

Multipole Superconductivity in Nonsymmorphic Sr_2IrO_4

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Discoveries of marked similarities to high- T_c cuprate superconductors point to the realization of superconductivity in the doped $J_{\text{eff}} = 1/2$ Mott insulator Sr_2IrO_4 . Contrary to the mother compound of cuprate superconductors, several stacking patterns of in-plane canted antiferromagnetic moments have been reported, which are distinguished by the ferromagnetic components as $-+ +-$, $+++$, and $-+ -+$. In this paper, we clarify unconventional features of the superconductivity coexisting with $-+ +-$ and $-+ -+$ structures. Combining the group theoretical analysis and numerical calculations for an effective $J_{\text{eff}} = 1/2$ model, we show unusual superconducting gap structures in the $-+ +-$ state protected by nonsymmorphic magnetic space group symmetry. Furthermore, our calculation shows that the Fulde-Ferrell-Larkin-Ovchinnikov superconductivity is inevitably stabilized in the $-+ -+$ state since the odd-parity magnetic $-+ -+$ order makes the band structure asymmetric by cooperating with spin-orbit coupling. These unusual superconducting properties are signatures of magnetic multipole order in nonsymmorphic crystal.

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A layered perovskite 5d transition metal oxide Sr_2IrO_4 has attracted recent attention because a lot of similarities to the high-temperature cuprate superconductors have been recognized. For example, Sr_2IrO_4 (La_2CuO_4) has one hole per Ir (Cu) ion, and shows a pseudospin-1/2 antiferromagnetic order [1]. Moreover, recent experiments on electron-doped Sr_2IrO_4 indicate the emergence of a pseudogap [2–4] and at low temperatures a d -wave gap [5], which strengthens the analogy with cuprates. Furthermore, d -wave superconductivity in Sr_2IrO_4 by carrier doping is theoretically predicted by several studies [6–9]. Distinct differences of Sr_2IrO_4 from cuprates are large spin-orbit coupling and nonsymmorphic crystal structure, both of which attract interest in the modern condensed matter physics. In this Letter, we predict exotic superconducting properties in Sr_2IrO_4 unexpected in cuprates.

Below $T_N \approx 230$ K, an antiferromagnetic order develops in undoped Sr_2IrO_4 . Large spin-orbit coupling and rotation of octahedra lead to canted magnetic moments from the a axis and induce a small ferromagnetic moment along the b axis (Fig. 1). Several magnetic structures for stacking along the c axis have been reported in response to circumstances. The magnetic ground states determined by resonant x-ray scattering [10–12], neutron diffraction [13,14], and second-harmonic generation [15] are summarized in a recent theoretical work [16]. In the undoped compound, the ferromagnetic component shows the stacking pattern $-+ +-$ [10,11,13], as illustrated in Fig. 1. On the other hand, the $+++$ pattern is suggested as the magnetic structure of Sr_2IrO_4 in a magnetic field directed in the ab plane [10] and of Rh-doped $\text{Sr}_2\text{Ir}_{1-x}\text{Rh}_x\text{O}_4$ [12,14]. The recent observation [15], however, advocates the $-+ -+$ magnetic pattern indicating an intriguing odd-parity hidden order in Sr_2IrO_4 (see Fig. 1).

The crystal space group of Sr_2IrO_4 was originally reported as $I4_1/acd$ from neutron powder diffraction experiments [17,18]. Very recently, however, the crystal structure has been revealed by single-crystal neutron diffraction to be rather $I4_1/a$ [14]. In either case, the symmetry of Sr_2IrO_4 is globally centrosymmetric and nonsymmorphic. On the other hand, the site symmetry of the Ir site is S_4 lacking local inversion symmetry. In such noncentrosymmetric systems, antisymmetric spin-orbit coupling (ASOC) entangles various internal degrees of freedom, such as spin, orbital, and sublattice, namely

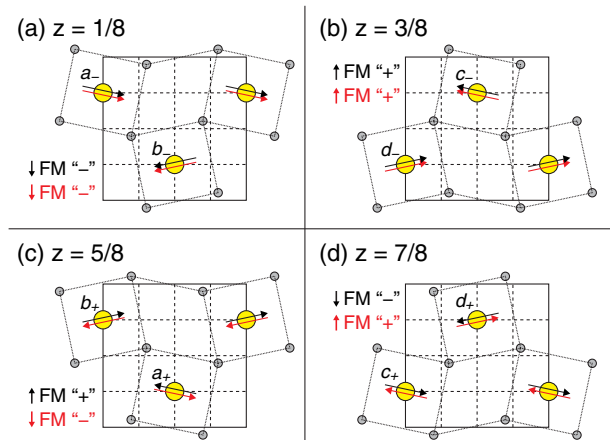


FIG. 1. Crystal and magnetic symmetries of Sr_2IrO_4 in the 4 IrO_2 planes: (a) $z = \frac{1}{8}$, (b) $z = \frac{3}{8}$, (c) $z = \frac{5}{8}$, and (d) $z = \frac{7}{8}$ [16]. The two magnetic patterns of interest, $-+ +-$ (black arrows) and $-+ -+$ (red arrows), are shown. They differ by the ferromagnetic in-plane component along the b axis. Iridium atoms (yellow circles) are labeled as $a_-, \dots, d_-, a_+, \dots, d_+$.

multipole degrees of freedom. As an intriguing consequence of the ASOC, locally noncentrosymmetric systems may realize odd-parity multipole order [19–26] beyond the paradigm of even-parity multipole order in d - and f -electron systems [27].

In noncentrosymmetric systems, exotic superconductivity such as the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state [28,29] has been expected to be realized by the external magnetic field [30]. Searches of the FFLO state have been an issue for more than five decades [31]. For example, a recent experiment tries to detect a hallmark of the FFLO state in κ -(BEDT-TTF)₂Cu(NCS)₂ [32]. However, it has been shown that in noncentrosymmetric systems the FFLO order parameter is hidden in vortex states [33,34]. Such difficulty of experimental researches may be resolved by odd-parity multipole order [35]. One of the purposes of this study is to propose material realization of the FFLO state free from disturbance by vortices.

Recent theories have shed light on mathematically rigorous properties ensured by nonsymmorphic crystal symmetry [36–41]. For nonsymmorphic superconductors, nodal-line superconductivity unexpected from existing classification based on the point group [42] was found by Norman in 1995 [43]. Unconventional superconductivity possessing such symmetry-protected line nodes is expected to appear in UPt₃ [43–48], UCoGe [49], and UPd₂Al₃ [49–51], due to the effect of spin-orbit coupling or magnetic order. However, nonsymmorphic superconductivity by multipole order has not been uncovered.

In this Letter, we show that Sr₂IrO₄ may be a platform realizing two unconventional superconducting states, assuming the coexistence with magnetic order [52]. First, superconductivity with nonsymmorphic symmetry-protected gap structures is induced by the $-+ +-$ order, which is regarded as a higher-order magnetic octupole (MO) order. Second, the FFLO superconductivity free from vortices is stabilized in the $-+ -+$ [magnetic quadrupole (MQ)] state. These results are evidenced by a combination of group theoretical analysis and numerical analysis of an effective $J_{\text{eff}} = 1/2$ model for Sr₂IrO₄.

$-+ +-$ state.—Now we consider the superconductivity in the $-+ +-$ state. We begin with the gap classification based on the space group (see the Supplemental Material [53]). The magnetic space group of the $-+ +-$ state, M_{-+++} , is a nonsymmorphic group P_1cca . We especially focus on the Cooper pairs on the basal planes (BPs) $k_{z,x,y} = 0$ and the zone faces (ZFs) $k_z = \pm\pi/c$ and $k_{x,y} = \pm\pi/a$. In these high-symmetry planes, the small representation γ_{-+++}^k can be calculated. Indeed, γ_{-+++}^k corresponds to the Bloch state with the crystal momentum \mathbf{k} .

In the superconducting state, the zero-momentum Cooper pairs have to be formed between the degenerate states present at \mathbf{k} and $-\mathbf{k}$ within the weak-coupling BCS theory. Therefore, these two states should be connected by some symmetry operations, such as space inversion. As a

TABLE I. The gap structure for A_{1g} and B_{2g} gap functions.

	$k_z = 0$	$k_z = \pm\pi/c$	$k_{x,y} = 0$	$k_{x,y} = \pm\pi/a$
A_{1g} (s -wave)	Gap	Node	Gap	Node
B_{2g} (d_{xy} -wave)	Gap	Node	Node	Gap

result, the representation of Cooper pair wave functions P_{-+++}^k can be constructed from the representations of the Bloch state γ_{-+++}^k [55–57].

We here calculate the character of the representation P_{-+++}^k , and then reduce P_{-+++}^k into irreducible representations (IRs) of the original crystal symmetry D_{4h} . The obtained results are summarized in the following:

(1) $k_z = 0, \pm\pi/c$

$$\begin{aligned} &A_{1g} + A_{2g} + B_{1g} + B_{2g} + 2A_{1u} \\ &\quad + 2A_{2u} + 2B_{1u} + 2B_{2u} + 2E_u \quad \text{BP} \\ &2E_g + A_{1u} + A_{2u} + B_{1u} + B_{2u} + 4E_u \quad \text{ZF} \end{aligned} \quad (1)$$

(2) $k_{x,y} = 0, \pm\pi/a$

$$\begin{aligned} &A_{1g} + B_{1g} + E_g + 2A_{1u} \\ &\quad + A_{2u} + 2B_{1u} + B_{2u} + 3E_u \quad \text{BP} \\ &A_{2g} + B_{2g} + E_g + 3A_{1u} + 3B_{1u} + 3E_u \quad \text{ZF} \end{aligned} \quad (2)$$

We find that possible IRs change from BPs to ZFs as a consequence of the nonsymmorphic symmetry. The gap functions should be zero, and thus, the gap nodes appear, if the corresponding IRs do not exist in these results of reductions [58–60]. Otherwise, the superconducting gap will open in general. From Eqs. (1) and (2), for instance, we find the gap structure of A_{1g} and B_{2g} superconducting states summarized in Table I.

We demonstrate the results of group theory (Table I) using a three-dimensional single-orbital tight-binding model for $J_{\text{eff}} = 1/2$ [53] manifold. Eight Ir atoms per unit cell and three types of ASOC [61] are taken into account. We consider the s -wave order parameter [64] which belongs to the A_{1g} representation of the point group D_{4h} ,

$$\hat{\Delta}^{(s)}(\mathbf{k}) = \Delta_0 \hat{1}_2 \otimes \hat{\sigma}_0^{(\text{layer})} \otimes \hat{\sigma}_0^{(\text{sl})} \otimes i\hat{\sigma}_y^{(\text{spin})}, \quad (3)$$

and the d_{xy} -wave order parameter [64] which belongs to the B_{2g} representation,

$$\hat{\Delta}^{(d)}(\mathbf{k}) = \Delta_0 \sin \frac{k_x a}{2} \sin \frac{k_y a}{2} \hat{1}_2 \otimes \hat{\sigma}_0^{(\text{layer})} \otimes \hat{\sigma}_x^{(\text{sl})} \otimes i\hat{\sigma}_y^{(\text{spin})}, \quad (4)$$

where $\hat{1}_M$ is a $M \times M$ identity matrix. $\hat{\sigma}_i^{(\text{spin})}$, $\hat{\sigma}_i^{(\text{sl})}$, and $\hat{\sigma}_i^{(\text{layer})}$ are the Pauli matrices representing the spin, sublattice, and layer degrees of freedom, respectively.

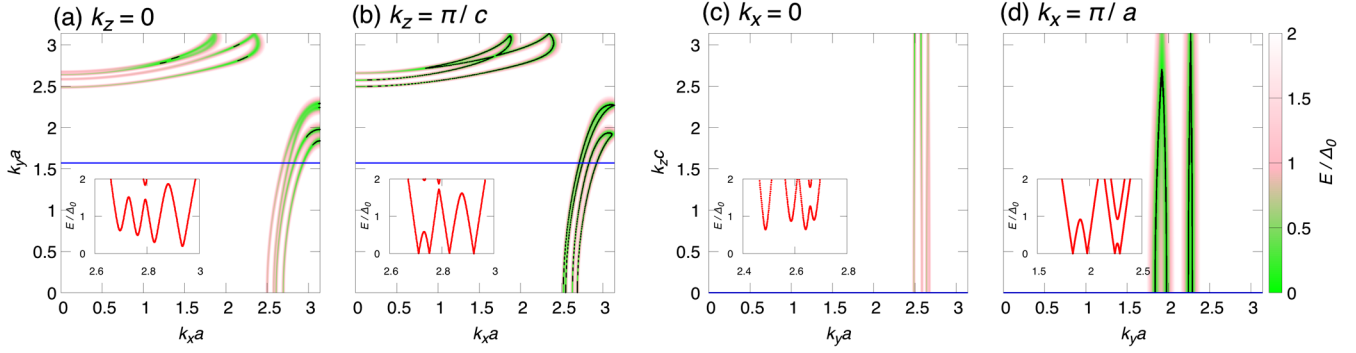


FIG. 2. The contour plot of quasiparticle energy dispersion E in the s -wave superconducting state normalized by the order parameter Δ_0 on (a) $k_z = 0$, (b) $k_z = \pm\pi/c$, (c) $k_x = 0$, and (d) $k_x = \pm\pi/a$. The insets in (a), (b), (c), and (d) show the dispersion E/Δ_0 along the respective blue line. Line nodes (black lines) appear on the ZF, $k_z = \pm\pi/c$ and $k_x = \pm\pi/a$.

The quasiparticle energy dispersion in the superconducting state $E = E(k_x, k_y, k_z)$ is obtained by diagonalizing the Bogoliubov–de Gennes (BdG) Hamiltonian [53],

$$\hat{H}_{\text{BdG}}(\mathbf{k}) = \begin{pmatrix} \hat{H}_n(\mathbf{k}) & \hat{\Delta}(\mathbf{k}) \\ \hat{\Delta}(\mathbf{k})^\dagger & -\hat{H}_n^T(-\mathbf{k}) \end{pmatrix}. \quad (5)$$

The chemical potential is chosen to set the electron density $n \sim 1.2$, around which the superconductivity has been predicted [7]. However, superconducting properties revealed below are independent of the electron density. The numerical results are shown in Figs. 2 and 3. Only $0 \leq E/\Delta_0 < 2$ region is colored, and especially nodal ($E \sim 0$) points are plotted by black.

The gap structure of the two superconducting states reproduces Table I. In both s -wave and d_{xy} -wave cases, the numerical results are consistent with the group theory. In other words, the gap nodes in Figs. 2 and 3 are protected by nonsymmorphic space group symmetry. Note that exceptional cases of the gap classification in Table I appear in some accidentally degenerate region [46]. For example, we see such unexpected gap structures on the $k_y = \pm\pi/a$ plane [53].

As introduced previously, both theory [6–9] and experiment [5] suggest d_{xy} -wave superconductivity analogous to cuprates [65]. In this case, a horizontal line node appears on the ZF ($k_z = \pm\pi/c$) in contrast to the usual d_{xy} -wave state. Moreover, the gap opening at the other ZFs ($k_{x,y} = \pm\pi/a$) is also nontrivial because the usual d_{xy} -wave order parameter vanishes not only at BPs but also at ZFs. These nontrivial gap structures are protected by the nonsymmorphic space group symmetry.

– + – + state.—We now turn to the – + – + state of Sr_2IrO_4 . In this case, the method of gap classification used above is not applicable since there is no symmetry operation connecting \mathbf{k} to $-\mathbf{k}$. Conversely, Cooper pairs do not need to be formed between \mathbf{k} and $-\mathbf{k}$ states, which indicates the emergence of the FFLO superconductivity. Indeed, the FFLO state is stabilized in the – + – + state as shown below.

Before going to the main result, here we show that the – + – + order can be regarded as an odd-parity MQ order, which results in the asymmetry in the band structure. Using a group theoretical analysis, it is determined that the – + – + order belongs to E_u representation of D_{4h} [53]. This IR permits time-reversal-odd basis functions: $\alpha y\hat{\sigma}_z + \beta z\hat{\sigma}_y$ in

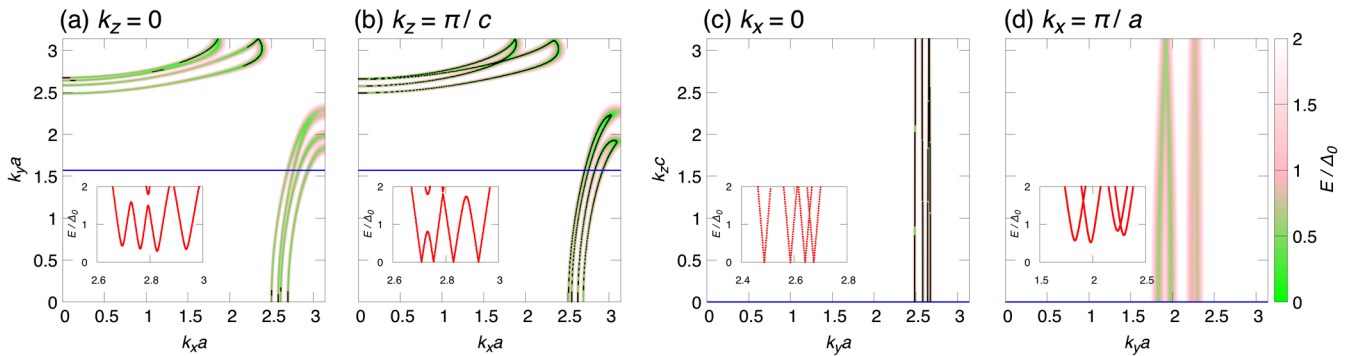


FIG. 3. The contour plot of quasiparticle energy dispersion E/Δ_0 for the d_{xy} -wave order parameter on (a) $k_z = 0$, (b) $k_z = \pm\pi/c$, (c) $k_x = 0$, and (d) $k_x = \pm\pi/a$. The insets show E/Δ_0 along the respective blue line. Line nodes (black lines) appear on the ZF $k_z = \pm\pi/c$ and the BP $k_x = 0$.

the real space, and k_x in the momentum space. In the real space, the basis function represents a rank-2 odd-parity MQ order [66],

$$\hat{M}_{2,1} + \hat{M}_{2,-1} \propto y\hat{z} + z\hat{y}, \quad (6)$$

where $\hat{M}_{l,m}$ is the magnetic multipole operator. Therefore, the $-+ -+$ order contains the component of a MQ order, though it may include a toroidal dipole order proportional to $y\hat{z} - z\hat{y}$ [19]. In the momentum space, the linear k_x function makes the band structure asymmetric along the k_x axis. We actually confirm the asymmetry of the band structure using our tight-binding model [53]. Then, we also notice a twofold degeneracy in the band structure protected by symmetry [53]. These features of band structure resemble the MQ state in the zigzag chain [20,35]. A similar analysis identifies the $-+ -+$ magnetic order as an even-parity MO order with $xy\hat{z} + yz\hat{x} + zx\hat{y}$.

Next, we study the superconductivity in the $-+ -+$ state. We can clarify the superconducting state near the transition temperature by linearizing the BdG equation while avoiding the numerical limitations of the full BdG equation. The linearized BdG equation is formulated by calculating the superconducting susceptibility $\chi_{mm'}(\mathbf{q}, i\Omega_n)$ [53], where $\Omega_n = 2n\pi T$ is the bosonic Matsubara frequency, and m represents the sublattice degrees of freedom. Here we assume the local s -wave superconductivity for simplicity. The 8×8 susceptibility matrix $\hat{\chi} = (\chi_{mm'})$ is obtained by the T -matrix approximation [67],

$$\hat{\chi}(\mathbf{q}, i\Omega_n) = \frac{\hat{\chi}^{(0)}(\mathbf{q}, i\Omega_n)}{\hat{1}_8 - U\hat{\chi}^{(0)}(\mathbf{q}, i\Omega_n)}, \quad (7)$$

where U is the s -wave on-site attraction, and $\hat{\chi}^{(0)}$ is the irreducible susceptibility.

The superconducting transition occurs at the temperature T_c where $\hat{\chi}(\mathbf{q}, i\Omega_n)$ diverges. Thus, the criterion of the superconducting instability is $\chi_{\max}^{(0)}(\mathbf{q}, i\Omega_n) = 1$, where $\chi_{\max}^{(0)}$ is the largest eigenvalue of $U\hat{\chi}^{(0)}$. Here $\chi_{\max}^{(0)}$ shows the maximum at $q_y = q_z = \Omega_n = 0$, since energy bands are symmetric with respect to k_y and k_z even in the $-+ -+$ state [53].

Figure 4 shows the q_x dependence of $\chi_{\max}^{(0)}(\mathbf{q}, 0)$ at $T \sim T_c$. In the normal state ($h = 0$), since the system preserves the inversion symmetry, $\chi_{\max}^{(0)}$ has a peak at $q_x = 0$ regardless of the presence or absence of the ASOC [Fig. 4(a)]. On the other hand, in the $-+ -+$ state ($h = 0.2$ and 0.8), $\chi_{\max}^{(0)}$ shows the maximum at a finite q_x when the ASOC exists, while the conventional $\mathbf{q} = \mathbf{0}$ state is stable in the absence of the ASOC [Figs. 4(b) and 4(c)]. This result reveals that the FFLO state is favored by the ASOC in the odd-parity $-+ -+$ magnetic ordered state, despite the absence of the macroscopic magnetization required for the conventional FFLO state [28–32]. Moreover in the large

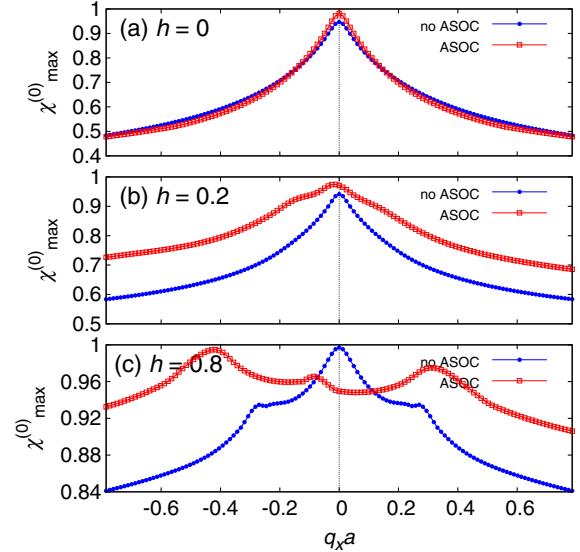


FIG. 4. The largest eigenvalue $\chi_{\max}^{(0)}$ in (a) the normal state ($h = 0$), (b) the small moment $-+ -+$ state ($h = 0.2$), and (c) the large moment $-+ -+$ state ($h = 0.8$). We fix the temperature $T = 0.01 \sim T_c$. For $h = 0, 0.2$, and 0.8 , the s -wave on-site interaction U is respectively assumed to be $0.26, 0.47$, and 1.45 in the absence of the ASOC, while it is $0.31, 0.55$, and 1.60 in the presence of the ASOC.

moment state ($h = 0.8$), three local maxima are observed in Fig. 4(c). The behavior resembles the band-dependent FFLO state in the one-dimensional zigzag chain [35]. Namely, a part of the bands mainly causes the superconductivity, while the other bands are weakly superconducting. The nonuniform state with a large $|q_x a| \sim 0.4$ should be regarded as a pair-density-wave state [68–70] rather than the FFLO state.

Summary.—In this Letter, we investigated the superconductivity of doped Sr_2IrO_4 in the two magnetic states, $-+ -+$ and $-+ -+$. In the $-+ -+$ (MO) state, both s -wave and d_{xy} -wave superconductivity shows nontrivial line nodes protected by nonsymmorphic symmetry on the BZ boundary. The nodal gap is analogous to that studied in toy models [47,48,50]. In a realistic model for Sr_2IrO_4 , however, we have clarified not only nontrivial line nodes but also an unexpected gap opening. In the case of d_{xy} -wave superconductivity, the gap opens on the vertical BZ face unlike the ordinary d_{xy} -wave superconductor. On the other hand, in the $-+ -+$ state identified as parity-violating odd-parity MQ state, the FFLO state is stabilized irrespective of the magnitude of the antiferromagnetic moment, because the band structure asymmetrically deforms. The asymmetric band structure and resulting FFLO superconductivity are regarded as magnetoelectric effects caused by odd-parity MQ order. The FFLO state caused by the MQ order does not need an external magnetic field, which means the “pure FFLO state,” namely the FFLO state free from vortices. Material realization in

Sr_2IrO_4 may enable experimental observation of FFLO superconductivity.

We suggest doped Sr_2IrO_4 as a platform of nonsymmorphic nodal superconductivity by magnetic multipole order. Furthermore, the realization of parity-violating multipole and FFLO superconductivity are proposed beyond the toy model [35]. These results point to nontrivial interplay of magnetic multipole order and superconductivity in the strongly spin-orbit coupled systems.

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