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Phys. Rev. Lett. **110**, 087202 — Published 21 February 2013

DOI: [10.1103/PhysRevLett.110.087202](https://doi.org/10.1103/PhysRevLett.110.087202)

Orbital Magnetization as a Local Property

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The modern expressions for polarization \mathbf{P} and orbital magnetization \mathbf{M} are \mathbf{k} -space integrals. But a genuine bulk property should also be expressible in \mathbf{r} -space, as unambiguous function of the ground-state density matrix, “nearsighted” in insulators, independently of the boundary conditions—either periodic or open. While \mathbf{P} —owing to its “quantum” indeterminacy—is *not* a bulk property in this sense, \mathbf{M} is. We provide its \mathbf{r} -space expression for any insulator, even with nonzero Chern invariant. Simulations on a model Hamiltonian validate our theory.

PACS numbers: 75.10.-b, 73.43.Cd, 77.84.-s

The macroscopic polarization \mathbf{P} and magnetization \mathbf{M} are essential ingredients of the in-medium Maxwell equations, but microscopic understanding of \mathbf{P} and of the orbital contribution to \mathbf{M} was achieved only in recent times [1–6]. Their elementary definition for a finite sample is

$$\mathbf{P} = \frac{\mathbf{d}}{V} = \frac{1}{V} \int d\mathbf{r} \mathbf{r} \rho^{(\text{micro})}(\mathbf{r}) \quad (1)$$

$$\mathbf{M} = \frac{\mathbf{m}}{V} = \frac{1}{2cV} \int d\mathbf{r} \mathbf{r} \times \mathbf{j}^{(\text{micro})}(\mathbf{r}). \quad (2)$$

Here and in the following we indicate with \mathbf{M} the orbital term only; $\rho^{(\text{micro})}(\mathbf{r})$ and $\mathbf{j}^{(\text{micro})}(\mathbf{r})$ are the microscopic charge and current densities, and V is the sample volume. The previous expressions are clearly dominated by surface contributions, while instead phenomenologically \mathbf{P} and \mathbf{M} are bulk properties: from this viewpoint, the two properties appear as closely analogous. The modern theories of polarization and magnetization (in their simplest formulations) address a crystalline system of independent electrons; therein, both \mathbf{P} and \mathbf{M} are expressed as a Brillouin-zone integral of Bloch-orbital matrix elements [1–8]; even the \mathbf{k} space expressions for \mathbf{P} and \mathbf{M} share many analogies. The modern theories are clearly based on periodic boundary conditions (PBCs): the sample has no boundary, and the properties are “bulk” by definition.

In this Letter we aim instead at \mathbf{r} -space definitions, but where—at variance with Eqs. (1) and (2)—the choice of the boundary conditions becomes irrelevant in the limit of a large sample. For a system of independent electrons the ground state is uniquely determined by the one-particle density matrix, a.k.a. ground-state projector $\mathcal{P}(\mathbf{r}, \mathbf{r}')$; it is a “nearsighted” [9–11] operator, exponentially decreasing with $|\mathbf{r} - \mathbf{r}'|$ in insulators even when the Chern invariant is nonzero [12]. Our aim is therefore to express \mathbf{P} and \mathbf{M} as local properties in \mathbf{r} space, directly in terms of $\mathcal{P}(\mathbf{r}, \mathbf{r}')$ in the bulk of a sample, *independently* of the boundary conditions. We show that such aim *cannot* be attained for \mathbf{P} , while we provide an explicit expression for \mathbf{M} , even for insulators with

nonzero Chern invariant (“Chern insulators”). Tinkering with the boundaries may alter the value of \mathbf{P} , but not of \mathbf{M} : this finding is in agreement with a very recent work by Chen and Lee, based on completely different arguments [13].

We validate our approach by means of simulations on a model Hamiltonian, performed on finite samples with *open* boundary conditions (OBCs). One outstanding virtue of our formula is that it converges to the bulk \mathbf{M} value much faster than the elementary definition of Eq. (2): see Fig. 4 below; another virtue is that it could be applied with no major changes to disordered and/or macroscopically inhomogeneous systems.

The modern theory of polarization addresses the difference in polarization $\Delta\mathbf{P}$ between two states of the material that can be connected by an adiabatic switching process. This is clearly a bulk property, provided the system remains insulating at all times: $\Delta\mathbf{P}$ in fact coincides with the integrated current flow across the material, which in turn is easily expressed in terms of the evolution of $\mathcal{P}(\mathbf{r}, \mathbf{r}')$ along the switching. But “ \mathbf{P} itself” is *not* a bulk property in the above sense: a basic tenet of the modern theory of polarization states that the bulk electron distribution determines \mathbf{P} only *modulo* a “quantum”, whose value depends on the boundary [1, 2]. Therefore it is impossible to evaluate \mathbf{P} for a homogeneous sample knowing $\mathcal{P}(\mathbf{r}, \mathbf{r}')$ in its bulk only: examples of systems with the same bulk and different \mathbf{P} values are e.g. in Ref. [14].

The modern theory of magnetization, instead, addresses “ \mathbf{M} itself” directly, and is not affected by any quantum indeterminacy. Therefore an expression for \mathbf{M} in terms of the bulk density matrix (either PBCs or OBCs), ergo boundary-independent, is not ruled out. Here we are providing such expression: in any macroscopically homogeneous sample \mathbf{M} is the macroscopic average—defined as in electrostatics [15]—of a local function $\mathfrak{M}(\mathbf{r})$, uniquely defined in terms of the density matrix in a neighborhood of \mathbf{r} . We draw attention to the fact that, in a polarized/magnetized solid, the charge and current densities $\rho^{(\text{micro})}(\mathbf{r})$ and $\mathbf{j}^{(\text{micro})}(\mathbf{r})$ are well defined,

while a “dipolar density” (either electric or magnetic) cannot be unambiguously defined [16, 17]; our $\mathfrak{M}(\mathbf{r})$ plays indeed the role of a magnetic dipolar density, although only its macroscopic average bears a physical meaning.

The main concepts are more clearly formulated in the simple two-dimensional (2D) case: electrons in the xy plane and magnetization M along z . Eq. (2) reads, for a 2D macroscopic flake of independent electrons

$$\begin{aligned} M &= -\frac{ie}{2\hbar cA} \sum_{\epsilon_n < \mu} \langle \varphi_n | \mathbf{r} \times [H, \mathbf{r}] | \varphi_n \rangle \\ &= -\frac{ie}{2\hbar cA} \sum_{\epsilon_n < \mu} (\langle \varphi_n | xHy | \varphi_n \rangle - \langle \varphi_n | yHx | \varphi_n \rangle) \end{aligned} \quad (3)$$

where A is the sample area, H is the single-particle Hamiltonian, $|\varphi_n\rangle$ are the orbitals, and μ is the Fermi level; single occupancy is assumed (“spinless electrons”). Eq. (3) only applies to a system that remains gapped (as a whole) in the large- A limit, and therefore does not apply, as such, to Chern insulators; more about this will be said below. Eq. (3) is a trace; since M is real,

$$M = \text{Im } iM = \frac{e}{\hbar cA} \text{Im Tr } \{ \mathcal{P}xHy\mathcal{P} \}, \quad (4)$$

where \mathcal{P} is the ground-state projector. In the following, we also need its complement \mathcal{Q} , i.e.

$$\mathcal{P} = \sum_{\epsilon_n < \mu} |\varphi_n\rangle\langle\varphi_n|, \quad \mathcal{Q} = 1 - \mathcal{P}. \quad (5)$$

If we write $H = \mathcal{P}H\mathcal{P} + \mathcal{Q}H\mathcal{Q}$, it is rather straightforward to transform Eq. (4) into

$$M = \frac{e}{\hbar cA} \text{Im Tr } \{ \mathcal{P}x\mathcal{Q}H\mathcal{Q}y\mathcal{P} - \mathcal{Q}x\mathcal{P}H\mathcal{P}y\mathcal{Q} \}. \quad (6)$$

A different derivation of the same expression is due to Souza and Vanderbilt [6]; they also show that Eq. (6) provides the link with the modern theory of magnetization. In fact the position operator \mathbf{r} is ill defined within PBCs [18], but becomes harmless and well defined within both OBCs and PBCs when “sandwiched” between a \mathcal{P} and a \mathcal{Q} . It is enough to perform the thermodynamic limit in Eq. (6), and then cast \mathcal{P} and \mathcal{Q} in terms of Bloch orbitals, in order to arrive at the \mathbf{k} -integral expression of the modern theory [3–5] for normal insulators (Chern number $C = 0$).

In order to get a local description we write Eq. (6) as

$$M = \frac{1}{A} \int d\mathbf{r} \mathfrak{M}_1(\mathbf{r}), \quad (7)$$

$$\begin{aligned} \mathfrak{M}_1(\mathbf{r}) &= \frac{e}{\hbar c} \text{Im } \langle \mathbf{r} | \mathcal{P}x\mathcal{Q}H\mathcal{Q}y\mathcal{P} | \mathbf{r} \rangle \\ &\quad - \frac{e}{\hbar c} \text{Im } \langle \mathbf{r} | \mathcal{Q}x\mathcal{P}H\mathcal{P}y\mathcal{Q} | \mathbf{r} \rangle. \end{aligned} \quad (8)$$

There is a paramount difference between our starting Eq. (3) and Eq. (7): while the former integral, like

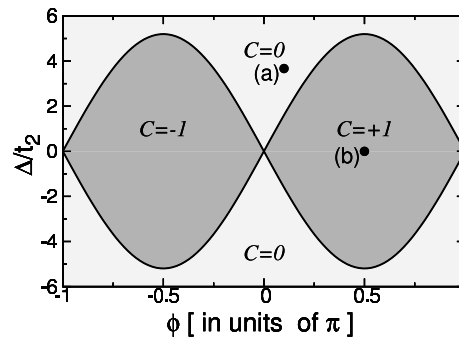


FIG. 1. Chern number C of the bottom band of the Haldane model as a function of the parameters φ and Δ/t_2 ($t_1 = 1, t_2 = 1/3$). The subsequent discussion and figures concern the points (a) and (b) only

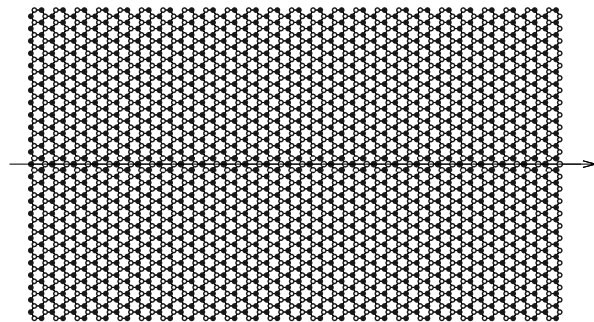


FIG. 2. A typical flake, with 2550 sites, showing the honeycomb lattice of the Haldane model [19]. The 50 sites on the horizontal line will be used in all the subsequent one-dimensional plots. Black and grey circles indicate nonequivalent sites (with onsite energies $\pm\Delta$)

Eq. (2), is dominated by boundary contributions, the latter expression is “bulk” in the above defined sense. In order to evaluate M for a macroscopically homogeneous region in the bulk of a sample, within either OBCs or PBCs, it is enough to take the macroscopic average of $\mathfrak{M}_1(\mathbf{r})$ in that region.

We demonstrate this key property of the local function $\mathfrak{M}_1(\mathbf{r})$ by performing simulations on the Haldane model Hamiltonian [19]; it is comprised of a 2D honeycomb lattice with two tight-binding sites per primitive cell with site energies $\pm\Delta$, real first-neighbor hoppings t_1 , and complex second-neighbor hoppings $t_2 e^{\pm i\varphi}$. This model has been previously used in several simulations, providing invaluable insight into orbital magnetization [4, 5, 20] as well as into nontrivial topological features of the electronic wavefunction [12, 19–23]. In the following, we invariably choose $t_1 = 1, t_2 = 1/3$. At half filling the system is insulating; it is either a normal insulator or a Chern insulator depending on the Δ and φ values, according to the phase diagram shown in Fig. 1.

We illustrate the case of a normal insulator ($C = 0$), choosing the point (a) in the phase diagram ($\Delta/t_2 = 3.67, \varphi = 0.1\pi$). We address, within OBCs, finite flakes

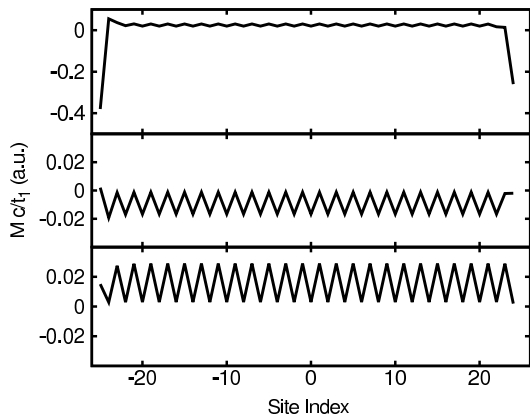


FIG. 3. Local magnetization for a normal insulator—point (a) in the phase diagram—along the line shown in Fig. 2. Top panel: site contributions to the trace in Eq. (4). Middle panel: first term in Eq. (8). Bottom panel: second term in Eq. (8). Notice the different scales.

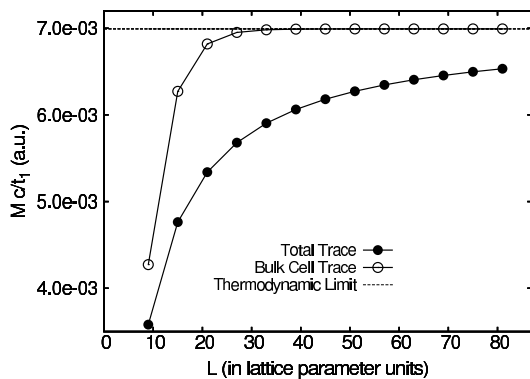


FIG. 4. Convergence of M with the flake size, point (a) in the phase diagram. Filled circles: total magnetic moment divided by the flake area, Eqs. (4) and (7). Open circles: average of $\mathfrak{M}(i)$ over the two central sites in the flake. All flakes have the same aspect ratio as in Fig. 2; the abscissae indicate the length of the arm-chair edge in lattice parameter units (75 in Fig. 2).

of rectangular shape cut from the bulk, as shown in Fig. 2. We separately plot in the two lowest panels of Fig. 3 the two terms in Eq. (8): they correspond to the “local circulation” and “itinerant circulation”, respectively, in the language of Refs. [4, 5]. It is easily realized that both are bulk, i. e. their average over any bulk cell coincides (in the large- A limit) with the average over the whole sample. The top panel of Fig. 3 shows, by contrast, the site contributions to Eq. (4). Although the trace is the same as in Eq. (7), the difference is striking: here most of the magnetization is due to the boundary. We then show in Fig. 4 the convergence of the computed M with the flake size. The figure shows that the macroscopic average of $\mathfrak{M}(i)$ in the flake center converges much faster than the trace, Eqs. (4) and (7). The former converges exponentially, owing to the density matrix decay; the

latter shows a $1/L$ convergence, because the number of bulk sites scales as L^2 , while the number of boundary sites scales as L .

Next, we address Chern insulators. Therein the spectrum of a finite sample within OBCs becomes gapless in the large sample limit; when μ is in the bulk gap, the bulk is insulating but M depends on μ , owing to boundary currents. We are going to prove that even this extra contribution to M is bulk in the above sense.

The macroscopic magnetization of a 2D macroscopic sample at fixed chemical potential is $M = -(1/A) \partial G / \partial B$, where G is the Gibbs grand potential. At zero temperature $G = U - \mu N$, and μ is the Fermi level:

$$M = -\frac{1}{A} \frac{\partial U}{\partial B} + \frac{\mu}{A} \frac{\partial N}{\partial B} = M_1 + M_2. \quad (9)$$

It is easy to show, using the Hellmann-Feynman theorem, that the first term M_1 in Eq. (9) coincides with Eq. (3), hence also with Eq. (7). Defining the areal density $n = N/A$, the second term in Eq. (9) is $M_2 = \mu \partial n / \partial B$; we then make contact with Středa’s formula [24]

$$\frac{\partial n}{\partial B} = \frac{eC}{2\pi\hbar c} = \frac{C}{\phi_0}, \quad (10)$$

where C is the Chern number and $\phi_0 = hc/e$ is the flux quantum. The formula was proved for a crystalline 2D system within PBCs [3, 25]. Its OBCs analogue displays subtle features, since for an isolated sample the number of electrons N stays constant: we are going to show that the boundary acts as a reservoir, in such a way that the density n in the bulk region obeys indeed Středa’s formula.

Even the Chern number admits a local description in real space [23], and can be directly expressed in terms of the ground state density matrix within either PBCs or OBCs. Here we define the dimensionless function [26]

$$\mathfrak{C}(\mathbf{r}) = 4\pi \text{Im} \langle \mathbf{r} | \mathcal{Q}x\mathcal{P}y\mathcal{Q} | \mathbf{r} \rangle, \quad (11)$$

whose macroscopic average in the bulk of a sample equals C . Therefore the magnetization of an insulator—either normal or Chern—obtains from the macroscopic average of $\mathfrak{M}(\mathbf{r}) = \mathfrak{M}_1(\mathbf{r}) + \mathfrak{M}_2(\mathbf{r})$ in some inner region of the sample, where $\mathfrak{M}_1(\mathbf{r})$ is the same as in Eq. (8), and

$$\mathfrak{M}_2(\mathbf{r}) = \frac{\mu}{\phi_0} \mathfrak{C}(\mathbf{r}) = \mu \frac{2e}{\hbar c} \text{Im} \langle \mathbf{r} | \mathcal{Q}x\mathcal{P}y\mathcal{Q} | \mathbf{r} \rangle. \quad (12)$$

We may also rewrite the local magnetization as

$$\begin{aligned} \mathfrak{M}(\mathbf{r}) &= \frac{e}{\hbar c} \text{Im} \langle \mathbf{r} | \mathcal{P}x\mathcal{Q}H\mathcal{Q}y\mathcal{P} | \mathbf{r} \rangle \\ &\quad - \frac{e}{\hbar c} \text{Im} \langle \mathbf{r} | \mathcal{Q}x\mathcal{P}(H - 2\mu)\mathcal{P}y\mathcal{Q} | \mathbf{r} \rangle. \end{aligned} \quad (13)$$

It is easy to verify that the macroscopic average of $\mathfrak{M}(\mathbf{r})$ is invariant by translation of the energy zero, i.e.

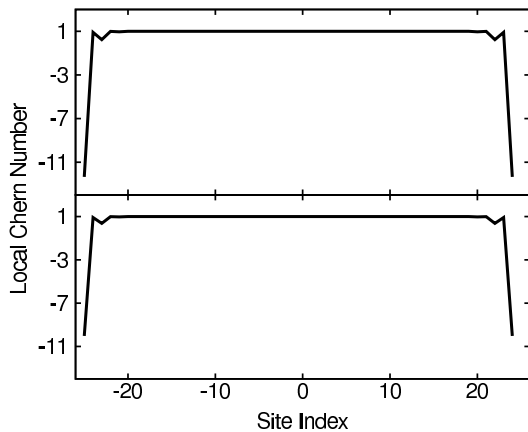


FIG. 5. Point (b) in the phase diagram. Top panel: local Chern number, Eq. (11). Bottom panel: B -derivative of the density in dimensionless units, Eq. (14).

is invariant under the transformation $H \rightarrow H + \Delta\epsilon$, $\mu \rightarrow \mu + \Delta\epsilon$, as it must be. If the thermodynamic limit is taken *before* the trace, Eq. (13) can be related to the known \mathbf{k} -space theory for the magnetization of a Chern insulator [5], and also to a recent reformulation and generalization to disordered systems [27].

We stress a very crucial feature. In Eq. (7) we have taken the trace of $\mathfrak{M}_1(\mathbf{r})$ over the whole sample within OBCs; we cannot do the same with $\mathfrak{M}_2(\mathbf{r})$, because such trace—as well as the trace of $\mathfrak{C}(\mathbf{r})$ —identically vanishes. We show in Fig. 5, top panel, a plot of $\mathfrak{C}(i)$: for the sake of simplicity we choose the very high symmetry point (b) in the phase diagram ($\Delta = 0, \varphi = \pi/2$), where the site occupancy in the bulk region is $n(i) = 1/2$, and $M_1 = 0$. It is perspicuous that the local Chern numbers $\mathfrak{C}(i)$ are equal to 1 in the bulk of the sample, while they deviate and become negative in the boundary region.

One gets the bulk magnetization M at fixed μ by taking the macroscopic average of Eq. (13) in the relevant sample region, for both normal and Chern insulators [28]. In the case of our tight-binding model, it is enough to take the average over the two central sites of the flake.

In order to evaluate M there is no need of running finite- B calculations; nonetheless it is worth showing how Středa’s formula works within OBCs. To this aim we use Eq. (10) in reverse: we give an alternative form for the local Chern number as

$$\tilde{\mathfrak{C}}(\mathbf{r}) = \phi_0 \frac{\partial n(\mathbf{r})}{\partial B}, \quad (14)$$

and we evaluate the B -derivative numerically. The result is shown in Fig. 5, bottom panel, for a B value such that the flux through the unit cell is $\phi = 0.001\phi_0$. The plot of the B -derivative of the density, as in Eq. (14), shows that Středa’s formula holds even *locally*, and confirms that the boundary region acts as an electron reservoir.

Our presentation has been limited to the 2D case for the sake of clarity; but the 3D theory is not conceptually

different, although it requires a more complex algebra. In conclusion, we have shown that the orbital magnetization \mathbf{M} in any macroscopically homogeneous region of an insulator—even topologically nontrivial—obtains as the macroscopic average of a magnetization density $\mathfrak{M}(\mathbf{r})$, Eq. (13), uniquely defined in terms of the density matrix in a neighborhood of \mathbf{r} , and insensitive to the conditions of the sample boundary. The approach applies with no major changes to disordered materials as well. Polarization \mathbf{P} behaves differently: a polarization density enjoying a similar property cannot be defined.

R. R. thanks H. Schulz-Baldes for bringing Ref. [27] to his attention. Work partially supported by the ONR Grant No. N00014-11-1-0145.

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- [1] R. D. King-Smith and D. Vanderbilt, Phys. Rev. B **47**, 1651 (1993).
 - [2] D. Vanderbilt and R. D. King-Smith, Phys. Rev. B **48**, 4442 (1993).
 - [3] D. Xiao, J. Shi, and Q. Niu, Phys. Rev. Lett. **95**, 137204 (2005).
 - [4] T. Thonhauser, D. Ceresoli, D. Vanderbilt, and R. Resta, Phys. Rev. Lett. **95**, 137205 (2005).
 - [5] D. Ceresoli, T. Thonhauser, D. Vanderbilt, and R. Resta, Phys. Rev. B **74**, 024408 (2006).
 - [6] I. Souza and D. Vanderbilt, Phys. Rev. B **77**, 054438 (2008).
 - [7] R. Resta, Rev. Mod. Phys. **66**, 899 (1994).
 - [8] R. Resta, J. Phys.: Condens. Matter **22**, 123201 (2010).
 - [9] W. Kohn, Phys. Rev. Lett. **76**, 3168 (1996).
 - [10] R. Resta, J. Chem. Phys. **124**, 104104 (2006).
 - [11] R. Resta, Eur. Phys. J. B **79**, 121 (2011).
 - [12] T. Thonhauser and D. Vanderbilt, Phys. Rev. B **74**, 235111 (2006).
 - [13] K.-T. Chen and P. A. Lee, Phys. Rev. B **86**, 195111 (2012).
 - [14] K. N. Kudin, R. Car, and R. Resta, J. Chem. Phys. **127**, 194902 (2007).
 - [15] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975).
 - [16] L. L. Hirst, Rev. Mod. Phys. **69**, 607 (1997).
 - [17] The statement does not apply to spin magnetization, where the “dipolar density” is trivially proportional to the spin density.
 - [18] R. Resta, Phys. Rev. Lett. **80**, 1800 (1998).
 - [19] F. D. M. Haldane, Phys. Rev. Lett. **61**, 2015 (1988).
 - [20] D. Ceresoli and R. Resta, Phys. Rev. B **76**, 012405 (2007).
 - [21] N. Hao *et al.*, Phys. Rev. B **78**, 075438 (2008).
 - [22] S. Coh and D. Vanderbilt, Phys. Rev. Lett. **102**, 107603 (2009).
 - [23] R. Bianco and R. Resta, Phys. Rev. B **84**, 241106(R) (2011).
 - [24] P. Středa, J. Phys. C **15**, L717 (1982).
 - [25] D. Xiao, M.-C. Chang, and Q. Niu, Rev. Mod. Phys. **82**, 1959 (2010).
 - [26] With respect to $\mathfrak{C}(\mathbf{r})$ as defined in Ref. [23], here it is expedient to exchange \mathcal{P} with \mathcal{Q} , and consequently to

- change sign.
- [27] H. Schulz-Baldes and S. Teufel, <http://arxiv.org/pdf/1201.4812v2.pdf>.
- [28] In the Chern case both $\mathfrak{M}_1(\mathbf{r})$ and $\mathfrak{M}_2(\mathbf{r})$ display a spurious boundary contribution.