Magnetic charge and indicators of the anomalous Hall effect in unconventional magnetic systems

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The anomalous Hall effect (AHE) can appear in certain antiferromagnetic metals when it is allowed by symmetry. Since the net magnetization is usually small in antiferromagnets, it is useful to have other physical indicators of the AHE that have the same symmetry properties as the latter and can be conveniently measured and calculated. Here we propose such indicators named as electronic chiralization (EC), which are constructed using spatial gradients of spin and charge densities in general periodic crystals, and can potentially be measured directly by scattering experiments. Such constructions particularly reveal the important role of magnetic charge in the AHE in unconventional magnetic systems with vanishing net magnetization. Guided by the EC we give two examples of the AHE when magnetic charge is explicitly present: A minimum honeycomb model inspired by the magnetic-charge-ordered phase of kagome spin ice, and skew scattering of two-dimensional (2D) Dirac electrons by magnetic charge.

Introduction—The anomalous Hall effect describes the transverse flow of charge currents driven by a longitudinal electric field in the absence of external magnetic fields [1–3]. The mechanisms of the AHE in ferromagnets have been well understood by now [3–8]. In recent years particular interests have been devoted to the AHE appearing in certain antiferromagnets with vanishing net magnetization [9–21], in contrast to the conventional wisdom that the anomalous Hall response is proportional to net magnetization. Although it is now clear that the AHE is generally nonvanishing as long as it is not forbidden by symmetry, it remains an open question whether one can find a convenient indicator of the AHE, that is similar to the net magnetization as a gauge-invariant observable but is not small because of energetic reasons in the AHE antiferromagnets. Such indicators, once identified, can help to understand the existence and variation of the AHE in inhomogeneous/disordered systems or across phase transitions, the dependence of the AHE on reorientation of the microscopic spin density field, and the scaling of the AHE with continuously tunable parameters such as temperature, doping, and strain, etc.

There have been a couple of proposals on constructing such indicators of the AHE in general magnetic crystals [22, 23], based on the idea of multipole expansion. Ref. 22 considered the magnetic (spherical) multipole moments of a finite atomic cluster having the same point group symmetry as the parent magnetic crystal. By decomposing the representation of a given point group in the basis of such cluster magnetic multipoles, the ones that resemble that formed by a magnetic dipole can be identified. The basis functions of such irreducible representations then transform in the same way as the magnetic dipole or the Hall conductivity vector under symmetry operations in the given point group, but not necessarily so under general O(3) operations applied on the whole magnetic crystal. More recently, Ref. 23 proposed to use the anisotropic magnetic dipole (AMD) as an indicator of the AHE. The AMD is a time-reversal-odd pseudovector and transforms in the same way as the Hall conductivity vector under general O(3) operations. However, to calculate the AMD for a given magnetic structure one still needs to first construct a finite atomic cluster using the approaches of [22] or [24].

The difficulty of defining multipole moments of an infinite crystal is long standing. In classical electromagnetism it is known that only the lowest-order nonvanishing multipole moment of a given charge or current distribution is independent of the choice of origin. Moreover, it has been realized through the studies on the electric polarization [25, 26], orbital magnetization [27–29], and magnetic toroidization [30, 31] that even these low-order multipole moments cannot be directly obtained from the local charge and current densities in a unit cell, but require the information of the ground state wavefunctions of the whole crystal. However, for the purpose of finding a physical indicator of the AHE in antiferromagnets, it is more convenient to base the construction on readily available data of the magnetic structure such as that from neutron and X-ray experiments. Such scattering experiments directly probe the Fourier components of spin and charge distributions in a periodic crystal which themselves are gauge-invariant quantities. Moreover, constructions based on scattering amplitudes may allow direct determination of the AHE indicators without having to first fix the magnetic order. Finally, the indicators may point out new mechanisms or prototypical examples for the AHE in unconventional magnetic systems.

In this Letter we propose a new class of indicators of the AHE, which we name as electronic chiralization (EC) due to resemblance to their optical counterparts [32–35], based on spatial gradients of periodic spin and charge densities of infinite crystals. We first introduce EC based on the symmetry properties of the AHE. Then we discuss their application in several model systems of AHE anti-
ferromagnets. Finally we give two nontrivial examples of the AHE inspired by the prominent role of magnetic charge in the EC: a minimal model based on the charge-ordered kagome spin ice [36–47] and skew scattering of 2D Dirac electrons by magnetic charge.

**Electronic chiralization as an indicator of the AHE**— We first discuss the symmetry properties of the AHE of a crystal. The AHE is described by the antisymmetric part of the conductivity tensor, or equivalently the anomalous Hall (pseudo)vector \( \sigma_{\text{AH}} \equiv \frac{1}{2} \epsilon_{\alpha\beta\gamma} \sigma_{\alpha\beta\gamma} \). \( \sigma_{\text{AH}} \) has definite transformation properties under time reversal (TR, acting on the equilibrium state of the system), O(3) operations, and continuous translation acting on the whole crystal, even if these operations are not in the symmetry group of the crystal. \( \sigma_{\text{AH}} \) changes sign under TR as a consequence of the Onsager relation; it rotates as a pseudovector under O(3) and is invariant under continuous translation operations because it is the response of a uniform current to a uniform electric field [48].

In addition to the system-independent properties above, \( \sigma_{\text{AH}} \) must also be invariant under any symmetry operations of the crystal, as dictated by Neumann’s principle. The symmetry operations that deserve special discussion are those combining a point group operation \( R \) with a spatial translation \( T \). Since \( \sigma_{\text{AH}} \) is invariant under continuous translation, it must be invariant under \( R \) even if \( R \) is not a symmetry of the crystal. An example of this is the vanishing of \( \sigma_{\text{AH}} \) in a collinear bipartite antiferromagnet whose magnetic unit cell is twice of the structural one.

Based on the discussion above, a suitable indicator of the AHE should be (1) a TR-odd pseudovector, and (2) invariant under all symmetry operations of the crystal. Then \( \sigma_{\text{AH}} \) will be linearly dependent this indicator to the lowest order of the latter. An important consequence of (1) is that the indicator must be translationally invariant, which is generally incompatible with multipole moments.

The TR-odd property of \( \sigma_{\text{AH}} \) is fundamentally due to the microscopic magnetization density \( m(r) \) in equilibrium. In ferromagnets the spatial average of \( m(r) \), \( \langle m \rangle \) serves as a suitable indicator of the AHE. When \( m \) nearly vanishes, it is reasonable to associate the AHE with the spatial variation of \( m(r) \). We thus propose indicators of the AHE constructed from the spatial gradients of \( m(r) \). For definiteness we only consider the first-order spatial derivative of \( m(r) \) in this work, although indicators based on higher orders in \( m \) or its derivatives can be constructed similarly and may be useful in different cases [48]. We start from a Cartesian tensor \( T_{ijk} \) defined as

\[
T_{ijk} \equiv \frac{1}{V} \int d^3r \partial_i \phi \partial_j m_k = \frac{1}{V_{uc}} \int d^3r \partial_i \phi \partial_j m_k
\]

where \( \phi \) is a TR-even scalar field observable of the crystal, which can be the charge density \( \rho(r) \) or the nonmagnetic potential \( V(r) \); \( V_{uc} \) stands for unit cell. We ignore any boundary contributions to \( m(r) \) and \( \phi(r) \) so that they have the same discrete translation symmetry as the infinite crystal. The inclusion of \( \phi \) is to ensure that \( T_{ijk} \) does not become a boundary term. It also signifies the role of orbital degrees of freedom in the AHE. Clearly \( T_{ijk} \) is a TR-odd rank-3 pseudotensor and is translationally invariant. It is also invariant under any symmetry operations of the crystal since both \( \phi(r) \) and \( m(r) \) are physical observables of the crystal. A pseudovector can be obtained from \( T_{ijk} \) by contracting it with Kronecker \( \delta \) or the Levi-Civita symbol \( \epsilon \). The latter must appear in pairs so that the obtained quantity is even under inversion. The only two independent pseudovectors obtained from this construction are

\[
\chi_1 \equiv \frac{1}{V} \int d^3r (\nabla \phi)(\nabla \cdot m), \quad (2)
\]

\[
\chi_2 \equiv \frac{1}{V} \int d^3r (\nabla \phi) \times (\nabla \times m).
\]

We name \( \chi_{1,2} \) generally as “electronic chiralization” to emphasize their electronic origin and pseudovector nature, analogous to the optical chirality (flow) in optics [32–35]. Several comments are in order: (i) One can define \( \chi_3 \equiv \frac{1}{V} \int d^3r (\nabla^2 \phi)m \) which is a linear combination of \( \chi_{1,2} \) using integration by parts. However, a nonzero \( \chi_3 \) suggests an AHE that is due to compensated \( m \) located on structurally inequivalent sites (different \( \nabla^2 \phi \)) and is relatively common. We thus focus on \( \chi_1 \) in this work only.

(ii) \( \chi_1 \) and \( \chi_2 \) are respectively related to the magnetic charge density \( \rho_m \equiv -\nabla \cdot m \) and the electric current density \( j = \nabla \times m \). When \( m \) can be approximated by \( g_{\mu B} s(r) \) the spin density, \( \nabla \times s(r) \) is the “spin current” contribution to the conserved charge current in the Dirac theory of electrons.

(iii) Using the electron charge density \( \rho(r) \) as the scalar field \( \phi \), one can potentially obtain \( \chi_{1,2} \) directly from magnetic neutron or X-ray diffraction data since it only requires the knowledge of \( \rho_{K_m} \mathbf{m}_K \), where \( K \) is a reciprocal lattice vector. Such a combination can appear, e.g. (for \( \chi_2 \), in the interference term of elastic neutron scattering cross-section between magnetic and electrostatic scatterings [49].

**EC in model examples of AHE antiferromagnets**— To start, we give equivalent expressions of Eq. (2) that are more suitable for tight-binding models. Denoting the net charge and magnetic moment of an ion as \( Q \) and \( M \), and assuming their spatial distribution about the nucleus are Gaussians \( g(r) \) with standard deviation \( \sigma \), we have

\[
\chi_1 = \frac{1}{V_{uc}} \sum_{nm} \sum_{\mathbf{R}} Q_n M_m \cdot \mathbf{a} \mathbf{R} + \mathbf{r}_n - \mathbf{r}_m. \quad (3)
\]

where the tensor \( I_{ij}(\mathbf{a}) \) is defined as

\[
I_{ij}(\mathbf{a}) = -\int d^3rg(r - \mathbf{a})\partial_i \partial_j g(r). \quad (4)
\]
Denoting $\mathbf{R} + \mathbf{r}_n - \mathbf{r}_m$ as $\mathbf{r}_{mn}$, we have
\[
\mathbf{T}(\mathbf{r}_{mn}) = \frac{1}{16\pi^2 \sigma^5} \left( \Pi - \mathbf{r}_{mn} \mathbf{r}_{mn} \right) e^{-\frac{r_{nn}^2}{2\sigma^2}}. \tag{5}
\]
It is interesting to note that the second term has the form of an electric quadruple. The main difference between the quadrupole moment here and and that in [23] is that the origin of the former is different for different pairs of ions. The trace of $\mathbf{T}$ gives rise to a weighted sum of electric charge surrounding a given magnetic moment $\mathbf{M}_m$. For a compensated ferrimagnet whose magnetic sublattices are inequivalent in the paramagnetic state, it will have a nonzero $\chi_1$ mainly due to this contribution. We will focus the traceless part of $\mathbf{T}$ below since it is less trivial. Separately, one can show that $\chi_2$ has a similar composition as $\chi_1$ [48]. Therefore it suffices to focus on $\chi_1$.

For simplicity we consider nearest neighbors only. Then
\[
\chi_1 = C \sum_m \mathbf{M}_m \cdot \mathbf{Q}_m \tag{6}
\]
where $C = -e^2/32\pi^2 \sigma^7 V_{uc}$ is a system dependent constant and $r_{nn}$ is the distance between a magnetic atom and its nearest neighbors. $\mathbf{Q}_m$ is the total electric quadrupole relative to the position of site $m$:
\[
\mathbf{Q}_m = \sum_{i \in \{m\}_m} Q_i \left( \mathbf{r}_{mi} \mathbf{r}_{mi} - \frac{1}{3} r_{mn}^2 \mathbf{1} \right) \tag{7}
\]
where $\{i\}_m$ stands for the set of nearest neighbors of site $m$, and $r_{nn}$ is the nearest-neighbor distance. It should be noted that $\mathbf{Q}_m$ has the same symmetry as a second-order, i.e. easy-axis or easy-plane, magnetic anisotropy. The origin of weak ferromagnetism and hence the AHE in systems with a nonzero $\chi_1$ is thus a site-dependent second order anisotropy, which applies to the known examples such as the noncollinear Mn$_3$X ($X = Ir, Pt, Sn, Ge, \text{etc.}$) and collinear AHE antiferromagnets [20, 21]. If such a site-dependent second order anisotropy is forbidden by symmetry, as in the case of hematite, one needs to consider EC that depends on higher-order spatial derivatives of $\rho$ and $\mathbf{m}$ [48].

We can now examine the behavior of Eq. (3) in a few model examples [10, 50]. For Mn$_3$Ir [Fig. 1 (a)], $\mathbf{Q}_m$ is diagonal with the principal axis of the largest eigenvalue along the four-fold axis on each Mn atom. One can then obtain the dependence of $\chi_1$ on rigid rotations of the sublattice moments, which as expected is very different from rotating the pseudovector $\chi_1$ directly. For example, the length of $\chi_1$ depends on the rotation about its direction as $|\cos \gamma|$ [Fig. 1 (b)], where $\gamma$ is the rotation angle, similar to $\sigma^{\text{AF}}$ and the orbital magnetization [50]. Such behavior is expected because $\chi_1$ is not required to transform as a pseudovector under separate rotations of the lattice and magnetic moments. For the magnetic structure of Mn$_3$Sn [Fig. 1 (c)] one can follow the same procedure and obtain a compact expression:
\[
\chi_1 \propto \cos ^2 \left( \frac{\beta }{2} \right) \left[ \sin (\alpha + \gamma ) \hat{x} + \cos (\alpha + \gamma ) \hat{y} \right] \tag{8}
\]
where $\alpha, \beta, \gamma$ are Euler angles about the $z, y', z''$ axes, respectively. It is interesting to note that the counterclockwise rotation about $z$ of all sublattice moments leads to clockwise rotation of $\chi_1$, same as the weak magnetization [12]. Moreover, rotation about the $y$ axis in Fig. 1 (c) by $\pi$ makes $\chi_1$ vanish [Fig. 1 (d)], since in this case the magnetic order become triangular rather than inverse triangular, and the AHE or weak ferromagnetism are forbidden by a $C_3$ symmetry.

Motivated by the way that $\nabla \phi$ and $\rho_m$ cooperatively give rise to finite $\chi_1$, we will next give two experimentally relevant model examples in which magnetic charge appears more explicitly and leads to the AHE.

**Minimal model of the AHE due to magnetic charge order**—We first consider a minimal tight-binding model having the essential ingredients for magnetic-charge-induced AHE. The model describes $s$ electrons hopping between nearest neighbors on a honeycomb lattice, with magnetic charge of opposite signs residing on the two
sublattices [Fig. 2 (a)]:

\[
H = -t \sum_{\langle ij \rangle \alpha} c_{i\alpha}^\dagger c_{j\alpha} - t_M \sum_{\langle ij \rangle \alpha\beta} \eta_{ij} \sigma_{\alpha\beta} \cdot \hat{\mathbf{r}}_{ij} c_{i\alpha}^\dagger c_{j\beta} + \Delta \sum_{\langle ij \rangle} \gamma_i c_{i\alpha}^\dagger c_{i\alpha}
\]

(9)

where the four terms respectively correspond to spin-independent hopping, spin-dependent hopping due to the magnetic charge, Rashba spin-orbit coupling, and an on-site potential breaking the sublattice symmetry; \( \eta_{ij} = +1(-1) \) if \( \hat{\mathbf{r}}_{ij} \) points from sublattice A (B) to B (A); \( \eta_{ij} \) together with \( \sigma \cdot \hat{\mathbf{r}}_{ij} \) capture the spin-dependent hopping due to the magnetic field (\( \mathbf{H} \) field) lines between neighboring magnetic charges; \( \gamma_i = +1(-1) \) on A (B) sublattice. The Rashba term is needed to provide the direction of \( \chi_1 \) along \( z \) as suggested by the expression of \( \chi_1 \), and the sublattice potential is needed to break the degeneracy between the opposite magnetic charges on the two sublattices.

\[\text{FIG. 2. (Color online) (a) Honeycomb lattice model with opposite magnetic charges residing on the two sublattices, respectively. The arrows correspond to the magnetic field lines. (b) Band structure (top) and Berry curvature summed over occupied bands (bottom) of the model. \( \mathbf{M}' = -\mathbf{M} \) and \( \mathbf{K}' = -\mathbf{K} \). The parameter values are: } t = 1, t_M = 0.7, \lambda_R = 0.2, \Delta = 0.5. \ (c) Berry curvature obtained using the same parameters as in (b) but plotted in the 2D momentum space.\]

is the gap opened by \( \Delta \), since the \( t_M \) term becomes identical to the Rashba term near \( K, K' \) [48]).

The minimal model can be connected with the magnetic-charge-ordered state of the kagome spin ice [37, 38, 45] by the duality between honeycomb and kagome lattices. The background magnetic field connecting neighboring magnetic charges can be regarded, as a first approximation, as the homogenized effect of the fluctuating magnetic dipole moments in the charge-ordered state of the kagome spin ice on itinerant electrons. In comparison with the models studied in [51, 52] where the local spins on the kagome lattice is noncoplanar, the present model has a vanishing net magnetization. More importantly, the essential symmetry breaking in the nonmagnetic part of model (9) is already present in the pyrochlore iridate \( \text{Pr}_2\text{Ir}_2\text{O}_7 \) [9], in which an AHE in the absence of long-range dipolar order and of net magnetization has been observed [53]. Although the ground state of \( \text{Pr}_2\text{Ir}_2\text{O}_7 \) may be elusive and the direct measurement [54] of scalar spin chirality by scattering techniques is challenging, it is possible to alternatively measure the EC which, if nonzero, can help solve the puzzle of the zero-field AHE in [53].

\textit{Skew scattering by magnetic charge}—In this section we predict an extrinsic contribution to the AHE by magnetic charge through skew scattering. Again motivated by the expression of \( \chi_1 \), we consider the following model of 2D Dirac electrons with Rashba-type spin-momentum-lowering scattered by a magnetic charge whose magnetic field is truncated at finite radius \( R \):

\[
H = -\hbar \lambda (\sigma_x \partial_y - \sigma_y \partial_x) - \frac{\Delta}{2\pi R} \cdot \sigma \Theta(R - r) \equiv H_D + H_\Delta
\]

(10)

where \( \Theta(r) \) is the step function. \( H_\Delta \) represents the Zee- man coupling between the electron spin and the magnetic field \( \mathbf{h}(r) = -\mathbf{m}(r) = \Delta \hat{x}/(2\pi r) \) generated by a magnetic charge located at the origin within a radius \( R \). Here we consider the analytically simpler case of \( \alpha \equiv \Delta/(2\pi \hbar \lambda) = 1 \) and relegate the more general solution to [48]. Assuming a positive chemical potential, the solution for \( r < R \) with energy \( E = \hbar \lambda k_0 > 0 \) is

\[
\Psi < = \sum_{n=-\infty}^{\infty} a_n \left( J_{n-1}(k_0 r) e^{in\theta} \right)
\]

(11)

while that for \( r > R \) with an incident plane wave traveling along \( \hat{x} \) is

\[
\Psi > = \frac{e^{ik_0 x}}{2\sqrt{2\pi}} \left( \frac{1}{1} + \sum_{n=-\infty}^{\infty} b_n \left( \frac{H_n(k_0 r) e^{in\theta}}{H_{n+1}(k_0 r) e^{i(n+1)\theta}} \right) \right)
\]

(12)

while \( \Psi_{\text{scatt}} \) does not include the origin where \( Y_n \) diverges.
Also the Hankel function of the 1st kind represents outgoing waves [5]. Solving $b_n$ from the boundary condition $\Psi_{\leq r=R} = R_{\leq r=R}$ and taking the large-distance asymptotic form of $\Psi_{\text{scatt}}$, we obtain the scattering cross section
\[
\sigma(\theta) \propto \langle \hat{j} \cdot \hat{r} \rangle_{\text{scatt}}(r, \theta) = \frac{4A}{\pi k_0 R^2} \sum_n b_n e^{i n (\theta - \frac{\pi}{2})} \left| b_n \right|^2. \tag{13}
\]
When $k_0 R \ll 1$ one can consider up to $p$-wave contributions to $\sigma(\theta)$. The Hall angle due to scattering by the magnetic charge only can be calculated as [55]
\[
\tan \theta_H = \frac{\int \sigma(\theta) \sin \theta d\theta}{\int \sigma(\theta) (1 - \cos \theta) d\theta} \approx \Re \left( \frac{b_{-1} + b_1}{b_0} \right) \tag{14}
\]
confirming the time-reversal-odd property of the AHE. Experimental detection of such an effect may be performed using topological insulator surface states [56] or 2D electron gas with large Rashba spin-orbit coupling that are proximate-coupled to magnetic textures [57, 58] having a nonzero 2D magnetic charge density. With $h \lambda \approx 10^3$ eV$\cdot$Å and $R \approx 1/k_0 \approx 100$ Å, the Zeeman coupling $\Delta/(2\pi R) \approx 0.1$ eV is reasonable to achieve experimentally.

Discussion — The object of electronic chiralization introduced in this work is a construction based on charge and spin densities that are themselves physical observables measured by scattering techniques. Therefore it does not directly correspond to a thermodynamic variable that is conjugate to a single external field configuration, such as the magnetization or toroidization. In this sense the EC is similar to other constructions such as the cluster multipole moments that are based on localized atomic magnetic moments. However, a corresponding thermodynamic variable for EC may be defined through the coupling with multipole moments of non-Gaussian electromagnetic waves. First-principles calculations of the EC is also straightforward, but to get meaningful values a proper treatment of the cutoff (e.g., by using an atomic form factor) may be essential due to the fluctuation of the gradients of charge and spin densities at high energies. Alternatively, one may use the expressions of EC derived by assuming localized atomic magnetic moments and charge.

Although we mainly focused on the magnetic-charge-related $\chi_1$, it is possible to predict the existence of the AHE in other systems based on the forms of $\chi_{2,3}$ and their generalizations [48]. Such predictions based on symmetry grounds do not necessarily yield universal microscopic mechanisms for the AHE, but may provide interesting new cases that in turn reveal new AHE mechanisms.

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