Optical Hall response in spin-orbit coupled metals: Comparative study of magnetic cluster monopole, quadrupole, and toroidal orders

Tatsuki Sato¹, Yuma Umimoto¹, Yusuke Sugita², Yasuyuki Kato², and Yukitoshi Motome²
¹Department of Advanced Materials Science, University of Tokyo, Kashiwa 277-8561, Japan
²Department of Applied Physics, University of Tokyo, Tokyo 113-8656, Japan

The optical Hall response is theoretically studied for spin-orbit coupled metals with ferroic orders of cluster-type magnetic multipoles. We find that different magnetic multipoles give rise to distinct spectra in the optical Hall conductivity. In the cases of monopole and quadrupole orders, the optical Hall response appears predominantly in high- and low-energy regions, which correspond to the energy scales of electron correlation and kinetic energy, respectively, while the response is dispersed and rather weak in the case of toroidal order. By decomposing the spectra into different interband contributions, we reveal selection rules stemming from the interplay between the antisymmetric spin-orbit coupling and the coupling of electrons to the underlying multipoles. Our results suggest that the optical Hall measurement is useful to detect and distinguish the cluster-type magnetic multipole orders.

I. INTRODUCTION

In condensed matter physics, multipoles provide a key concept to understand physical properties through classification of spatial distributions of electrons with charge, spin, and orbital degrees of freedom. Typical examples are the multipoles defined in the atomic scale, which have been used to characterize electronic and magnetic phases in f-electron systems [1, 2]. Multipoles can also be defined in an extended scale over several atomic sites and such cluster-type extensions have recently garnered great attention as a source of intriguing phenomena [3–11]. For instance, a magnetic toroidal dipole induces the second harmonic generation in LiCoPO4 [12], and the magneto-optical Kerr effect in UNi4B [6, 13] and Ce5TiBi5 [14], a magnetic octupole plays a crucial role in the anomalous Hall effect [15], the anomalous Nernst effect [16], and the magneto-optical Kerr effect [17] in Mn3Sn; and a magnetic quadrupole causes the magnetoelectric effect in A(TiO)Cu4(PO4)4 (A=Ba, Sr, and Pb) [18–21]. Thus, it is useful to identify relevant multipoles for predicting the electronic, magnetic, transport, and optical properties. At the same time, measurement of these properties enables us to identify the relevant multipoles. For instance, in the linear response, the diagonal, traceless symmetric, and antisymmetric parts of the magnetoelectric effect have one-to-one correspondence with the magnetic monopole, quadrupole, and toroidal dipole, respectively [4]. Such studies have been extensively performed in the DC limit for metallic systems. Although the AC responses would also serve as useful tools as shown for the optical responses in insulating materials, they have not been studied systematically thus far.

In the present study, we theoretically study the optical Hall response for different types of cluster-type magnetic multipoles. For a minimal model defined on a layered structure where spatial inversion symmetry is broken at each lattice site, we compute the electronic band structure and the optical Hall conductivity in the presence of ferroic orders of the cluster-type magnetic monopole, quadrupole, and toroidal dipole (toroidal). We reveal that despite the similarity in the band structure, the optical Hall responses exhibit distinct frequency dependence for the three types of the multipoles. By decomposing the responses into the interband contributions and analyzing them with the atomic bases, we show that the distinct behaviors can be understood from optical selection rules arising from the interplay between the antisymmetric spin-orbit coupling and the coupling of electrons to the underlying multipole orders.

This paper is organized as follows. In Sec. II we introduce the model with the cluster-type magnetic multipole orders. We present the results for the electronic band structure in Sec. III A and the optical Hall conductivity in Sec. III B. From the decomposition into the interband contributions, we find optical selection rules in Sec. III C. In Sec. IV we discuss the origin of the optical selection rules by applying the perturbation theory in the atomic limit. Section V is devoted to the summary. We also study a variant of the model in Appendix to confirm the generality of the optical selection rules.

II. MODEL

We consider a minimal model with ferroic orders of cluster-type magnetic multipoles. We adopt a single-band tight-binding model on a layered lattice structure, where each layer consists of a periodic array of four-site square clusters, as shown in Fig. I(a). Each square cluster can accommodate magnetic cluster multipoles composed of four spins, such as monopole, quadrupole, and toroidal, as shown in Figs. I(c), I(d), and I(e), respectively. Note that spatial inversion symmetry is broken at each lattice site, while it is retained at the centers of square plaquettes in each layer and of cuboids defined by two squares in adjacent layers. A similar model with hexagonal clusters was discussed in the previous study [6] (see Appendix). The Hamiltonian of our model is given by

\[ H = H_l + H_{ASOC} + H_{MF} + H_{Zeeman}, \] (1)
where
\[ H_t = - \sum_{i,j} t_{ij} (c_i^\dagger c_j + c_j^\dagger c_i + h.c.), \]
\[ H_{ASOC} = 2 \sum_{k,l} [s_{kl} \times D_{kl}]_z, \]
\[ H_{MF} = - \sum_i M_i \cdot s_i, \]
\[ H_{Zeeman} = -B \cdot \sum_i s_i. \]

\( H_t \) in Eq. (2) describes the hoppings of electrons. \( c_i^\dagger \) \( (c_i) \) is the creation (annihilation) operator for an electron at site \( i \) with spin \( \sigma \). We take into account three types of transfer integrals between neighboring sites: the intralayer ones \( t_1 \) and \( t_2 \) within and between the clusters, respectively, and the interlayer one \( t_z \) [see Fig. 1(a)]. All of the other transfer integrals between further-neighbor sites are assumed to be zero.

\( H_{ASOC} \) in Eq. (3) describes the antisymmetric spin-orbit coupling. It originates from the interplay among the atomic spin-orbit coupling, off-site orbital hybridization, and the crystalline electric field \( s_{kl} \). \( s_{kl} \) is the Fourier transform of the spin operator at site \( i \), \( s_i = \frac{1}{2} \sum_{\sigma, \sigma'} c_i^\dagger \sigma \sigma' c_i \). \( M_i \) is the vector of the Pauli matrices; \( k \) and \( l \) denote momentum and sublattice, respectively. \( D_{kl} \) represents a sublattice-dependent vector antisymmetric with respect to \( k_z \), which is given by
\[ D_{kl} = D_l \sin(k_z c), \]
with
\[ D_l = D \left( \cos \theta_l^D, \sin \theta_l^D, 0 \right). \]

Here, \( D \) is the coupling constant and
\[ \theta_l^D = \frac{\pi}{2} n_l - \frac{3\pi}{4}, \]
where \( n_l = 0, 1, 2, \) and \( 3 \) correspond to the sublattices \( l = \alpha, \beta, \gamma, \) and \( \delta \), respectively. The directions of \( D_l \) are shown by the gray arrows in Fig. 1(b). The \( k_z \) dependence in Eq. (5) comes from the off-site orbital hybridization along the \( z \) axis, where \( c \) is the lattice constant in the \( z \) direction [6].

\( H_{MF} \) in Eq. (4) describes the exchange coupling between itinerant electron spins and magnetic multipoles at the mean-field level. The multipoles are composed of magnetic moments \( M_i \), which can be regarded as localized moments coupled to itinerant electrons or mean fields decoupled from the Coulomb interaction between itinerant electrons. We assume ferroic orders of three types of cluster-type magnetic multipoles, monopole, quadrupole, and toroidal [see Figs. 1(c)-1(e)]. Then, \( M_i \) depends only on the sublattice \( l \) as
\[ M_i = M (\cos \theta_i^M, \sin \theta_i^M, 0), \]
where \( M \) denotes the magnitude of the magnetic moments and
\[ \theta_i^M = \frac{\pi}{2} n_l - \frac{3\pi}{4} \quad \text{for monopole}, \]
\[ \theta_i^M = \frac{\pi}{2} n_l - \frac{\pi}{4} \quad \text{for quadrupole}, \]
\[ \theta_i^M = \frac{\pi}{2} n_l - \frac{\pi}{4} \quad \text{for toroidal}. \]

\( H_{Zeeman} \) in Eq. (5) represents the Zeeman coupling to an external magnetic field \( B \). We assume that the magnetic field couples only to the electron spins and neglect a canting of the magnetic moments \( M_i \) by \( B \).
III. RESULT

In this section, we present the results of the electronic and transport properties for the model in Eq. (1) in the presence of ferroic orders of the cluster-type magnetic multipoles. In Sec. IIIA we show that the electronic band structures look similar to each other for the monopole, quadrupole, and toroidal orders. Despite the similarity, however, we show that the frequency dependences of the optical Hall conductivity are substantially different in Sec. IIIB. In Sec. IIIC, we discuss the origin of the differences, we analyze the contributions from different interband processes. All the following calculations in this section are obtained for the model parameters, \( a = c = 1, \tilde{a} = 0.35, t_1 = 1.25, t_2 = 0.75, t_z = 1, D = 0.5, M = 8, \) and \( B = (0.5, 0, 0), \) which correspond to the strongly correlated metal under a small magnetic field.

A. Electronic band structure

Figure 2 shows the electronic band structures of the model in Eq. (1) in the presence of ferroic orders of the cluster-type magnetic multipoles: (a) monopole, (b) quadrupole, and (c) toroidal. In all cases, we obtain eight bands corresponding to the four sublattices and spin degrees of freedom. The eight bands are split into two groups by the exchange coupling to the magnetic multipoles, \( H_{MF} \) in Eq. (1): four lower (higher)-energy bands correspond to the bands with spins \( s = \uparrow (\uparrow \downarrow) \) parallel to the magnetic moments \( M_i. \) Further splitting of each four into two groups is brought by \( H_t \) in Eq. (2) and the smallest splitting is caused by \( H_{Zeeman} \) in Eq. (5) (see the discussion in Sec. IV). The overall band structures are similar to each other for the three types of multipoles, but nevertheless they lead to distinct optical Hall responses as shown in the next subsection. We note that the band bottom is shifted along the \( k_z \) direction (\( \Gamma-Z \)) in the case of the toroidal order, which is parallel to the toroidal moment, as shown in Fig. 2(c) [3, 6], whereas no such a shift is seen for the monopole and quadrupole orders as shown in Figs. 2(a) and 2(b), respectively.

B. Optical Hall conductivity

We calculate the optical Hall conductivity \( \sigma^{\mu z}(\omega) \) for an electric current in the \( \mu \) direction induced by that in the \( z \) direction. It is obtained by using the Kubo formula as

\[
\sigma^{\mu z}(\omega) = \sum_{m,n} \sigma^{\mu z}_{m,n}(\omega),
\]

where

\[
\sigma^{\mu z}_{m,n}(\omega) = \sum_k \frac{e^2}{\hbar} \frac{1}{iV} \frac{f(\epsilon_{nk}) - f(\epsilon_{mk})}{\epsilon_{nk} - \epsilon_{mk}} \frac{\langle nk|j_{k\mu}^{\mu}|mk\rangle \langle mk|j_{k\mu}^{\mu}|nk\rangle}{\hbar\omega + \epsilon_{nk} - \epsilon_{mk} + i\delta}.
\]

Here, \( V \) is the system volume, \( f(\epsilon) \) is the Fermi-Dirac distribution function, \( \epsilon_{mk} \) and \( |mk\rangle \) are the eigenvalue and eigenstate of \( H \) for band \( m \) with momentum \( k, \) respectively (we label the bands \( m = 1, 2, \cdots, 8 \) from the lowest energy to the highest one), and \( j_{k\mu}^{\mu} = -\partial H_k/\partial k_{\mu \mu} \) is the current operator in the \( \mu \) direction with momentum \( k, \) where \( H_k \) is the Fourier component of \( H \) defined as \( H = \sum_k H_k. \) In the following, we take the elementary charge \( e = 1, \) the Dirac constant \( \hbar = 1, \) the tempera-
ture $k_B T = 0.1$ ($k_B$ is the Boltzmann constant), and the broadening factor $\delta = 0.02$.

In our model, the optical Hall conductivity becomes nonzero for $\sigma^{yz}(\omega)$ in $B \parallel [100]$ or $\sigma^{xz}(\omega)$ in $B \parallel [010]$ and has the antisymmetric relation $\sigma^{yz}(\omega) = -\sigma^{xz}(\omega)$ from the fourfold rotational symmetry in the $xy$ plane. We therefore focus on the results of $\sigma^{yz}(\omega)$ in the following. Figure 3 shows $\sigma^{yz}(\omega)$ as a function of the energy $\omega$ for the (a) monopole, (b) quadrupole, and (c) toroidal orders. The electron filling $n_e = \sum_{i,\sigma} \langle c_i^\dagger \sigma c_i \sigma \rangle$ is set to 0.1 so that the chemical potential lies in the lowest two bands with the energies $\varepsilon_{1k}$ and $\varepsilon_{2k}$ ($N$ is the total number of lattice sites). We find that the optical Hall conductivity exhibits distinct $\omega$ dependence for different types of the multipole orders. For the monopole and quadrupole orders, $\sigma^{yz}(\omega)$ shows its primary responses in rather high-energy ($\omega \gtrsim 6$) and low-energy ($0 \leq \omega \lesssim 6$) regions, as shown in Figs. 3(a) and 3(b), respectively. On the other hand, in the toroidal ordered state, the optical Hall responses in the low- and high-energy regions are comparable to each other, and the overall amplitude is strongly suppressed, as shown in Fig. 3(c).

In order to demonstrate that the distinct responses are generic for any electron filling $n_e$, we compute the integrated intensities $I_{\text{low}}$ and $I_{\text{high}}$ of the absolute values of the real part of $\sigma^{yz}(\omega)$ in the low- and high-energy ranges,

$$I_{\text{low}} = \int_0^{\omega_1} d\omega \left| \text{Re} \sigma^{yz}(\omega) \right| \text{,}$$

FIG. 3. Optical Hall conductivity $\sigma^{yz}(\omega)$ as a function of the energy $\omega$ in the presence of the multipole orders of (a) monopole, (b) quadrupole, and (c) toroidal. The green solid (black dashed) line indicates the real (imaginary) part of $\sigma^{yz}(\omega)$. The results are computed at the electron filling of $n_e = 0.1$ with $k_B T = 0.1$ and $\delta = 0.02$ in Eqs. 13 and 14. The other model parameters are common to those in Fig. 2.

FIG. 4. Integrated intensities of $|\text{Re} \sigma^{yz}(\omega)|$ as functions of the electron filling $n_e$ in the presence of (a) monopole, (b) quadrupole, and (c) toroidal orders [see Eqs. 15 and 16]. The parameters except for $n_e$ are common to those in Fig. 3.
I_{\text{high}} = \int_{\omega_1}^{\omega_2} d\omega |\text{Re}\sigma_{\text{yz}}^m(\omega)|, \quad (16)

respectively, where we take \( \omega_1 = 6 \) and \( \omega_2 = 20 \). Figure 4 shows the results as functions of \( n_e \). They are symmetric with respect to the half filling \( n_e = 1/2 \) because of the particle-hole symmetry between the states of \((k_x, k_y, k_z, \sigma)\) and \((-k_x, -k_y, -k_z, +\pi, -\sigma)\). The optical Hall response vanishes at the half filling as well as empty and full fillings, where the system becomes insulating. For generic filling, however, \( \sigma_{\text{yz}}^m(\omega) \) becomes nonzero. \( I_{\text{high}} \) and \( I_{\text{low}} \) are predominant for the monopole and quadrupole orders as shown in Figs. 4(a) and 4(b), respectively, while both responses are comparable to each other and relatively weak for the toroidal order as shown in Fig. 4(c).

C. Decomposition into interband contributions

In order to clarify which electronic bands play an important role in the optical Hall responses, we decompose the integrated intensities into the interband contributions as

\[
I_{m,n}^{\text{low}} = \int_{0}^{\omega_1} d\omega |\text{Re}\sigma_{\text{yz}}^{m,n}(\omega)|, \quad (17)
\]

We focus on the cases with \( n = 1, 2 \) (partially occupied bands) and \( m = 3, 4, \ldots, 8 \) (unoccupied bands) at \( n_e = 0.1 \). The results are plotted in Fig. 5. For the monopole order, the large values of \( I_{m,n}^{\text{high}} \) are found for \((m,n) = (6,1)\) and \((5,2)\), as shown in Figs. 5(a) and 5(b), respectively. Meanwhile, for the quadrupole order, the dominant contributions in \( I_{m,n}^{\text{low}} \) appear for \((m,n) = (3,1)\) and \((4,2)\), as shown in Figs. 5(c) and 5(d), respectively. For the toroidal case shown in Figs. 5(e) and 5(f), \( I_{m,n}^{\text{low}} \) is distributed for \((m,n) = (4,1)\), \((5,1)\), \((3,2)\), and \((6,2)\), while \( I_{m,n}^{\text{high}} \) is concentrated on \((5,1)\) and \((6,2)\).

We confirm that the distinct \( \omega \) dependences shown in Fig. 3 are qualitatively explained by these dominant interband contributions. For the monopole order, as shown in Fig. 6(a), the large response in the high-energy region is well accounted for by the dominant contributions from

![Fig. 5. Histogram of the integrated intensities of the interband contributions. The blue and red bars represent the low- and high-energy intensities, \( I_{m,n}^{\text{low}} \) in Eq. (17) and \( I_{m,n}^{\text{high}} \) in Eq. (18), respectively, in the presence of (a), (b) monopole, (c), (d) quadrupole, and (e), (f) toroidal orders. The results are shown for (a), (c), (e) \( n = 1 \) and (b), (d), (f) \( n = 2 \). The parameters are common to those in Fig. 3.](image)

![Fig. 6. Dominant interband contributions \( \sigma_{\text{yz}}^{m,n}(\omega) \) in the presence of (a) monopole, (b) quadrupole, and (c) toroidal orders. The parameters are common to those in Fig. 3.](image)
In the quadrupole order, the low-energy and high-energy states. By using the basis of the eight Bloch states in a four-site cluster into the fourfold low-energy ones with the eigenenergy of $-M/2$ and the other fourfold high-energy ones with the eigenenergy of $+M/2$. The eigenstates are given by \( \langle \psi_{k_{a}} \rangle \), \( \langle \psi_{k_{b}} \rangle \), \( \langle \psi_{k_{c}} \rangle \), \( \langle \psi_{k_{d}} \rangle \), \( \langle \psi_{k_{e}} \rangle \), \( \langle \psi_{k_{f}} \rangle \), \( \langle \psi_{k_{g}} \rangle \), \( \langle \psi_{k_{h}} \rangle \), respectively [see Fig. 1(d)].

Next, we discuss the effect of electron hopping $H_{t}$ on the four low-energy states. In the following treatment of $H_{t}$ and $H_{Zeeman}$, we neglect the hybridization between the low-energy and high-energy states. By using the basis set of \( \langle \psi_{k_{a}} \rangle \), \( \langle \psi_{k_{b}} \rangle \), \( \langle \psi_{k_{c}} \rangle \), \( \langle \psi_{k_{d}} \rangle \), \( \langle \psi_{k_{e}} \rangle \), \( \langle \psi_{k_{f}} \rangle \), \( \langle \psi_{k_{g}} \rangle \), \( \langle \psi_{k_{h}} \rangle \), respectively $H_{t}$, which is defined as $H_{t} = \sum_{k} H_{t}(k)$, is expressed in the matrix form of

\[
\frac{1}{\sqrt{2}} \begin{pmatrix}
0 & \tau_{x}^{*} & 0 & \tau_{y}^{*} \\
\tau_{x}^{*} & 0 & -\tau_{y}^{*} & 0 \\
0 & -\tau_{y} & 0 & \tau_{x} \\
\tau_{y} & 0 & \tau_{x} & 0
\end{pmatrix} + \tau_{z} I,
\]

(19)

where

\[
\tau_{x} = -t_{1}e^{ik_{x}a} - t_{2}e^{-ik_{x}(a - \bar{a})},
\]

(20)

\[
\tau_{y} = -t_{1}e^{ik_{y}a} - t_{2}e^{-ik_{y}(a - \bar{a})},
\]

(21)

\[
\tau_{z} = -2t_{z} \cos(k_{z}c),
\]

(22)

and $I$ is the $4 \times 4$ identity matrix. The four eigenstates of Eq. (19) are split into two manifolds, each of which is doubly degenerate. One has the eigenvalue of $-C_{xy}/\sqrt{2} + \tau_{z}$ and the eigenstates of

\[
\frac{1}{\sqrt{2}} \begin{pmatrix}
\cos \rho e^{i\phi} & 0 \\
\sin \rho e^{i\phi} & -1
\end{pmatrix},
\]

while the other has the eigenvalue of $C_{xy}/\sqrt{2} + \tau_{z}$ and the eigenstates of

\[
\frac{1}{\sqrt{2}} \begin{pmatrix}
\cos \rho e^{i\phi} & 0 \\
\sin \rho e^{i\phi} & 1
\end{pmatrix},
\]

Here, $C_{xy} = \sqrt{|r_{xy}|^2 + |r_{yz}|^2} > 0$, $\tau_{x} = C_{xy} \sin \rho e^{i\phi}$, and $\tau_{y} = C_{xy} \cos \rho e^{i\phi}$, with $\rho \in [0, \pi/2]$, $(\phi, \eta) \in [0, 2\pi)$. Note that $PT$ symmetry of $H_{t} + H_{ASOC} + H_{MF}$ results in the twofold degeneracy in Eqs. (23) and (24).

The remaining degeneracy is lifted by the Zeeman coupling $H_{Zeeman}$. In the first-order perturbation, $H_{Zeeman}$ is given by a $2 \times 2$ matrix for each Kramers doublet as

\[
\frac{B}{2} \cos \rho \sin \rho e^{i(-\phi + \eta)} - \cos \rho e^{i\phi + \eta}.
\]

(25)

By diagonalizing Eq. (25), we obtain the eigenstates:

\[
|1k\rangle = \frac{1}{\sqrt{2}} \begin{pmatrix}
\epsilon^{\theta^{*}_{+}} & \epsilon^{-\theta^{*}_{-}} \\
\epsilon^{-\theta^{*}_{+}} & \epsilon^{\theta^{*}_{-}}
\end{pmatrix}, \quad |2k\rangle = \frac{1}{\sqrt{2}} \begin{pmatrix}
-\epsilon^{\theta^{*}_{+}} & \epsilon^{-\theta^{*}_{-}} \\
\epsilon^{-\theta^{*}_{+}} & \epsilon^{\theta^{*}_{-}}
\end{pmatrix}.
\]

(26)

for Eq. (23) and

\[
|3k\rangle = \frac{1}{\sqrt{2}} \begin{pmatrix}
\epsilon^{\varphi^{*}_{+}} & \epsilon^{-\varphi^{*}_{-}} \\
\epsilon^{-\varphi^{*}_{+}} & \epsilon^{\varphi^{*}_{-}}
\end{pmatrix}, \quad |4k\rangle = \frac{1}{\sqrt{2}} \begin{pmatrix}
-\epsilon^{\varphi^{*}_{+}} & \epsilon^{-\varphi^{*}_{-}} \\
\epsilon^{-\varphi^{*}_{+}} & \epsilon^{\varphi^{*}_{-}}
\end{pmatrix},
\]

(27)

for Eq. (24), where $\epsilon = \cos(\rho/2)$, $\delta = \sin(\rho/2)$, and $\theta_{\pm} = \exp[i(\phi \pm \eta)/2]$.

Similar procedures of the degeneracy lifting by $H_{MF}$, $H_{t}$, and $H_{Zeeman}$ hold for the high-energy four states. Consequently, the eightfold degenerate states in the four-site cluster are split into \( |1k\rangle, |2k\rangle, \ldots, |8k\rangle \), as schematically shown in Fig. 2. The eight states yield the band structure shown in Fig. 2(b).

Since the chemical potential is set in the lowest two bands ($n = 1, 2$) in the present calculations, dominant contributions to $\sigma_{mn}^{\pm}(k, \omega)$ come from the matrix elements of $\langle m|j_{k}^{\pm}|n\rangle$ in Eq. (11) for $n = 1$ or 2, and $m \neq n$. Among the four terms in Eq. (11), only $H_{t}$ and $H_{ASOC}$ contribute to $j_{k}^{\pm} = -\partial H_{t}/\partial k_{z}$ as

\[
j_{k} = -\frac{\partial H_{t}}{\partial k_{z}} = -2t_{z} \sin(k_{z}c) \sum_{n} \epsilon_{n}^{1} c_{n}^{\dagger} c_{n},
\]

(28)
and
\[ j_{\text{ASOC}} = -\frac{\partial H_{\text{ASOC}}}{\partial k_z} = 2c \cos(k_z c) \sum_i \Delta_i \cdot s_{ki}, \tag{29} \]

respectively, where \( H_{\text{ASOC}} = \sum_k H_{\text{ASOC}} \) and \( \Delta_i \) is given by \( (D_i^y, -D_i^y, 0) \) within the unit cell \( [D_i^y] \) is the \( \mu \) component of \( D_i \); see Eq. (7). Note that \( \Delta_i \) is regarded as an effective magnetic field which has a toroidal-like configuration at the four sublattices. Since \( j_{\text{ASOC}} \) in Eq. (28) is diagonal in momentum space, it has only nonzero values for the intraband contributions \( (1k|j_{\text{ASOC}}^x|1k) \) and \( (2k|j_{\text{ASOC}}^y|2k) \). On the other hand, \( j_{\text{ASOC}} \) in Eq. (29) has interband contributions. The nonzero values of \( \langle m|j_{\text{ASOC}}^y|n \rangle \) are found only for \( n = 1 \) or \( 2 \) and \( 1 \leq m \leq 4 \) within the present approximation because all of the bases \( \{ \eta \}_{k\alpha \gamma}, \{ \gamma \}_{k\beta \gamma} \), and \( \{ \eta \}_{k\delta} \) (the low-energy eigenstates of \( H_{\text{MF}} \)) are the eigenstates of \( j_{\text{ASOC}} \) with the toroidal-like \( \Delta_i \) which is parallel or antiparallel to the quadrupole order \( M_i \) at each sublattice. For the basis set of the four low-energy states, \( j_{\text{ASOC}} \) is written in the matrix form of
\[ -2c \cos(k_z c) \begin{pmatrix} D & 0 & 0 & 0 \\ 0 & -D & 0 & 0 \\ 0 & 0 & D & 0 \\ 0 & 0 & 0 & -D \end{pmatrix}. \tag{30} \]

Consequently, in the quadrupole ordered state, we obtain the selection rule for the interband contributions:
\[ \langle m|j_{\text{ASOC}}^y|n \rangle = \begin{cases} -2Dc \cos(k_z c) & \text{for } (m,n) = (3,1), (4,2) \\ 0 & \text{otherwise.} \end{cases} \tag{31} \]

The other matrix element \( \langle nk|j_{\text{ASOC}}^y|mk \rangle \) in \( \sigma_{m,n}^y(k,\omega) \) is also estimated by the same basis set. Considering Eq. (31), the important contributions are calculated as
\[ \langle 1k|j_{\text{ASOC}}^y|3k \rangle = -\langle 2k|j_{\text{ASOC}}^y|4k \rangle = \frac{i}{2\sqrt{2}} \text{Im} \left( \frac{\partial \tau_y}{\partial k_y} \right)^* e^{i\eta}. \tag{32} \]

Combining Eqs. (31) and (32), \( \sigma_{m,n}^y(k,\omega) \) for the quadrupole ordered state is approximately given as
\[ \sigma_{m,n}^y(k,\omega) = \begin{cases} \{ f(\varepsilon_{1k}) - f(\varepsilon_{3k}) \} \Xi(k,\omega) & \text{for } (m,n) = (3,1) \\ \{-f(\varepsilon_{2k}) - f(\varepsilon_{4k}) \} \Xi(k,\omega) & \text{for } (m,n) = (4,2) \\ 0 & \text{otherwise,} \end{cases} \tag{33} \]

where
\[ \Xi(k,\omega) = \frac{Dc \cos(k_z c) \text{Im}[(\partial \tau_y/\partial k_y)^* e^{i\eta}]}{\sqrt{2}C_{xy}(\hbar \omega - \sqrt{2}C_{xy} + i\delta)}. \tag{34} \]

The results explain well the dominant interband contributions found in Sec. IIIC: the dominant contributions appear only for \( (m,n) = (3,1) \) and \( (4,2) \) with opposite sign in rather low-energy regions where \( \hbar \omega \sim \sqrt{2}C_{xy} \) in the denominator in Eq. (31) [see Figs. (5c), (5d), and (6a)]. Thus, the optical selection rule for the quadrupole ordered state is rooted in the selection rule of \( \langle nk|j_{\text{ASOC}}^y|nk \rangle \) in Eq. (31).

Next, we discuss the monopole case. A difference between the quadrupole and monopole orders lies in the relative angles between the magnetic moments \( M_i \) and the effective magnetic field \( \Delta_i \) in Eq. (29); while \( M_i \) is parallel or antiparallel to \( \Delta_i \) for the quadrupole order, it is perpendicular to \( \Delta_i \) for the monopole order. Thus, the eigenstates of \( H_{\text{MF}} \) for the monopole order, \( \{ \eta \}_{k\alpha \gamma}, \{ \gamma \}_{k\beta \gamma} \), and \( \{ \eta \}_{k\delta} \) spin flipped by \( j_{\text{ASOC}} \). This means that the matrix elements become nonzero for the interband processes with \( m \) belonging to the four high-energy levels split by \( H_{\text{MF}} \). Consequently, the selection rule for this case is given by
\[ \langle m|j_{\text{ASOC}}^y|n \rangle = \begin{cases} -2Dc \cos(k_z c) & \text{for } (m,n) = (6,1), (5,2) \\ 0 & \text{otherwise.} \end{cases} \tag{35} \]

Following a similar procedure to the quadrupole case above, we end up with
\[ \sigma_{m,n}^y(k,\omega) = \begin{cases} f(\varepsilon_{1k}) \Xi(k,\omega) & \text{for } (m,n) = (6,1) \\ -f(\varepsilon_{2k}) \Xi(k,\omega) & \text{for } (m,n) = (5,2) \\ 0 & \text{otherwise,} \end{cases} \tag{36} \]

where
\[ \Xi_{\pm}(k,\omega) = \frac{Dc \cos(k_z c) \text{Im}[(\partial \tau_y/\partial k_y)^* e^{i\eta}]}{\sqrt{2}M_{\pm}(\hbar \omega - M_{\pm} + i\delta)}, \tag{37} \]

\[ M_{\pm} = M \pm \frac{1}{\sqrt{2}} B \cos \rho. \tag{38} \]

Thus, the optical Hall responses in the monopole ordered state appear dominantly in rather high-energy regions corresponding the energy scale of \( H_{\text{MF}} \), namely,
\( \hbar \omega \sim M_\pm \). The result explains well again the findings in Figs. 5(a), 5(b), and 5(c), as in the case of the quadrupole order.

Finally, in the case of the toroidal order, \( M_l \) is in the same direction to \( \Delta_l \). This means that \( j_{\text{ASOC}}^z \) is proportional to an identity matrix in the four low-energy eigenstates. Hence, \( j_{\text{ASOC}}^z \) as well as \( j_k^z \) does not lead to any interband excitations, resulting in \( \sigma_{m,n}^{yz}(k, \omega > 0) = 0 \) for all \( (m,n) \) within the present approximation. This explains the small responses found in Figs. 5(e), 5(f), and 6(c); they originate in the contributions beyond the present approximation.

Since the optical selection rules discussed here are based on the atomic bases under strong correlation, they are generic to spin-orbit coupled metals under strong influence of the cluster multipole orders, irrespective of the lattice structures and detailed electronic band structures. To confirm this, we study a honeycomb-lattice variant in Appendix, and obtain optical Hall spectra obeying similar optical selection rules.

V. SUMMARY

In summary, we have theoretically investigated the optical Hall responses in spin-orbit coupled metals with ferroic orders of cluster-type magnetic multipoles. Taking a minimal model with monopole, quadrupole, and toroidal orders, we unveiled that the optical Hall conductivity shows distinct frequency dependence for the three types of multipoles. In the cases of the monopole and quadrupole orders, the predominant response appears in high- and low-energy regions, which correspond to characteristic energy scales of electron correlation and kinetic energy, respectively. Meanwhile, in the case of the toroidal order, the response is spread over both energy regions with relatively suppressed intensity. Careful analysis on the interband contributions showed that these distinct optical Hall responses are rooted in the optical selection rules coming from the interplay between the antisymmetric spin-orbit coupling and the underlying cluster multipole ordering.

Our results indicate that the careful investigation of the optical Hall conductivity would be helpful to probe and distinguish the magnetic multipole orders in experiments. It would also be interesting to extend our study to electric multipoles, which are often more difficult to detect compared to the magnetic ones. While our model includes the essential ingredients for the spin-orbit coupled metals, further realistic models would be necessary to discuss candidate materials, such as UNi₂B [6, 13, 22], Cd₂Re₂O₇ [23, 24], and PbRe₂O₆ [25]. Our work would serve as a starting point for such future studies.

ACKNOWLEDGMENTS

T.S. and Y.S. were supported by the Japan Society for the Promotion of Science through Program for Leading Graduate Schools (MERIT). Y.S. was also supported by the Japan Society for the Promotion of Science through a research fellowship for young scientists. This research was supported by Grant-in-Aid for Scientific Research Grants Number JP19H05822 and JST CREST (JP-MJCR18T2). T.S. and Y.U. contributed equally to this work.

Appendix: Layered honeycomb lattice

In order to show the generality of the optical selection rules, we calculate the optical Hall conductivity \( \sigma_{\mu z}(\omega) \) in Eq. (13) for the layered honeycomb lattice schemati-

FIG. 8. Schematics of (a) a perspective view and (b) a top view of the layered honeycomb lattice. In (a), the transfer integrals \( t \) and \( t_z \) are shown. \( a \) and \( c \) are the lattice constants in and out of the plane, respectively. In (b), the dotted hexagon indicates the six-sublattice magnetic unit cell. The gray arrows denote the directions of \( D_l \) at each sublattice; see Eqs. (2) and (A.1). (c)-(e) Schematics of cluster-type magnetic multipoles composed of six magnetic dipoles (red arrows): (c) monopole (d) quadrupole-type, and (e) toroidal.
A.2 shows the optical Hall conductivities $\sigma^{yz}(\omega)$ and (b),(d),(f) $\sigma^{zz}(\omega)$ as functions of the energy $\omega$ in the presence of the multipole orders of (a),(b) monopole, (c),(d) quadrupole-type, and (e),(f) toroidal. The green solid (black dashed) line indicates the real (imaginary) part of the optical Hall conductivities. The results are obtained at $a = c = 1$, $t = t_z = 1$, $D = 0.5$, $M = 8$, $k_BT = 0.1$, and $\delta = 0.02$. The electron filling is set at $n_e = 0.04$. The magnetic field is applied along the $x$ and $y$ direction for $\sigma^{yz}(\omega)$ and $\sigma^{zz}(\omega)$ as $B = (0.5, 0.0)$ and $B = (0.0, 0.5)$, respectively.

By adopting a similar Hamiltonian to Eq. (1), in Eq. (2), we consider two types of transfer integrals: the intralayer one $t$ and the interlayer one $t_z$ [see Fig. 8(a)]. All of the other transfer integrals between further-neighbor sites are assumed to be zero. Note that despite an uniform bond length $a$ and the uniform transfer integral $t$ within each layer, spatial inversion symmetry is broken at each lattice site in the honeycomb case, in contrast to the square case in Fig. 1. We choose $\theta_l^0$ in Eq. (7) as

$$\theta_l^0 = \frac{\pi}{3} n_l,$$

and $\theta_l^M$ in Eq. (9) as

$$\theta_l^M = \frac{\pi}{3} n_l \quad \text{for monopole},$$

$$\theta_l^M = -\frac{\pi}{3} n_l \quad \text{for quadrupole-type},$$

$$\theta_l^M = \frac{\pi}{3} n_l + \frac{\pi}{2} \quad \text{for toroidal},$$

where $n_l = 0, 1, 2, 3, 4, 5$ identify the six sublattices in the magnetic unit cell surrounded by the dashed hexagon in Fig. 8(b). The directions of $D_l$ specified by Eq. (A.1) are shown by the gray arrows in Fig. 8(b), and those of $M_l$ specified by Eqs. (A.2)–(A.4) are displayed by the red arrows in Figs. 8(c)–8(e), respectively. The model is an extension of that considered in Ref. [6].

Figure 9 shows the optical Hall conductivities $\sigma^{yz}(\omega)$ and $\sigma^{zz}(\omega)$ in a magnetic field applied along the $x$ and $y$ axis, respectively, as functions of the energy $\omega$ for the (a),(b) monopole, (c),(d) quadrupole-type, and (e),(f) toroidal orders. The electron filling $n_e$ is set to 0.04 so that the chemical potential lies in the lowest two bands. We find that $\sigma^{yz}(\omega)$ and $\sigma^{zz}(\omega)$ show distinct $\omega$ dependence for different types of the multipole orders in a similar manner to the square lattice case in Sec. III B.

In contrast, the trend is common to those in Fig. 11 which supports that similar optical selection rules to those discussed in Sec. IV are applicable to this honeycomb case.
[1] Y. Kuramoto, H. Kusunose, and A. Kiss, J. Phys. Soc. Jpn. 78, 072001 (2009).
[2] P. Santini, S. Carretta, G. Amoretti, R. Caciullo, N. Magnani, and G. H. Lander, Rev. Mod. Phys. 81, 807 (2009).
[3] C. Ederer and N. A. Spaldin, Phys. Rev. B 76, 214404 (2007).
[4] N. A. Spaldin, M. Fiebig, and M. Mostovoy, J. Phys.: Condens. Matter 20, 434203 (2008).
[5] Y. Yanase, J. Phys. Soc. Jpn. 83, 014703 (2014).
[6] S. Hayami, H. Kusunose, and Y. Motome, Phys. Rev. B 90, 024432 (2014).
[7] M.-T. Suzuki, T. Koretsune, M. Ochi, and R. Arita, Phys. Rev. B 95, 094406 (2017).
[8] Y. Gao, D. Vanderbilt, and D. Xiao, Phys. Rev. B 97, 134423 (2018).
[9] S. Hayami, M. Yatsushiro, Y. Yanagi, and H. Kusunose, Phys. Rev. B 98, 165110 (2018).
[10] H. Watanabe and Y. Yanase, Phys. Rev. B 98, 245129 (2018).
[11] M.-T. Suzuki, T. Nomoto, R. Arita, Y. Yanagi, S. Hayami, and H. Kusunose, Phys. Rev. B 99, 174407 (2019).
[12] B. B. V. Aken, J.-P. Rivera, H. Schmid, and M. Fiebig, Nature (London) 449, 702 (2007).
[13] H. Saito, K. Uenishi, N. Miura, C. Tabata, H. Hidaka, T. Yanagisawa, and H. Amitsuka, J. Phys. Soc. Jpn. 87, 033702 (2018).
[14] M. Shinozaki, G. Motoyama, M. Tsubouchi, M. Sezaki, J. Gouchi, S. Nishigori, T. Mutou, A. Yamaguchi, K. Fujiwara, K. Miyoshi, and Y. Uwatoko, J. Phys. Soc. Jpn. 89, 033703 (2020).
[15] S. Nakatsuji, N. Kiyohara, and T. Higo, Nature (London) 527, 212 (2015).
[16] M. Ikhlas, T. Tomita, T. Koretsune, M.-T. Suzuki, D. Nishio-Hamane, R. Arita, Y. Otani, and S. Nakatsuji, Nat. Phys. 13, 1085 (2017).
[17] T. Higo, H. Man, D. B. Gopman, L. Wu, T. Koretsune, O. M. van’t Erve, Y. P. Kabanov, D. Rees, Y. Li, M.-T. Suzuki, S. Patankar, M. Ikhlas, C. L. Chien, R. Arita, R. D. Shull, J. Orenstein, and S. Nakatsuji, Nat. Photon. 12, 73 (2018).
[18] K. Kimura, P. Babkevich, M. Sera, M. Toyoda, K. Yamamura, G. S. Tucker, J. Martius, T. Fennell, P. Manuel, D. D. Khalyavin, R. D. Johnson, T. Nakano, Y. Nozue, H. M. Rønnow, and T. Kimura, Nat. Commun. 7, 13039 (2016).
[19] Y. Kato, K. Kimura, A. Miyake, M. Tokunaga, A. Matsuo, K. Kindo, M. Akaki, M. Hagiwara, M. Sera, T. Kimura, and Y. Motome, Phys. Rev. Lett. 118, 107601 (2017).
[20] K. Kimura, Y. Kato, K. Yamamura, A. Miyake, M. Tokunaga, A. Matsuo, K. Kindo, M. Akaki, M. Hagiwara, S. Kimura, M. Toyoda, Y. Motome, and T. Kimura, Phys. Rev. Mater. 2, 104415 (2018).
[21] Y. Kato, K. Kimura, A. Miyake, M. Tokunaga, A. Matsuo, K. Kindo, M. Akaki, M. Hagiwara, S. Kimura, T. Kimura, and Y. Motome, Phys. Rev. B 99, 024415 (2019).
[22] S. A. M. Mentink, A. Drost, G. J. Nieuwenhuys, E. Friekke, A. A. Menovsky, and J. A. Mydosh, Phys. Rev. Lett. 73, 1031 (1994).
[23] J.-i. Yamaura and Z. Hiroi, J. Phys. Soc. Jpn. 71, 2598 (2002).
[24] J.-i. Yamaura, K. Takeda, Y. Ikeda, N. Hirao, Y. Ohishi, T. C. Kobayashi, and Z. Hiroi, Phys. Rev. B 95, 020102(R) (2017).
[25] Z. Hiroi, J.-i. Yamaura, T. C. Kobayashi, Y. Matsubayashi, and D. Hirai, J. Phys. Soc. Jpn. 87, 024702 (2018).
[26] S. Hayami, Y. Yanagi, H. Kusunose, and Y. Motome, Phys. Rev. Lett. 122, 147602 (2019).
[27] S. Tajima, D. Hirai, T. Yajima, D. Nishio-Hamane, Y. Matsubayashi, and Z. Hiroi, J. Solid State Chem. 288, 121359 (2020).