

Bond-dependent interactions and ill-ordered state in the honeycomb cobaltate $\text{BaCo}_2(\text{AsO}_4)_2$

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The ground state and Hamiltonian of the honeycomb lattice material $\text{BaCo}_2(\text{AsO}_4)_2$ hosting magnetic Co^{2+} have been debated for decades. The recent proposal for anisotropic bond-dependent interactions in such honeycomb cobaltates has raised the prospect of revisiting its Hamiltonian in the context of Kitaev physics. To test this hypothesis, we have combined magnetization, ac susceptibility, and neutron scattering measurements on a $\text{BaCo}_2(\text{AsO}_4)_2$ single crystal, together with advanced modeling. Our experimental results highlight a collinear magnetic ground state with intrinsic disorder associated with an average incommensurate propagation vector. Monte Carlo simulations and linear spin wave calculations were performed to obtain a spin model compatible with this unusual ground state, the dispersion of magnetic excitations, and a magnetization plateau under magnetic field. We thus show that bond-dependent anisotropic interactions, including Kitaev-like interactions, are necessary to account for the puzzling properties of this long-explored material, and are hence a general ingredient in the cobaltates.

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In transition metal oxides, the interplay between crystal field, spin-orbit coupling, and lattice geometry can induce a rich variety of magnetic properties. Recent focus has been drawn toward $4d$ and $5d$ heavy transition metals displaying a large spin-orbit coupling, following the exciting proposal on Kitaev bond-dependent interactions [1,2]. This has led to numerous experimental and theoretical studies of a large panel of iridate/ruthenate materials hosting unconventional properties from potential spin liquids to topological and superconducting phases [3,4]. $3d$ transition metals, in particular cobalt oxides, in which spin-orbit coupling and electron correlations directly compete, are also of interest since they are known to display various exotic phases, some of which have been studied for many years [5–9]. In these systems, the Co^{2+} ions host a sufficiently large spin-orbit coupling to stabilize a spin-orbital $j_{\text{eff}} = 1/2$ Kramers ground state and it has been argued that, in contrast to the iridate and ruthenate counterparts, due to the presence of additional spin-active e_g electrons in Co^{2+} , the Heisenberg term can be suppressed to the benefit of

Kitaev interactions [10–25]. Experimental evidence for Kitaev interactions has been reported in some cobaltate materials but always competing with other terms in the Hamiltonian such as Heisenberg, off-diagonal, further neighbor interactions, or single-ion anisotropy. Even though these additional ingredients push the investigated cobaltates away from a Kitaev spin liquid, the presence of bond-dependent interactions produces interesting physics, in particular, original magnetic ground states such as the triple-q orders proposed in $\text{Na}_2\text{Co}_2\text{TeO}_6$ and $\text{Na}_3\text{Co}_2\text{SbO}_6$ [21,22,25–27].

Among the investigated cobaltates, $\text{BaCo}_2(\text{AsO}_4)_2$ (BCAO) is one example of a honeycomb magnet that has been extensively studied since the 1980s [5–9,28], yet open questions remain about its magnetic ground state and excitations. Contradicting reports concerning its Hamiltonian, the presence or not of bond-dependent interactions, and their influence on the properties have been evoked recently. However, none of the proposed models seem to account for all observations, from the field-induced magnetization to the zero-field spin wave dispersion. The latter, a stringent test to elucidate the physical parameters at play, has not been achieved yet.

In this work, we address the long-standing issue around the nature of BCAO's magnetic ground state and excitations, and highlight the role of bond-dependent anisotropic interactions in stabilizing its original spin arrangement by combining neutron scattering and very low temperature magnetic measurements on a single crystal, along with Monte Carlo and linear spin wave calculations. Our results also question the

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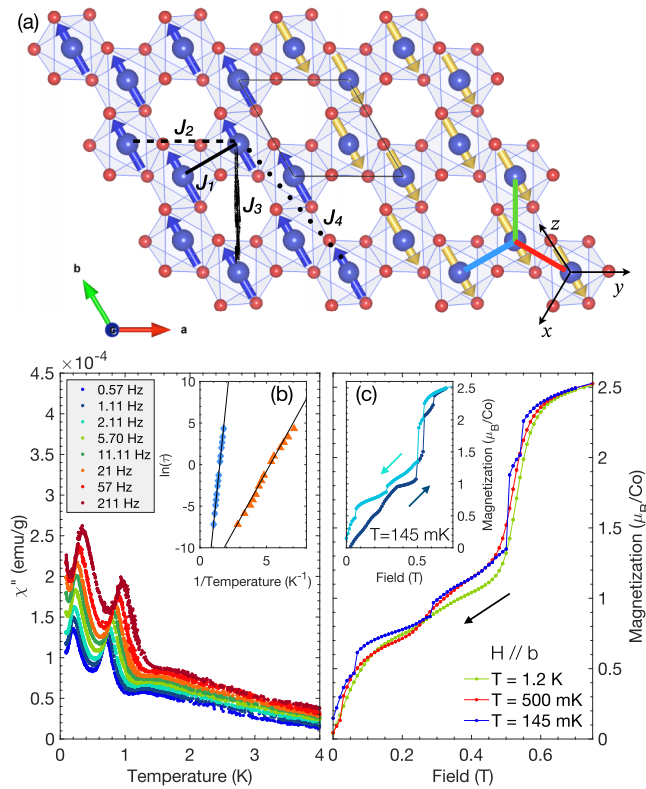


FIG. 1. (a) Honeycomb plane in BCO, with the proposed uudd sequence as the building block of the magnetic structure. The considered Heisenberg interactions are displayed in black. The red, green, and blue lines refer to the *x*, *y*, and *z* bonds, respectively, for the anisotropic interactions defined in the local *xyz* orthogonal frame. (b) Imaginary part of the ac susceptibility χ'' plotted as a function of temperature for various frequencies with $H_{ac} = 3.3$ Oe along *b*. Inset: Relaxation times τ associated with the low/high-temperature χ'' peaks (in orange/blue, respectively) vs inverse temperature. The lines represent fits to Arrhenius laws. (c) Magnetization curves as a function of decreasing magnetic field at various temperatures. Inset: Hysteresis observed at 145 mK for increasing and decreasing fields.

ubiquity of Kitaev-like interactions in cobaltate materials in general and their role in stabilizing exotic spin textures.

BCAO crystallizes in the hexagonal $R\bar{3}$ space group and comprises honeycomb layers of magnetic Co^{2+} ions in the (*a*, *b*) plane, with the interplane distance more than twice the intraplane Co-Co distance, leading to a quasi-two-dimensional system. Previous neutron studies show that, in zero field, BCO undergoes a sharp magnetic transition below $T_N = 5.3$ K and orders with a propagation vector $\mathbf{k} = (0.27, 0, -4/3)$ [5,7]. One particular unsettled issue concerns BCO magnetic order: while first thought as a helical spin structure with *XY* anisotropy, neutron spherical polarimetry measurements by L.-P. Regnault *et al.* eventually suggested that the magnetic order may rather be collinear, made of ferromagnetic spin chains with magnetic moments aligned close to the *b* direction, the chains being organized in an antiferromagnetic pattern along the *a** direction [Fig. 1(a)]. This pattern would correspond to an up-up-down-down (uudd) configuration with inserted chain defects allowing to match the incommensurate propagation vector ($q_h \approx 0.27$ instead of 0.25 expected for

a uudd commensurate order). In favor of the presence of disorder, the chains are weakly coupled along the *a** direction with anisotropic correlation lengths ξ measured by neutron scattering within the honeycomb plane ($\xi_b/\xi_{a^*} \approx 2.7$) [8,9]. Since this work, no further experimental evidence has confirmed this ill-ordered collinear magnetic state.

Such a state is expected to display slow dynamics associated with the chain defects inserted in the (uudd) arrangement that can be probed by ac susceptibility measurements. Figure 1(b) displays the temperature dependence of the imaginary part of the ac susceptibility χ'' , down to 100 mK at various frequencies *f* below T_N . Two peaks are observed, whose position varies with frequency, confirming that slow dynamics are present in the ordered state. As shown in the inset of Fig. 1(b), the corresponding relaxation times $\tau = 1/2\pi f$ follow Arrhenius laws $\tau = \tau_0 \exp(E/T)$, illustrating that both are associated with thermally activated processes over an energy barrier *E*, with a characteristic relaxation time τ_0 . The fits yield $E_{LT} = 2.8(1)$ K, $\tau_{0LT} = 2.5(8) \times 10^{-7}$ s and $E_{HT} = 15.5(1)$ K, $\tau_{0HT} \sim 10^{-10}$ s for the low- and high-temperature peaks, respectively.

Given the characteristic relaxation time, consistent with the spin correlation length observed along the chains [9], we can tentatively ascribe the highest temperature peak in χ'' to the reversal/creation of chainlike defects [29–33] whose presence in the collinear pattern allows the system to accommodate the incommensurate magnetic structure [see Fig. 3(a)]. The lowest-temperature peak is characterized by a longer relaxation time and a smaller energy barrier, suggesting that it is related to the dynamics of larger objects, i.e., domain walls separating configurational (180° or 120°) domains. It is worth noting that beyond these two well-defined processes, χ'' presents a broad nonzero background, which indicates the existence of a large number of relaxation channels, with a huge relaxation time distribution. These complex dynamics also manifest in magnetization curves as a function of magnetic field at very low temperature [see Fig. 1(c)]. At 1.2 K, our $\mathbf{H} \parallel \mathbf{b}$ measurements recover the magnetization plateau at about one-third of the saturated magnetization between 0.1 and 0.55 T, previously reported in higher temperature measurements [5,28,34]. When decreasing further the temperature at 500 mK and below, i.e., below the higher temperature χ'' peak, magnetization steps occur at intermediate magnetic field values, whose number increases with temperature lowering. In addition, the step positions depend on the field sweeping direction and rate [35]. This behavior is characteristic of small magnetic avalanches that often occur at low temperature and are usually characteristic of domain reorganizations. In that sense, these steps may be the signature under applied magnetic field of the processes observed in the ac susceptibility. Finally, the hysteresis opening, which was only observed up to the plateau at higher temperature, is present up to saturation at 500 mK and below [see inset of Fig. 1(c)], indicating the existence of metastable states over the whole field range. Macroscopic magnetic measurements at very low temperature thus provide experimental evidence for a magnetic ground state encompassing magnetic defects associated with thermally activated processes. In the proposed picture, this disorder is intrinsically related to the observed

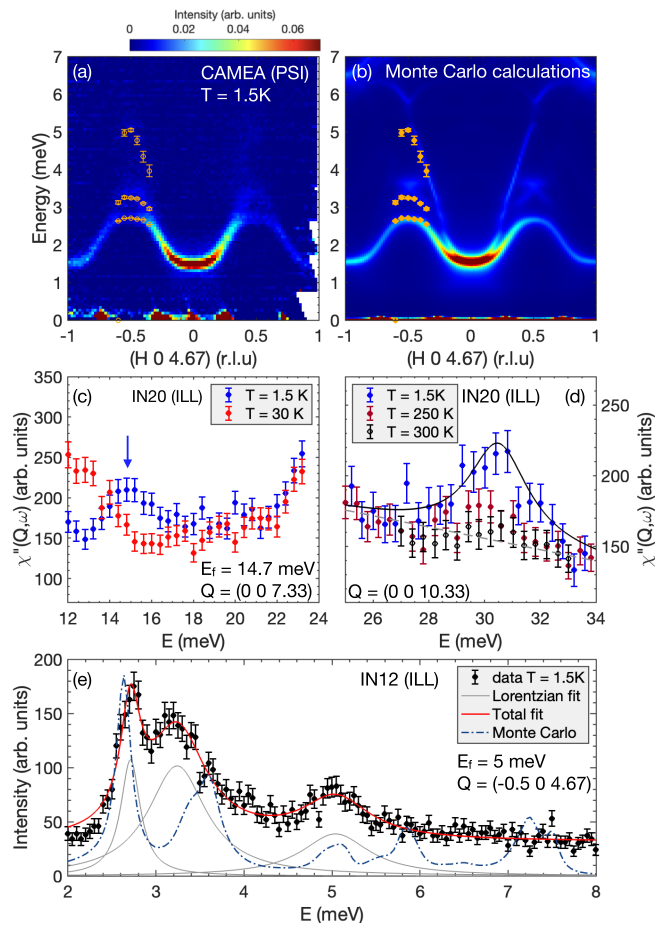


FIG. 2. (a) Magnetic excitation dispersion in the $(H\ 0\ 4.67 \pm 0.1)$ direction from neutron inelastic scattering measurements performed on CAMEA at $T = 1.5$ K. (b) Comparison with $S(Q, \omega)$ calculated from Monte Carlo simulations, using the model given in Table I. (c), (d) Constant- Q energy scans measured on IN20 at different temperatures, revealing higher energy magnetic excitations. The generalized susceptibility χ'' corresponds to $S(Q, \omega)$ corrected from the Bose factor. (e) Energy scan measured at the Brillouin zone boundary on IN12 showing several magnetic modes fitted by Lorentz functions (gray and red lines) and compared to the Monte Carlo simulation (blue dotted line). The orange dots in panels (a) and (b) result from the fits.

incommensurate propagation vector that is not compatible with a long-range (uudd) arrangement of collinear spins.

To gain more insight into the microscopic parameters leading to this original magnetic state, the spin excitations were measured in zero field using neutron inelastic scattering [35]. Figure 2(a) displays the spin wave dispersion of BCO measured on CAMEA [35] along the $(H\ 0\ 4.67)$ reciprocal space direction at $T = 1.5$ K in its ordered phase. A well-defined mode is observed, with a rather flat bottom above an energy gap of about 1.45 meV at the magnetic Brillouin zone center, on the order of the highest energy gap determined from ac susceptibility. This mode disperses to reach about 3 meV at the zone boundaries where it separates into two branches while getting broader in energy. At higher energy, between 3.5 and 6 meV, a broad signal can also be observed in the continuity of the lower spin wave branches, as highlighted

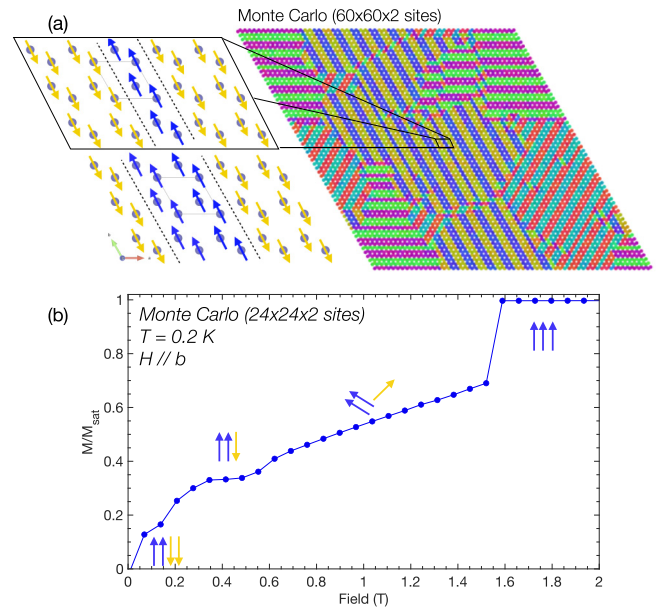


FIG. 3. Monte-Carlo simulations using the model given in Table I: (a) spin structure where the colors represent different orientations of the spins, which are antiparallel within each three 120° domains. The inset illustrates the defective uudd pattern for one domain and the flipping of a chain defect. (b) Calculated magnetization curve as a function of magnetic field applied along b with a sketch of the spin arrangements of the different phases.

in the constant- Q cut measured on IN12 [Fig. 2(e) and orange dots in panel (a)]. The absence of dispersion along the c axis confirms the two-dimensional nature of the magnetic order [35]. Consistent with other neutron studies [28], another weakly dispersing mode is visible around 15 meV, as shown in the constant- Q cut [Fig. 2(c)] measured on IN20. It disappears above T_N , thus confirming its magnetic nature. A broad excitation is also detected around 30 meV, which is not correlated with T_N , but disappears at 300 K [see panel (d)]. This is attributed to the spin-orbit excitation corresponding to the $j_{\text{eff}} = 3/2$ multiplet, usually observed in this energy range in other cobaltates [14,15,20,22]. It confirms the $j_{\text{eff}} = 1/2$ ground-state picture in BCO and the assignment of the low-energy spectrum to excitations within this manifold. One intriguing feature of the spin waves resides in the absence of a Goldstone mode stemming from the incommensurate magnetic Bragg positions. The global gap, confirmed by another neutron study along other directions [28], is not compatible with a helical arrangement of spins with an XY anisotropy in the ab plane, hinting for the presence of an additional anisotropy within this easy plane.

Note that this additional in-plane anisotropy that aligns the spins along a direction close to the b axis cannot originate from single-ion physics. Indeed, in BCO, the distorted CoO_6 octahedra are in a trigonal antiprismatic configuration, with a conserved local C_3 symmetry at the Co^{2+} ion sites (Wyckoff position 6c) [35]. Therefore, from symmetry arguments, the single-ion anisotropy in BCO is purely easy plane since no in-plane easy-axis anisotropy that would break the C_3 symmetry is allowed [9,28,36–39]. To account for an

additional in-plane anisotropy, other ingredients must then be considered, which naturally leads to the possibility of bond-dependent interactions.

To proceed with the modeling of the neutron inelastic data, we considered the generic $JK\Gamma\Gamma'$ Kitaev-Heisenberg matrix involving bond-dependent interactions [13,35]. In first approximation, we only considered anisotropic interactions for the first nearest neighbors as it was previously suggested that the contribution of such interactions for further neighbors should be weak [15,28,38,40,41]. In the local xyz frame of the octahedra [see Fig. 1(a)], the symmetry allowed matrix terms have this form:

$$H_{(1)}^z = \begin{pmatrix} J - \eta & \Gamma & \Gamma'_1 \\ \Gamma & J + \eta & \Gamma'_2 \\ \Gamma'_1 & \Gamma'_2 & J + K \end{pmatrix}$$

expressed for the z bond, where, among the six independent parameters, J is the isotropic Heisenberg coupling, K is a bond-dependent (Kitaev) interaction, and Γ and Γ' are two off-diagonal bond-dependent symmetric exchange couplings. For simplicity, we fix $\eta = 0$ and $\Gamma' = \Gamma'_1 = \Gamma'_2$, thus neglecting the tiny octahedral distortion resulting in the loss of the local twofold axis [38]. Our model also includes Heisenberg interactions beyond first neighbors up to the fourth ones [Fig. 1(a)] and a gyromagnetic tensor with $g_z/g_{xy} = 0.5$, to be consistent with the single-ion physics of Co^{2+} in BCAO [35].

In order to find a spin wave model that reproduces the excitation dispersion, the parameter space was carefully explored by varying all the parameters. First, we determined regions in the parameter space that stabilize a $(k_x, 0, 0)$ magnetic phase with k_x close to the incommensurate value, by calculating phase diagrams using the Luttinger-Tisza method [35,42]. Linear spin wave calculations were then performed using selected sets of parameters. Those giving the best agreement with the experimental dispersion were tested using Monte Carlo algorithm in order to check whether they yield (1) a 1/3-magnetization plateau as a function of magnetic field and (2) a uudd spin arrangement disrupted by magnetic defects. Indeed, the Monte Carlo method is better adapted for ill-ordered magnetic states, complementing the linear spin wave calculations, performed on periodic ($k_x = 0.25$) and ordered spin arrangements. Considering the abovementioned constraints, several models are in close agreement with the experimental dispersion but only a few of them are also consistent with the disordered spin structure and 1/3-magnetization plateau within the observed magnetic field range.

Figures 2(b), 2(e), and 3 show the results of the model yielding the best agreement with the experimental data [see the Supplemental Material [35] for other models]. The set of parameters given in Table I reproduces well the low-energy excitation dispersion, the energy gap, and the diffuse intensity above 4 meV at the zone boundaries [Fig. 2(b)]. It also generates optical branches although at an energy (≈ 7 meV) lower than the observed higher magnon one around 15 meV [Fig. 2(c)]. Most importantly, as shown in Fig. 3(a), the stabilized spin structure corresponds to ferromagnetic chains with spins aligned along the b axis, which are stacked along the a^* direction in a uudd sequence including defective chains,

TABLE I. Set of interactions (in meV) corresponding to the best agreement with the magnetic structure, the magnetization vs H , and the dispersion of magnetic excitations in BCAO. Plus/minus signs correspond to antiferromagnetic/ferromagnetic interactions.

J_1	J_2	J_3	J_4	K	Γ'	Γ
$-3.3(1)$	$-0.50(2)$	$0.60(5)$	$0.15(2)$	$2.0(15)$	$-0.9(1)$	$3.3(1)$

resulting in an average incommensurate propagation vector equal to 0.27 as observed experimentally [35]. Finally, the calculated magnetization curve qualitatively reproduces the 1/3-magnetization plateau above $H_{\text{th1}} = 0.2$ T [Fig. 3(d)] observed in the experiment at $H_{c1} = 0.1$ T [Fig. 1(c)] with a uudd spin arrangement. Note that, in the calculation, between 0.6 and 1.6 T, the calculations favor a “spin flop” configuration with the antiferromagnetic spin component perpendicular to the field, as also reported in another theoretical work [43]. This additional phase could explain the slope observed experimentally on the plateau and will deserve further investigations.

The proposed model includes a subdominant antiferromagnetic Kitaev term K [38,39], thus contrasting with recent studies, including delafossite-type honeycomb cobaltates [20,25,44]. It also implies significant off-diagonal Γ' and Γ terms arising from CoO_6 distortions [35], the latter being as large as the Kitaev term. Other Hamiltonians displaying a rather large distribution of parameters have been proposed in the literature to explain BCAO magnetic properties [28,38–41]. Some of these models are obviously insufficient since they predict a helical ground state [28,39]. Most of them agree with a large ferromagnetic J_1 , a weaker antiferromagnetic Kitaev K interaction, and a non-negligible antiferromagnetic J_3 interaction, as in our results. We considered in addition second and fourth neighbors Heisenberg interactions. The latter, although weak, is essential in our semiclassical approach to stabilize the uudd defective magnetic ground state, as well as shaping the Brillouin zone center excitation and stabilizing a 1/3-magnetization plateau. Note that a uudd ground state has been predicted in other studies by adding either quantum fluctuations [39,45] or anisotropic interactions on the third neighbor path [40]. These additional parameters cannot be excluded. Quantum effects could explain some broadening of our measured spin wave excitations (which could also be attributed to disorder) and question the nature of the magnetic excitations beyond linear spin waves. A recent study shows that the 15 meV mode, which our model fails to reproduce, lies near an optical phonon branch and therefore may arise from electromagnetovibrational scattering [46]. It was also suggested that anisotropic third neighbor interactions may improve the high-energy part of the magnetic excitations, in particular yielding the 15 meV mode [43]. However, it is to be emphasized that none of the cited models were shown to fully reproduce the spin wave dispersion in zero field, whereas our set of parameters accounts rather well simultaneously for the low-energy magnetic excitations, the 1/3-magnetization plateau, and the peculiar ground state.

This intrinsically disordered magnetic structure emanates from a delicate balance between the competing interactions at

play and the in-plane easy-axis anisotropy. Some hallmarks of this ground state have been observed in other systems, in particular with Co^{2+} , displaying strong Ising-like anisotropy and frustration, such as the anisotropic next-nearest-neighbor interaction model yielding a devil's staircase physics on a triangular lattice. In some of these cobalt oxides, multiple field-induced magnetization steps, frequency-dependent ac susceptibility, and complex phase diagrams including short-range orders have been reported [47–55]. B CAO represents here an original example where the Ising-like spin anisotropy stems directly from anisotropic interactions. Note that similar signatures of magnetic defects have also been recently reported in the sister material $\text{BaCo}_2(\text{PO}_4)_2$ via magnetostriction measurements at low temperature [56] and in another cobalt honeycomb magnet, $\text{Na}_3\text{Co}_2\text{SbO}_6$. In the latter, the measured strong in-plane anisotropy may reflect significant contribution from bond-dependent interactions [57].

In conclusion, our study highlights, for the first time, the existence of an original ill-ordered magnetic state in B CAO, with further evidence for chainlike magnetic defects, and establishes the role of anisotropic bond-dependent interactions in stabilizing such exotic spin texture. Our Monte Carlo calculations show that a spin Hamiltonian including

bond-dependent interactions is able to describe the magnetic structure, magnetization data, and excitations in B CAO, thus providing a comprehensive understanding of this material. Beyond Kitaev physics, this highlights the relevance of such contributions in the description of 3d transition metal magnetism and opens prospects for exploring exotic magnetic states in these materials.

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Data availability. The data that support the findings of this Letter are openly available [58].

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