

Magnetic evolution, phase transitions, and electronic band structure of the ferrotoroidic candidate $\text{Ba}_6\text{Cr}_2\text{S}_{10}$

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Ferrotoroidic materials, which break both time- and space-reversal symmetries to enhance magnetoelectric responses, are of great interest for applications. $\text{Ba}_6\text{Cr}_2\text{S}_{10}$, a recently synthesized quasi-one-dimensional ferrotoroidic candidate, combines polarization, magnetization, and toroidal moment, yet its electronic band structure, magnetic evolution, and phase transitions remain incompletely understood to date. Here, we investigate $\text{Ba}_6\text{Cr}_2\text{S}_{10}$, using electrical transport, direct current magnetization, alternating current susceptibility, specific heat, and muon spin rotation (μSR) measurements, along with various first-principles calculations. Electrical transport measurements and density functional theory confirm a narrow band gap (~ 0.707 eV, experimentally; ~ 0.680 eV, theoretically). μSR measurements reveal that static magnetism dominates down to 2 K, with an ordered volume fraction of $\sim 89\%$ at this temperature. Two distinct transitions are identified: one around the Néel temperature [$T_N = (8 \pm 1)$ K] and another corresponding to a phase transition near 30 K, as consistently indicated by our complementary experimental techniques. The absence of dynamic critical behaviors and the lack of a λ -type specific heat anomaly are consistent with a first-order-like transition at T_N . Based on various complementary experimental results and theoretical calculations, we propose a hypothesis that the ~ 30 K broad feature arises from the complex interplay between crystal structure distortion and magnetic coupling, which is potentially directly linked to ferrotoroidicity. As the first study integrating μSR with complementary experiments and first-principles calculations to explore ferrotoroidicity, this work provides key insights into the underlying physics of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ and related compounds in this area.

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I. INTRODUCTION

In the solid state, the spontaneous ordering of orbital, charge, and spin degrees of freedom gives rise to phenomena such as metal-to-insulator transitions and colossal magnetoresistance [1–3]. Unlike the ferromagnetic state, characterized

by magnetic order and broken time-reversal symmetry, and the ferroelectric state, defined by electric polarization and broken space-reversal symmetry, ferrotoroidic order violates both symmetries, thereby enhancing the magnetoelectric response [4–7]. A key technological advantage of these materials lies in the tunability of their order parameter. Consequently, extensive research efforts have been devoted to discovering multiferroic materials (e.g., BiFeO_3) that exhibit multiple ferroic orders, owing to their unique properties and potential applications. The toroidal moment—an order parameter for ferrotoroidicity—describes a head-to-tail configuration of magnetic moments [8] and can be achieved through engineered electromagnetic metamaterials. Surface currents in toroidal structures can generate static toroidal moments, which have been observed in single-molecule compounds with wheel-shaped topologies, such as Dy_6 wheels [9,10], Dy_4 squares [11], and Dy_3 triangles [12]. In crystalline

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solids, the static toroidal moment is often found in multiferroics or materials with a linear magnetoelectric effect, such as LiCoPO_4 [4] and $\text{LiFeSi}_2\text{O}_6$ [6]. LiCoPO_4 serves as a prime example of this, as toroidal domains can be detected using optical second harmonic generation and manipulated by an applied toroidal field, exhibiting hysteretic behavior. In its magnetically ordered state, LiCoPO_4 has two pairs of Co^{2+} spins at positions $(1/4 + \epsilon, 1/4, -\delta)$ that generate opposite yet unequal toroidal moments. Notably, two antiparallel spins can also form the simplest toroidal moment. A model of an antiparallel-periodic-aligned (APA) spin chain, with distances $a_1 \neq a_2$, breaks both spatial and time-reversal symmetries. Here, a_1 and a_2 are defined as nearest distance in the dimerized and antiferromagneticlike spin chain [see Fig. 1(g) in Ref. [8]; a_1 and a_2 also correspond to J_1 and J_2 , respectively, in Fig. 6(a) of the present study]. Therefore, this one-dimensional (1D) “spin-pairing” APA spin chain is expected to exhibit ferrotoroidicity. Recently, $\text{Ba}_6\text{Cr}_2\text{S}_{10}$, a quasi-one-dimensional ferrotoroidic candidate, was synthesized under high-temperature and high-pressure conditions [8]. It uniquely combines polarization, magnetization, and toroidal moment, as demonstrated by direct current (dc) magnetization measurements, neutron powder diffraction, x-ray absorption spectroscopy, and electric polarization studies. Nevertheless, the electronic band structure, magnetic evolution, phase transitions, and critical phenomena of this material remain incompletely understood to date. More importantly, the relationship between the characteristics of ferrotoroidicity and structural-magnetic coupling constitutes a central question to resolve, highlighting the necessity for investigating the static and dynamic magnetic properties—particularly via local probe techniques and theoretical calculations and simulations.

To address these unresolved issues, the present study adopts a comprehensive approach combining dc magnetization, alternating current (ac) susceptibility, specific heat, muon spin rotation (μSR), and electrical transport measurements and various first-principles calculations for $\text{Ba}_6\text{Cr}_2\text{S}_{10}$. Our key findings are as follows: (1) Electrical transport measurements reveal a narrow band gap of 0.707 eV, which is corroborated by density functional theory (DFT) calculations yielding an estimated value of 0.680 eV. (2) Longitudinal field (LF)- μSR spectra demonstrate that static magnetism dominates the magnetic ground state down to 2 K. As temperature decreases, the static ordered volume fraction initially increases gradually, followed by a substantial rise around (8 ± 1) K (Néel temperature, T_N), and ultimately reaches 89% at 2 K—a hallmark of intrinsic magnetism. The experimentally derived local magnetic field magnitude aligns well with neutron-scattering data and theoretical calculations of muon sites and local fields. (3) The absence of dynamic critical behaviors (from ac susceptibility and μSR measurements) and the lack of a λ -type specific heat anomaly are consistent with a first-order-like transition at T_N . (4) A peak emerges around 30 K (T^*) in magnetization, ac susceptibility, and specific heat data; we hypothesize that the ~ 30 K broad peak originates from the complex interplay between crystal structural distortions and magnetic coupling. This feature may be directly related to ferrotoroidicity, given that the unique crystal and magnetic structures of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ serve as a key foundation for such toroidal order.

II. RESULTS AND DISCUSSION

A. Electronic band structure

Figure 1(a) presents the top view (left panel) and side view (right panel) of the crystal structure of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$. This compound features face-sharing CrS_6 octahedral chains that are aligned along the c axis, arranged in a triangular lattice within the ab plane, and separated by barium (Ba) and sulfur (S) ions. For the calculations of the electronic band structure, the crystal structure of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ was fixed based on the experimental parameters [8], with lattice constants $a = b = 9.12$ Å and $c = 12.36$ Å. Bader charge analysis shows that each Cr atom loses 1.33 electrons, indicating that its oxidation state is approximately +2.66. The band structure of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ is illustrated in Fig. 1(b), revealing a band gap of 0.680 eV. To validate this result, electrical transport measurements were conducted, as depicted in Fig. 1(c). Due to the constraints imposed by the upper limit of the resistance measurement range in our setup, the dataset is confined to the temperature interval of 375–400 K. Despite this limitation, an approximate band gap value of (0.707 ± 0.009) eV was extracted, as depicted in Fig. 1(d). This experimentally derived value shows good agreement with the theoretically predicted band gap of 0.680 eV.

B. Magnetic evolution and phase transitions

The μSR technique has been widely and effectively employed to probe both static and dynamic magnetic properties in diverse materials—including cuprate- [13,14] and iron-based superconductors [15,16], frustrated magnetic systems [17,18], Mott insulators [19–21], diluted magnetic semiconductors [22–24], and spin glasses [25,26]—owing to its exceptional sensitivity and wide applicability of various conditions [27,28]. Herein, we extend the application of the μSR technique to probe the ferrotoroidic properties of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ polycrystals from a local probe perspective, utilizing systematic measurements under zero-field (ZF), weak transverse field (wTF), and LF configurations across a wide range of temperatures and external field conditions, as presented in Figs. 2(a)–2(c) and 3. All the fitting models and details are provided in the materials and methods part of this study with further explanations.

At the base temperature of 2 K, the external longitudinal field was able to almost fully decouple the LF- μSR spectra [Fig. 2(a)], indicating that random static local fields dominate the magnetic response [27,28], in contrast to the opposite spin fluctuation case (see Fig. 3 of Ref. [18]). Given the rapid decay of the signal within the first 1 μs , it is more robust to adopt the conventional analysis method for the temperature-dependent LF- μSR spectra measured under 100 G [Fig. 2(b)], with the summarized results presented in Fig. 2(c). No abnormality of $1/T_1$ was observed near T_N . As a complementary technique to μSR , ac susceptibility serves as a valuable direct probe for investigating magnetic dynamics in the low-frequency regime, which is characterized by correlation times ranging from approximately 10^{-5} to 1 s [29]. As presented in Fig. 2(d), frequency- and temperature-dependent ac susceptibility measurements were performed on $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ polycrystals. No abnormalities in either the real (M') or imaginary (M'') components were observed near T_N . Consequently, combining the

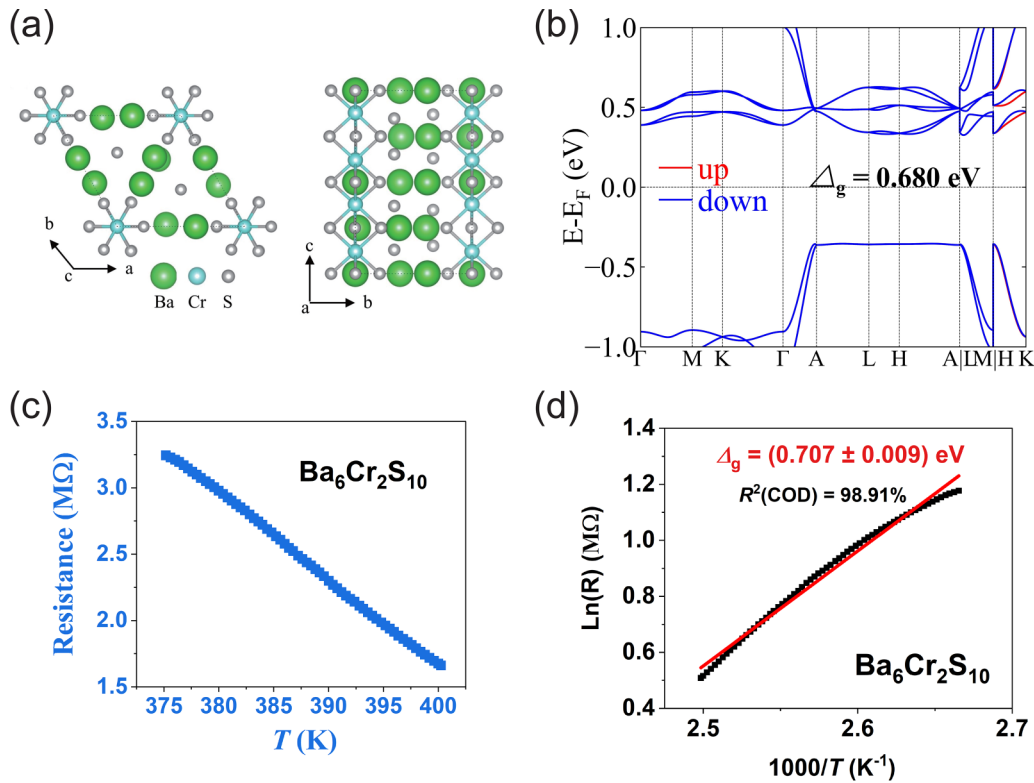


FIG. 1. Crystal structure and electronic band structure of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$. (a) Top view (left panel) and side view (right panel) of the crystal structure. (b) The spin-polarized band structure, obtained through DFT calculations. This band structure corresponds to the AFM1 configuration in Fig. 6(b) within PBE + U ($U = 2.5$ eV for Cr), where the spin-up (red) and spin-down (blue) bands of the antiferromagnetic state almost completely overlap. (c) The temperature dependence of resistivity in the range from 375 to 400 K. (d) The experimental data presented in panel (c) were fitted using the model $\text{Ln}(R) = \text{Ln}(R_0) + \Delta g/2k_B T$, yielding an electronic band gap of (0.707 ± 0.009) eV. Here, R denotes the resistance, R_0 represents the temperature-independent resistance term, Δg is the electronic band gap, k_B is the Boltzmann constant, and T is the absolute temperature. The black dots correspond to the raw data points, while the red lines illustrate the outcomes of the linear analysis. Although the coefficient of determination [$R^2(\text{COD})$] is 98.91%, which is close to 100%, this analysis should be considered an approximate estimation. This limitation arises from the narrow temperature range and the complex band structure of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$, which complicate direct comparison with theoretical calculations.

results obtained from μSR and ac susceptibility measurements, we conclude that no dynamic critical behavior emerges around T_N .

Figure 3(c) shows the summary of temperature-dependent analysis results, including the static ordered volume fraction (f_M), the fast relaxation rate (a_s), and the slow relaxation rate (λ), which was extracted from ZF μSR spectra [Fig. 3(a)] and wTF μSR spectra [Fig. 3(b)], respectively. As temperature decreases, the f_M initially increases gradually, followed by a substantial rise around (8 ± 1) K (T_N), and ultimately reaches $\sim 89\%$ at 2 K—an important signature of intrinsic magnetism in the system. A similar temperature-dependent trend is also observed for a_s . To provide full transparency of the analysis procedure, we additionally include the temperature dependence of λ [the bottom panel of Fig. 3(c)]. Note that the value of λ is approximately one order of magnitude smaller than that of a_s [the upper panel of Fig. 3(c)]. More importantly, this parameter is not reliable for investigating critical behaviors under ZF and wTF configurations [27,28], as it incorporates contributions from the longitudinal component of a_s around T_N . This is clearly shown in the bottom panel of Fig. 3(c), where the value of λ extracted from wTF μSR spectra around T_N is effectively suppressed by the

applied field, when compared to the value obtained from the ZF μSR spectra. This limitation highlights the critical role of LF- μSR measurements, which effectively decouple static magnetic contributions to enable more accurate analysis, as reported in Figs. 2(a)–2(c).

Specific heat measurements were performed on polycrystalline $\text{Ba}_6\text{Cr}_2\text{S}_{10}$, as presented in Fig. 4(a). Notably, no λ -type anomaly—a characteristic feature of second-order magnetic phase transitions—was observed near the T_N [(8 ± 1) K], and this trend is consistent with previous reports [8]. When combined with the absence of dynamic critical behaviors (shown by μSR and ac susceptibility measurements, as discussed earlier) and the lack of a λ -type specific heat anomaly, the transition around T_N is consistent with a first-order-like transition [30]. Here, we should mention that the absence of a λ peak anomaly or the presence of a very weak anomaly in the specific heat data at a second-order phase transition of a low-dimensional magnet is not uncommon (see Fig. 1 in Ref. [31]). This is due to the onset of correlations that set in well above the transition to a three-dimensional magnetic order that the muons are sensitive to. Consequently, more theoretical and experimental efforts are needed to reveal the nature of this transition.

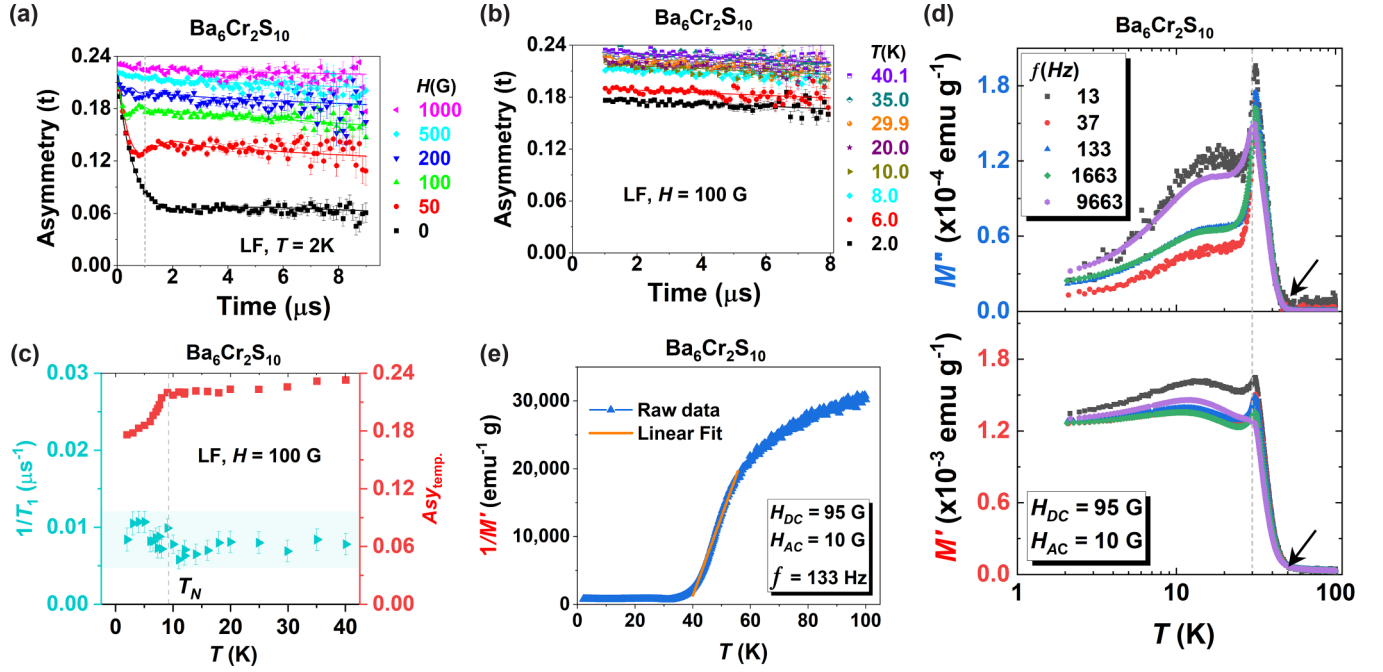


FIG. 2. Longitudinal field μSR and ac susceptibility measurements of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$. (a) Field dependence of LF- μSR spectra at $T = 2\text{ K}$. (b) Temperature dependence of μSR spectra under an applied longitudinal field of 100 G. (c) $1/T_1$ and corresponding temperature-dependent asymmetry Asy_{temp} extracted from the LF- μSR time spectra in panel (b). (d) Frequency- and temperature-dependent real (M' , lower panel) and imaginary (M'' , upper panel) components of the ac susceptibility. Note that the background dc field (H_{DC}) is 95 G, while the alternating field (H_{AC}) is 10 G. The distinction between H_{DC} and H_{AC} exists only in ac susceptibility measurements. In other cases, such as dc magnetization, μSR , and specific heat measurements, the field refers to the dc field; for simplicity, we use H instead of H_{DC} within this study. The black arrow indicates the temperature at which the ac susceptibility starts to increase abruptly. (e) A linear fit to the reciprocal of the real component ($1/M'$) of the ac susceptibility at a fixed frequency of 133 Hz reveals that the sharp phase transition at 30 K is indicative of ferromagnetic coupling.

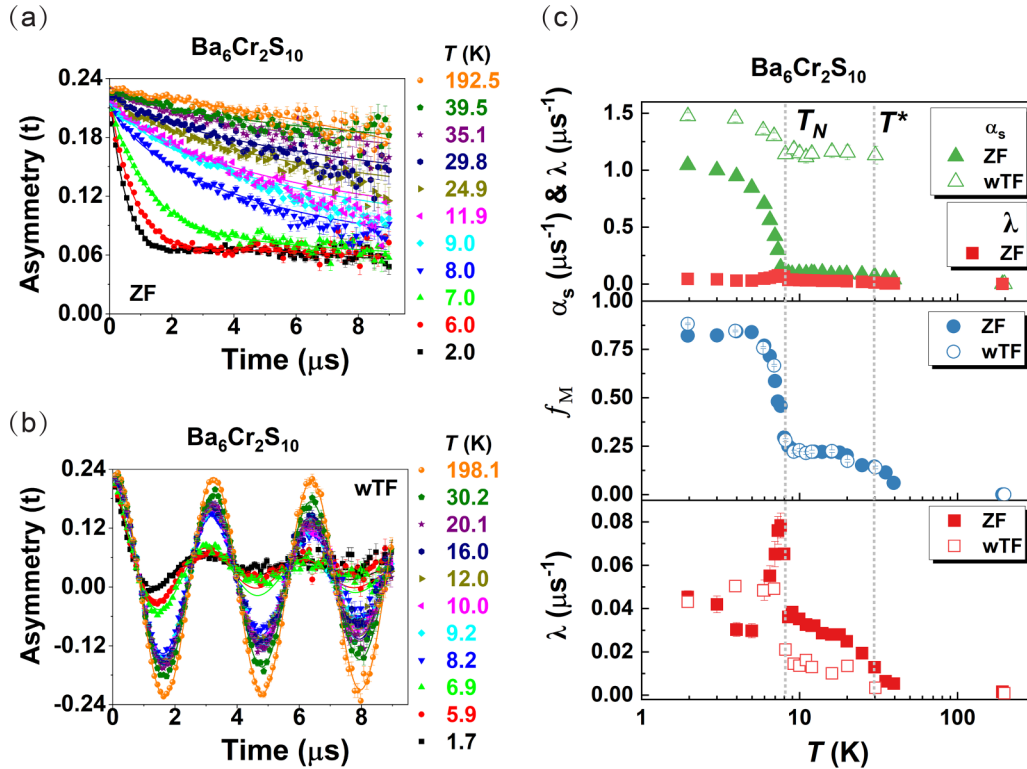


FIG. 3. μSR time spectra and summary of analysis results for polycrystalline $\text{Ba}_6\text{Cr}_2\text{S}_{10}$. (a) Zero-field μSR spectra. (b) Weak transverse field μSR spectra. (c) Summary of temperature-dependent analysis results, including the static ordered volume fraction f_M , the fast relaxation rate α_s , and the slow relaxation rate λ .

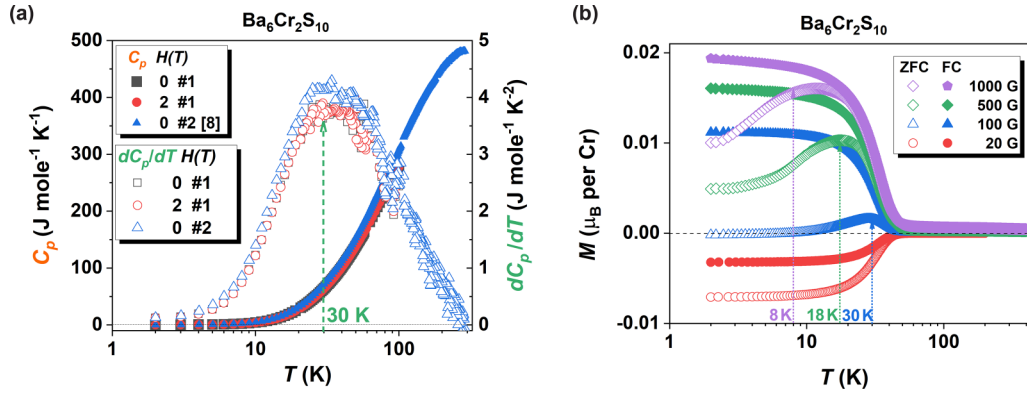


FIG. 4. Specific heat analysis and dc magnetization of polycrystalline $\text{Ba}_6\text{Cr}_2\text{S}_{10}$. (a) Specific heat (C_p) and its first-order derivative with respect to temperature (dC_p/dT) analysis: Note that “#1” corresponds to Sample #1, while “#2” corresponds to Sample #2, with the data for Sample #2 extracted from Ref. [8]. (b) Field- and temperature-dependent dc magnetization of polycrystalline $\text{Ba}_6\text{Cr}_2\text{S}_{10}$. Note that the results measured under zero-field cooling (ZFC) and field cooling (FC) procedures are denoted by empty symbols and solid symbols, respectively.

Furthermore, a considerable broad peak in dC_p/dT (spanning the temperature range from 2 to 300 K) is evident around 30 K—This feature corresponds directly to the static magnetic signal observed in dc magnetization measurements under 100 G [empty symbols in Fig. 4(b)] and the dynamic magnetic response characterized by ac susceptibility [Fig. 2(d)], linking the specific heat signature to both the static and dynamic magnetic behaviors of polycrystalline $\text{Ba}_6\text{Cr}_2\text{S}_{10}$. Notably, this same ~ 30 K broad peak has also been recently observed in time-resolved ultrafast pump-probe spectroscopy measurements [32], further validating the universality of this temperature-dependent feature across distinct experimental characterization techniques. This 30 K anomaly has the following features: (1) The static ordered volume fraction f_M exhibits a well-defined temperature dependence, reaching $\sim 14\%$ at 30 K [middle panel of Fig. 3(c)]. (2) The ac susceptibility values at 30 K are higher than those at (8 ± 1) K [Fig. 2(d)]. (3) No specific heat C_p anomaly was observed even at 2 T [Fig. 4(a)], whereas the dc magnetization [Fig. 4(b)] can be effectively tuned by external fields, with peaks at (30 ± 1) K (100 G), (18 ± 1) K (500 G), and (9 ± 1) K (1000 G), respectively.

One possibility for the 30 K anomaly could stem from “unidentified magnetic impurities,” as suggested by the authors in Ref. [32], yet they evaded detection by x-ray diffraction (XRD) [8] due to the technical resolution limit. Here, we propose another hypothesis that the broad peak around 30 K originates from the complex interplay between crystal structural distortions and magnetic coupling based on these complementary characterizations in the present study (e.g., dc magnetization, ac susceptibility, specific heat, and μSR measurements). Specifically, (1) as temperature decreases from the high-temperature region, crystal structural distortions occur gradually; consequently, dC_p/dT changes progressively and reaches its maximum around 30 K [Fig. 4(a)]. These structural distortions that break central symmetry—the origin of ferroelectricity—give rise to a spontaneous polarization reversible by an external electric field (see Fig. 5 of Ref. [8]). The emergence of ferroelectricity is one consequence of space-symmetry breaking in ferrotoroidicity. (2) The magnetic coupling of the antiferromagnetic phase 1

(AFM1, confirmed by neutron and calculations in Ref. [8]) evolves gradually and competes with thermal fluctuations in $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ since it contains spins. Owing to quantum effects, a portion of the sample enters a short-range magnetic ordered phase even at temperatures well above T_N . This behavior can account for the gradual increases in dc magnetization [Fig. 4(b)], f_M and a_s [both in Fig. 3(c)], and the dynamic susceptibility [Fig. 2(d)]. This short-range magnetic ordered phase can be effectively tuned by an external field for which we provided further evidence from dc magnetization measurements [Fig. 4(b)]. The emergence of antiferromagnetic ordering below T_N is one consequence of time-symmetry breaking in ferrotoroidicity. (3) Crystal structural distortions [e.g., charge density wave (CDW) ordering] are not uncommon in low-dimensional materials, such as the quasi-1D compound $\text{K}_{0.3}\text{MnO}_3$ [33]. In certain quasi-1D compound instances, such as $\text{K}_{0.3}\text{MnO}_3$, the emergence of the CDW ordering can lead to a reduction in magnetic susceptibility (see Fig. 1 in Ref. [33]). An analogous behavior is also evident in the dc magnetization measurements of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ performed under an applied magnetic field of 20 G [see red symbols in Fig. 4(b)]. Additionally, a theoretical preprint on arXiv supports the argument that complex interplay between CDW ordering and antiferromagnetic coupling exists in this quasi-1D $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ system [34]. Besides, no field dependence was observed for either C_p or the first-order derivative with respect to temperature (dC_p/dT)—This observation indicates that magnetic contributions do not dominate the specific heat response in the measured temperature and field ranges. In other words, the lattice-specific heat component is the primary contributor to the overall specific heat response of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ system. To be rigorous, we should point out that this “structural-magnetic coupling” hypothesis, while it can well explain these observed data, requires further validation.

C. Muon site estimation and local field calculations

According to the neutron results [8], the ordered moment size is $1.03 \mu_B$ under the AFM1-type spin structure. To estimate the muon stopping site, we calculated the elec-

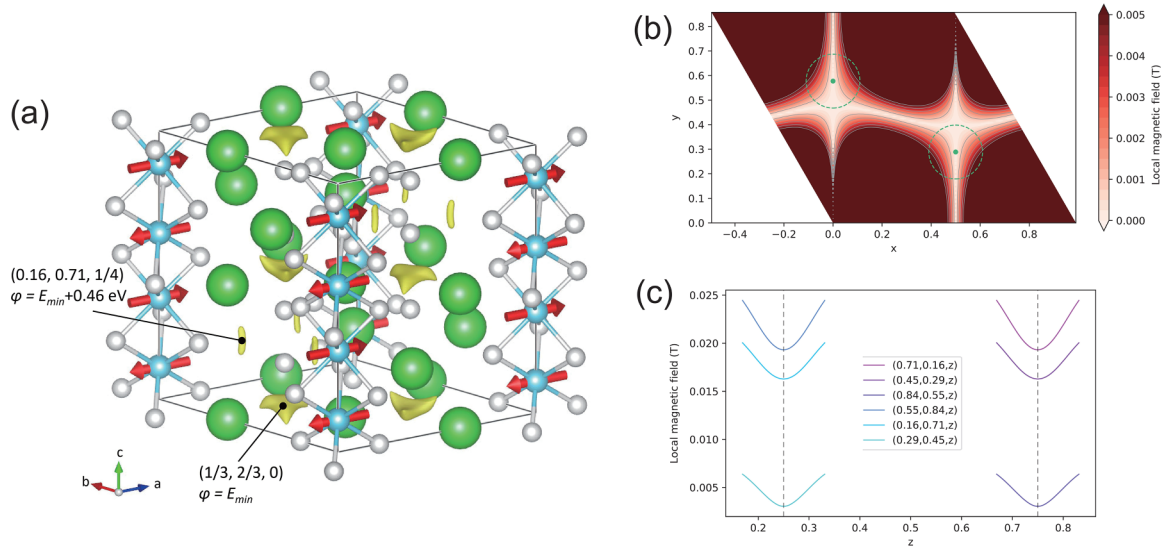


FIG. 5. Muon site estimation and local field calculations. (a) Possible muon sites based on the electrostatic potential approach. The yellow isosurfaces at $\phi = E_{\min} + 0.5$ eV enclose regions of lower potential. (b) Two-dimensional distribution of the local field magnitude in the $(x + \frac{1}{\sqrt{3}}y, \frac{2}{\sqrt{3}}y, 0)$ plane calculated for the AFM1-type spin structure. The Cartesian frame is defined so that the crystallographic a axis lies along x and the c axis along z . The green dot and surrounding dashed circle represent the potential minimum ($\phi = E_{\min}$) site and the region within 1 Å from it. (c) Line profiles of the local field magnitude calculated along the c axis within ± 1 Å around the points at $\phi = E_{\min} + 0.46$ eV.

trostatic potential $\phi(r)$ at interstitial positions in the perfect lattice using the Vienna Ab Initio Simulation Package (VASP). A single-point self-consistent field calculation was converged with the same parameters as those employed in the band-structure calculations described above. $\phi(r)$ was then obtained by summing the contributions from the resulting electron density distribution and the ionic cores. Figure 5(a) shows the unit cell with the $\phi(r) = E_{\min} + 0.5$ eV isosurfaces. Here, $E_{\min}$ denotes the global minimum value of the potential, which is found at sites equivalent to $(1/3, 2/3, 0)$. The enclosed volume containing this most probable muon stopping site extends within the ab plane. Another enclosed volume is centered at positions equivalent to $(0.16, 0.71, 1/4)$ and elongated along the c axis. ϕ at the center of this second candidate site is 0.46 eV higher than $E_{\min}$. The AFM1-type spin structure was imposed as indicated by the arrows in Fig. 5(a), and the distribution of spontaneous local magnetic fields around the possible muon sites was calculated using the MUESR package [35]. The ordered moment size was set to $1 \mu_B$, and a simple dipole sum was performed over the magnetic moments contained within a Lorentz sphere with a radius of 97 Å to evaluate the magnitude of the local field at the center.

Figure 5(b) displays the distribution of the local field magnitude in the $(x + \frac{1}{\sqrt{3}}y, \frac{2}{\sqrt{3}}y, 0)$ plane. At the potential minimum ($\phi = E_{\min}$) site marked by the green dot, the dipole fields are completely canceled. Even so, considering that the enclosed volume containing the $\phi = E_{\min}$ site in Fig. 5(a) spreads within the ab plane, and that in real systems the muon induces an asymmetric local lattice distortion, it is possible that the muons probe finite local fields of several millitesla at positions slightly offset from the zero-field site within the plane. Figure 5(c) shows the line profiles of the local field magnitude calculated along the c axis within ± 1 Å around the points at $\phi = E_{\min} + 0.46$ eV. At around this

second candidate site, local fields of several tens of millitesla are expected to arise. Positive muons undergo Larmor precession around the direction of the local magnetic field at a frequency determined by the muon gyromagnetic ratio, $\gamma_\mu = 2\pi \times 135.534 \text{ Mrad(s/T)} \simeq 852 \text{ Mrad(s/T)}$. Utilizing the experimentally fitted relaxation rate, $a_s = 1.13 \mu\text{s}^{-1}$ (where $a_s = \gamma_\mu B_{\text{loc}}$) obtained from ZF μSR experiments, the local magnetic field was calculated to be approximately 1.33 mT. This value is consistent with the proposed muon site, as illustrated in Fig. 5(b). It is important to note that the ZF μSR spectra presented in Fig. 3(a) exhibit no oscillations, indicating that the muons experience a distribution of local fields rather than a single-valued B_{loc} . The broad, exponential-like relaxation without a discernible minimum—contrary to the expectation from a static Kubo-Toyabe function—likely reflects either dynamic spin fluctuations or magnetic field inhomogeneity. These observations suggest that the local field (B_{loc}) represents an average over the spatial inhomogeneity of the magnetic environment. This strong agreement confirms the reliability of our local field estimation and the consistency between theoretical predictions and experimental observations.

D. Magnetic specific heat

To further deepen our understanding of the magnetic specific heat response, we adapted the calculations of the Heisenberg-type Hamiltonian for $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ as follows:

$$H = \sum_{\langle i,j \rangle \in J_1, J_2, J_3} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + J_4 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + A \sum_i (S_i^z)^2 + E_0, \quad (1)$$

where the spin operators for Cr atoms at sites i and j are $\mathbf{S}_i$ and $\mathbf{S}_j$, respectively. J_1 , J_2 , and J_3 are the exchange coupling constants between the first, second, and third nearest chromium

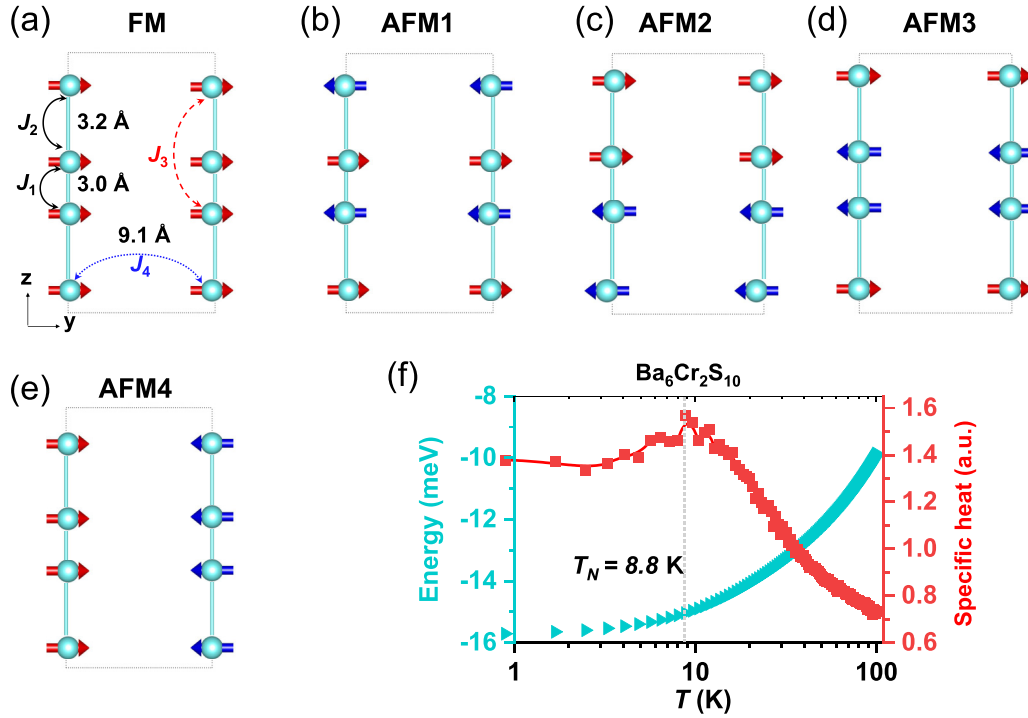


FIG. 6. Schematic diagram of magnetic configurations and Monte Carlo results of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$. To determine the magnetic coupling constants, we consider a ferromagnetic configuration [panel (a)] and four antiferromagnetic configurations [panels (b)–(e)], where the AFM1 configuration corresponds to the ground state. Panel (f) presents Monte Carlo results for the energy and specific heat as functions of temperature, from which a T_N of 8.8 K is derived for a Hubbard U value of 2.5 eV.

(Cr) atoms in a chain, with distances of 3.0, 3.2, and 6.2 Å, respectively. J_4 is the coupling constants between Cr atoms in nearest chains with a distance of 9.1 Å. The parameter A represents the single-ion magnetic anisotropy, defined as the difference between the energies of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ with out-of-plane and in-plane magnetization. The result obtained is $A = 0.42$ meV/Cr, which is consistent with the experimental findings [8]. To obtain coupling constants $J_1 \sim J_4$, we employed the energy mapping method [36] and considered five different configurations, as shown in Figs. 6(a)–6(e), including four antiferromagnetic (AFM) configurations and one ferromagnetic (FM) configuration. Among them AFM1 [Fig. 6(b)] is the ground state. The energies for these different magnetic configurations can be expressed as follows:

$$\begin{aligned}
 E_{\text{FM}} &= 4J_1S^2 + 4J_2S^2 + 8J_3S^2 + 24J_4S^2 + E_0, \\
 E_{\text{AFM1}} &= -4J_1S^2 - 4J_2S^2 + 8J_3S^2 + 24J_4S^2 + E_0, \\
 E_{\text{AFM2}} &= -4J_1S^2 + 4J_2S^2 - 8J_3S^2 + 24J_4S^2 + E_0, \\
 E_{\text{AFM3}} &= 4J_1S^2 - 4J_2S^2 - 8J_3S^2 + 24J_4S^2 + E_0, \\
 E_{\text{AFM4}} &= 4J_1S^2 + 4J_2S^2 + 8J_3S^2 - 8J_4S^2 + E_0. \quad (2)
 \end{aligned}$$

The exchange coupling parameters of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ can be calculated by solving this equation. According to our DFT results with a Hubbard U of 2.5 eV, the relative energies of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ in the FM, AFM1, AFM2, AFM3, and AFM4 states are 250.6, 0, 32.2, 227.4, and 250.7 meV, respectively, indicating AFM1 [Fig. 6(b)] as the ground state. The calculated exchange interactions J_1S^2 , J_2S^2 , J_3S^2 , and J_4S^2 are 33.435, 5.427, -0.137 , and 0.0006 meV, respectively. The Monte

Carlo simulation yields a T_N of 8.8 K, which agrees with the experimental value of (8 ± 1) K [Fig. 3(c)] and previously reported neutron-scattering measurements [8]. In addition, no anomalous behavior was observed near 30 K when the crystal parameters were fixed for calculations. This observation provides further support for the possibility of crystal structural distortions, as proposed in our previous discussion.

III. MATERIALS AND METHODS

Materials synthesis and characteristics. Polycrystalline $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ samples were synthesized under high-temperature and high-pressure conditions, as detailed in Ref. [8]. To eliminate any interference from residual radioactivity resulting from previous neutron-scattering experiments, all μSR measurements in this study were conducted on a separately prepared, nonirradiated set of samples. Alternating current susceptibility, specific heat, and electrical transport measurements were performed using a physical property measurement system, while dc magnetization measurements were carried out with a magnetic property measurement system. μSR spectra were obtained at the TRIUMF in Vancouver, Canada, with the LAMPF spectrometer under ZF, LF, and TF conditions. Measurements were taken with a helium gas-flow cryostat capable of reaching ~ 2 K.

μSR analysis. The μSR analysis was conducted in the time domain using the MUSR-FIT software, based on CERN's ROOT framework for particle physics data analysis [37]. The μSR data consistently display features indicative of field inhomogeneity and possibly slow dynamics, including the absence

of oscillations, the smooth monotonic relaxation in ZF data, and the lack of a well-defined minimum typical of a static Kubo-Toyabe response. This suggests that the fluctuating on a timescale is either comparable to or slower than the muon lifetime. Until now, there is no well-established model to analyze the μ SR time spectra in the quasi-one-dimensional antiferromagnet in the textbook and published literature to address this case. Although several materials (e.g., $\text{CeOs}_4\text{Sb}_{12}$ [38,39], CePt_3Si [40,41]) have exhibited signatures of magnetic Bragg peaks in neutron-scattering experiments, they do not display oscillations in μ SR measurements. In the present study, following numerous comparative attempts, the model presented below proves one of the most suitable options. ZF and wTF μ SR spectra were analyzed using functions (3) and (4) within the time range of 0–9 μs , to achieve an approximate fit to the experimental data. Note that to better trace the intrinsic signature of static ordered magnetism under ZF and wTF conditions, we employed a stretched Kubo-Toyabe function, where the stretched parameter β was fixed at 1 for ZF measurements and set to 1.369 for wTF measurements, respectively. Here, the stretched exponent ($\beta = 1.369$) provides a continuous between the diluted moment Lorentzian case ($\beta = 1$) and the dense moments Gaussian case ($\beta = 2$) [28]. We infer that it may be directly related to the quasi-one-dimensional characteristics of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ under the transverse field.

Asymmetry(t)

$$= \text{Asy}_{\text{total}} \left\{ f_M \left\{ \frac{1}{3} + \frac{2}{3} [1 - (a_s t)^\beta] e^{-(a_s t)^\beta / \beta} \right\} + (1 - f_M) \left[\frac{1}{3} + \frac{2}{3} (1 - (\Delta t)^2) e^{-(\Delta t)^2 / 2} \right] \right\} e^{-(\lambda t)^{0.5}} \quad (3)$$

Asymmetry(t)

$$= \text{Asy}_{\text{total}} \left\{ f_M \left\{ \frac{1}{3} + \frac{2}{3} [1 - (a_s t)^\beta] e^{-(a_s t)^\beta / \beta} \right\} + (1 - f_M) \cos(\gamma_\mu H_{\text{ext}} t + \phi) \right\} e^{-(\lambda t)^{0.5}} \quad (4)$$

The total asymmetry ($\text{Asy}_{\text{total}}$) and the nuclear dipolar relaxation rate (Δ) remain constant and are typically determined at the highest temperature within the paramagnetic state. The static ordered volume fraction (f_M), the amplitude of static random fields (a_s), and the slow relaxation rate (λ) are treated as free parameters. It is noteworthy to mention that the value of λ is more than an order of magnitude smaller than that of a_s in the $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ system, reflecting the amplitude of the dynamic depolarization rate and/or the static longitudinal slow decay. Consequently, these two contributions could not be distinguished. To address this issue, LF- μ SR measurements were conducted and analyzed using function (5) over a time range of 1–9 μs .

$$\text{Asymmetry}(t) = \text{Asy}_{\text{temp}} e^{-t/T_1} \quad (5)$$

The temperature dependence asymmetry, referred to as $\text{Asymmetry}_{\text{temp}}$, is treated as a free parameter across the entire temperature range. Additionally, the spin fluctuation rate, denoted as $1/T_1$, is also considered a free parameter.

TABLE I. Results of magnetic coupling constants and T_N calculated with $U = 0$ –5 eV.

U (eV)	Coupling constants (meV)				Ground	
	$J_1 S^2$	$J_2 S^2$	$J_3 S^2$	$J_4 S^2$	state	T_N (K)
0	67.738	17.371	1.468	0.014		23.0
1	47.536	10.334	0.352	0.006		20.1
2	33.435	5.427	−0.137	0.0006	AFM1	12.4
2.5	27.859	3.466	−0.282	−0.002		8.8
3	23.000	1.748	−0.387	−0.003		6.2
4	14.928	−1.142	−0.529	−0.008	AFM2	2.5
5	8.491	−3.474	−0.614	−0.011		9.5

Computational details and Monte Carlo simulations. All calculations utilized DFT in the vasp [42]. The exchange-correlation potential is defined by the Perdew-Burke-Ernzerhof (PBE) form with the generalized gradient approximation (GGA) [43]. The electron-ion potential is represented by the projector-augmented wave method [44]. The plane-wave cutoff energy was set to 650 eV. A $3 \times 3 \times 2$ Γ -centered K -point mesh was employed for Brillouin zone sampling. The convergence criteria for energy and force were set to 10^{-6} eV and 10^{-2} eV/Å, respectively. We carried out the calculation of GGA + U with $U = 2.5$ eV for Cr in all DFT calculations. $U = 2.5$ eV was chosen since it gives a ground state AFM1 and a $T_N = 8.8$ K close to the experimental value (8 ± 1 K). The ground-state is AFM1 with T_N values of 23.0, 20.1, 12.4, 8.8, and 6.2 K for $U = 0, 1, 2, 2.5$, and 3 eV, respectively. In contrast, for $U = 4$ and 5 eV, the ground state switches to AFM2, which deviates from the experimental findings. Their coupling constants are given in Table I. A Heisenberg-type Monte Carlo simulation was conducted on a $20 \times 20 \times 20$ lattice with 16 000 magnetic points for $\text{Ba}_6\text{Cr}_2\text{S}_{10}$, involving 3×10^6 steps at each temperature. The last one-third of the steps were used to calculate temperature-dependent physical quantities.

IV. CONCLUSIONS

In summary, we comprehensively characterized the quasi-1D ferrotoroidic candidate $\text{Ba}_6\text{Cr}_2\text{S}_{10}$, via experiments (electrical transport, dc magnetization, ac susceptibility, specific heat, μ SR) and first-principles calculations. We confirmed a narrow band gap (~ 0.707 eV, experimentally; ~ 0.680 eV, theoretically), found static magnetism dominates down to 2 K ($\sim 89\%$ volume fraction) with a first-order-like magnetic transition at $T_N = (8 \pm 1)$ K, and attributed the ~ 30 K (T^*) peak to crystal structure-magnetic coupling distortion, potentially linked to ferrotoroidicity. As the first work integrating μ SR with complementary experiments and theory to investigate ferrotoroidicity, this study clarifies the fundamental phase transition around T_N and provides insights to advances the understanding of $\text{Ba}_6\text{Cr}_2\text{S}_{10}$ and its analogs. Moving forward, more extensive experimental studies—utilizing diverse techniques—and theoretical investigations are needed, particularly focusing on compounds in single-crystal form, to explore the structure-magnetic coupling in this field more broadly and deeply.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [45].

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