-q}{\hbar c}\,{\rm Im}\int_{\rm BZ}\frac{d^2k}{(2\pi)^2}\,\langle \frac{\partial u_{\mathbf{k}}}{\partial k_x}\Big|\,H_{\mathbf{k}}\,\Big|\frac{\partial u_{\mathbf{k}}}{\partial k_y}\rangle$$

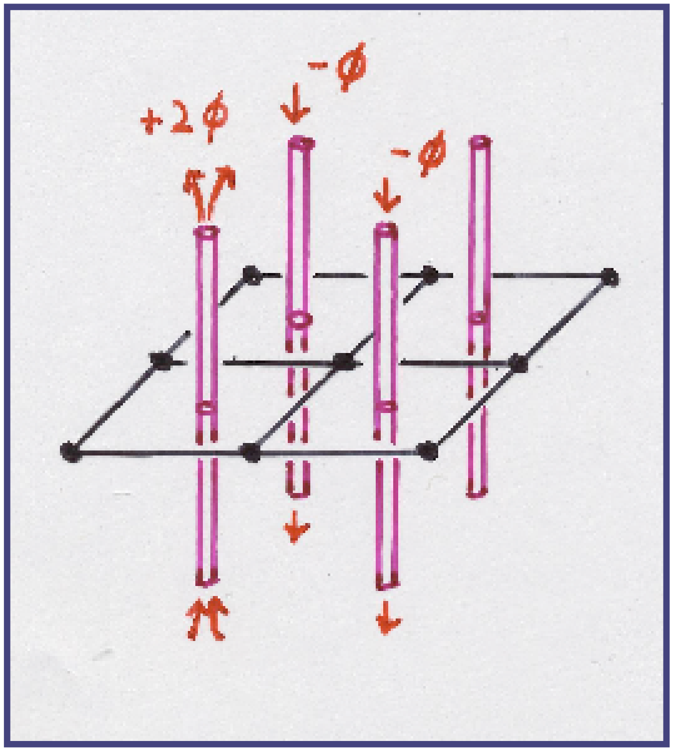
?

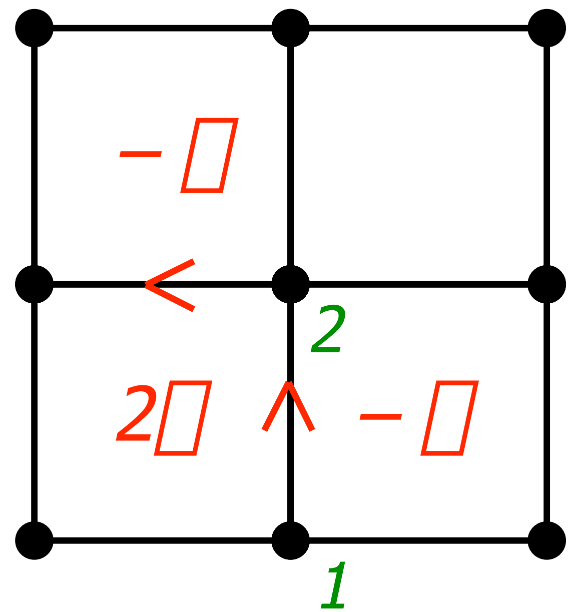
Numerical Tests: Haldane model

Tight-binding model of Haldane, 1988



Complex hoppings and flux tubes



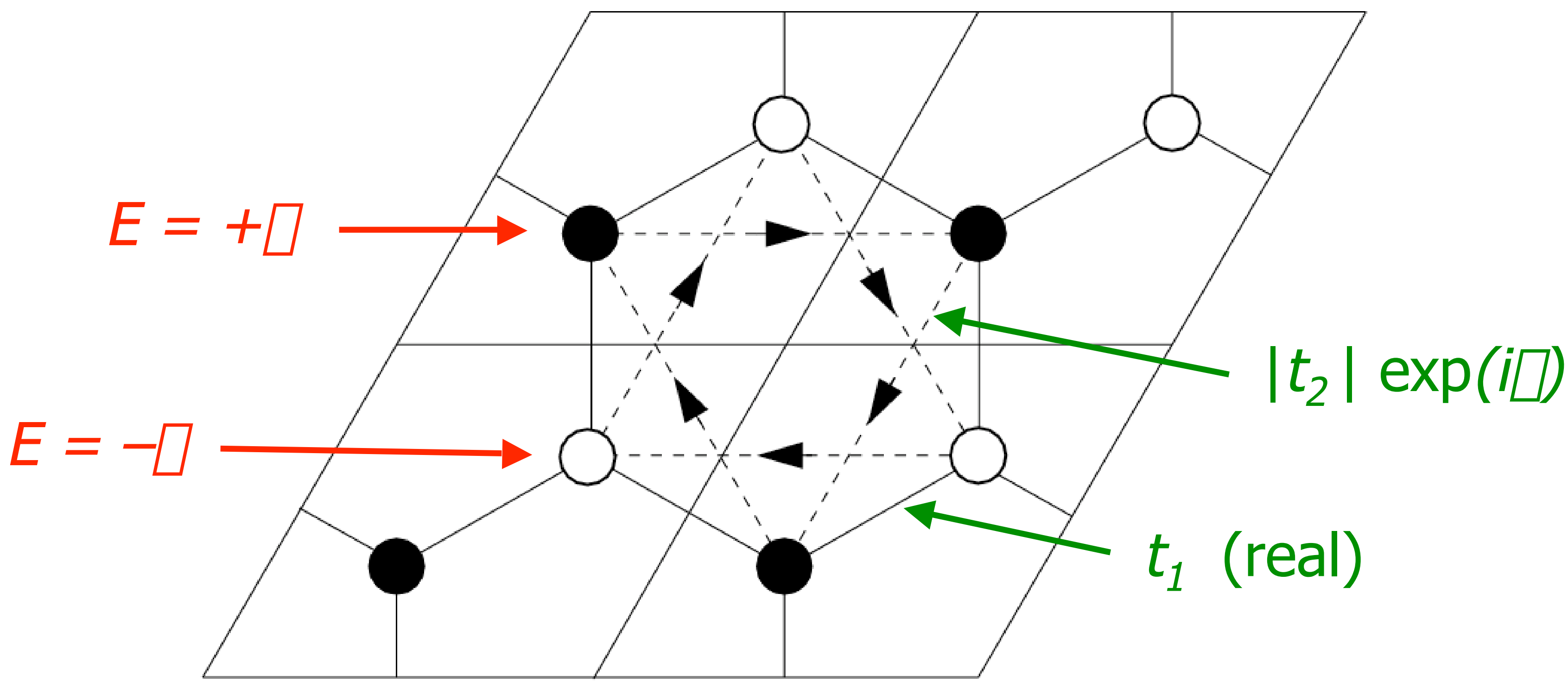


$$t_{12} = |t_0|\,\exp(+i\phi)$$

$$t_{21} = |t_0|\,\exp(-i\phi)$$

Numerical Tests: Haldane model

Tight-binding model of Haldane, 1988



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Numerical Tests: Haldane model

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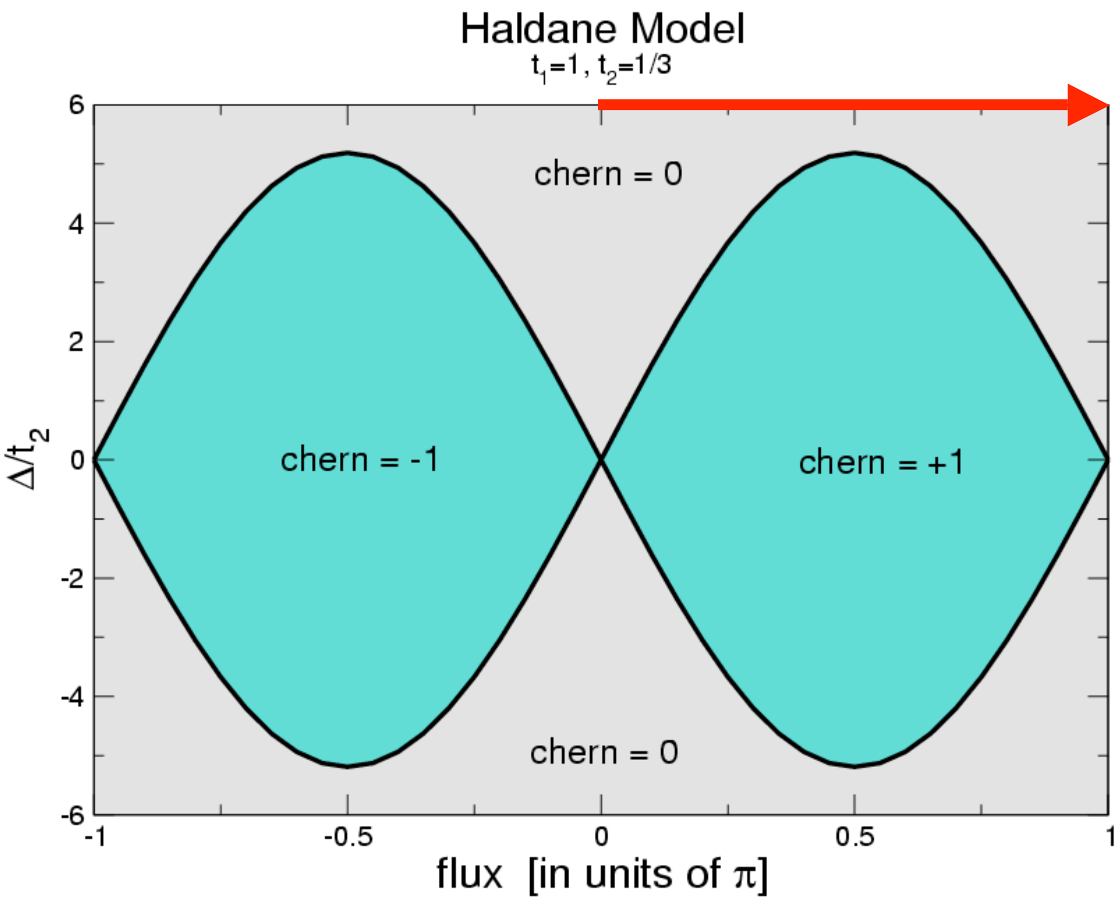
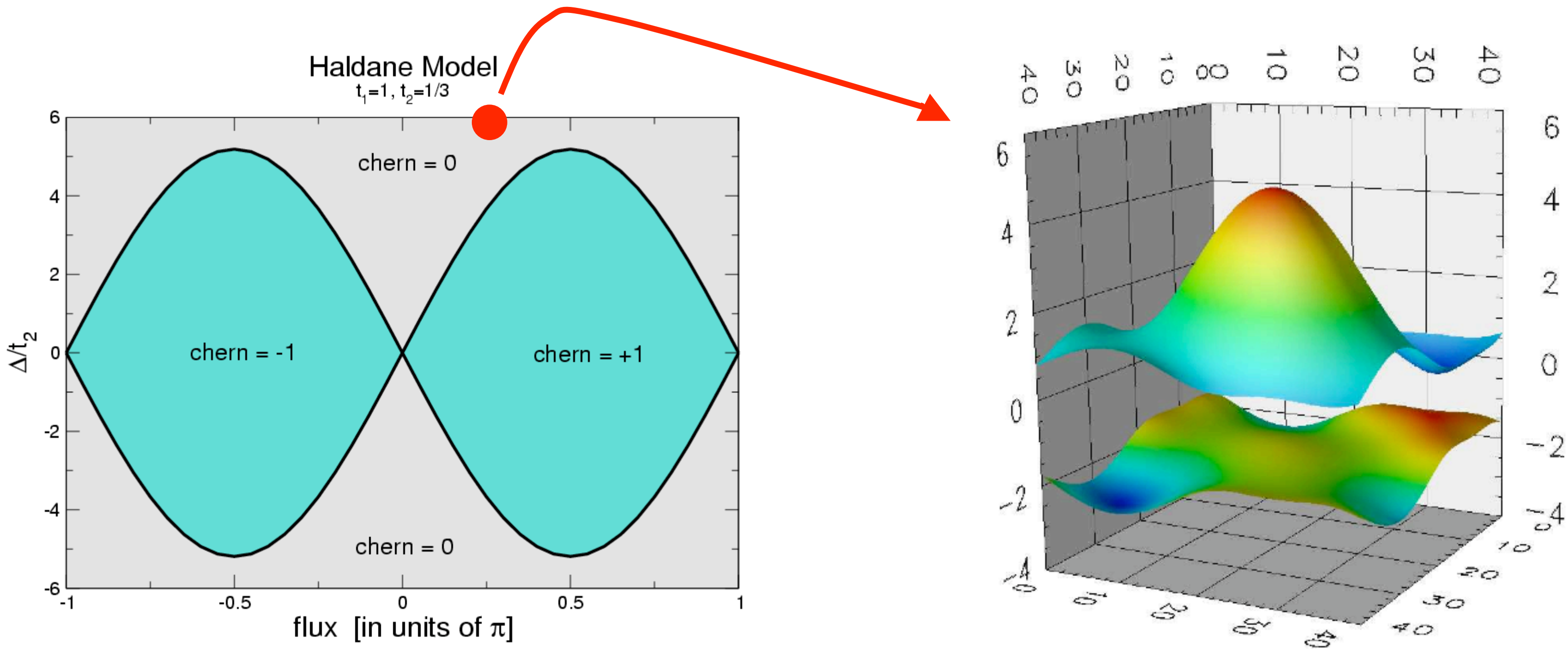
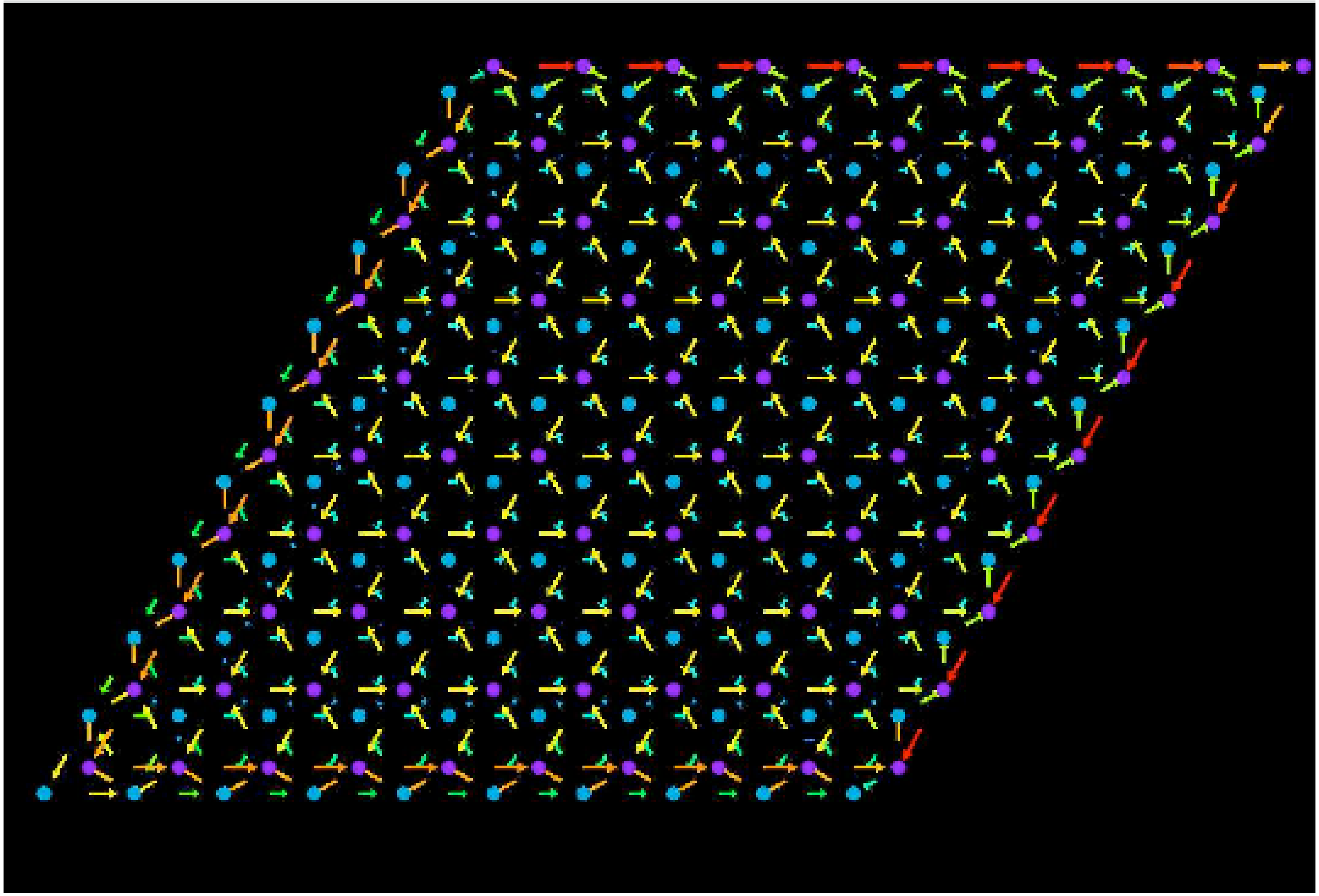


Numerical Tests: Haldane model

Tight-binding model of Haldane, 1988



Numerical Tests: Haldane model



Derivation of Orbital Magnetization?

Magnetization of finite sample

$$M = \frac{q}{2Ac} \sum_j \langle \psi_j | x v_y - y v_x | \psi_j \rangle$$

$$= \frac{-iq}{2\hbar Ac} \sum_m \langle w_m | \, x[y,H] - y[x,H] \, | w_m \rangle$$

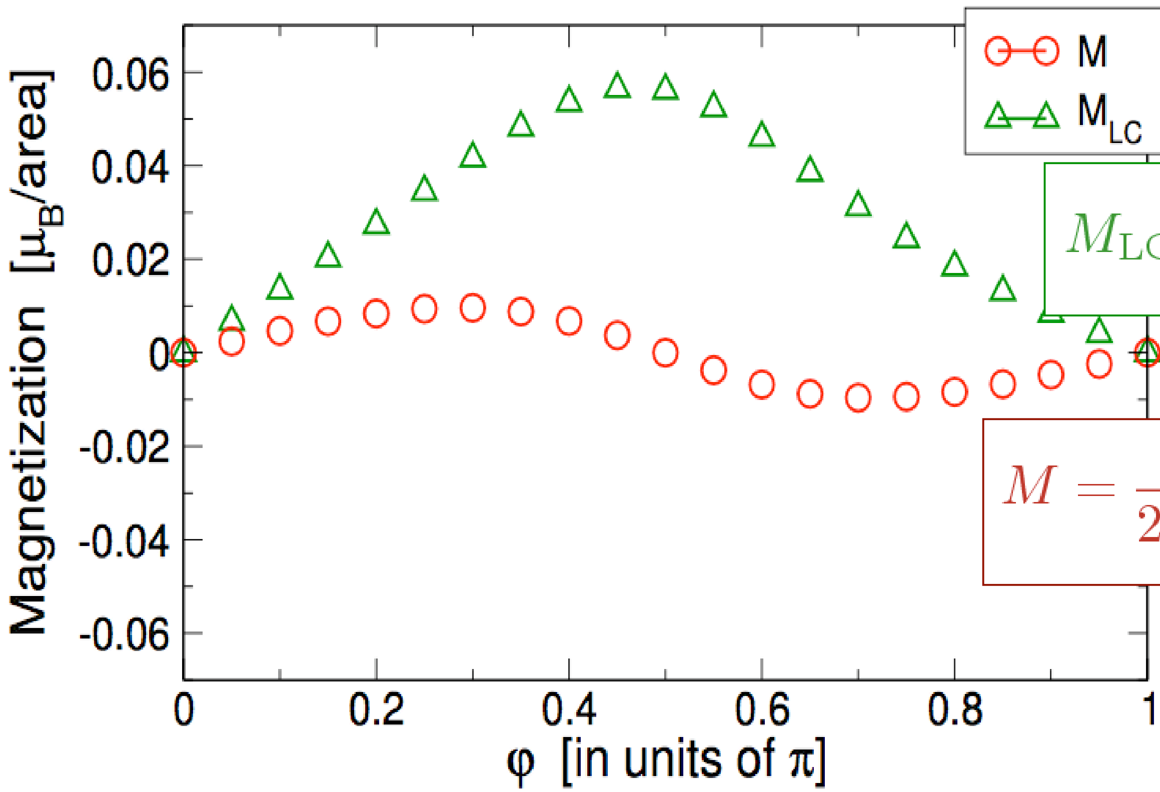
$$= \frac{-q}{\hbar Ac} \, \mathrm{Im} \sum_m \langle w_m | \, xHy \, | w_m \rangle$$

$M = M_{LC}$?

Magnetization in thermodynamic limit

$$M_{\mathrm{LC}} = \frac{-q}{\hbar c A_0} \, \mathrm{Im} \, \langle \mathbf{0} | xHy | \mathbf{0} \rangle$$

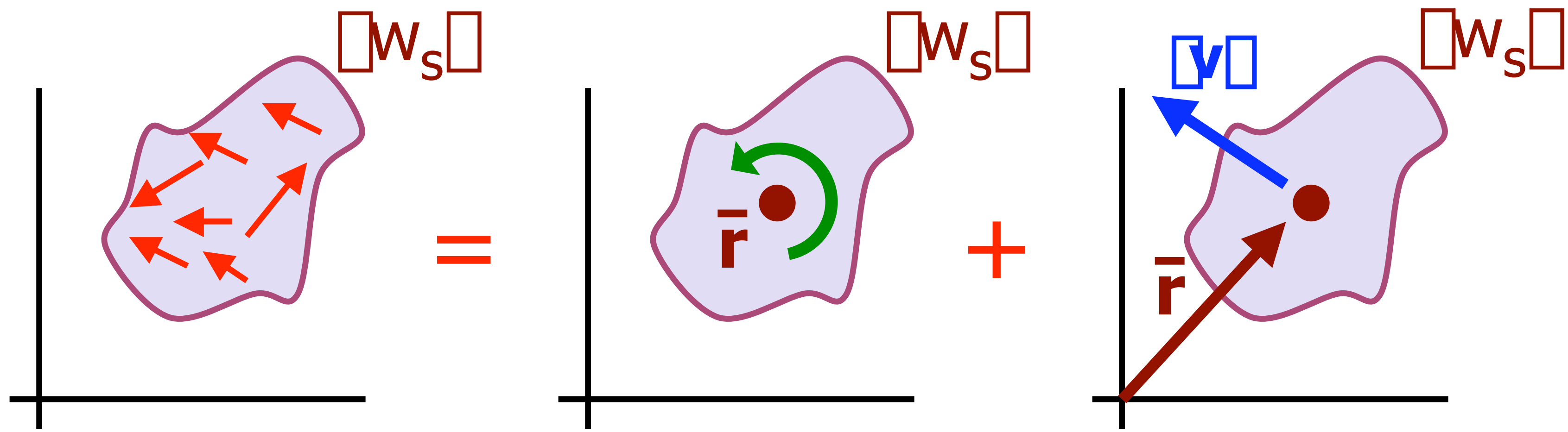
Numerical Tests: Haldane model



$$M_{\rm LC} = \frac{-q}{\hbar A_0 c}\,{\rm Im}\,\langle \mathbf{0}|xHy|\mathbf{0}\rangle$$

$$M = \frac{q}{2Ac}\sum_j \langle \psi_j | x v_y - y v_x | \psi_j \rangle$$

What is missing?

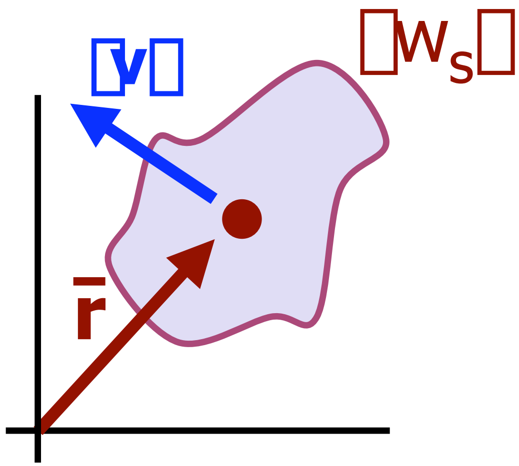


$$\langle w_s | \, \mathbf{r} \times \mathbf{v} \, | w_s \rangle = \langle w_s | \, (\mathbf{r} - \bar{\mathbf{r}}) \times \mathbf{v} \, | w_s \rangle + \bar{\mathbf{r}} \times \langle w_s | \mathbf{v} | w_s \rangle$$

Local Circulation
(LC)

Itinerant Circulation
(IC)

Itinerant Circulation

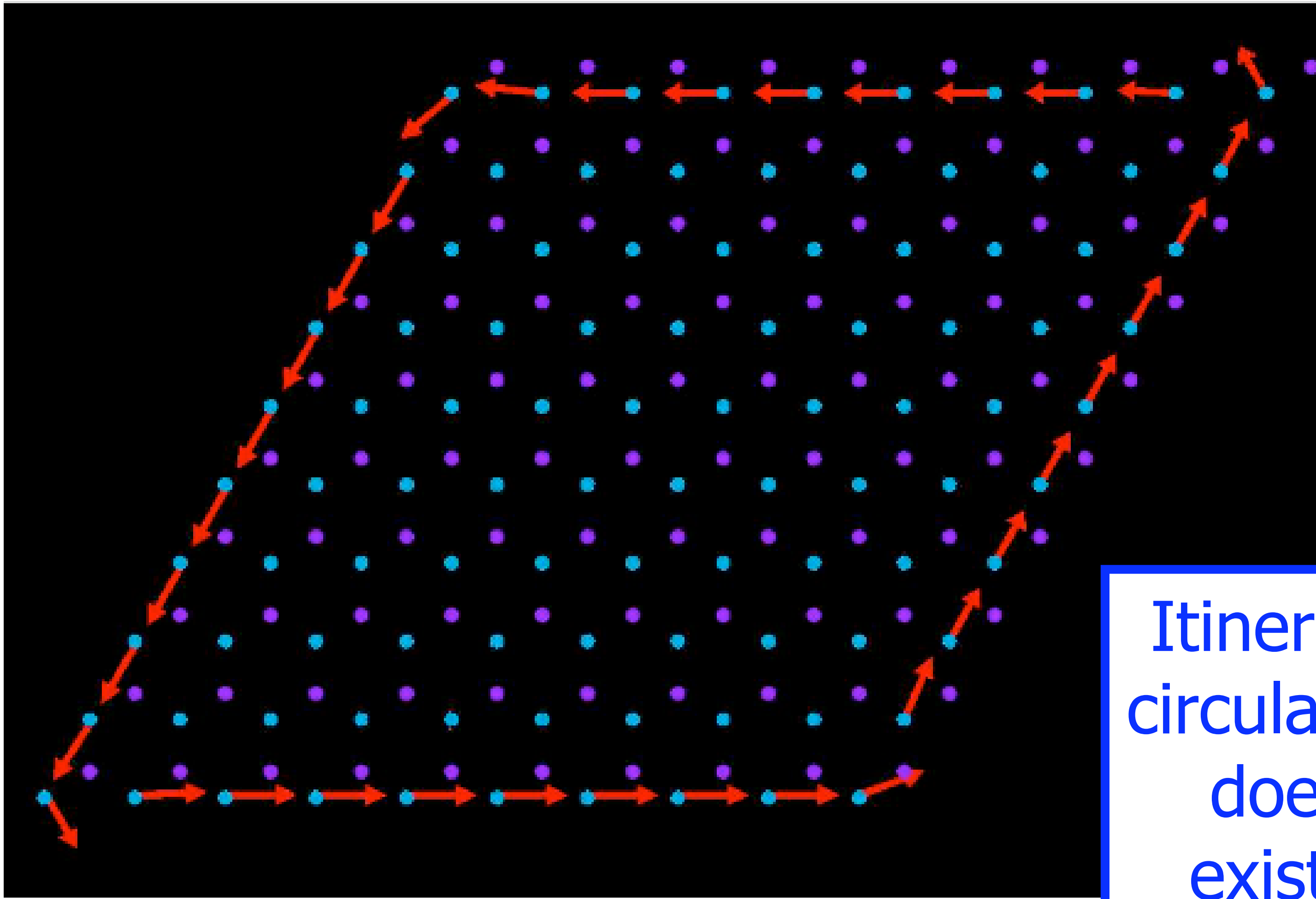


$$\underbrace{\bar{\mathbf{r}} \times \langle w_s|\mathbf{v}|w_s\rangle}$$

Itinerant Circulation (IC)

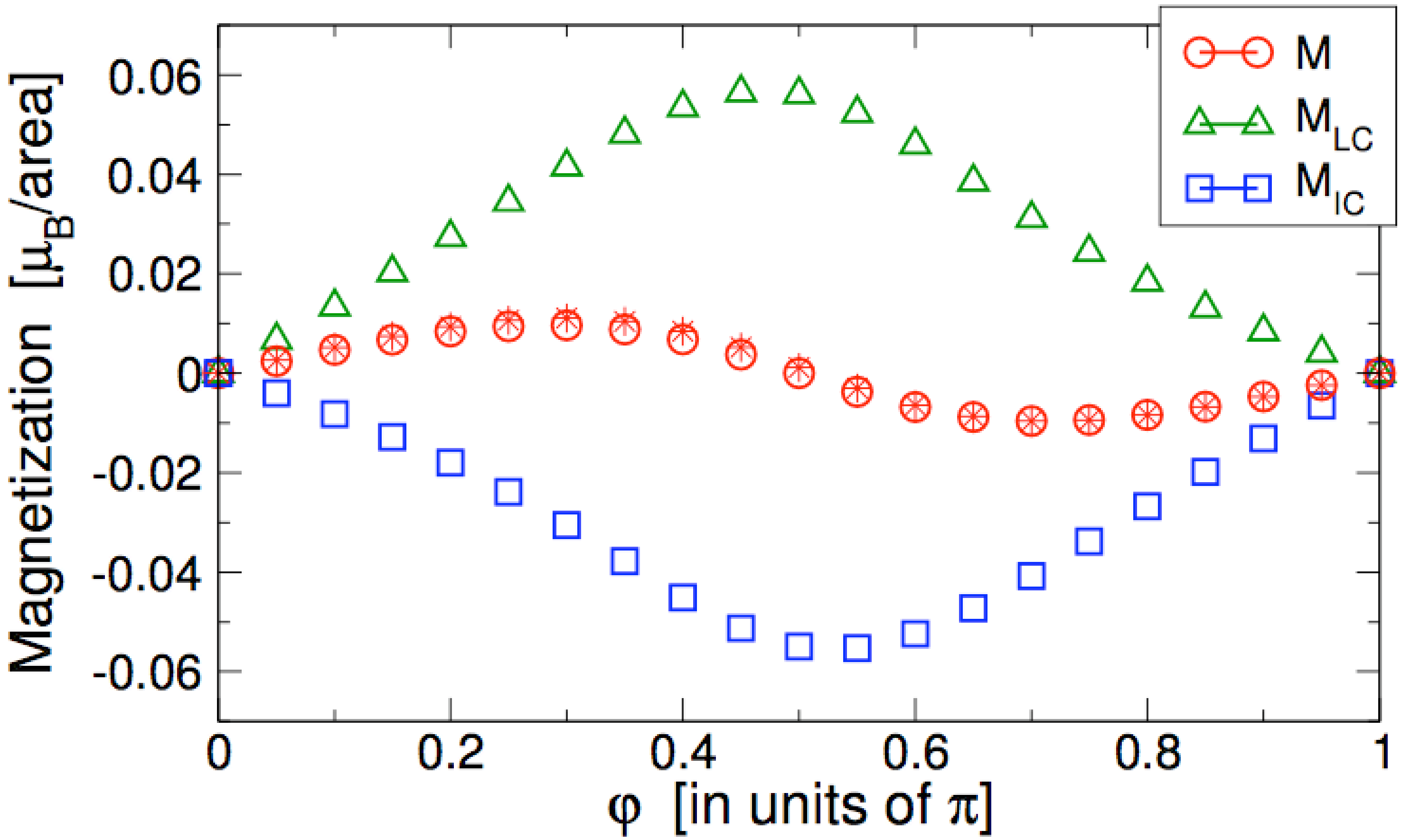
- Bulk WF:
  - Bulk band carries no net current
  - So $\langle \mathbf{v} \rangle = 0$
  - So $\bar{\mathbf{r}} \times \langle \mathbf{v} \rangle = 0$
- But what about a surface WF ?

Numerical Tests: Haldane model



Itinerant circulation does exist !

Numerical Tests: Haldane model



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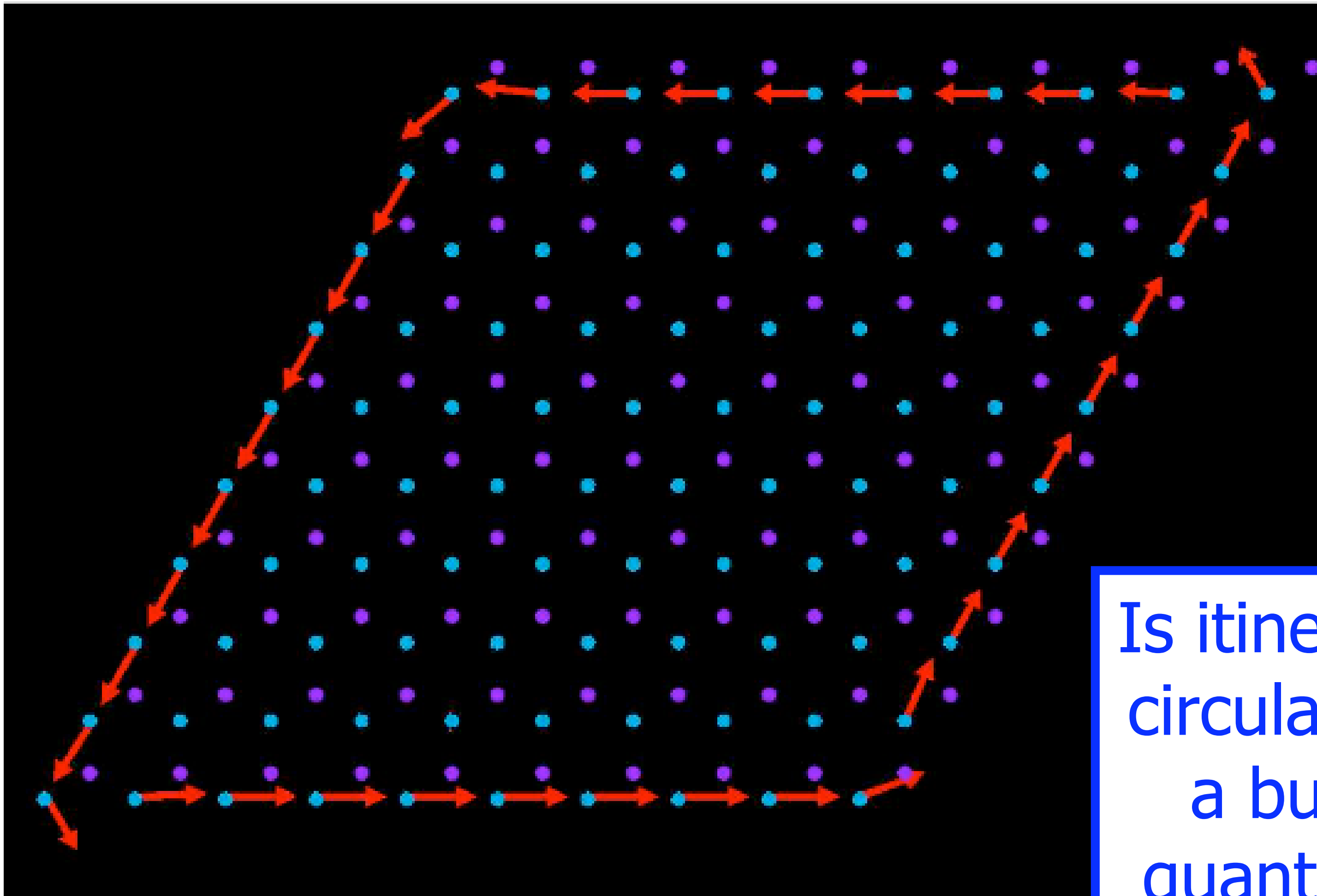
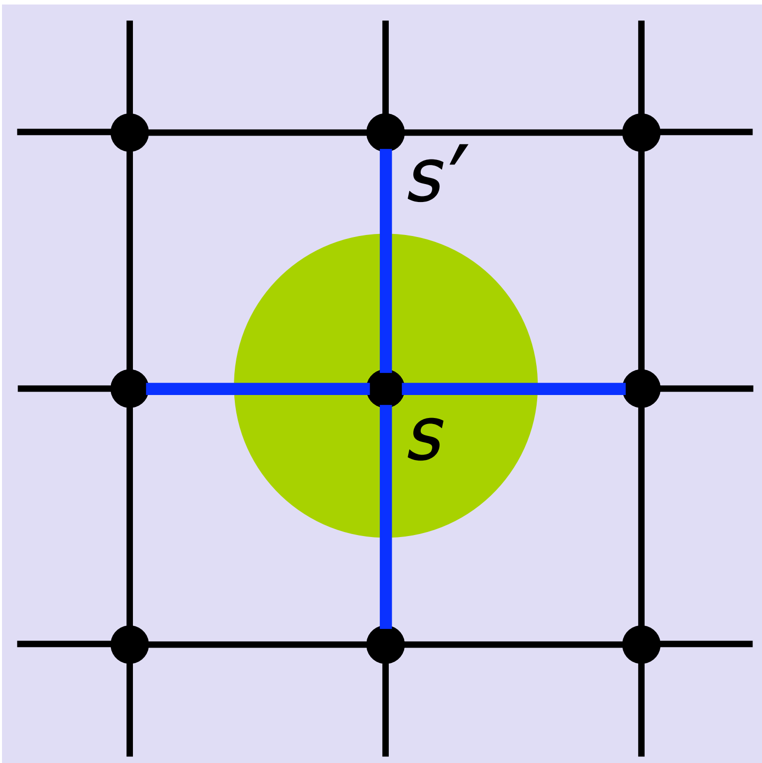
Numerical Tests: Haldane model



Is itinerant circulation a bulk quantity?

Understanding Itinerant Circulation



$$I_s = q \langle w_s | \mathbf{v} | w_s \rangle$$

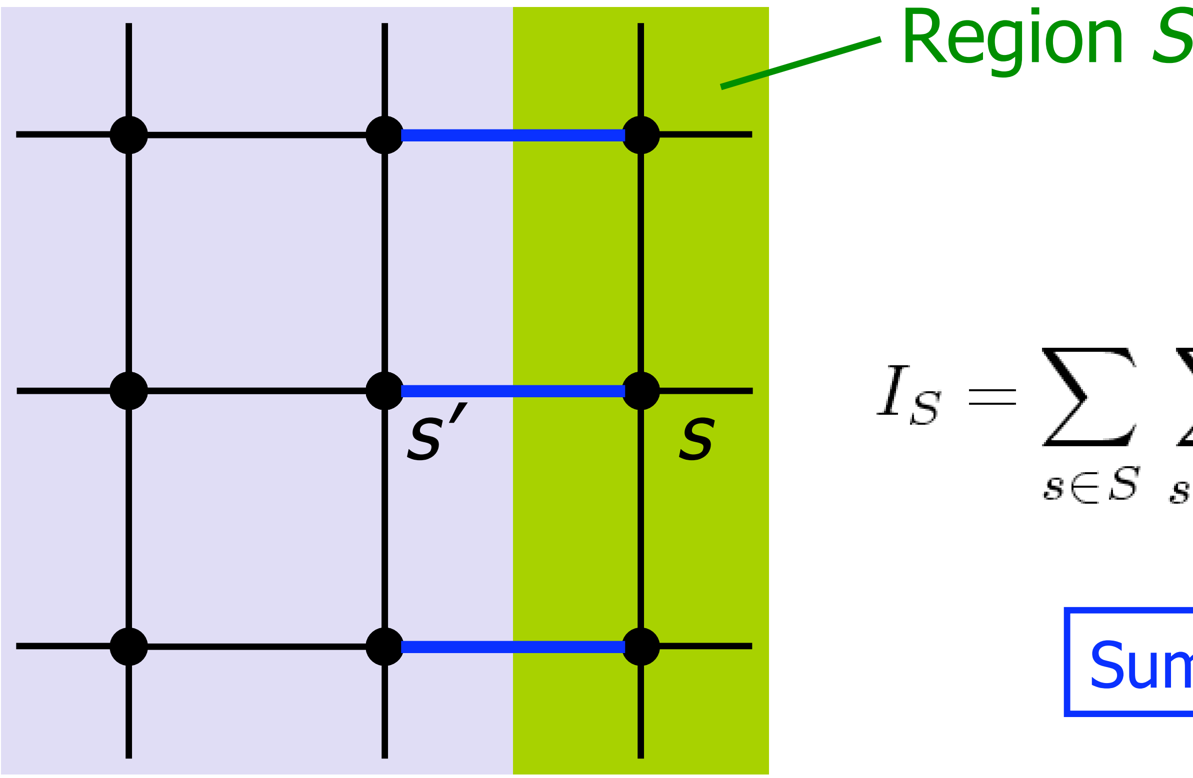
$$= \frac{-iq}{\hbar} \langle w_s | \, [r, H] \, | w_s \rangle$$

$$\sum_{s'} |w_{s'}\rangle\langle w_{s'}|$$

$$= \frac{2q}{\hbar} \sum_{s' \neq s} \mathrm{Im} \, \langle w_s | \mathbf{r} | w_{s'} \rangle \, \langle w_{s'} | H | w_s \rangle$$

$$= \frac{2q}{\hbar} \sum_{s' \neq s} \mathrm{Im} \, \mathbf{r}_{ss'} \, H_{s's}$$

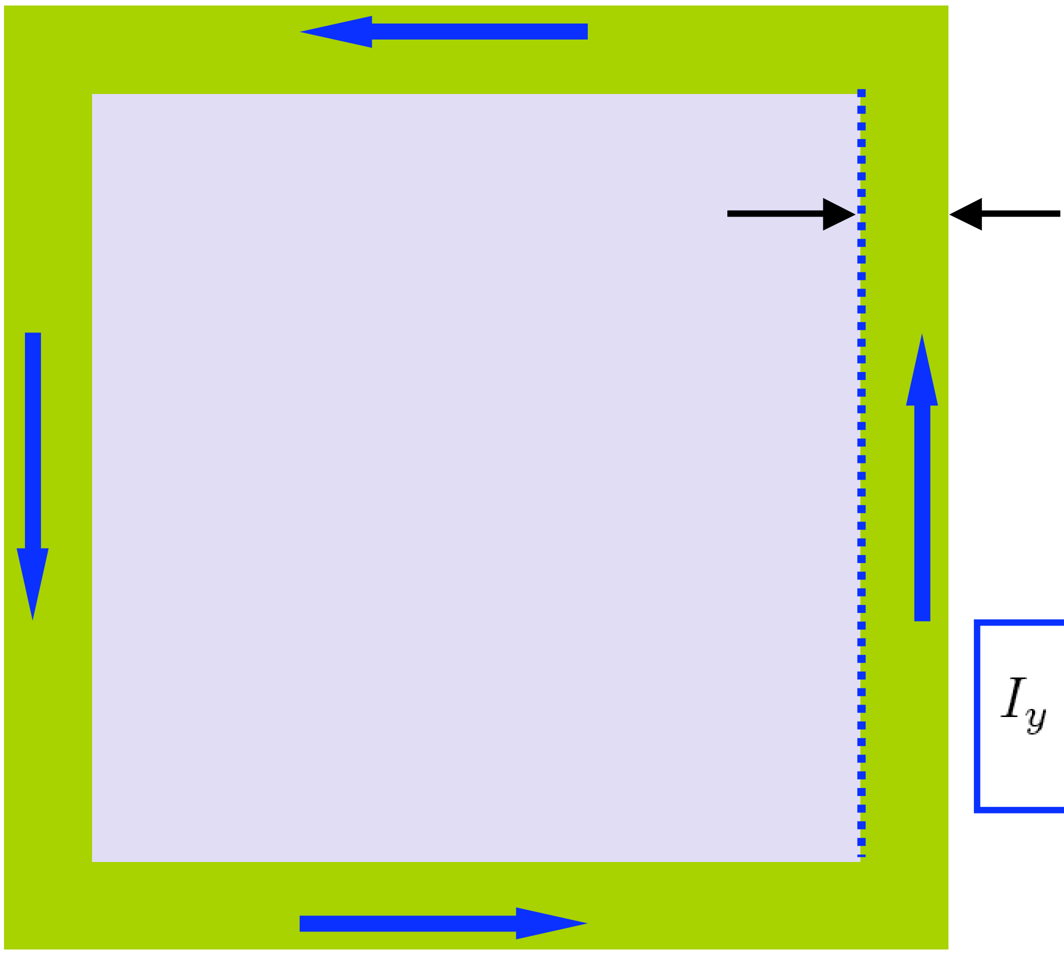
Understanding Itinerant Circulation



$$I_S = \sum_{s \in S} \sum_{s' \notin S} \frac{2q}{\hbar} \, \mathrm{Im}\, \mathbf{r}_{ss'} \, H_{s's}$$

Sum over blue links only

Understanding Itinerant Circulation



Thickness:

<< Sample size

>> Unit cell

$$I_y = \frac{q}{A_0 \hbar} \sum_{\mathbf{R}} R_x \, \mathrm{Im} \, y_{\mathbf{R},\mathbf{0}} H_{\mathbf{0},\mathbf{R}}$$

Understanding Itinerant Circulation

$$I_y = \frac{q}{A_0 \hbar} \sum_{\mathbf{R}} R_x \, \mathrm{Im} \, y_{\mathbf{R},\mathbf{0}} \, H_{\mathbf{0},\mathbf{R}}$$

$$\boxed{M_{\mathrm{IC}} = \frac{-q}{2 A_0 \hbar c} \sum_{\mathbf{R}} \mathrm{Im} \, \left(R_x \, y_{\mathbf{0},\mathbf{R}} \, H_{\mathbf{R},\mathbf{0}} - R_y \, x_{\mathbf{0},\mathbf{R}} \, H_{\mathbf{R},\mathbf{0}} \right)}$$

M_{IC} can be written in terms of WFs !

□ M_{IC} is a bulk quantity !

Understanding Itinerant Circulation

$$M_{\mathrm{IC}} = \frac{-q}{2A_0\hbar c} \sum_{\mathbf{R}} \mathrm{Im}\ \left(R_x\, y_{\mathbf{0},\mathbf{R}}\, H_{\mathbf{R},\mathbf{0}} - R_y\, x_{\mathbf{0},\mathbf{R}}\, H_{\mathbf{R},\mathbf{0}} \right)$$

$$\langle \mathbf{0}|x|\mathbf{R}\rangle = \frac{A_0}{(2\pi)^2} \int d^2k\ A_x(\mathbf{k})\, e^{-i\mathbf{k}\cdot\mathbf{R}}$$

$$\langle \mathbf{0}|y|\mathbf{R}\rangle = \frac{A_0}{(2\pi)^2} \int d^2k\ A_y(\mathbf{k})\, e^{-i\mathbf{k}\cdot\mathbf{R}}$$

$$\langle \mathbf{0}|H|\mathbf{R}\rangle = \frac{A_0}{(2\pi)^2} \int d^2k\ E(\mathbf{k})\, e^{-i\mathbf{k}\cdot\mathbf{R}}$$

$$M_{\mathrm{IC}} = \frac{q}{2\hbar c} \int \frac{d^2k}{(2\pi)^2}\, E(\mathbf{k})\, \nabla \times \mathbf{A}(\mathbf{k})$$

$$\boxed{M_{\mathrm{IC}} = \frac{q}{2\hbar c} \int \frac{d^2k}{(2\pi)^2}\ E(\mathbf{k})\, \Omega(\mathbf{k})}$$

Two Contributions to the Magnetization

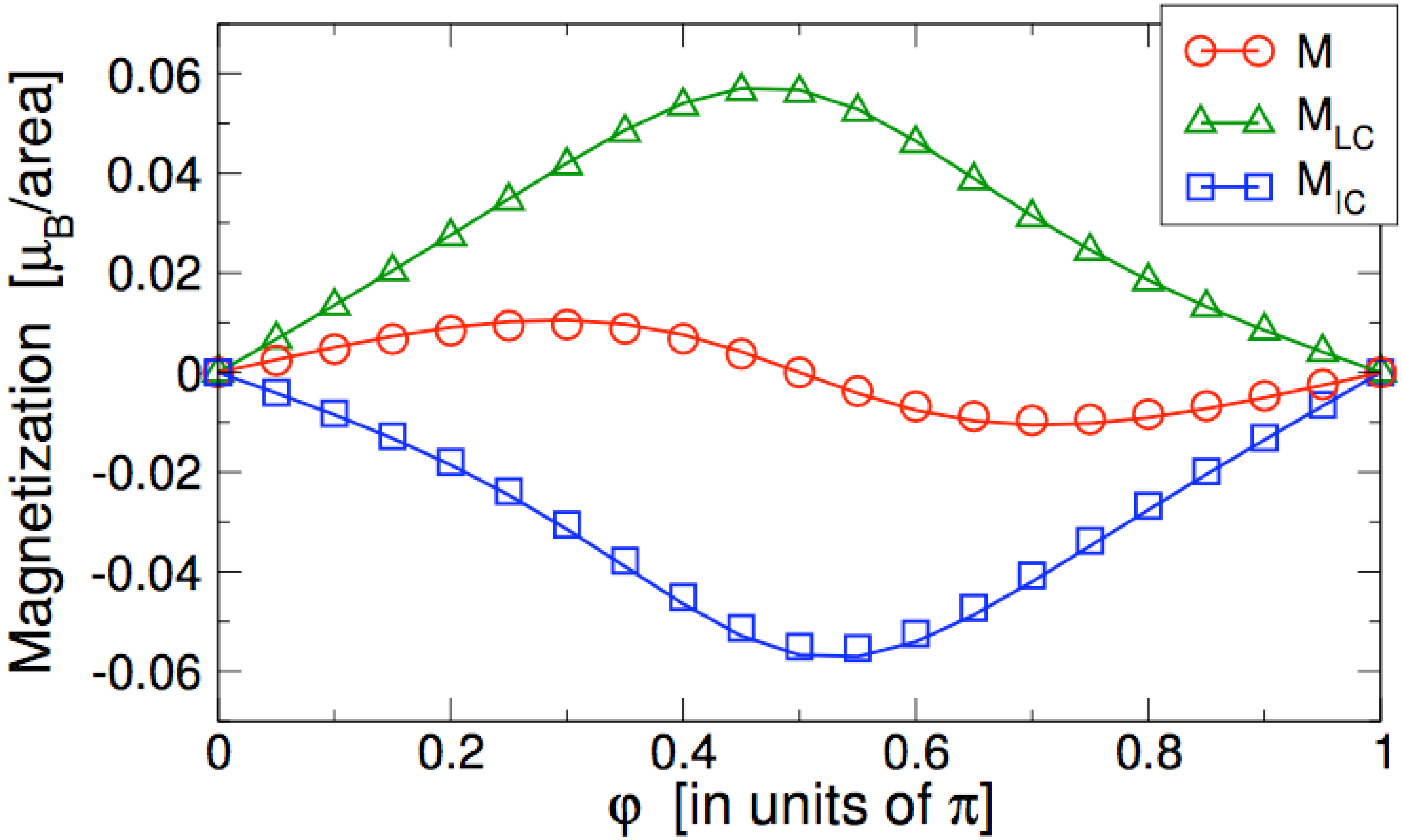
$$M = M_{\mathrm{LC}} + M_{\mathrm{IC}}$$

$$M_{\mathrm{LC}} = \frac{-q}{\hbar c}\,\mathrm{Im}\int_{\mathrm{BZ}} \frac{d^2k}{(2\pi)^2}\,\langle\frac{\partial u_{\mathbf{k}}}{\partial k_x}|\,H_{\mathbf{k}}\,|\frac{\partial u_{\mathbf{k}}}{\partial k_y}\rangle$$

$$M_{\mathrm{IC}} = \frac{q}{2\hbar c}\int \frac{d^2k}{(2\pi)^2}\,E(\mathbf{k})\,\Omega(\mathbf{k})$$

$$M = \frac{-q}{\hbar c}\,\mathrm{Im}\int_{\mathrm{BZ}} \frac{d^2k}{(2\pi)^2}\,\langle\frac{\partial u_{\mathbf{k}}}{\partial k_x}|\,H_{\mathbf{k}} + E_{\mathbf{k}}\,|\frac{\partial u_{\mathbf{k}}}{\partial k_y}\rangle$$

Numerical Tests: Haldane model



Two Contributions to the Magnetization

$$M = M_{\mathrm{LC}} + M_{\mathrm{IC}}$$

$$M = \frac{-q}{\hbar c}\,\mathrm{Im}\int_{\mathrm{BZ}} \frac{d^2k}{(2\pi)^2} \left\langle \frac{\partial u_{\mathbf{k}}}{\partial k_x} \right| H_{\mathbf{k}} + E_{\mathbf{k}} \left| \frac{\partial u_{\mathbf{k}}}{\partial k_y} \right\rangle$$

- Each contribution invariant under *H* □ *H* + □*E*
- Each contribution gauge invariant ($|u_{\boldsymbol{k}}$□□ $e^{i□(\boldsymbol{k})}|u_{\boldsymbol{k}}$□)
- Consistent with result of Xiao et al.

$$\boldsymbol{M} = \frac{e}{2\hbar}\int^{\mu_0} \frac{\mathrm{d}\boldsymbol{k}}{(2\pi)^d}\mathrm{i}\left\langle \frac{\partial u}{\partial \boldsymbol{k}} \right| \times [2\mu_0 - \varepsilon_0(\boldsymbol{k}) - \hat{H}_0] \left| \frac{\partial u}{\partial \boldsymbol{k}} \right\rangle$$

↑ Needed for metals or non-zero Chern

Future Challenges

- One-particle Hamiltonian $[H, TR] \neq 0$
- $B_{macro} = 0$ (or commensurate)

} 1-particle states labeled by ***k***

- Insulator
- Chern number $C = 0$

} Wannier representable

- Spinless electrons
- 2D
- Isolated occupied band

} For simplicity of presentation

- Tight-binding models

} For tests

Summary

- Orbital magnetization is a bulk property
- Expandable in terms of bulk band-structure properties
- Closely related to Berry phases and Berry curvature
- Sum of two distinct contributions
- Suitable for calculation using standard band-structure codes
- Generalizable for metals, Chern insulators?