



Spontaneous antisymmetric spin splitting in noncollinear antiferromagnets without spin-orbit coupling

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(Received 23 December 2019; revised manuscript received 11 March 2020; accepted 13 May 2020; published 2 June 2020)

We propose a realization of an antisymmetric spin-split band structure through magnetic phase transitions without spin-orbit coupling. It enables us to utilize it for a variety of magnetic-order-driven cross-correlated and nonreciprocal transport phenomena as similar to those in spin-orbit-coupling oriented systems. We unveil its general condition as an emergence of a bond-type magnetic toroidal multipole (polar tensor) in a triangular unit with noncollinear 120° antiferromagnetic structures. By using the concept of augmented multipoles, we systematically analyze the phenomena in terms of an effective multipole coupling. Our multipole description is ubiquitously applied to any trigonal and hexagonal structures including triangular, kagome, and breathing kagome structures, and provides a way to design and engineer materials with a giant antisymmetric spin splitting and its physical responses even without spin-orbit coupling.

DOI: [10.1103/PhysRevB.101.220403](https://doi.org/10.1103/PhysRevB.101.220403)

Antisymmetric spin splitting in electronic band structures, which is an opposite spin polarization at opposite wave vectors, has drawn considerable interest in noncentrosymmetric materials, since it is a fundamental origin of rich spintronic functionalities, nonreciprocal transports, and magnetoelectric effects [1–3]. It is typically realized in polar materials with a relatively large spin-orbit coupling (SOC), for instance, the nonmagnetic Rashba compound BiTeI [4–6] and monolayer transition-metal dichalcogenides MX_2 ($M = \text{Mo, W}$ and $X = \text{S, Se}$) [7–10].

Even though a crystal structure is centrosymmetric, a magnetic transition actualizes the antisymmetric spin splitting by an interplay between the kinetic motion of electrons and the magnetic structure via SOC [11,12]. A spiral magnetic order is a typical example, which induces a linear magnetoelectric effect in the presence of nonzero vector spin chirality [13–15]. Another example is found in CoNb_3S_6 and $\text{Co}_4\text{Nb}_2\text{O}_9$ showing giant anomalous Hall and angle-dependent magnetoelectric effects [16–21], respectively. It is emphasized that the emergent antisymmetric spin splitting through the magnetic phase transition is more flexibly controllable, i.e., the spin splitting driven by magnetic orders can be varied or even switched on and off by external fields, pressure, and temperature. The complex interplay can be understood in a transparent manner by introducing the concept of an augmented multipole [22–24].

Since the above fascinating phenomena usually rely on the presence of SOC, candidate materials are limited to those constituted by moderately heavier elements in a crystal structure under low space-group symmetry. Such a limitation motivates a search for an alternative mechanism to exhibit spin splitting without relying on SOC. This can be done by considering appropriate magnetic structures which break

crystalline symmetry in addition to time-reversal symmetry [15,25,26]. For example, a collinear-type antiferromagnetic (AFM) phase transition in a nonsymmorphic organic compound, $\kappa\text{-(BETD-TTF)}_2\text{Cu[N(CN)}_2\text{]Cl}$ [27,28], and a distorted tetragonal compound, RuO_2 [29,30], result in spin-current generation. However, in the absence of SOC it is proven that the collinear magnetic order leads only to *symmetric* spin splitting in momentum space even with broken spatial inversion symmetry due to $\text{SU}(2)$ symmetry in spin space [26,27].

In the present Rapid Communication, we propose a realization of *antisymmetric* spin splitting by focusing on a triangular unit with noncollinear 120° -AFM structures, and clarify the microscopic conditions for the emergent spin splitting from a general point of view by introducing a multipole description [23,31]. The condition we found is that the magnetic toroidal (MT) multipoles are present in the hopping Hamiltonian and they couple with the noncollinear AFM order parameters within the same irreducible representation in a high-temperature series expansion. We also predict possible cross-correlated and nonreciprocal transport phenomena in terms of an effective coupling among multipole degrees of freedom, which can be modified by an external magnetic field, for instance.

Our multipole description is ubiquitously applied to any trigonal and hexagonal structures including the triangular, kagome, and breathing kagome structures. Our proposal is demonstrated for the trigonal noncollinear AFM $\text{Ba}_3\text{MnNb}_2\text{O}_9$ based on the density-functional theory (DFT) calculation. The present mechanism provides potentially gigantic antisymmetric spin splitting due to its kinetic-motion origin without SOC, which can be directly detected in spin- and angle-resolved photoemission spectroscopy.

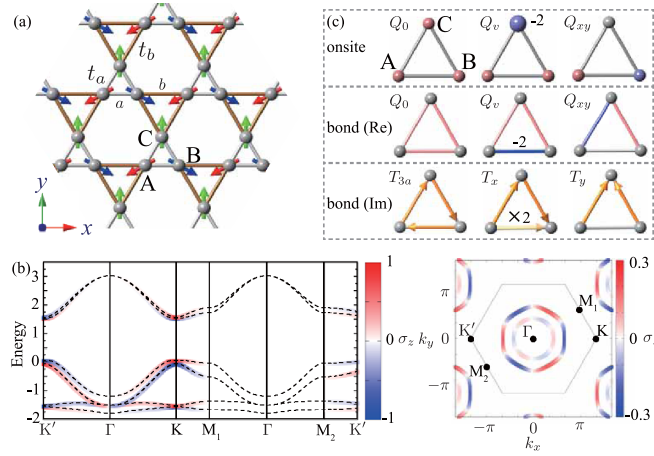


FIG. 1. (a) Noncollinear 120° -AFM pattern in the breathing kagome structure. (b) The band structure of the model in Eq. (1) at $t_a = 1$, $t_b = 0.5$, and $m = 0.3$ (left panel) and the isoenergy surfaces at $\mu = -1$ (right panel). The dashed lines show the band dispersions and the color map shows the spin polarization of the z components at each wave vector. (c) Schematic pictures of the triangular-unit multipoles. The red (blue) circles represent the positive (negative) on-site potential, and the red (blue) lines and orange arrows on each bond represent the positive (negative) real and imaginary hoppings, respectively. The gray lines represent no hoppings.

We start by considering a breathing kagome system with the noncollinear 120° -AFM structure in Fig. 1(a), which is an intuitive example showing an antisymmetric spin splitting. The positions of the three sublattice sites are defined by $\mathbf{r}_A = (0, 0, 0)$, $\mathbf{r}_B = a(1, 0, 0)$, and $\mathbf{r}_C = a(1/2, \sqrt{3}/2, 0)$ and the lattice constant $a + b$ is set to be unity. The tight-binding Hamiltonian is given by

$$\mathcal{H} = \left(t_a \sum_{\sigma, \langle ij \rangle}^{\in \Delta} + t_b \sum_{\sigma, \langle ij \rangle}^{\in \nabla} \right) c_{i\sigma}^\dagger c_{j\sigma} + \sum_{i\sigma\sigma'} \mathbf{m}_i \cdot c_{i\sigma}^\dagger \boldsymbol{\sigma}_{\sigma\sigma'} c_{i\sigma'}, \quad (1)$$

where $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) is the creation (annihilation) operator for site i and spin $\sigma = \uparrow, \downarrow$. The first term represents the hoppings within upward triangles t_a and downward triangles t_b . The second term represents the mean-field term corresponding to the magnetic order. We assume a noncollinear three-sublattice 120° -AFM structure in the xy plane with the order-parameter amplitude m , i.e., $\mathbf{m}_A = m(-\sqrt{3}/2, -1/2, 0)$, $\mathbf{m}_B = m(\sqrt{3}/2, -1/2, 0)$, and $\mathbf{m}_C = m(0, 1, 0)$ in Fig. 1(a), which can be naturally stabilized by the frustrated exchange interactions in the triangle unit, e.g., in the single-band Hubbard model on a triangular lattice [32]. We also consider the presence of the implicit small magnetic anisotropy due to magnetic dipole-dipole interactions and/or the SOC for the stabilization of the in-plane 120° -AFM structure. Regardless of these stabilization mechanisms, the following properties are accounted for by the simple model in Eq. (1).

Figure 1(b) shows the band structure at $t_a = 1$, $t_b = 0.5$, and $m = 0.3$ where the color map shows the spin polarization of the z component. The results clearly exhibit antisymmetric z -component spin polarization despite the AFM structure in the xy plane; the spin polarization along the Γ - K line is opposite to that along the Γ - K' line, while no spin polarization

is found in the Γ - $M_{1,2}$ line. The isoenergy surfaces at the chemical potential $\mu = -1$ in the right panel in Fig. 1(b) indicate that the antisymmetric spin polarization keeps the threefold rotational symmetry.

This threefold out-of-plane antisymmetric spin splitting has a close resemblance to that observed in monolayer dicalcogenides with SOC, which is the so-called Ising-type spin splitting [33–35]. However, the microscopic origin is totally different, i.e., the present case is of non-SOC origin, although the resultant antisymmetric spin splitting becomes a source of various cross-correlated and transport phenomena, such as the magnetoelectric effect and nonreciprocal transport, as discussed below. The out-of-plane antisymmetric spin splitting can be detected by using spin- and angle-resolved photoemission spectroscopy [36].

The origin of the antisymmetric spin splitting can be intuitively captured by applying the multipole description to the model in Eq. (1) [23,26], as the type of an additional crystalline symmetry breaking to the time-reversal symmetry is essential. To demonstrate this, first we introduce the triangular unit with three sublattices A–C, and define the nine multipole degrees of freedom as shown in Fig. 1(c). Then, the spinless hopping matrix in the three-sublattice basis can be spanned by these multipoles. Moreover, the mean-field magnetic structure is also described by the multipoles, which are known as cluster multipoles [37,38].

The Hamiltonian in Eq. (1) is Fourier transformed with respect to the unit cell as

$$\mathcal{H} = \sum_{\mathbf{k}\sigma\sigma' l l'} c_{\mathbf{k}l\sigma}^\dagger \left[\delta_{\sigma\sigma'} (H_l^Q + H_l^T)^{ll'} + \delta_{ll'} H_m^{\sigma\sigma'} \right] c_{\mathbf{k}l'\sigma'}, \quad (2)$$

where $c_{\mathbf{k}l\sigma}^\dagger$ ($c_{\mathbf{k}l\sigma}$) is the Fourier transform of $c_{i\sigma}^\dagger$ ($c_{i\sigma}$) at wave vector \mathbf{k} and sublattice l . H_l^Q and H_l^T stand for the real and imaginary hopping matrices, respectively, which comes from the first term in Eq. (1), and H_m is the mean-field matrix from the second term in Eq. (1). The matrices H_l^Q , H_l^T , and H_m are decomposed in terms of the triangular-unit multipoles defined in Table I as

$$\begin{aligned} H_l^Q &= Q_0(\mathbf{k})\hat{Q}_0^{(1)} + Q_v(\mathbf{k})\hat{Q}_v^{(1)} + Q_{xy}(\mathbf{k})\hat{Q}_{xy}^{(1)}, \\ H_l^T &= T_{3a}(\mathbf{k})\hat{T}_{3a}^{(1)} + T_x(\mathbf{k})\hat{T}_x^{(1)} + T_y(\mathbf{k})\hat{T}_y^{(1)}, \\ H_m &= -m(\hat{Q}_{xy}^{(0)}\hat{\sigma}_x + \hat{Q}_v^{(0)}\hat{\sigma}_y). \end{aligned} \quad (3)$$

Here, three on-site potentials and three bonds with real hoppings are described by a linear combination of the electric monopole $\hat{Q}_0^{(n)}$ and two electric quadrupoles ($\hat{Q}_v^{(n)}$, $\hat{Q}_{xy}^{(n)}$), whereas three bonds with imaginary hoppings by two MT dipoles ($\hat{T}_x^{(1)}$, $\hat{T}_y^{(1)}$) and a MT octupole $\hat{T}_{3a}^{(1)}$, where the superscripts $n = 0$ and 1 stand for on-site and bond indices, respectively [23,26]. Note that the MT multipoles are defined as the bond degree of freedom in contrast to the conventional definition by the vector products of the spin (orbital) angular momentum and the position vector, where both definitions share the same symmetry properties. [39–42]. We use the standard Gell-Mann matrices to express each multipole in Table I, and their schematic pictures are shown in Fig. 1(c). Each multipole is normalized as $\text{Tr}[\hat{X}^2] = 1$. By using the molecular-orbital basis in the triangular unit [26,43], we identify the symmetry of each multipole as indicated by

TABLE I. Multipole degrees of freedom in the triangular unit. The on-site potentials and nearest-neighbor hoppings are described by these multipoles. $\hat{\lambda}_\alpha$ ($\alpha = 0-8$) are the Gell-Mann matrices. We use the abbreviated notations, $\tilde{k}_x = k_x/2$, $\tilde{k}_y = \sqrt{3}k_y/2$, $p_a = t_a$, and $p_b = -t_b$.

Electric	\hat{Q}_0	\hat{Q}_v	\hat{Q}_{xy}
On site	$\frac{1}{\sqrt{3}}\hat{\lambda}_0$	$\frac{1}{\sqrt{2}}\hat{\lambda}_8$	$\frac{1}{\sqrt{2}}\hat{\lambda}_3$
Real bond	$\frac{1}{\sqrt{6}}(\hat{\lambda}_1 + \hat{\lambda}_4 + \hat{\lambda}_6)$	$\frac{\sqrt{3}}{6}(\hat{\lambda}_4 + \hat{\lambda}_6 - 2\hat{\lambda}_1)$	$\frac{1}{2}(-\hat{\lambda}_4 + \hat{\lambda}_6)$
Form factor	$\sqrt{\frac{2}{3}}\sum_\eta t_\eta (\cos k_x \eta + 2 \cos \tilde{k}_x \eta \cos \tilde{k}_y \eta)$	$\frac{2}{\sqrt{3}}\sum_\eta t_\eta (\cos \tilde{k}_x \eta \cos \tilde{k}_y \eta - \cos k_x \eta)$	$2\sum_\eta t_\eta \sin \tilde{k}_x \eta \sin \tilde{k}_y \eta$
Magnetic	\hat{T}_{3a}	\hat{T}_x	\hat{T}_y
Imaginary bond	$\frac{1}{\sqrt{6}}(\hat{\lambda}_2 - \hat{\lambda}_5 + \hat{\lambda}_7)$	$\frac{\sqrt{3}}{6}(\hat{\lambda}_7 - \hat{\lambda}_5 - 2\hat{\lambda}_2)$	$-\frac{1}{2}(\hat{\lambda}_5 + \hat{\lambda}_7)$
Form factor	$-\sqrt{\frac{2}{3}}\sum_\eta p_\eta (\sin k_x \eta - 2 \sin \tilde{k}_x \eta \cos \tilde{k}_y \eta)$	$\frac{2}{\sqrt{3}}\sum_\eta p_\eta (\sin k_x \eta + \sin \tilde{k}_x \eta \cos \tilde{k}_y \eta)$	$2\sum_\eta p_\eta \cos \tilde{k}_x \eta \sin \tilde{k}_y \eta$

the subscript. The linear coefficients, the electric and MT multipoles $Q(\mathbf{k})$ and $T(\mathbf{k})$, represent the form factors, which are even and odd functions of \mathbf{k} , respectively. Note that their \mathbf{k} dependences are consistent with the general definition of multipoles in momentum representation [23].

In the multipole description, the active odd-rank MT bond multipoles (imaginary hoppings) can become the origin of the antisymmetric spin splitting, once the effective coupling between them and the mean-field multipoles is activated under spontaneous magnetic orders [44]. Such an effective coupling is systematically obtained from the high-temperature expansion of the quantity at wave vector \mathbf{k} ,

$$\text{Tr}\left[e^{-\beta\hat{H}_k}\hat{\sigma}_\mu\right] = \sum_s \frac{(-\beta)^s}{s!} g_s^\mu(\mathbf{k}), \quad (4)$$

where $\mu = 0, x, y, z$, $\hat{H} = \sum_{\mathbf{k}} \hat{H}_{\mathbf{k}}$, and β is the inverse temperature. By means of the s th-order expansion coefficient of the μ component, $g_s^\mu(\mathbf{k})$, the corresponding effective multipole coupling is given by $g_s^\mu(\mathbf{k})\hat{\sigma}_\mu/2$.

The contribution to the antisymmetric spin splitting for the z component is obtained at the fifth order in Eq. (4) as

$$g_5^z(\mathbf{k}) = \sqrt{\frac{2}{3}}m^2 \left\{ Q_0(\mathbf{k})[Q_{xy}(\mathbf{k})T_y(\mathbf{k}) - Q_v(\mathbf{k})T_x(\mathbf{k})] \right. \\ \left. + Q_0^2(\mathbf{k})T_{3a}(\mathbf{k}) + \frac{1}{3\sqrt{2}}T_x(\mathbf{k})[T_x^2(\mathbf{k}) - 3T_y^2(\mathbf{k})] \right\}. \quad (5)$$

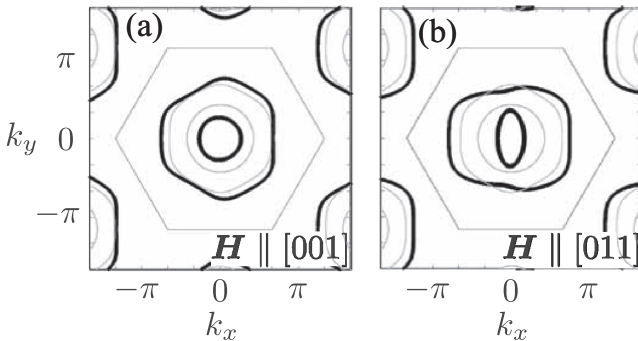


FIG. 2. The band deformations in the breathing kagome AFM at $|\mathbf{H}| = 0.3$ along (a) [001] and (b) [011] directions. The thin gray lines represent the isoenergy surfaces at $|\mathbf{H}| = 0$.

Around $\mathbf{k} = \mathbf{0}$, Eq. (5) reduces to $-m^2 t_a t_b (t_a - t_b) k_x (k_x^2 - 3k_y^2)(a + b)^3/2$, which captures the qualitative behavior of the antisymmetric spin splitting in Fig. 1(b). It provides microscopic ingredients about the antisymmetric spin splitting. The first is that giant antisymmetric spin splitting could occur in the strong interaction regime, since the mean field m in Eq. (3) is proportional to the repulsive interaction in the Hubbard model. The second is that the spin splitting is proportional to the square of the order parameter m^2 , which implies that the two spin components, i.e., the noncollinear spin structure, is necessary to induce spin splitting. Moreover, m^2 dependence indicates the AFM domain formation is irrelevant to this spin splitting, although the opposite chirality reverses the sign in Eq. (5). The third is that the spin splitting occurs for $t_a \neq 0$, $t_b \neq 0$, and $t_a \neq t_b$: The breathing structure (a -, b -bond inequivalency) is important.

Furthermore, the effective multipole coupling in Eq. (5) is a source of multiferroic responses, since each multipole is related to specific response tensors [23]. For example, the effective coupling $Q_0^2(\mathbf{k})T_{3a}(\mathbf{k})\hat{\sigma}_z \sim k_x(k_x^2 - 3k_y^2)\hat{\sigma}_z$ in Eq. (5) implies that a spontaneous threefold rotational nonreciprocity is induced by a magnetic field along the z direction if one divides it as $k_x(k_x^2 - 3k_y^2) \times \hat{\sigma}_z$. Similarly, the spin current along the x direction with the z -spin component is expected by the $(x^2 - y^2)$ -type strain field by dividing the effective coupling $Q_v(\mathbf{k})T_x(\mathbf{k})\hat{\sigma}_z$ as $k_x \hat{\sigma}_z \times (k_x^2 - k_y^2)$.

The analyses are straightforwardly extended to include an external magnetic field with the Zeeman coupling, $-\mathbf{H} \cdot$

TABLE II. Effective multipole couplings under an external magnetic field [45]. The lowest-order band deformations $\mathbf{g}_s(\mathbf{k}) \cdot \hat{\sigma}$, the wave-vector \mathbf{k} dependences around $\mathbf{k} = \mathbf{0}$, and relevant physical responses are shown, where \mathbf{Q} and \mathbf{T} are the electric polarization and magnetic toroidalization. ME and NR indicate magnetoelectric and nonreciprocal responses, respectively.

\mathbf{H}	$\mathbf{g}_s(\mathbf{k}) \cdot \hat{\sigma}$	$\mathbf{k} \rightarrow \mathbf{0}$ limit	Response
[100]	$m^3 H_x Q_{xy}(\mathbf{k}) T_{3a}(\mathbf{k}) \hat{\sigma}_z$	$k_y^5 \hat{\sigma}_z \sim Q_x$	ME
[010]	$m^3 H_y Q_v(\mathbf{k}) T_{3a}(\mathbf{k}) \hat{\sigma}_z$	$k_x^5 \hat{\sigma}_z \sim Q_y$	ME
[001]	$m^2 H_z T_{3a}(\mathbf{k}) \hat{\sigma}_0(\mathbf{k})$	$k_x(k_x^2 - 3k_y^2) \hat{\sigma}_0 \sim T_{3a}$	NR
	$m^3 H_z (Q_{xy}(\mathbf{k}) T_{3a}(\mathbf{k}) \hat{\sigma}_x$	$k_x^5 \hat{\sigma}_y - k_y^5 \hat{\sigma}_x \sim Q_z$	ME
	$+ Q_v(\mathbf{k}) T_{3a}(\mathbf{k}) \hat{\sigma}_y)$		
[011]	$m^3 H_y H_z Q_v(\mathbf{k}) T_{3a}(\mathbf{k}) \hat{\sigma}_0$	$k_x^5 \hat{\sigma}_0 \sim T_x$	NR
[101]	$m^3 H_x H_z Q_{xy}(\mathbf{k}) T_{3a}(\mathbf{k}) \hat{\sigma}_0$	$k_y^5 \hat{\sigma}_0 \sim T_y$	NR

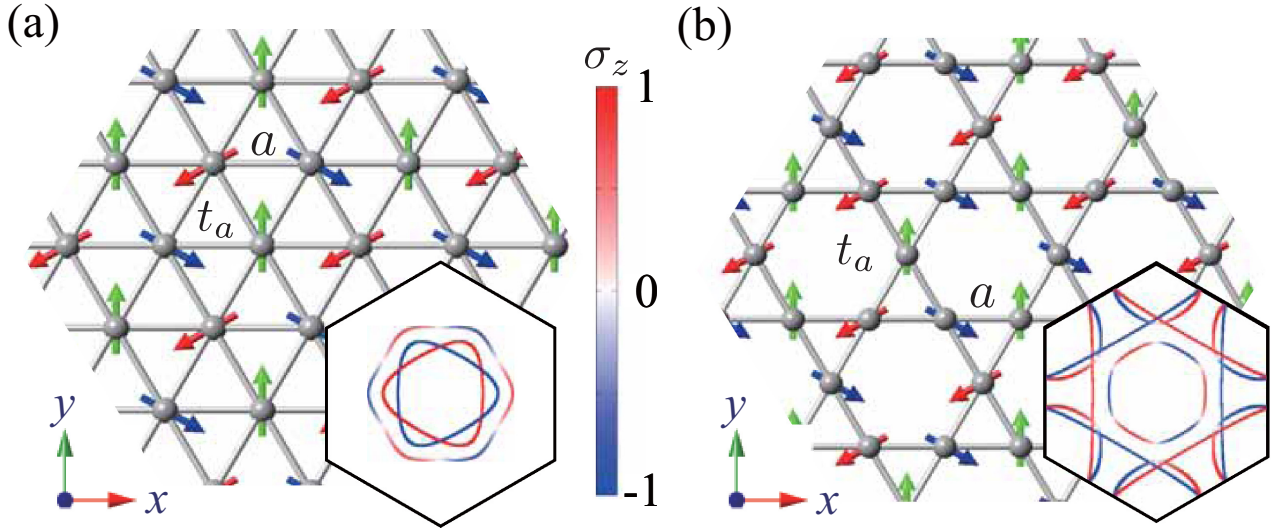


FIG. 3. Schematic pictures of the 120° AFM on (a) triangular and (b) kagome lattices with the $\sqrt{3} \times \sqrt{3}$ structures. In the inset, corresponding isoenergy surfaces where the contour shows the z -spin component are presented. The model parameters are given by (a) $t_a = 1$, $m = 0.5$, and $\mu = -2.5$ and (b) $t_a = 1$, $m = 0.5$, and $\mu = 0$.

$\sum_{i\sigma\sigma'} c_{i\sigma}^\dagger \sigma_{\sigma\sigma'} c_{i\sigma'}$, yielding a rich variety of band deformations depending on the field direction. For $\mathbf{H} \parallel [100]$, the directional antisymmetric spin splitting with $k_y^5 \hat{\sigma}_z$ is induced by the coupling between \hat{T}_{3a} , \hat{Q}_{xy} , and $\hat{\sigma}_z$. This band deformation describes the emergent magnetoelectric (ME) effect where the electric polarization along the x direction, Q_x , is induced by H_x , since $k_y^5 \hat{\sigma}_z$ is the same symmetry as Q_x [23]. In a similar way, different ME couplings are obtained for $\mathbf{H} \parallel [010]$ and $\mathbf{H} \parallel [001]$: the $k_x^5 \hat{\sigma}_z$ -type band deformation corresponding to Q_y for $\mathbf{H} \parallel [010]$ and the $k_x^5 \hat{\sigma}_y - k_y^5 \hat{\sigma}_x$ -type band deformation corresponding to Q_z for $\mathbf{H} \parallel [001]$. Thus, the 120°-AFM order in the breathing kagome system exhibits the longitudinal ME effect ($\mathbf{Q} \parallel \mathbf{H}$). Note that qualitatively similar results are also obtained by the symmetry analysis based on the cluster multipole theory [38], although our approach is apparent for microscopic conditions to induce antisymmetric spin splitting in a systematic way. We summarize the effective coupling and relevant responses under the magnetic fields in Table II [45].

Interestingly, spin-independent antisymmetric band deformations are realized when the magnetic field is applied along the z direction as shown in Fig. 2(a), where the effective multipole coupling is expressed as $m^2 H_z T_{3a}(\mathbf{k}) \hat{\sigma}_0 \sim m^2 H_z k_x (k_x^2 - 3k_y^2) \hat{\sigma}_0$. This type of antisymmetric band deformation becomes a microscopic source of angle-dependent nonreciprocal transport. Moreover, when \mathbf{H} is rotated from [001] to [011], the additional contribution $k_x^5 \hat{\sigma}_0$ appears due to the effective multipole coupling as $m^3 H_y H_z Q_v(\mathbf{k}) T_{3a}(\mathbf{k}) \hat{\sigma}_0$ [Fig. 2(b)], which means that the magnetic field can induce the MT dipole T_x . Similar nonreciprocal dispersions have been studied in the localized spin model [46,47].

So far, we have considered a specific breathing kagome structure. Similar analyses can be directly applied to any other systems with a triangular unit, such as the triangular and kagome systems. For example, as the multipoles $\hat{Q}_0^{(1)}$ and $\hat{T}_{3a}^{(1)}$ are active in the three-sublattice 120°-AFM order on a triangular lattice, antisymmetric spin splitting is expected

[Fig. 3(a)]. Note that there are no additional antisymmetric band deformations induced by the magnetic field, since there are no active $\hat{Q}_v^{(1)}$ and $\hat{Q}_{xy}^{(1)}$ (see also Table II). The nearly 120°-AFM materials, such as CsFeCl₃ [48] and PdCrO₂ [49,50], are candidates to exhibit antisymmetric spin splitting. In a similar way, antisymmetric spin splitting is expected for the $\sqrt{3} \times \sqrt{3}$ AFM order on a simple kagome structure, where only $\hat{Q}_0^{(1)}$ and $\hat{T}_{3a}^{(1)}$ are active multipoles as shown in Fig. 3(b). Moreover, the lower-symmetry trigonal material also shows spin-split physics, such as trimer and triangular tube magnets, LuFeO₃ [51] and CsCrF₄ [52,53], which possesses the same active multipoles as the breathing kagome structure.

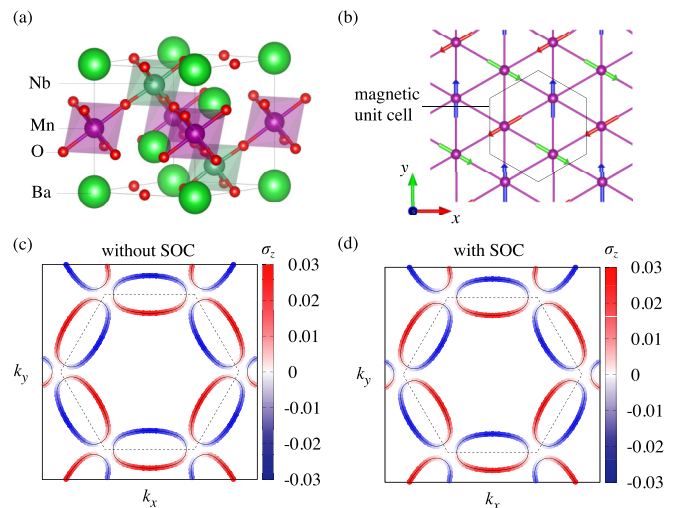


FIG. 4. (a) Crystal and (b) magnetic structures of Ba₃MnNb₂O₉. The isoenergy surfaces on the $k_z = 0$ plane at $\mu = -0.05$ eV (c) without and (d) with SOC in the AFM state, where the top of the valence band is set to 0 eV.

Finally, we demonstrate emergent antisymmetric spin splitting in $\text{Ba}_3\text{MnNb}_2\text{O}_9$ [54]. This compound belongs to the trigonal space group $P\bar{3}m1$ (No. 164), and the high-spin state ($S = 5/2$) of Mn^{2+} ions exhibits the 120° -AFM structure with out-of-plane cantings on the triangular lattice at low temperatures as shown in Figs. 4(a) and 4(b) [54]. We calculate the expected AFM band structures of $\text{Ba}_3\text{MnNb}_2\text{O}_9$ with and without SOC based on the DFT with the generalized gradient approximation plus U method [55,56] by using the Vienna *ab initio* simulation package (VASP) [57,58], where we employ the projector augmented-wave (PAW) potentials [59,60] and set $U = 3.0$ eV for Mn $3d$ orbitals according to a previous study [54]. Figure 4(c) shows isoenergy surfaces without SOC projected onto the σ_z component in the AFM state at zero magnetic field. The results are consistent with the analysis in the simple triangular AFM in Fig. 3(a), i.e., the antisymmetric z -spin polarization in the form of $k_y(3k_x^2 - k_y^2)$. Note that the SOC for the Mn $3d$ orbitals is small and does not have any significant impact on the antisymmetric spin splitting as shown in Fig. 4(d). In addition, we also confirmed that the isoenergy surfaces are deformed antisymmetrically for $\mathbf{H} \parallel [001]$. Therefore, this compound can be an archetypal example of the antisymmetric SOC physics induced by a noncollinear magnetic ordering.

In summary, we clarified the general conditions for emergent antisymmetric spin-split band structures in noncollinear magnets. The following three conditions are enough to obtain *antisymmetric* spin splitting in the band structure without SOC: (1) a triangular unit with a 120° -AFM structure, (2) inversion symmetry breaking, and (3) active MT multipoles (imaginary hopping) in the one-body Hamiltonian. We also demonstrated the origin of cross-correlated coupling and non-reciprocal transport is attributed to effective microscopic multipole couplings. As our analysis on the basis of the multipole description is ubiquitously applied to any systems with a triangular unit, the result will shed light on potential candidate materials with a giant spin splitting even without SOC.

This research was supported by JSPS KAKENHI Grants No. JP15H05885, No. JP18H04296 (J-Physics), No. JP18K13488, No. JP19K03752, No. JP19H01834, and No. JP20K05299. This work was also supported by the Toyota Riken Scholarship. DFT calculations were performed using the MAterial science Supercomputing system for Advanced MUlti-scale simulations towards NExt-generation-Institute for Materials Research (MASAMUNE-IMR) of the Center for Computational Materials Science, Institute for Materials Research, Tohoku University.

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