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Unconventional Hall effect and magnetoresistance induced by metallic ferroaxial ordering

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Transport property under metallic ferroaxial ordering in an external magnetic field is theoretically investigated. After presenting the relation between the magnetoconductivity tensor and ferroaxial moment from the symmetry viewpoint, we analyze the behavior of the unconventional Hall effect and magnetoconductivity for a general five d -orbital tight-binding model under the point group C_{4h} , where the ferroaxial moment is activated. We show that the crystalline electric field that arises from the symmetry reduction from D_{4h} to C_{4h} is essential for the ferroaxial-related magnetotransport, while the relativistic spin-orbit coupling is not required. We also compare the unconventional Hall effect driven by the ferroaxial moment with the conventional Hall effect, the latter of which does not require the ferroaxial moment. The present results provide characteristic transport properties in the ferroaxial systems, which can be observed in various candidate materials like $\text{Ca}_5\text{Ir}_3\text{O}_{12}$.

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I. INTRODUCTION

Ferroaxial order has attracted much attention as a new quantum state of matter, which is qualitatively distinct from ferromagnetic, ferroelectric, and ferromagnetoelectric (magnetic toroidal) orders in terms of spatial inversion and time-reversal parities [1–3]. Its order parameter is characterized by a time-reversal-even axial vector, which means that the ferroaxial moment can be activated even in the paramagnetic state with centrosymmetric lattice structures. Such ferroaxial order has been mainly studied in dielectric materials, where the vortex structure of the electric dipole moments corresponding to a signature of the ferroaxial moment emerges in nanodisks and nanorods [4–8]. So far, several materials have been identified to be ferroaxial ordered states in bulk systems, such as $\text{Co}_3\text{Nb}_2\text{O}_8$ [9], $\text{CaMn}_7\text{O}_{12}$ [10], $\text{RbFe}(\text{MoO}_4)_2$ [11,12], NiTiO_3 [12–15], $\text{Ca}_5\text{Ir}_3\text{O}_{12}$ [16–19], BaCoSiO_4 [20], $\text{K}_2\text{Zr}(\text{PO}_4)_2$ [21], $\text{Na}_2\text{Hf}(\text{BO}_3)_2$ [22], and Na-superionic conductors [23].

The microscopic origin of ferroaxial ordering is described by odd-rank electric toroidal multipoles, such as the electric toroidal dipole and octupole, whose expression at the classical level is obtained by performing the multipole expansion under the assumption that the magnetic charge (monopole) exists in Maxwell’s equation [24–27]. In this case, the electric toroidal dipole is described by the outer product of the position vector and the electric dipole. Meanwhile, recent theoretical studies revealed an atomic-scale description of the electric toroidal dipole at the quantum-mechanical level by introducing the symmetry-adapted multipole basis [28], where it can be also described by the outer product of the orbital and spin angular momenta [29]. Such fundamental progress in terms of the microscopic description of the electric toroidal dipole provides the possibility of the atomic-scale ferroaxial order and a way of the quantitative evaluation based on the *ab initio* calculations [30].

In parallel with the development of microscopic expressions, physical phenomena driven by the ferroaxial moment have been extensively investigated [3,31–36]. As the ferroaxial moment breaks the mirror symmetry parallel to its direction, it acts as a rotator against external stimuli [35]. For example, when the ferroaxial moment lies in the z direction, an input field along the x (y) direction induces the response of the conjugate physical quantities along the y ($-x$) direction. In other words, the ferroaxial moment becomes a source of transverse responses between the input and output fields/currents with the same symmetry. These intriguing features would open a new research field with the use of the ferroaxial moment, i.e., “ferroaxiality,” as with ferromagnetism and ferroelectricity.

In the present study, we further investigate physical phenomena under metallic ferroaxial ordering. We especially focus on two transport phenomena under an external magnetic field: one is the unconventional Hall effect and the other is the magnetoconductivity, both of which are caused by a ferroaxial moment as nanometric rotator. The former is characterized by the off-diagonal antisymmetric part of the linear conductivity tensor proportional to an external magnetic field H , while the latter is by its symmetric part proportional to H^2 . We perform symmetry and model analyses for a general five d -orbital model consisting of electron hopping, relativistic spin-orbit coupling, and tetragonal crystalline electric field under the point group C_{4h} . As a result, we find that the crystalline electric field arising from the symmetry reduction from D_{4h} to C_{4h} leads to the unconventional Hall effect and magnetoconductivity characteristic of the metallic ferroaxial moment. Our results indicate that the ferroaxial system exhibits characteristic transport properties, which would be observed in ferroaxial materials, such as $\text{Ca}_5\text{Ir}_3\text{O}_{12}$ [37].

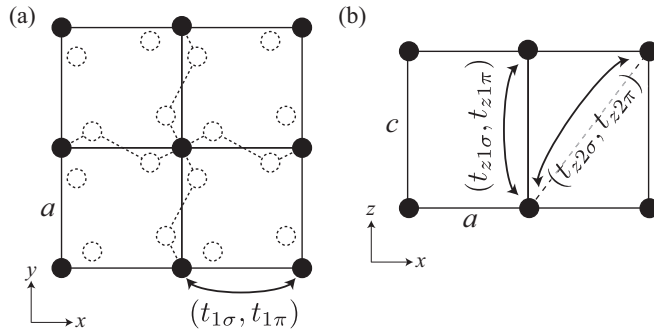


FIG. 1. Lattice structure with the lattice constants a and c under the point group C_{4h} viewed from (a) the z axis and (b) the y axis. The black-filled circles represent the d -orbital sites, while the black-dashed circles represent the ligand sites. The hopping parameters, $(t_{1\sigma}, t_{1\pi}, t_{z1\sigma}, t_{z1\pi}, t_{z2\sigma}, t_{z2\pi})$, are presented.

The rest of the paper is organized as follows. In Sec. II, we present the tight-binding model in the tetragonal lattice structure. We also introduce the ferroaxial moments described by the electric toroidal dipole, octupole, and electric hexadecapole. Among them, we discuss the behavior of the field-induced electric toroidal dipole. We give symmetry analysis and numerical results in terms of the magnetoconductivity tensor under an external magnetic field in Secs. III and IV. In Sec. III, we discuss the behavior of the unconventional Hall conductivity with an odd function of H and, in Sec. IV, we discuss that of the magnetoconductivity with an even function of H . Section V is devoted to a summary of this paper. We present the essential model parameters for the field-induced electric toroidal dipole, unconventional Hall conductivity, and magnetoconductivity in Appendix A. In Appendix B, we show the result of the optical unconventional Hall conductivity.

II. SETUP

In this section, we introduce the tight-binding model in the three-dimensional tetragonal lattice system under the point group C_{4h} in Sec. II A. Then, we discuss the behavior of the atomic-scale electric toroidal dipole induced by the external magnetic field in Sec. II B.

A. Model

We consider the tight-binding model consisting of five d orbitals, where the lattice structure is set as a three-dimensional square-lattice structure belonging to the point group C_{4h} ; the vertical mirror plane perpendicular to the xy plane is broken. We suppose here that such a lack of vertical mirror symmetry is caused by the ligand ions (the black-dashed circles) around the d -orbital sites (the black-filled circles), as shown in Fig. 1(a); the point group is D_{4h} if the ligand ions are absent. The square layers in Fig. 1(a) are stacked along the z direction, as shown in Fig. 1(b). We set the lattice constants as $a = c = 1$ for simplicity. Then, the Hamiltonian is given by

$$\mathcal{H} = \mathcal{H}_t + \mathcal{H}_{\text{SOC}} + \mathcal{H}_{\text{CEF}} + \mathcal{H}_V + \mathcal{H}_Z, \quad (1)$$

$$\mathcal{H}_t = \sum_{ij\alpha\beta\sigma} (t_{\alpha\beta}^{ij} c_{i\alpha\sigma}^\dagger c_{j\beta\sigma} + \text{H.c.}), \quad (2)$$

$$\mathcal{H}_{\text{SOC}} = \lambda \sum_i \mathbf{l}_i \cdot \mathbf{s}_i, \quad (3)$$

$$\mathcal{H}_{\text{CEF}} = \sum_{i\sigma} (\Delta_1 c_{ixy\sigma}^\dagger c_{ixy\sigma} + \Delta_2 c_{iv\sigma}^\dagger c_{iv\sigma} + \Delta_3 c_{iu\sigma}^\dagger c_{iu\sigma}), \quad (4)$$

$$\mathcal{H}_V = V \sum_{i\sigma} (c_{ixy\sigma}^\dagger c_{iv\sigma} + c_{iv\sigma}^\dagger c_{ixy\sigma}), \quad (5)$$

$$\mathcal{H}_Z = - \sum_i \mathbf{H} \cdot (\mathbf{l}_i + 2\mathbf{s}_i), \quad (6)$$

where $c_{i\alpha\sigma}^\dagger$ ($c_{i\alpha\sigma}$) is the creation (annihilation) operator of an electron with site i , orbital $\alpha = (u = 3z^2 - r^2, v = x^2 - y^2, yz, zx, xy)$ for five d orbitals ($d_u, d_v, d_{yz}, d_{zx}, d_{xy}$), and spin σ .

The first term in Eq. (1) represents the hopping Hamiltonian \mathcal{H}_t in Eq. (2). We consider nonzero hopping elements allowed from the symmetry based on the Slater-Koster table; we parametrize the nearest-neighbor hopping in the xy plane, $(t_{1\sigma}, t_{1\pi})$ [Fig. 1(a)], and the nearest-neighbor and next-nearest-neighbor hoppings along the z direction, $(t_{z1\sigma}, t_{z1\pi})$ and $(t_{z2\sigma}, t_{z2\pi})$ [Fig. 1(b)], where we ignore the δ component of the Slater-Koster parameters for simplicity. We set $t_{1\sigma} = -1$, $t_{1\pi} = 0.5$, $t_{z1\sigma} = -0.8$, $t_{z1\pi} = 0.4$, $t_{z2\sigma} = -0.5$, and $t_{z2\pi} = 0.25$; $t_{1\sigma}$ is the energy unit of the model. The second term in Eq. (1) represents the spin-orbit coupling Hamiltonian \mathcal{H}_{SOC} in Eq. (3), where \mathbf{l}_i and \mathbf{s}_i represent the orbital and spin angular momentum operators, respectively; we set $\lambda = 2$ unless otherwise stated by implicitly considering the $5d$ materials where λ tends to be comparable to the hopping parameters [38]. The third term in Eq. (1) represents the Hamiltonian for the crystalline electric field (CEF) under the point group D_{4h} \mathcal{H}_{CEF} in Eq. (4). We take $\Delta_1 = 0.5b$, $\Delta_2 = 6.5b$, and $\Delta_3 = 10.5b$ with $b = \sqrt{3}/2$, where the atomic-orbital energy levels E_α satisfy $E_{yz} = E_{zx} < E_{xy} < E_v < E_u$. The fourth term in Eq. (1) represents the additional CEF Hamiltonian in Eq. (5) to represent the symmetry lowering from D_{4h} to C_{4h} . The CEF parameter V mixes the xy and x^2-y^2 orbitals; we set $V = 0.7$. The choice of the model parameters does not alter qualitative features in the following sections. In particular, a different ratio of the spin-orbit coupling and CEF as found in $3d$, $4d$, and $5d$ materials does not affect the following results at the qualitative level; we demonstrate this argument by performing the method to extract the essential model parameters in response tensors in Appendix A. The last term in Eq. (1) represents the Zeeman Hamiltonian in Eq. (6) for an external magnetic field $\mathbf{H} = (H_x, 0, H_z) = H(\sin \theta_H, 0, \cos \theta_H)$ for $0 \leq \theta_H \leq \pi/2$.

B. Ferroaxial moment

In the model in Eq. (1), there are $100 = 10 \times 10$ independent electronic degrees of freedom in the Hilbert space, which are composed of five orbital and two spin degrees of freedom ($5 \times 2 = 10$). In spinless space, there are $25 = 5 \times 5$ electronic degrees of freedom. As we consider d orbital with the orbital angular momentum $l = 2$, the spinless Hilbert space is spanned by the rank 0–4 multipoles [27,29,39,40]: the electric monopole (Q_0), magnetic dipole (M_x, M_y, M_z), electric quadrupole ($Q_u, Q_v, Q_{yz}, Q_{zx}, Q_{xy}$), magnetic octupole ($M_{xyz}, M_{3a}, M_{3b}, M_z^\alpha, M_{3u}, M_{3v}, M_z^\beta$), and electric

TABLE I. Relationship between the irreducible representation (irrep) and multipole activated in spinless and spinful spaces under the point groups D_{4h} and C_{4h} . The sign of the superscript in the irreducible representation denotes the time-reversal parity (+: even; -: odd). The multipole degrees of freedom belonging to the A_{2g}^+ representation correspond to the ferroaxial moment.

Irrep (D_{4h})	(C_{4h})	Spinless	Multipole	Spinful	Multipole
A_{1g}^+	A_g^+	4	Q_0, Q_u, Q_4, Q_{4u}	5	$Q_0, 2Q_u, Q_4, Q_{4u}$
A_{2g}^+	A_g^+	1	Q_{4z}^α	3	$Q_{4z}^\alpha, G_z, G_z^\alpha$
B_{1g}^+	B_g^+	2	Q_v, Q_{4v}	4	$2Q_v, Q_{4v}, G_{xyz}$
B_{2g}^+	B_g^+	2	Q_{xy}, Q_{4z}^β	4	$2Q_{xy}, Q_{4z}^\beta, G_z^\beta$
E_g^+	E_g^+	3	$Q_{yz}, Q_{4x}^\alpha, Q_{4x}^\beta$ $Q_{zx}, Q_{4y}^\alpha, Q_{4y}^\beta$	7	$2Q_{yz}, Q_{4x}^\alpha, Q_{4x}^\beta, G_x, G_{3a}, G_{3u}$ $2Q_{zx}, Q_{4y}^\alpha, Q_{4y}^\beta, G_y, G_{3b}, G_{3v}$
A_{1g}^-	A_g^-	0		4	M_{5u}, T_u, T_4, T_{4u}
A_{2g}^-	A_g^-	2	M_z, M_z^α	7	$2M_z, 2M_z^\alpha, M_{5z}^{\alpha 1}, M_{5z}^{\alpha 2}, T_{4z}^\alpha$
B_{1g}^-	B_g^-	1	M_{xyz}	5	$2M_{xyz}, M_{5v}, T_v, T_{4v}$
B_{2g}^-	B_g^-	1	M_z^β	5	$2M_z^\beta, M_{5z}^\beta, T_{xy}, T_{4z}^\beta$
E_g^-	E_g^-	3	$M_x, M_x^\alpha, M_x^\beta$ $M_y, M_y^\alpha, M_y^\beta$	12	$2M_x, 2M_{3a}, 2M_{3u}, M_{5x}^{\alpha 1}, M_{5x}^{\alpha 2}, M_{5x}^\beta, T_{yz}, T_{4x}^\alpha, T_{4x}^\beta$ $2M_y, 2M_{3b}, 2M_{3v}, M_{5y}^{\alpha 1}, M_{5y}^{\alpha 2}, M_{5y}^\beta, T_{zx}, T_{4y}^\alpha, T_{4y}^\beta$

hexadecapole ($Q_4, Q_{4u}, Q_{4v}, Q_{4x}^\alpha, Q_{4x}^\beta, Q_{4z}^\alpha, Q_{4z}^\beta, Q_{4y}^\alpha, Q_{4y}^\beta$), i.e., $1 + 3 + 5 + 7 + 9 = 25$. Among them, only Q_{4z}^α belongs to the A_{2g}^+ representation of D_{4h} point group (+ in the superscript means the time-reversal even), which is related to the ferroaxial moment. The matrix of \mathcal{H}_V in Eq. (5) corresponds to Q_{4z}^α . The other 75 electronic degrees of freedom activated in spinful Hilbert space are as follows: one electric monopole, two magnetic dipoles, two electric quadrupoles, two magnetic octupoles, one electric hexadecapole, one magnetic dotriacontapole ($M_{5u}, M_{5v}, M_{5x}^{\alpha 1}, M_{5x}^{\alpha 2}, M_{5y}^{\alpha 1}, M_{5y}^{\alpha 2}, M_{5z}^{\alpha 1}, M_{5z}^{\alpha 2}, M_{5x}^\beta, M_{5y}^\beta, M_{5z}^\beta$), one electric toroidal dipole (G_x, G_y, G_z), one magnetic toroidal quadrupole ($T_u, T_v, T_{yz}, T_{zx}, T_{xy}$), one electric toroidal octupole ($G_{xyz}, G_{3a}, G_{3b}, G_z^\alpha, G_{3u}, G_{3v}, G_z^\beta$), and one magnetic toroidal hexadecapole ($T_4, T_{4u}, T_{4v}, T_{4x}^\alpha, T_{4x}^\beta, T_{4y}^\alpha, T_{4y}^\beta$).

$T_{4y}^\beta, T_{4z}^\beta$), i.e., $1 + 2 \times 3 + 2 \times 5 + 2 \times 7 + 9 + 11 + 3 + 5 + 7 + 9 = 75$. Among them, there are three multipole degrees of freedom belonging to the A_{2g}^+ representation of D_{4h} ; Q_{4z}^α, G_z , and G_z^α . The relationship between the irreducible representation under the point groups D_{4h} and C_{4h} and multipoles activated in spinless and spinful spaces are summarized in Table I.

The matrix elements of four multipoles belonging to the A_{2g}^+ representation are given by

$$Q_{4z}^\alpha = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \end{pmatrix} \sigma_0, \quad (7)$$

$$G_z = \frac{1}{2} \begin{pmatrix} 0 & 0 & \sqrt{3}i & 0 & 0 \\ 0 & 0 & i & 0 & 0 \\ -\sqrt{3}i & -i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & i \\ 0 & 0 & 0 & -i & 0 \end{pmatrix} \sigma_y - \frac{1}{2} \begin{pmatrix} 0 & 0 & 0 & -\sqrt{3}i & 0 \\ 0 & 0 & 0 & i & 0 \\ 0 & 0 & 0 & 0 & -i \\ \sqrt{3}i & -i & 0 & 0 & 0 \\ 0 & 0 & i & 0 & 0 \end{pmatrix} \sigma_x, \quad (8)$$

$$G_z^\alpha = \begin{pmatrix} 0 & 0 & 2i & 0 & 0 \\ 0 & 0 & -\sqrt{3}i & 0 & 0 \\ -2i & \sqrt{3}i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\sqrt{3}i \\ 0 & 0 & 0 & \sqrt{3}i & 0 \end{pmatrix} \sigma_y - \begin{pmatrix} 0 & 0 & 0 & -2i & 0 \\ 0 & 0 & 0 & -\sqrt{3}i & 0 \\ 0 & 0 & 0 & 0 & \sqrt{3}i \\ 2i & \sqrt{3}i & 0 & 0 & 0 \\ 0 & 0 & -\sqrt{3}i & 0 & 0 \end{pmatrix} \sigma_x, \quad (9)$$

$$Q_{4z}^\alpha = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -i & 0 & 0 \\ 0 & i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & i \\ 0 & 0 & 0 & -i & 0 \end{pmatrix} \sigma_y + \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & i & 0 \\ 0 & 0 & 0 & 0 & i \\ 0 & -i & 0 & 0 & 0 \\ 0 & 0 & -i & 0 & 0 \end{pmatrix} \sigma_x, \quad (10)$$

for the basis ($d_u, d_v, d_{yz}, d_{zx}, d_{xy}$); $\sigma = (\sigma_x, \sigma_y, \sigma_z)$ is the vector of the Pauli matrices and σ_0 is the 2×2 unit matrix in spin space [35,41]. To distinguish the electric hexadecapole in spinless and spinful bases, we denote the latter as Q_{4z}^α .

By numerically calculating the expectation values of ($Q_{4z}^\alpha, Q_{4z}^\alpha, G_z, G_z^\alpha$) for the Hamiltonian in Eq. (1) at zero field, one finds that $\langle Q_{4z}^\alpha \rangle, \langle Q_{4z}^\alpha \rangle$, and $\langle G_z^\alpha \rangle$ become nonzero but $\langle G_z \rangle$

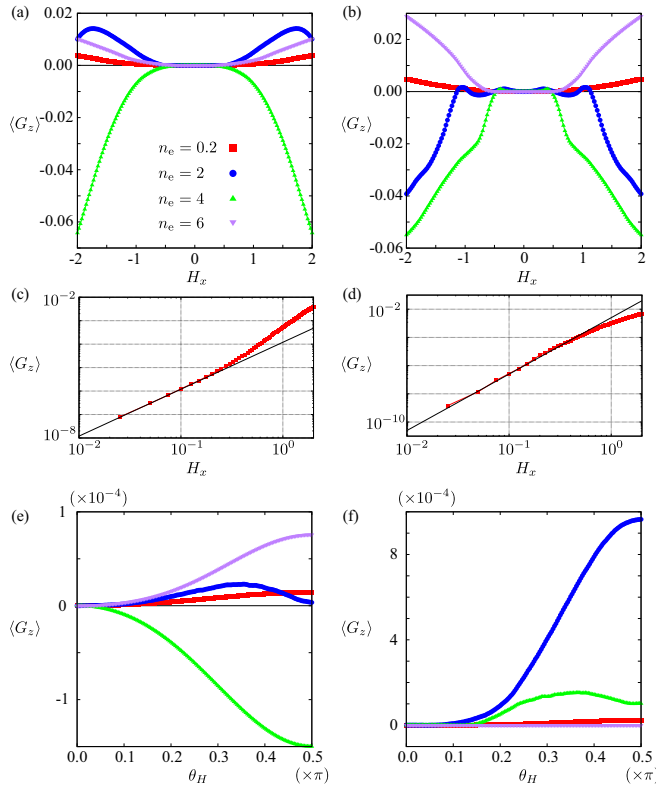


FIG. 2. (a), (b) H_x dependence of the expectation values of G_z , $\langle G_z \rangle$, for several electron fillings n_e at (a) $\lambda = 2$ and (b) $\lambda = 0$. (c), (d) The double logarithmic plot for $H_x > 0$ corresponding to (a) and (b) at $n_e = 0.2$. The black lines are lines proportional to (c) H_x^2 and (d) H_x^4 . (e), (f) θ_H dependence of $\langle G_z \rangle$ at (e) $\lambda = 2$ and (f) $\lambda = 0$ for $H = 0.3$ and several n_e .

vanishes ($\langle \dots \rangle$ stands for the statistical average) [35]. In other words, the dipole component of the electric toroidal multipole is not activated in the model Hamiltonian in Eq. (1) even when it belongs to the totally symmetric irreducible representation for $V \neq 0$.

Meanwhile, we find that $\langle G_z \rangle$ becomes nonzero by introducing an external magnetic field in Eq. (6). Figure 2(a) shows the H_x dependence of $\langle G_z \rangle$ for electron fillings $n_e = 0.2, 2, 4$, and 6; $n_e = 10$ means the full filling. We take the summation of the momentum \mathbf{k} over 360^3 grid points in the first Brillouin zone. The results at $n_e = 0.2$ and 2 are metallic and those at $n_e = 4$ and 6 are insulating. The data show that $\langle G_z \rangle$ becomes nonzero irrespective of metals and insulators for $H_x \neq 0$ at all n_e and its H_x dependence is symmetric. Similarly, such behavior is also found in the case of $\lambda = 0$, as shown in Fig. 2(b), which indicates that the spin-orbit coupling is not important in driving the field-induced electric toroidal dipole. Meanwhile, the H_x dependence of $\langle G_z \rangle$ is different in each case; $\langle G_z \rangle$ is proportional to H_x^2 (H_x^4) for $\lambda \neq 0$ ($\lambda = 0$), as shown in Fig. 2(c) [Fig. 2(d)]. One can analytically obtain such a difference by performing essential-model-parameter calculations in Appendix A [42].

We also show the θ_H dependence of $\langle G_z \rangle$ for $H = 0.3$ at $\lambda = 2$ in Fig. 2(e) and at $\lambda = 0$ in Fig. 2(f). The result indicates that $\langle G_z \rangle$ becomes nonzero except for $\theta_H = 0$, i.e., the z direction. Moreover, one finds that $\langle G_z \rangle$ tends to be

larger as θ_H approaches $\pi/2$. Thus the atomic-scale electric hexadecapole (Q_{4z}^α) that arises from V in Eq. (5) is related to the electric toroidal dipole via the in-plane component of the applied magnetic field.

The above result that $\langle G_z \rangle$ is symmetric against the magnetic field is understood from the time-reversal symmetry. Since the time-reversal parity of G_z is $+1$, it can couple to the even order of the magnetic field, whose time-reversal parity is given as -1 . Meanwhile, when the time-reversal symmetry of the system is broken so as to accommodate the time-reversal odd and spatial-inversion even multipoles, such as the magnetic toroidal monopole and quadrupole, $\langle G_z \rangle$ behaves antisymmetric against the magnetic field [43].

III. UNCONVENTIONAL HALL EFFECT

In this section, we discuss the unconventional Hall effect under the ferroaxial ordering. We first discuss the general symmetry condition of the Hall effect in Sec. III A. Next, we examine the numerical results by comparing the conventional Hall effect without the ferroaxial moment in Sec. III B.

A. Hall conductivity tensor

The Hall conductivity tensor $\sigma_{\mu\nu;\eta}^H$ is defined by

$$J_\mu = \sum_{\nu\eta} \sigma_{\mu\nu;\eta}^H E_\nu H_\eta, \quad (11)$$

where J_μ , E_ν , and H_η are electric current, electric field, and magnetic field for $\mu, \nu, \eta = (x, y, z)$, respectively. Since $\sigma_{\mu\nu;\eta}^H$ corresponds to the antisymmetric part of the linear conductivity tensor proportional to H , it satisfies the relation of $\sigma_{\mu\nu;\eta}^H = -\sigma_{\nu\mu;\eta}^H$ from the Onsager reciprocal relations. Then, there are nine independent tensor components in $\sigma_{\mu\nu;\eta}^H$, which are represented by

$$\begin{aligned} \sigma^H &= \begin{pmatrix} \sigma_{yz;x}^H & \sigma_{yz;y}^H & \sigma_{yz;z}^H \\ \sigma_{zx;x}^H & \sigma_{zx;y}^H & \sigma_{zx;z}^H \\ \sigma_{xy;x}^H & \sigma_{xy;y}^H & \sigma_{xy;z}^H \end{pmatrix} \\ &= \begin{pmatrix} Q_0 - Q_u + Q_v & Q_{xy} + G_z & Q_{zx} - G_y \\ Q_{xy} - G_z & Q_0 - Q_u - Q_v & Q_{yz} + G_x \\ Q_{zx} + G_y & Q_{yz} - G_x & Q_0 + 2Q_u \end{pmatrix}. \end{aligned} \quad (12)$$

Here, the second line of Eq. (12) describes the component of σ^H in terms of multipoles. Since the antisymmetric $\sigma_{\mu\nu;\eta}^H$ is the second-rank polar tensor, the relevant multipoles are the rank 0–2 time-reversal even multipoles, i.e., electric monopole Q_0 , electric toroidal dipole (G_x, G_y, G_z), and electric quadrupole ($Q_u, Q_v, Q_{yz}, Q_{zx}, Q_{xy}$) [29]. Once any of the multipoles belong to the totally symmetric irreducible representation under the point group we focus on, the corresponding tensor component can be nonzero.

In the case of the point group D_{4h} , Q_0 and Q_u belong to the totally symmetric irreducible representation. Then, the

nonzero tensor component is represented by

$$\sigma^H(D_{4h}) = \begin{pmatrix} Q_0 - Q_u & 0 & 0 \\ 0 & Q_0 - Q_u & 0 \\ 0 & 0 & Q_0 + 2Q_u \end{pmatrix}. \quad (13)$$

The induced component corresponds to the conventional Hall effect, where the transverse current occurs perpendicular to both the input electric field and the magnetic field.

Meanwhile, when the symmetry is lowered from D_{4h} to C_{4h} so that the ferroaxial moment is induced, the component corresponding to the electric toroidal dipole G_z can be nonzero. The additional tensor component is represented by

$$\sigma^H(\text{FAO}) = \begin{pmatrix} 0 & G_z & 0 \\ -G_z & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \quad (14)$$

Thus $\sigma_{zx}^H = -\sigma_{yz}^H$ is expected under the ferroaxial ordering. Both σ_{zx}^H and σ_{yz}^H mean the transverse conductivity under the magnetic field perpendicular to the ferroaxial moment; all J_μ , E_ν , and H_η lie in the same plane, which corresponds to the unconventional Hall effect [44] rather than the planar Hall effect [45–48].

The emergence of the unconventional Hall effect under the ferroaxial ordering is intuitively understood from the nature of the ferroaxial moment as a nanometric rotator that causes the transverse responses between the conjugate physical quantities [31,35]. We consider the case of σ_{zx}^H as an example. When the magnetic field is applied in the x direction, the system exhibits the magnetization along the perpendicular y direction in addition to the parallel x direction [36]. Such an induced magnetization along the y axis can contribute to the ordinary Hall effect in the zx plane. In the end, one can find the emergence of the unconventional Hall effect σ_{zx}^H under the ferroaxial ordering. In addition, one finds that σ_{zx}^H is an odd function of the magnetic field owing to the time-reversal parity of the ferroaxial ordering. In the following, we focus on the behavior of σ_{zx}^H induced by the ferroaxial ordering.

B. Numerical results

We compute $\sigma_{\mu\nu}^H(H_\eta)$ by using the Kubo formula within the linear response theory [49] as

$$\sigma_{\mu\nu}^H(H_\eta) = \frac{e^2}{\hbar} \frac{1}{i\bar{V}} \sum_{m,n,k} \frac{f(\epsilon_{nk}) - f(\epsilon_{mk})}{\epsilon_{nk} - \epsilon_{mk}} \frac{J_{\mu k}^{nm} J_{\nu k}^{mn}}{\epsilon_{nk} - \epsilon_{mk} + i\delta}, \quad (15)$$

where e is the elementary charge, $\hbar = h/(2\pi)$ is the reduced Planck constant, \bar{V} is the system volume, δ is the broadening factor, $f(\epsilon)$ is the Fermi distribution function, and $J_{\mu k}^{nm} = \langle n\mathbf{k} | J_\mu | m\mathbf{k} \rangle$ is the matrix element of the current operator in the direction $\mu = (x, y, z)$, $J_\mu \cdot \epsilon_{mk}$ and $|m\mathbf{k}\rangle$ are the m th eigenvalue and eigenstate of \mathcal{H} , respectively; we include the effect of an external magnetic field as the Zeeman coupling in \mathcal{H} (η represents the magnetic-field direction) and hence the higher-order contribution of \mathbf{H} is included in the conductivity tensor. We set $e^2/\hbar = 1$, $\delta = 0.01$, and temperature $T = 0.001$. The summation of \mathbf{k} is taken over the 360^3 grid points in the Brillouin zone.

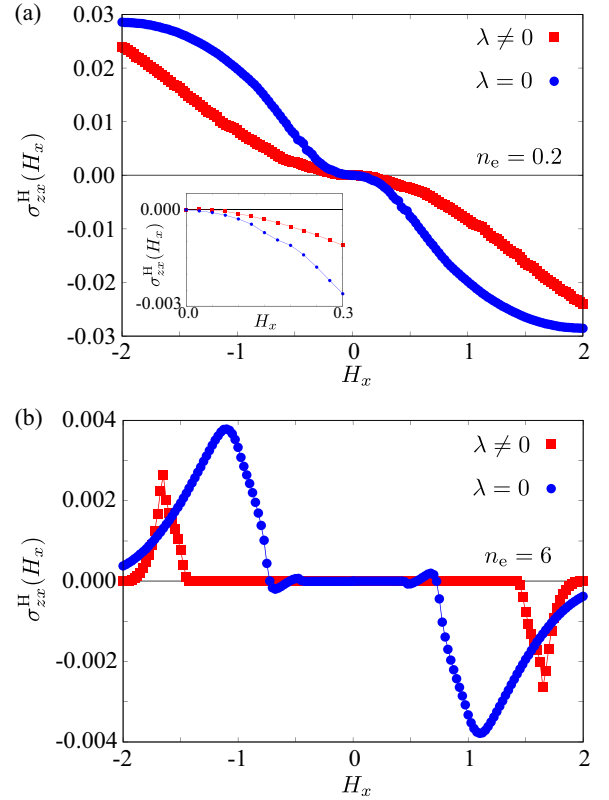


FIG. 3. H_x dependence of the unconventional Hall conductivity $\sigma_{zx}^H(H_x)$ at $\lambda = 0$ and 2 for (a) $n_e = 0.2$ and (b) $n_e = 6$. The inset of (a) represents $\sigma_{zx}^H(H_x)$ for small positive H_x .

Figure 3(a) shows the H_x dependence of $\sigma_{zx}^H(H_x)$ at $\lambda = 0$ and 2 for $n_e = 0.2$. As expected from the symmetry consideration in Sec. III A, $\sigma_{zx}^H(H_x)$ becomes nonzero and is an odd function of H_x , as the conventional Hall effect. We also confirm the relation of $\sigma_{zx}^H(H_x) = -\sigma_{yz}^H(H_y)$, although $\sigma_{zx}^H(H_x)$ is not linear to H_x for small H_x , as shown in the inset of Fig. 3(a); this might be ascribed to the higher-order effect in terms of H_x included in Eq. (15). Furthermore, one finds that the spin-orbit coupling λ does not play an intrinsic role in inducing $\sigma_{zx}^H(H_x)$, as shown in the comparison of the results at $\lambda = 0$ and 2. Since a larger Hall response is obtained for larger $|H_x|$, a larger magnetic field is preferred in order to detect it in experiments. Note that the interband process with $\epsilon_{mk} \neq \epsilon_{nk}$ in Eq. (15) contributes to $\sigma_{zx}^H(H_x)$.

In order to analytically obtain the essential model parameters to cause $\sigma_{zx}^H(H_x)$, we perform an expansion method to $\sigma_{zx}^H(H_x)$ [42]. As a result, we find that $\sigma_{zx}^H(H_x)$ is proportional to $VH_x^{2m+1}g(t_{z2\sigma}, t_{z2\pi})$; m is integer and $g(t_{z2\sigma}, t_{z2\pi})$ is an appropriate function depending on $t_{z2\sigma}$ and $t_{z2\pi}$; $g(0, 0) = 0$, which is consistent with both the symmetry and the numerical results. From the above expression, we find that the next-nearest-neighbor hopping along the z direction ($t_{z2\sigma}, t_{z2\pi}$) is important to obtain nonzero $\sigma_{zx}^H(H_x)$. The details of the expansion method are presented in Appendix A.

A similar behavior is found in the insulating case at $n_e = 6$, as shown in Fig. 3(b). Although $\sigma_{zx}^H(H_x) = 0$ for $|H_x| \lesssim 1.4$ at $\lambda = 2$ and for $|H_x| \lesssim 0.45$ at $\lambda = 0$ in the insulating region, H_x -odd behavior becomes prominent once the band gap is

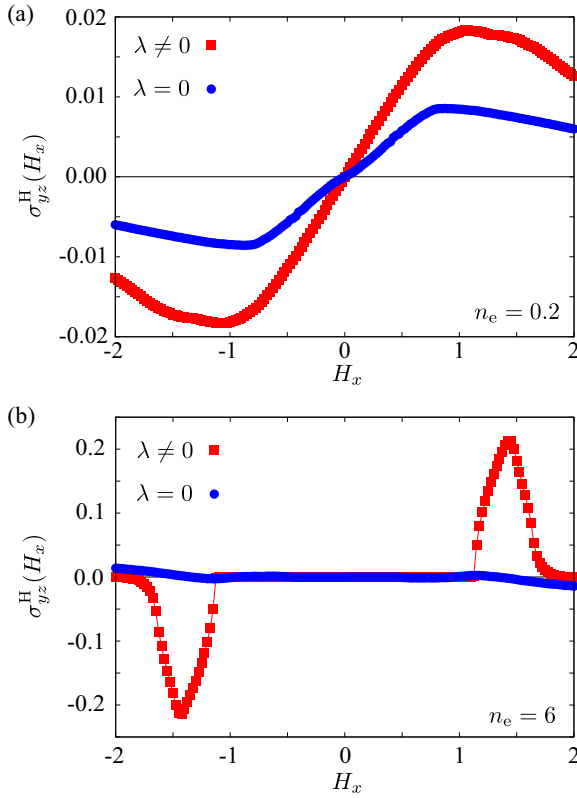


FIG. 4. H_x dependence of the conventional Hall conductivity $\sigma_{yz}^H(H_x)$ at $\lambda = 0$ and 2 for (a) $n_e = 0.2$ and (b) $n_e = 6$.

closed by a larger magnetic field. In other words, it is difficult to observe the Hall response in the insulating case in experiments, since the large magnetic field comparable to the band gap is necessary; such an issue is evaded by considering the optical Hall conductivity by applying an ac electric field instead of a dc one. We also present the result of the optical unconventional Hall conductivity, which can give a signal of ferroaxial ordering in the insulating case, in Appendix B.

We also calculate the ordinary nonplanar Hall conductivity $\sigma_{yz}^H(H_x)$ while the field direction is kept. Figures 4(a) and 4(b)

show the H_x dependence of $\sigma_{yz}^H(H_x)$ at $n_e = 0.2$ and $n_e = 6$, respectively. As compared to the result of the unconventional Hall effect in Fig. 3, the behaviors are similar to each other; $\sigma_{yz}^H(H_x)$ becomes nonzero for $H_x \neq 0$ and an odd function of H_x . On the other hand, we find the difference in their essential model parameters; the unconventional Hall conductivity $\sigma_{zx}^H(H_x)$ needs the CEF parameter V , while the conventional Hall conductivity $\sigma_{yz}^H(H_x)$ does not, as detailed in Appendix A.

IV. MAGNETOCONDUCTIVITY

We discuss the magnetoconductivity under the ferroaxial ordering. Similar to the unconventional Hall effect in Sec. III, we show the symmetry condition and the relation to the multipoles in Sec. IV A. Then, we compute the magnetoconductivity tensor based on the Kubo formula in Sec. IV B.

A. Magnetoconductivity tensor

The magnetoconductivity tensor corresponding to the rank-4 polar tensor is defined by

$$J_\mu = \sum_{\nu\eta\gamma} \sigma_{\mu\nu;\eta\gamma}^{\text{MC}} E_\nu H_\eta H_\gamma, \quad (16)$$

where $\sigma_{\mu\nu;\eta\gamma}^{\text{MC}}$ is the symmetric tensor as $\sigma_{\mu\nu;\eta\gamma}^{\text{MC}} = \sigma_{\nu\mu;\eta\gamma}^{\text{MC}} = \sigma_{\mu\nu;\gamma\eta}^{\text{MC}}$. The independent 36 components in $\sigma_{\mu\nu;\eta\gamma}^{\text{MC}}$ are represented by the 6×6 matrix as follows:

$$\sigma^{\text{MC}} = \begin{pmatrix} \sigma_{xx;xx}^{\text{MC}} & \sigma_{xx;yy}^{\text{MC}} & \sigma_{xx;zz}^{\text{MC}} & \sigma_{xx;yz}^{\text{MC}} & \sigma_{xx;zx}^{\text{MC}} & \sigma_{xx;xy}^{\text{MC}} \\ \sigma_{yy;xx}^{\text{MC}} & \sigma_{yy;yy}^{\text{MC}} & \sigma_{yy;zz}^{\text{MC}} & \sigma_{yy;yz}^{\text{MC}} & \sigma_{yy;zx}^{\text{MC}} & \sigma_{yy;xy}^{\text{MC}} \\ \sigma_{zz;xx}^{\text{MC}} & \sigma_{zz;yy}^{\text{MC}} & \sigma_{zz;zz}^{\text{MC}} & \sigma_{zz;yz}^{\text{MC}} & \sigma_{zz;zx}^{\text{MC}} & \sigma_{zz;xy}^{\text{MC}} \\ \sigma_{yz;xx}^{\text{MC}} & \sigma_{yz;yy}^{\text{MC}} & \sigma_{yz;zz}^{\text{MC}} & \sigma_{yz;yz}^{\text{MC}} & \sigma_{yz;zx}^{\text{MC}} & \sigma_{yz;xy}^{\text{MC}} \\ \sigma_{zx;xx}^{\text{MC}} & \sigma_{zx;yy}^{\text{MC}} & \sigma_{zx;zz}^{\text{MC}} & \sigma_{zx;yz}^{\text{MC}} & \sigma_{zx;zx}^{\text{MC}} & \sigma_{zx;xy}^{\text{MC}} \\ \sigma_{xy;xx}^{\text{MC}} & \sigma_{xy;yy}^{\text{MC}} & \sigma_{xy;zz}^{\text{MC}} & \sigma_{xy;yz}^{\text{MC}} & \sigma_{xy;zx}^{\text{MC}} & \sigma_{xy;xy}^{\text{MC}} \end{pmatrix} \\ = \sigma^{\text{MC(M)}} + \sigma^{\text{MC(D)}} + \sigma^{\text{MC(Q)}} + \sigma^{\text{MC(O)}} + \sigma^{\text{MC(H)}}, \quad (17)$$

where $\sigma^{\text{MC(M)}}$, $\sigma^{\text{MC(D)}}$, $\sigma^{\text{MC(Q)}}$, $\sigma^{\text{MC(O)}}$, and $\sigma^{\text{MC(H)}}$ denote the monopole, dipole, quadrupole, octupole, and hexadecapole components in σ^{MC} . The correspondence between the tensor components and multipoles is given by

$$\sigma^{\text{MC(M)}} = \begin{pmatrix} 4Q_0 + Q'_0 & -2Q_0 + Q'_0 & -2Q_0 + Q'_0 & 0 & 0 & 0 \\ -2Q_0 + Q'_0 & 4Q_0 + Q'_0 & -2Q_0 + Q'_0 & 0 & 0 & 0 \\ -2Q_0 + Q'_0 & -2Q_0 + Q'_0 & 4Q_0 + Q'_0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 3Q_0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 3Q_0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 3Q_0 \end{pmatrix}, \quad (18)$$

$$\sigma^{\text{MC(D)}} = \begin{pmatrix} 0 & 0 & 0 & 0 & -2G_y & 2G_z \\ 0 & 0 & 0 & 2G_x & 0 & -2G_z \\ 0 & 0 & 0 & -2G_x & 2G_y & 0 \\ 0 & -2G_x & 2G_x & 0 & -G_z & G_y \\ 2G_y & 0 & -2G_y & G_z & 0 & -G_x \\ -2G_z & 2G_z & 0 & -G_y & G_x & 0 \end{pmatrix}, \quad (19)$$

$$\sigma^{\text{MC(Q)}} = \begin{pmatrix} \tilde{Q}_u + \tilde{Q}_v & -\tilde{Q}_u^{(+)} - \tilde{Q}_u^{(-)} + 2\tilde{Q}_v^{(-)} & \tilde{Q}_u^{(+)} + \tilde{Q}_v^{(-)} & \tilde{Q}_{yz}^{(+)} & \tilde{Q}'_{zx}^{(+)} & \tilde{Q}'_{xy}^{(+)} \\ -\tilde{Q}_u^{(+)} - \tilde{Q}_u^{(-)} + 2\tilde{Q}_v^{(+)} & \tilde{Q}_u - \tilde{Q}_v & \tilde{Q}_u^{(+)} - \tilde{Q}_v^{(-)} & \tilde{Q}'_{yz}^{(+)} & \tilde{Q}_{zx}^{(+)} & \tilde{Q}'_{xy}^{(+)} \\ \tilde{Q}_u^{(-)} + \tilde{Q}_v^{(+)} & \tilde{Q}_u^{(-)} - \tilde{Q}_v^{(+)} & -2\tilde{Q}_u & \tilde{Q}'_{yz}^{(+)} & \tilde{Q}'_{zx}^{(+)} & \tilde{Q}'_{xy}^{(+)} \\ \tilde{Q}_{yz}^{(-)} & \tilde{Q}'_{yz}^{(-)} & \tilde{Q}'_{yz}^{(-)} & 3Q_u - 3Q_v & 3Q_{xy} & 3Q_{zx} \\ \tilde{Q}_{zx}^{(-)} & \tilde{Q}'_{zx}^{(-)} & \tilde{Q}'_{zx}^{(-)} & 3Q_{xy} & 3Q_u + 3Q_v & 3Q_{yz} \\ \tilde{Q}'_{xy}^{(-)} & \tilde{Q}'_{xy}^{(-)} & \tilde{Q}'_{xy}^{(-)} & 3Q_{zx} & 3Q_{yz} & -6Q_u \end{pmatrix}, \quad (20)$$

$$\sigma^{\text{MC(O)}} = \begin{pmatrix} 0 & G_{xyz} & -G_{xyz} & -2G_x^\beta & G_y^\alpha + G_y^\beta & -G_z^\alpha + G_z^\beta \\ -G_{xyz} & 0 & G_{xyz} & -G_x^\alpha + G_x^\beta & -2G_y^\beta & G_z^\alpha + G_z^\beta \\ G_{xyz} & -G_{xyz} & 0 & G_x^\alpha + G_x^\beta & -G_y^\alpha + G_y^\beta & -2G_z^\beta \\ 2G_x^\beta & G_x^\alpha - G_x^\beta & -G_x^\alpha - G_x^\beta & 0 & -2G_z^\alpha & 2G_y^\alpha \\ -G_y^\alpha - G_y^\beta & 2G_y^\beta & G_y^\alpha - G_y^\beta & 2G_z^\alpha & 0 & -2G_x^\alpha \\ G_z^\alpha - G_z^\beta & -G_z^\alpha - G_z^\beta & 2G_z^\beta & -2G_y^\alpha & 2G_x^\alpha & 0 \end{pmatrix}, \quad (21)$$

$$\sigma^{\text{MC(H)}} = \begin{pmatrix} 2Q_4 - Q_{4u} + Q_{4v} & -Q_4 + 2Q_{4u} & -Q_4 - Q_{4u} - Q_{4v} & 2Q_{4x}^\beta & -Q_{4y}^\alpha - Q_{4y}^\beta & Q_{4z}^\alpha - Q_{4z}^\beta \\ -Q_4 + 2Q_{4u} & 2Q_4 - Q_{4u} - Q_{4v} & -Q_4 - Q_{4u} + Q_{4v} & Q_{4x}^\alpha - Q_{4x}^\beta & 2Q_{4y}^\beta & -Q_{4z}^\alpha - Q_{4z}^\beta \\ -Q_4 - Q_{4u} - Q_{4v} & -Q_4 - Q_{4u} + Q_{4v} & 2Q_4 + 2Q_{4u} & -Q_{4x}^\alpha - Q_{4x}^\beta & Q_{4y}^\alpha - Q_{4y}^\beta & 2Q_{4z}^\beta \\ 2Q_{4x}^\beta & Q_{4x}^\alpha - Q_{4x}^\beta & -Q_{4x}^\alpha - Q_{4x}^\beta & -Q_4 - Q_{4u} + Q_{4v} & 2Q_{4z}^\beta & 2Q_{4y}^\beta \\ -Q_{4y}^\alpha - Q_{4y}^\beta & 2Q_{4y}^\beta & Q_{4y}^\alpha - Q_{4y}^\beta & 2Q_{4z}^\beta & -Q_4 - Q_{4u} - Q_{4v} & 2Q_{4x}^\beta \\ Q_{4z}^\alpha - Q_{4z}^\beta & -Q_{4z}^\alpha - Q_{4z}^\beta & 2Q_{4z}^\beta & 2Q_{4y}^\beta & 2Q_{4x}^\beta & -Q_4 + 2Q_{4u} \end{pmatrix}, \quad (22)$$

where

$$\begin{aligned} (\tilde{Q}_u, \tilde{Q}_u^{(\pm)}) &\equiv (-4Q_u - 2Q_u^{(+)}, -4Q_u + Q_u^{(+)} \pm 3Q_u^{(-)}), \\ (\tilde{Q}_v, \tilde{Q}_v^{(\pm)}) &\equiv (4Q_v + 2Q_v^{(+)}, -4Q_v + Q_v^{(+)} \pm Q_v^{(-)}), \\ (\tilde{Q}_\zeta^{(\pm)}, \tilde{Q}'_{\zeta}^{(\pm)}) &\equiv (-4Q_\zeta + Q_\zeta^{(+)} \pm Q_\zeta^{(-)}, 2Q_\zeta + Q_\zeta^{(+)} \pm Q_\zeta^{(-)}), \end{aligned} \quad (23)$$

for $\zeta = (yz, zx, xy)$. Thus rank-0–4 multipoles can contribute to σ^{MC} .

For the point group D_{4h} , the multipoles belonging to the totally symmetric irreducible representation up to rank 4 are Q_0 , Q_u , Q_4 , and Q_{4u} . Then, nonzero components of σ^{MC} under D_{4h} are given by

$$\sigma^{\text{MC}(D_{4h})} = \begin{pmatrix} Q_1 & Q_3 & Q_4 & 0 & 0 & 0 \\ Q_3 & Q_1 & Q_4 & 0 & 0 & 0 \\ Q_5 & Q_5 & Q_2 & 0 & 0 & 0 \\ 0 & 0 & 0 & Q_6 & 0 & 0 \\ 0 & 0 & 0 & 0 & Q_6 & 0 \\ 0 & 0 & 0 & 0 & 0 & Q_7 \end{pmatrix}, \quad (24)$$

where

$$Q_1 = 4Q_0 + Q'_0 + \tilde{Q}_u + 2Q_4 - Q_{4u}, \quad (25)$$

$$Q_2 = 4Q_0 + Q'_0 - 2\tilde{Q}_u + 2Q_4 + 2Q_{4u}, \quad (26)$$

$$Q_3 = -2Q_0 + Q'_0 - \tilde{Q}_u^{(+)} - \tilde{Q}_u^{(-)} - Q_4 + 2Q_{4u}, \quad (27)$$

$$Q_4 = -2Q_0 + Q'_0 + \tilde{Q}_u^{(+)} - Q_4 - Q_{4u}, \quad (28)$$

$$Q_5 = -2Q_0 + Q'_0 + \tilde{Q}_u^{(-)} - Q_4 - Q_{4u}, \quad (29)$$

$$Q_6 = 3Q_0 + 3Q_u - Q_4 - Q_{4u}, \quad (30)$$

$$Q_7 = 3Q_0 - 6Q_u - Q_4 + 2Q_{4u}. \quad (31)$$

There are seven independent components of σ^{MC} .

For the point group C_{4h} , the electric toroidal octupole G_z^α and electric hexadecapole Q_{4z}^α belong to the totally symmetric irreducible representation in addition to the electric toroidal dipole G_z . Then, the additional nonzero components of σ^{MC} are given by

$$\sigma^{\text{MC}(FAO)} = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & G_2 \\ 0 & 0 & 0 & 0 & 0 & -G_2 \\ 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -G_3 & 0 \\ 0 & 0 & 0 & G_3 & 0 & 0 \\ G_1 & -G_1 & 0 & 0 & 0 & 0 \end{pmatrix}, \quad (32)$$

where $G_1 = -(2G_z - G_z^\alpha - Q_{4z}^\alpha)$, $G_2 = 2G_z - G_z^\alpha + Q_{4z}^\alpha$, and $G_3 = G_z + 2G_z^\alpha$. Thus $\sigma^{\text{MC}(FAO)}$ has three independent components: $\sigma_{xy;xx}^{\text{MC}} = -\sigma_{xy;yy}^{\text{MC}}$, $\sigma_{xx;xy}^{\text{MC}} = -\sigma_{yy;xy}^{\text{MC}}$, and $\sigma_{zx;yz}^{\text{MC}} = -\sigma_{yz;zx}^{\text{MC}}$. We focus on $\sigma_{xy;xx}^{\text{MC}}$ in the following calculations, since other components, $\sigma_{xx;xy}^{\text{MC}}$ and $\sigma_{zx;yz}^{\text{MC}}$, also show a similar behavior to $\sigma_{xy;xx}^{\text{MC}}$.

B. Numerical results

We calculate σ^{MC} by using the Kubo formula in Eq. (15), where the effect of the magnetic field is included in the Hamiltonian. In contrast to the unconventional Hall effect, the intraband process proportional to $1/\delta$ contributes to the magnetoconductivity tensor. Figure 5(a) shows the H_x dependence of $\sigma_{xy}^{\text{MC}}(H_x)$ in the metallic ferroaxial ordered state at $n_e = 2$ for $\lambda = 0$ and 2. Similar to the unconventional Hall effect in Sec. III B, $\sigma_{xy}^{\text{MC}}(H_x)$ becomes nonzero for $H_x \neq 0$ irrespective of λ . On the other hand, it is the even function of

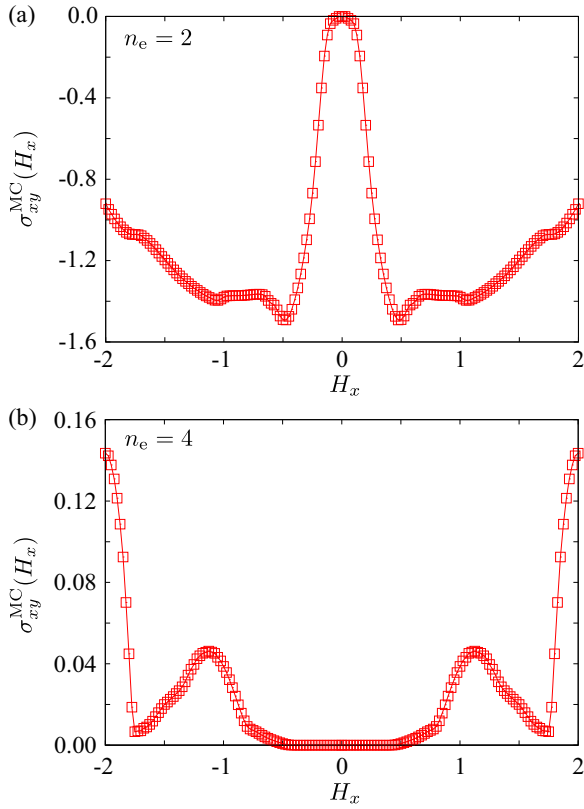


FIG. 5. H_x dependence of σ_{xy}^{MC} at $\lambda = 2$ for (a) $n_e = 2$ and (b) $n_e = 4$.

H_x owing to the opposite time-reversal parity of σ^{H} . Indeed, the essential model parameters of $\sigma_{xy}^{\text{MC}}(H_x)$ are obtained in the form of VH_x^{2m} , as shown in Appendix A. These features are common to the insulating ferroaxial ordered state, as shown in Fig. 5(b).

V. SUMMARY

To summarize, we have investigated metallic ferroaxial ordering, which is characterized by mirror symmetry breaking without the breaking of spatial inversion and time-reversal symmetries, by focusing on its transport property in the external magnetic field. We analyzed a fundamental five d -orbital model to include the electric toroidal dipole, electric toroidal octupole, and electric hexadecapole degrees of freedom corresponding to the ferroaxial moment under the tetragonal lattice structure. As a result, we find three characteristic features under the magnetic field. First, the electric toroidal dipole is induced by the in-plane magnetic field. Second, the unconventional Hall effect is induced as an odd function of the applied magnetic field. Third, the magnetoconductivity as an even function of the applied magnetic field occurs. In all the cases, we derive the essential model parameters out of the

hoppings, spin-orbit coupling, and crystalline electric field. Our results indicate that the metallic ferroaxial ordered state becomes the source of unconventional magnetotransport phenomena in which the spin-orbit coupling is not necessary; it is noted that similar transport phenomena can be also expected in the insulating ferroaxial ordered state by applying an ac electric field. The present transport property can be detected in various materials without mirror symmetry parallel to the ferroaxial moment, such as $\text{Co}_3\text{Nb}_2\text{O}_8$ [9], $\text{CaMn}_7\text{O}_{12}$ [10], $\text{RbFe}(\text{MoO}_4)_2$ [11,12], NiTiO_3 [12–15], $\text{Ca}_5\text{Ir}_3\text{O}_{12}$ [16–18], BaCoSiO_4 [20], $\text{K}_2\text{Zr}(\text{PO}_4)_2$ [21], $\text{Na}_2\text{Hf}(\text{BO}_3)_2$ [22], and Na-superionic conductors [23]. By using tight-binding model parameters obtained from density functional theory (DFT) calculations, one can quantitatively evaluate the transport tensors in these materials. Since such model parameters were obtained for $\text{Ca}_5\text{Ir}_3\text{O}_{12}$ [38], it is intriguing to examine the transport behavior in the ferroaxial system, which is left for future study.

ACKNOWLEDGMENTS

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APPENDIX A: ESSENTIAL MODEL PARAMETERS

1. Hamiltonian in terms of electronic multipoles

In this section, we present the model Hamiltonian expressed as the linear combination of the symmetry-adapted multipole basis (SAMB) [41,50], which enables us to clarify the hidden multipole degrees of freedom in the present system. First, we introduce the atomic multipole basis defined in the spinful d orbitals at a single site and the momentum multipole basis defined as the function of the wave vector \mathbf{k} [28,29,51]. Then, we show that the SAMB is decomposed as the linear combination of the products of these two multipole bases.

a. Atomic multipole basis

Let us first introduce the atomic multipole basis within the spinful d orbitals. There are 25 (75) independent spinless (spinful) atomic multipoles. Here, we only show the multipoles that appeared in the Hamiltonian and some relevant ones discussed in the main text, Q_{4z}^α , Q_{4z}^β , G_z , and G_z^α . The explicit expressions of these multipoles are summarized in Table II and their matrix elements in the five d orbitals (d_u , d_v , d_{yz} , d_{zx} , d_{xy}) are given by

TABLE II. Operator expressions of the atomic multipole basis in the point group D_{4h} . Only the multipoles appearing in the Hamiltonian and some relevant ones are summarized. E, M, ET, and MT stand for electric, magnetic, electric toroidal, and magnetic toroidal, respectively. The superscript (a) denotes the atomic multipole. \mathbf{l} and $\boldsymbol{\sigma}/2$ represent the dimensionless orbital and spin angular-momentum operators. The upper and lower parts separated by the horizontal line represent the spinless and spinful multipoles, respectively.

Rank	Type	Irrep	Symbol	Expression
0	E	A_{1g}^+	$Q_0^{(a)}$	1
1	M	A_{2g}^-	$M_z^{(a)}$	l_z
	M	E_g^-	$M_x^{(a)}, M_y^{(a)}$	l_x, l_y
2	E	A_{1g}^+	$Q_u^{(a)}$	$3z^2 - r^2$
	E	B_{1g}^+	$Q_v^{(a)}$	$x^2 - y^2$
	E	E_g^+	$Q_{yz}^{(a)}, Q_{zx}^{(a)}$	yz, zx
	E	B_{2g}^+	$Q_{xy}^{(a)}$	xy
3	M	E_g^-	$M_{3a}^{(a)}, M_{3b}^{(a)}$	$(x^2 - y^2)l_x - 2xy l_y, 2xy l_x + (x^2 - y^2)l_y$
	M	E_g^-	$M_{3u}^{(a)}, M_{3v}^{(a)}$	$(5z^2 - r^2)l_x + 2x(5z l_z - \mathbf{r} \cdot \mathbf{l}), (5z^2 - r^2)l_y + 2y(5z l_z - \mathbf{r} \cdot \mathbf{l})$
4	E	A_{1g}^+	$Q_4^{(a)}$	$x^4 - 3x^2y^2 - 3x^2z^2 + y^4 - 3y^2z^2 + z^4$
	E	A_{1g}^+	$Q_{4u}^{(a)}$	$-(x^4 - 12x^2y^2 + 6x^2z^2 + y^4 + 6y^2z^2 - 2z^4)$
	E	B_{1g}^+	$Q_{4v}^{(a)}$	$(x^2 - y^2)(x^2 + y^2 - 6z^2)$
	E	A_{2g}^+	$Q_{4z}^{\alpha(a)}$	$xy(x^2 - y^2)$
	E	B_{2g}^+	$Q_{4z}^{\beta(a)}$	$xy(7z^2 - r^2)$
	E	E_g^+	$Q_{4x}^{\alpha(a)}, Q_{4y}^{\alpha(a)}$	$yz(y^2 - z^2), zx(z^2 - x^2)$
	E	E_g^+	$Q_{4x}^{\beta(a)}, Q_{4y}^{\beta(a)}$	$yz(7x^2 - r^2), zx(7y^2 - r^2)$
0	E	A_{1g}^+	$Q_0^{(a)}$	$\mathbf{l} \cdot \boldsymbol{\sigma}$
1	ET	A_{2g}^+	$G_z^{(a)}$	$l_x \sigma_y - l_y \sigma_x$
3	ET	A_{2g}^+	$G_z^{\alpha(a)}$	$M_{3u}^{(a)} \sigma_y - M_{3v}^{(a)} \sigma_x$
4	E	A_{2g}^+	$Q_{4z}^{\alpha(a)}$	$M_{3a}^{(a)} \sigma_y + M_{3b}^{(a)} \sigma_x$

$$Q_0^{(a)} = \frac{1}{\sqrt{10}} \begin{pmatrix} 1 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix} \sigma_0,$$

$$M_z^{(a)} = \frac{\sqrt{5}}{10} \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -2i \\ 0 & 0 & 0 & i & 0 \\ 0 & 0 & -i & 0 & 0 \\ 0 & 2i & 0 & 0 & 0 \end{pmatrix} \sigma_0, \quad M_x^{(a)} = \frac{\sqrt{5}}{10} \begin{pmatrix} 0 & 0 & \sqrt{3}i & 0 & 0 \\ 0 & 0 & i & 0 & 0 \\ -\sqrt{3}i & -i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & i \\ 0 & 0 & 0 & -i & 0 \end{pmatrix} \sigma_0,$$

$$M_y^{(a)} = \frac{\sqrt{5}}{10} \begin{pmatrix} 0 & 0 & 0 & -\sqrt{3}i & 0 \\ 0 & 0 & 0 & i & 0 \\ 0 & 0 & 0 & 0 & -i \\ \sqrt{3}i & -i & 0 & 0 & 0 \\ 0 & 0 & i & 0 & 0 \end{pmatrix} \sigma_0,$$

$$Q_u^{(a)} = \frac{\sqrt{7}}{14} \begin{pmatrix} 2 & 0 & 0 & 0 & 0 \\ 0 & -2 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & -2 \end{pmatrix} \sigma_0, \quad Q_v^{(a)} = \frac{\sqrt{7}}{14} \begin{pmatrix} 0 & -2 & 0 & 0 & 0 \\ -2 & 0 & 0 & 0 & 0 \\ 0 & 0 & -\sqrt{3} & 0 & 0 \\ 0 & 0 & 0 & \sqrt{3} & 0 \\ 0 & 0 & 0 & 0 & 0 \end{pmatrix} \sigma_0,$$

$$Q_{yz}^{(a)} = \frac{\sqrt{7}}{14} \begin{pmatrix} 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & -\sqrt{3} & 0 & 0 \\ 1 & -\sqrt{3} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & \sqrt{3} \\ 0 & 0 & 0 & \sqrt{3} & 0 \end{pmatrix} \sigma_0, \quad Q_{zx}^{(a)} = \frac{\sqrt{7}}{14} \begin{pmatrix} 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & \sqrt{3} & 0 \\ 0 & 0 & 0 & 0 & \sqrt{3} \\ 1 & \sqrt{3} & 0 & 0 & 0 \\ 0 & 0 & \sqrt{3} & 0 & 0 \end{pmatrix} \sigma_0,$$

$$\begin{aligned}
Q_{xy}^{(a)} &= \frac{\sqrt{7}}{14} \begin{pmatrix} 0 & 0 & 0 & 0 & -2 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \sqrt{3} & 0 \\ 0 & 0 & \sqrt{3} & 0 & 0 \\ -2 & 0 & 0 & 0 & 0 \end{pmatrix} \sigma_0, \\
M_{3a}^{(a)} &= \frac{\sqrt{2}}{4} \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -i & 0 & 0 \\ 0 & i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & i \\ 0 & 0 & 0 & -i & 0 \end{pmatrix} \sigma_0, & M_{3b}^{(a)} &= \frac{\sqrt{2}}{4} \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & i & 0 \\ 0 & 0 & 0 & 0 & i \\ 0 & -i & 0 & 0 & 0 \\ 0 & 0 & -i & 0 & 0 \end{pmatrix} \sigma_0, \\
M_{3u}^{(a)} &= \frac{\sqrt{10}}{20} \begin{pmatrix} 0 & 0 & 2i & 0 & 0 \\ 0 & 0 & -\sqrt{3}i & 0 & 0 \\ -2i & \sqrt{3}i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -\sqrt{3}i \\ 0 & 0 & 0 & \sqrt{3}i & 0 \end{pmatrix} \sigma_0, & M_{3v}^{(a)} &= \frac{\sqrt{10}}{20} \begin{pmatrix} 0 & 0 & 0 & -2i & 0 \\ 0 & 0 & 0 & -\sqrt{3}i & 0 \\ 0 & 0 & 0 & 0 & \sqrt{3}i \\ 2i & \sqrt{3}i & 0 & 0 & 0 \\ 0 & 0 & -\sqrt{3}i & 0 & 0 \end{pmatrix} \sigma_0, \\
Q_4^{(a)} &= \frac{\sqrt{15}}{30} \begin{pmatrix} 3 & 0 & 0 & 0 & 0 \\ 0 & 3 & 0 & 0 & 0 \\ 0 & 0 & -2 & 0 & 0 \\ 0 & 0 & 0 & -2 & 0 \\ 0 & 0 & 0 & 0 & -2 \end{pmatrix} \sigma_0, & Q_{4u}^{(a)} &= \frac{\sqrt{21}}{42} \begin{pmatrix} 3 & 0 & 0 & 0 & 0 \\ 0 & -3 & 0 & 0 & 0 \\ 0 & 0 & -2 & 0 & 0 \\ 0 & 0 & 0 & -2 & 0 \\ 0 & 0 & 0 & 0 & 4 \end{pmatrix} \sigma_0, \\
Q_{4v}^{(a)} &= \frac{\sqrt{7}}{14} \begin{pmatrix} 0 & -\sqrt{3} & 0 & 0 & 0 \\ -\sqrt{3} & 0 & 0 & 0 & 0 \\ 0 & 0 & 2 & 0 & 0 \\ 0 & 0 & 0 & -2 & 0 \\ 0 & 0 & 0 & 0 & 0 \end{pmatrix} \sigma_0, \\
Q_{4z}^{\alpha(a)} &= \frac{1}{2} \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \end{pmatrix} \sigma_0, \\
Q_0^{(a)} &= \frac{1}{\sqrt{3}} M^{(a)} \cdot \boldsymbol{\sigma}, \\
G_z^{(a)} &= \frac{1}{\sqrt{2}} (M_x^{(a)} \sigma_y - M_y^{(a)} \sigma_x), & G_z^{\alpha(a)} &= \frac{1}{\sqrt{2}} (M_{3u}^{(a)} \sigma_y - M_{3v}^{(a)} \sigma_x), & Q_{4z}^{\alpha(a)} &= \frac{1}{\sqrt{2}} (M_{3a}^{(a)} \sigma_y + M_{3b}^{(a)} \sigma_x), \tag{A1}
\end{aligned}$$

where the superscript (a) denotes the atomic multipole, σ_i ($i = x, y, z$) are the Pauli matrices, and σ_0 is the 2×2 identity matrix in spin space. In this Appendix, these atomic multipoles are normalized as $\text{Tr}(X_i^{(a)} Y_j^{(a)}) = \delta_{XY} \delta_{ij}$ ($X, Y = Q/M/G$).

b. Momentum multipole basis

To decompose the real and imaginary parts of the nearest-neighbor hopping in the xy plane, we introduce the electric and magnetic toroidal momentum multipole bases as

$$A_{1g}^+ : Q_0^{(1)}(\mathbf{k}) = \cos(k_x) + \cos(k_y), \tag{A2}$$

$$B_{1g}^+ : Q_v^{(1)}(\mathbf{k}) = \cos(k_x) - \cos(k_y), \tag{A3}$$

$$E_u^- : T_x^{(1)}(\mathbf{k}) = \sqrt{2} \sin(k_x), \quad T_y^{(1)}(\mathbf{k}) = \sqrt{2} \sin(k_y), \tag{A4}$$

where the superscript (1) denotes the nearest-neighbor hopping within the xy plane.

Similarly, the electric and magnetic toroidal momentum multipole bases for the nearest-neighbor hoppings along the z direction are given by

$$A_{1g}^+ : Q_0^{(2)}(\mathbf{k}) = \sqrt{2} \cos(k_z), \tag{A5}$$

$$A_{2u}^- : T_z^{(2)}(\mathbf{k}) = \sqrt{2} \sin(k_z), \tag{A6}$$

and those for the next-nearest-neighbor hoppings along the z direction are given by

$$A_{1g}^+ : Q_0^{(3)}(\mathbf{k}) = \sqrt{2} [\cos(k_x) + \cos(k_y)] \cos(k_z), \tag{A7}$$

$$B_{1g}^+ : Q_v^{(3)}(\mathbf{k}) = \sqrt{2} [\cos(k_x) - \cos(k_y)] \cos(k_z), \tag{A8}$$

$$E_g^+ : Q_{yz}^{(3)}(\mathbf{k}) = 2 \sin(k_y) \sin(k_z),$$

$$Q_{zx}^{(3)}(\mathbf{k}) = 2 \sin(k_z) \sin(k_x), \tag{A9}$$

$$A_{2u}^- : T_z^{(3)}(\mathbf{k}) = \sqrt{2}[\cos(k_x) + \cos(k_y)] \sin(k_z), \quad (\text{A10})$$

$$B_{2u}^- : T_{3v}^{(3)}(\mathbf{k}) = \sqrt{2}[\cos(k_x) - \cos(k_y)] \sin(k_z), \quad (\text{A11})$$

$$\begin{aligned} E_u^- : T_x^{(3)}(\mathbf{k}) &= 2 \sin(k_x) \cos(k_z), \\ T_y^{(3)}(\mathbf{k}) &= 2 \sin(k_y) \cos(k_z), \end{aligned} \quad (\text{A12})$$

where the superscripts (2) and (3) represent the nearest-neighbor and next-nearest-neighbor hoppings along the z direction. The momentum multipoles are normalized as $\sum_{\mathbf{k}} \text{Tr}[X_i^{(m)}(\mathbf{k})Y_j^{(n)}(\mathbf{k})] = \delta_{XY}\delta_{ij}$ ($X, Y = Q/T, n = 1, 2, 3$).

c. Multipole decomposition of the tight-binding Hamiltonian

Next, we express the model Hamiltonian by using the atomic and momentum multipoles given in Secs. A 1 a and A 1 b, respectively [41,51–53]. Through this procedure, we can clarify the microscopic multipole degrees of freedom in the present system.

Let us consider the total Hamiltonian given in the main text, $h(\mathbf{k}) \equiv \mathcal{H}_t + \mathcal{H}_{\text{SOC}} + \mathcal{H}_{\text{CEF}} + \mathcal{H}_V$. The crystalline electric field \mathcal{H}_{CEF} is expressed by using the spinless atomic multipoles belonging to the A_{1g}^+ irreducible representation as

$$\mathcal{H}_{\text{CEF}} = \epsilon_1 Q_u^{(a)} + \epsilon_2 Q_4^{(a)} + \epsilon_3 Q_{4u}^{(a)}. \quad (\text{A13})$$

$$\mathcal{H}_t^{(1)} = t_1 Q_0^{(a)} Q_0^{(1)}(\mathbf{k}) + t_2 Q_u^{(a)} Q_0^{(1)}(\mathbf{k}) + t_3 Q_v^{(a)} Q_v^{(1)}(\mathbf{k}) + t_4 Q_4^{(a)} Q_0^{(1)}(\mathbf{k}) + t_5 Q_{4u}^{(a)} Q_0^{(1)}(\mathbf{k}) + t_6 Q_{4v}^{(a)} Q_v^{(1)}(\mathbf{k}),$$

$$\mathcal{H}_t^{(2)} = t_7 Q_0^{(a)} Q_0^{(2)}(\mathbf{k}) + t_8 Q_u^{(a)} Q_0^{(2)}(\mathbf{k}) + t_9 Q_4^{(a)} Q_0^{(2)}(\mathbf{k}) + t_{10} Q_{4u}^{(a)} Q_0^{(2)}(\mathbf{k}),$$

$$\begin{aligned} \mathcal{H}_t^{(3)} &= t_{11} Q_0^{(a)} Q_0^{(3)}(\mathbf{k}) + t_{12} Q_u^{(a)} Q_0^{(3)}(\mathbf{k}) + t_{13} Q_v^{(a)} Q_v^{(3)}(\mathbf{k}) + t_{14} \frac{1}{\sqrt{2}} (Q_{yz}^{(a)} Q_{yz}^{(3)}(\mathbf{k}) + Q_{zx}^{(a)} Q_{zx}^{(3)}(\mathbf{k})) + t_{15} Q_4^{(a)} Q_0^{(3)}(\mathbf{k}) + t_{16} Q_{4u}^{(a)} Q_0^{(3)}(\mathbf{k}) \\ &+ t_{17} Q_{4v}^{(a)} Q_v^{(3)}(\mathbf{k}) + t_{18} \frac{1}{\sqrt{2}} (Q_{4x}^{\alpha(a)} Q_{yz}^{(3)}(\mathbf{k}) + Q_{zx}^{\alpha(a)} Q_{zx}^{(3)}(\mathbf{k})) + t_{19} \frac{1}{\sqrt{2}} (Q_{4x}^{\beta(a)} Q_{yz}^{(3)}(\mathbf{k}) + Q_{zx}^{\beta(a)} Q_{zx}^{(3)}(\mathbf{k})). \end{aligned} \quad (\text{A17})$$

By using the Slater-Koster parameters given in the main text, $t_{1\sigma}, t_{1\pi}, t_{z1\sigma}, t_{z1\pi}, t_{z2\sigma}, t_{z2\pi}, t_i$ ($i = 1-19$) are represented by

$$\begin{aligned} t_1 &= \frac{4\sqrt{5}}{5}(2t_{1\sigma} + t_{1\pi}), \\ t_2 &= -\frac{\sqrt{14}}{7}(4t_{1\sigma} + t_{1\pi}), \\ t_3 &= -\frac{\sqrt{42}}{7}(4t_{1\sigma} + t_{1\pi}), \\ t_4 &= \frac{4\sqrt{30}}{15}(3t_{1\sigma} - t_{1\pi}), \\ t_5 &= -\frac{2\sqrt{42}}{21}(3t_{1\sigma} - t_{1\pi}), \\ t_6 &= -\frac{2\sqrt{14}}{7}(3t_{1\sigma} - t_{1\pi}), \end{aligned} \quad (\text{A18})$$

By using Δ_1, Δ_2 , and Δ_3 defined in the main text, ϵ_1, ϵ_2 , and ϵ_3 are expressed as follows:

$$\begin{aligned} \epsilon_1 &= \frac{1}{\sqrt{7}}(-3\Delta_1 - 3\Delta_2 + \Delta_3), \\ \epsilon_2 &= \frac{\sqrt{15}}{3}(\Delta_2 + \Delta_3), \\ \epsilon_3 &= \frac{1}{\sqrt{21}}(6\Delta_1 - \Delta_2 + 5\Delta_3). \end{aligned} \quad (\text{A14})$$

The atomic spin-orbit coupling, $\mathcal{H}_{\text{SOC}} = (\lambda/2)\mathbf{l} \cdot \boldsymbol{\sigma}$, is represented by using the spinful atomic monopole $Q_0^{(a)}$ in A_{1g}^+ as

$$\mathcal{H}_{\text{SOC}} = 2\sqrt{15}\lambda Q_0^{(a)}. \quad (\text{A15})$$

Similarly, the symmetry-breaking term \mathcal{H}_V in D_{4h} is represented by using the spinless atomic hexadecapole $Q_{4z}^{\alpha(a)}$ belonging to A_{2g}^+ irreducible representation,

$$\mathcal{H}_V = 2V Q_{4z}^{\alpha(a)}. \quad (\text{A16})$$

Since the kinetic energy term, $\mathcal{H}_t = \mathcal{H}_t^{(1)} + \mathcal{H}_t^{(2)} + \mathcal{H}_t^{(3)}$, is also fully symmetric for all the symmetry operations in the point group D_{4h} , only the independent products of the atomic and momentum multipoles belonging to A_{1g}^+ irreducible representation contribute to \mathcal{H}_t . Considering $A_{1g}^{\pm} \otimes A_{1g}^{\pm} = A_{2g}^{\pm} \otimes A_{2g}^{\pm} = A_{1g}^+$, we obtain

$$\begin{aligned} t_7 &= \frac{\sqrt{10}}{5}(2t_{z1\pi} + t_{z1\sigma}), \\ t_8 &= 2\frac{\sqrt{7}}{7}(t_{z1\pi} + t_{z1\sigma}), \\ t_9 &= -\frac{\sqrt{15}}{15}(4t_{z1\pi} - 3t_{z1\sigma}), \\ t_{10} &= -\frac{\sqrt{21}}{21}(4t_{z1\pi} - 3t_{z1\sigma}), \\ t_{11} &= \frac{2\sqrt{10}}{5}(2t_{z2\pi} + t_{z2\sigma}), \\ t_{12} &= \frac{\sqrt{7}}{7}(t_{z2\pi} + t_{z2\sigma}), \\ t_{13} &= -\frac{\sqrt{21}}{7}(t_{z2\pi} + t_{z2\sigma}), \\ t_{14} &= \frac{2\sqrt{21}}{7}(t_{z2\pi} + t_{z2\sigma}), \end{aligned} \quad (\text{A19})$$

$$\begin{aligned}
t_{15} &= \frac{\sqrt{15}}{30}(4t_{z2\pi} - 3t_{z2\sigma}), \\
t_{16} &= \frac{5\sqrt{21}}{84}(4t_{z2\pi} - 3t_{z2\sigma}), \\
t_{17} &= -\frac{5\sqrt{7}}{28}(4t_{z2\pi} - 3t_{z2\sigma}), \\
t_{18} &= 0, \\
t_{19} &= \frac{\sqrt{7}}{7}(4t_{z2\pi} - 3t_{z2\sigma}). \tag{A20}
\end{aligned}$$

Notably, although there are 19 independent parameters allowed by the symmetry, only the six independent Slater-Koster parameters appear in the hopping Hamiltonian. This is because the Slater-Koster approach assumes the axial symmetry along the bond direction and neglects the surrounding environment of the actual system.

2. Essential model parameters in response tensors

In this section, we show the essential model parameters to give a nonzero thermal average of the electric toroidal dipole under the external magnetic field along the x direction $H_x \neq 0$ and the linear electric conductivity tensors under the external magnetic fields, by using the systematic analysis method given in Refs. [51] and [42], which has been used to analyze the essential model parameters of nonlinear (spin) transport [42,54–58] and nonreciprocal magnon dispersion [59].

a. Thermal average of the electric toroidal dipole

Let us begin with the essential model parameters for the thermal average of the electric toroidal dipole $G_z^{(a)}$ given in Eq. (8) in the main text. The essential model parameters for $\langle G_z^{(a)} \rangle \neq 0$ under $H_x \neq 0$ are extracted by analytically evaluating the low-order contributions of the following quantity [42,51]:

$$\Gamma^i(G_z^{(a)}) = \sum_{\mathbf{k}} \text{Tr}[G_z^{(a)} h^i(\mathbf{k})], \tag{A21}$$

where $h^i(\mathbf{k})$ denotes the i th power of the Hamiltonian matrix including the Zeeman coupling with $H_x \neq 0$ at wave vector \mathbf{k} . The summation of the momentum \mathbf{k} is taken over a 10^3 grid.

The lowest- and next-lowest-order contributions $i = 5$ and $i = 6$ are explicitly given by

$$\Gamma^5(G_z^{(a)}) = -120VH_x^4, \tag{A22}$$

$$\begin{aligned}
\Gamma^6(G_z^{(a)}) &= -6V\lambda \\
&\times [100H_x^4 + (-8t_{1\pi}t_{1\sigma} - t_{z2\pi}t_{z2\sigma} - 32t_{1\sigma}^2 - t_{z2\sigma}^2)H_x^2]. \tag{A23}
\end{aligned}$$

As a result, V is the essential model parameter for $\langle G_z^{(a)} \rangle \neq 0$ under $H_x \neq 0$. Notably, the second term in Eq. (A23) indicates that the coupling between V and λ gives rise to the additional contributions proportional to H_x^2 in $\langle G_z^{(a)} \rangle$. These results are consistent with the numerical calculation results shown in Figs. 2(a) and 2(b) in the main text.

b. Unconventional Hall conductivity tensors

We also identify the essential model parameters in the unconventional Hall conductivity tensors $\sigma_{\mu\nu}^H(H_x)$ given in Eq. (11) in the main text by evaluating the following quantity [42]:

$$\Gamma_{\mu\nu}^{ij}(H_\eta) = \sum_{\mathbf{k}} \text{Tr}[J_{\mu\mathbf{k}} h^i(\mathbf{k}) J_{\nu\mathbf{k}} h^j(\mathbf{k})], \tag{A24}$$

where $J_{\mu\mathbf{k}}$ is the μ directional electric current operators at \mathbf{k} and $h(\mathbf{k})$ includes the Zeeman coupling with $H_\eta \neq 0$. The summation of the momentum \mathbf{k} is taken over a 10^3 grid. Since the essential model parameters are contained in any pairs of (i, j) in Eq. (A24), we only show several lowest-order contributions to Eq. (A24).

First, we focus on the $\Gamma_{zx}^{ij}(H_x)$ that corresponds to the unconventional Hall conductivity tensor $\sigma_{zx}^H(H_x)$. The lowest-order contributions to $\text{Im}[\Gamma_{zx}^{ij}(H_x)] = -\text{Im}[\Gamma_{xz}^{ij}(H_x)]$ that are proportional to H_x and H_x^3 are explicitly given by

$$\begin{aligned}
\text{Im}[\Gamma_{zx}^{03}(H_x)] &= -\frac{3\sqrt{2}}{10}VH_x(8t_{1\pi}t_{z1\pi}t_{z2\pi} - 3t_{1\pi}t_{z1\pi}t_{z2\sigma} \\
&\quad - 12t_{1\sigma}t_{z1\pi}t_{z2\sigma} - 12t_{1\sigma}t_{z1\sigma}t_{z2\pi} - 6t_{1\sigma}t_{z1\sigma}t_{z2\sigma}), \tag{A25}
\end{aligned}$$

$$\begin{aligned}
\text{Im}[\Gamma_{zx}^{05}(H_x)] &= -\frac{3\sqrt{2}}{10}VH_x^3(148t_{1\pi}t_{z1\pi}t_{z2\pi} - 36t_{1\pi}t_{z1\pi}t_{z2\sigma} \\
&\quad + 12t_{1\pi}t_{z1\sigma}t_{z2\pi} + 9t_{1\pi}t_{z1\sigma}t_{z2\sigma} - 138t_{1\sigma}t_{z1\pi}t_{z2\sigma} \\
&\quad - 180t_{1\sigma}t_{z1\sigma}t_{z2\pi} - 72t_{1\sigma}t_{z1\sigma}t_{z2\sigma}). \tag{A26}
\end{aligned}$$

We find that all the terms in $\text{Im}[\Gamma_{zx}^{ij}(H_x)]$ are proportional to VH_x^{2m+1} and hence $\sigma_{zx}^H(H_x)$ is an odd function of H_x :

$$\begin{aligned}
\sigma_{zx}^H(H_x) &= V \sum_{m=0} H_x^{2m+1} \\
&\quad \times F_m(V, \lambda, \Delta_1, \Delta_2, \Delta_3, t_{1\pi}, t_{1\sigma}, t_{z1\pi}, t_{z1\sigma}, t_{z2\pi}, t_{z2\sigma}), \tag{A27}
\end{aligned}$$

where F_m is a function of the parameters. Therefore, the essential model parameters for $\sigma_{zx}^H(H_x) \neq 0$ are V under $H_x \neq 0$. These results are consistent with the numerical calculation results shown in Figs. 3(a) and 3(b) in the main text.

On the other hand, the lowest-order contributions to $\text{Im}[\Gamma_{yz}^{ij}(H_x)] = -\text{Im}[\Gamma_{zy}^{ij}(H_x)]$ that correspond to the conventional Hall conductivity tensor $\sigma_{yz}^H(H_x)$ are explicitly given by

$$\begin{aligned}
\text{Im}[\Gamma_{yz}^{02}(H_x)] &= -\frac{3\sqrt{2}}{5}H_x(4t_{1\pi}t_{z1\pi}t_{z2\pi} + 3t_{1\pi}t_{z1\sigma}t_{z2\sigma} \\
&\quad + 12t_{1\sigma}t_{z1\pi}t_{z2\sigma} + 12t_{1\sigma}t_{z1\sigma}t_{z2\sigma}), \tag{A28}
\end{aligned}$$

$$\begin{aligned}
\text{Im}[\Gamma_{yz}^{04}(H_x)] &= -\frac{12\sqrt{2}}{5}H_x^3(8t_{1\pi}t_{z1\pi}t_{z2\pi} + 9t_{1\pi}t_{z1\sigma}t_{z2\sigma} \\
&\quad + 36t_{1\sigma}t_{z1\pi}t_{z2\sigma} + 39t_{1\sigma}t_{z1\sigma}t_{z2\sigma}). \tag{A29}
\end{aligned}$$

We find that all the terms in $\text{Im}[\Gamma_{yz}^{ij}(H_x)]$ are proportional to the odd power of H_x . Unlike the unconventional Hall conductivity tensor $\sigma_{zx}^H(H_x)$, V is not essential for the conventional Hall conductivity tensor $\sigma_{yz}^H(H_x) \neq 0$. These results are consistent with the numerical calculation results shown in Figs. 4(a) and 4(b) in the main text.

c. Magnetoconductivity tensors

We show here the essential model parameters in the magnetoconductivity tensors $\sigma_{\mu\nu}^{\text{MC}}(H)$ for $H_\eta = H_\gamma = H$ given in Eq. (15) in the main text by evaluating Eq. (A24).

First, we focus on the $\Gamma_{xy}^{ij}(H_x)$ that corresponds to the magnetoconductivity tensor $\sigma_{xy}^{\text{MC}}(H_x)$ by setting $\eta = \gamma = x$. The lowest-order contributions to $\text{Re}[\Gamma_{xy}^{ij}(H_x)] = \text{Re}[\Gamma_{yx}^{ij}(H_x)]$ that are proportional to H_x^2 and H_x^4 are explicitly given by

$$\begin{aligned} \text{Re}[\Gamma_{xy}^{05}(H_x)] = & -\frac{1}{512}VH_x^2(2560t_{1\pi}^2t_{z2\pi}^2 - 1536t_{1\pi}^2t_{z2\pi}t_{z2\sigma} - 192t_{1\pi}^2t_{z2\sigma}^2 + 6144t_{1\pi}t_{1\sigma}t_{z2\pi}^2 - 14592t_{1\pi}t_{1\sigma}t_{z2\pi}t_{z2\sigma} \\ & - 8448t_{1\pi}t_{1\sigma}t_{z2\sigma}^2 + 6144t_{1\sigma}^2t_{z2\pi}^2 - 30720t_{1\sigma}^2t_{z2\pi}t_{z2\sigma} - 11520t_{1\sigma}^2t_{z2\sigma}^2 + 768t_{z2\pi}^4 \\ & - 1728t_{z2\pi}^3t_{z2\sigma} - 4632t_{z2\pi}^2t_{z2\sigma}^2 - 1644t_{z2\pi}t_{z2\sigma}^3 - 135t_{z2\sigma}^4), \end{aligned} \quad (\text{A30})$$

$$\begin{aligned} \text{Re}[\Gamma_{xy}^{07}(H_x)] = & -\frac{3}{256}VH_x^4(12544t_{1\pi}^2t_{z2\pi}^2 - 7296t_{1\pi}^2t_{z2\pi}t_{z2\sigma} - 1248t_{1\pi}^2t_{z2\sigma}^2 + 29696t_{1\pi}t_{1\sigma}t_{z2\pi}^2 \\ & - 68224t_{1\pi}t_{1\sigma}t_{z2\pi}t_{z2\sigma} - 39552t_{1\pi}t_{1\sigma}t_{z2\sigma}^2 + 26624t_{1\sigma}^2t_{z2\pi}^2 - 145408t_{1\sigma}^2t_{z2\pi}t_{z2\sigma} \\ & - 52992t_{1\sigma}^2t_{z2\sigma}^2 + 4544t_{z2\pi}^4 - 8640t_{z2\pi}^3t_{z2\sigma} - 23328t_{z2\pi}^2t_{z2\sigma}^2 - 8256t_{z2\pi}t_{z2\sigma}^3 - 747t_{z2\sigma}^4). \end{aligned} \quad (\text{A31})$$

We find that all the terms in $\text{Re}[\Gamma_{xy}^{ij}(H_x)]$ are proportional to VH_x^{2m} and hence $\sigma_{xy}^{\text{MC}}(H_x)$ is an even function of H_x :

$$\sigma_{xy}^{\text{MC}}(H_x) = V \sum_{m=0} H_x^{2m} F_m(V, \lambda, \Delta_1, \Delta_2, \Delta_3, t_{1\pi}, t_{1\sigma}, t_{z1\pi}, t_{z1\sigma}, t_{z2\pi}, t_{z2\sigma}), \quad (\text{A32})$$

where F_m is a function of the parameters. Therefore, the essential model parameters for $\sigma_{xy}^{\text{MC}}(H_x) \neq 0$ are V under $H_x \neq 0$. These results are consistent with the numerical calculation results shown in Figs. 5(a) and 5(b) in the main text.

Finally, let us discuss the lowest-order contributions to $\text{Re}[\Gamma_{zx}^{ij}(H)] = \text{Re}[\Gamma_{xz}^{ij}(H)]$ that correspond to the magnetoconductivity tensor $\sigma_{zx}^{\text{MC}}(H)$ for $H_x = H_z = H$ and are explicitly given by

$$\text{Re}[\Gamma_{zx}^{02}(H)] = -\frac{72}{5}H^2(t_{z2\pi}^2 + t_{z2\sigma}^2), \quad (\text{A33})$$

$$\text{Re}[\Gamma_{zx}^{04}(H)] = -\frac{144}{5}H^4(11t_{z2\pi}^2 + 10t_{z2\sigma}^2). \quad (\text{A34})$$

Unlike the magnetoconductivity tensor $\sigma_{xy}^{\text{MC}}(H_x)$, $\sigma_{zx}^{\text{MC}}(H) \neq 0$ can be realized without the ferroaxial ordering term V .

There are other contributions, $\text{Im}[\Gamma_{zx}^{ij}(H)] = -\text{Im}[\Gamma_{xz}^{ij}(H)]$, that correspond to the unconventional Hall conductivity tensor $\sigma_{zx}^{\text{H}}(H_x)$ with $H \neq 0$. The lowest-order contributions are explicitly given by

$$\text{Im}[\Gamma_{zx}^{03}(H)] = -\frac{3\sqrt{2}}{10}VH(8t_{1\pi}t_{z1\pi}t_{z2\pi} - 3t_{1\pi}t_{z1\pi}t_{z2\sigma} - 12t_{1\sigma}t_{z1\pi}t_{z2\sigma} - 12t_{1\sigma}t_{z1\sigma}t_{z2\pi} - 6t_{1\sigma}t_{z1\sigma}t_{z2\sigma}), \quad (\text{A35})$$

$$\begin{aligned} \text{Im}[\Gamma_{zx}^{05}(H)] = & -\frac{3\sqrt{2}}{10}VH^3(296t_{1\pi}t_{z1\pi}t_{z2\pi} - 90t_{1\pi}t_{z1\pi}t_{z2\sigma} + 12t_{1\pi}t_{z1\sigma}t_{z2\pi} + 9t_{1\pi}t_{z1\sigma}t_{z2\sigma} + 48t_{1\sigma}t_{z1\pi}t_{z2\pi} - 366t_{1\sigma}t_{z1\pi}t_{z2\sigma} \\ & - 360t_{1\sigma}t_{z1\sigma}t_{z2\pi} - 162t_{1\sigma}t_{z1\sigma}t_{z2\sigma}). \end{aligned} \quad (\text{A36})$$

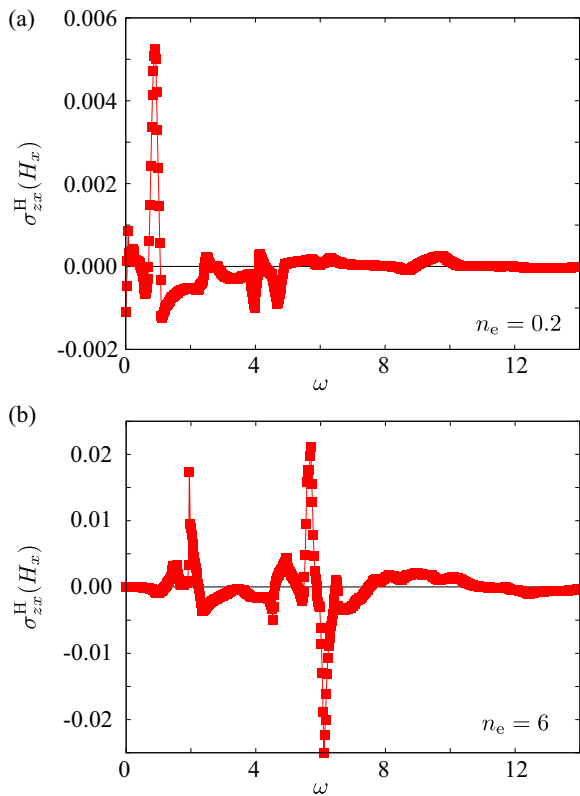


FIG. 6. ω dependence of $\sigma_{zx}^H(H_x)$ at $\lambda = 2$ for (a) $n_e = 0.2$ and (b) $n_e = 6$.

Similar to Eq. (A27), all the terms in $\text{Im}[\Gamma_{zx}^{ij}(H)]$ are proportional to VH^{2m+1} .

APPENDIX B: OPTICAL UNCONVENTIONAL HALL CONDUCTIVITY

We show the optical unconventional Hall conductivity $\sigma_{zx}^H(H_x)$ by considering finite frequency ω in Fig. 6. The data are calculated at fixed $H_x = 0.3$ and $\lambda = 2$ for $n_e = 0.2$ in Fig. 6(a) and $n_e = 6$ in Fig. 6(b). As shown in Figs. 6(a) and 6(b), $\sigma_{zx}^H(H_x)$ becomes nonzero for finite frequency ω , although it shows a complicated behavior including the sign change with changing ω . In the insulating case in Fig. 6(b), $\sigma_{zx}^H(H_x)$ remains zero for a low frequency smaller than the band gap, but it becomes nonzero when the frequency is larger than the band gap. Thus the optical unconventional Hall conductivity is one of the measurements with which to identify the ferroaxial ordering when the materials are insulating.

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