Persistent Spin Dynamics Intrinsic to Amplitude-Modulated Long-Range Magnetic Order

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An incommensurate elliptical helical magnetic structure in the frustrated coupled-spin-chain system $\text{FeTe}_2\text{O}_5\text{Br}$ is surprisingly found to persist down to 53(3) mK ($T/T_N \sim 1/200$), according to neutron scattering and muon spin relaxation. In this state, finite spin fluctuations at $T \rightarrow 0$ are evidenced by muon depolarization, which is in agreement with specific-heat data indicating the presence of both gapless and gapped excitations. We thus show that the amplitude-modulated magnetic order intrinsically accommodates contradictory persistent spin dynamics and long-range order and can serve as a model structure to investigate their coexistence.

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Geometrical frustration is a common precursor for exotic magnetic ground states—from long-range ordered (LRO) incommensurate (IC) spiral states to highly disordered frozen spin-glass states [1]. It can even lead to spin liquids, where spin fluctuations may endure down to zero temperature—persistent spin dynamics (PSD) [2] is present. Such behavior is typically found in highly frustrated pyrochlore and kagome spin systems with macroscopically degenerate ground states [3]. Since spin fluctuations hinder the onset of extended static correlations, PSD and LRO are generally considered mutually exclusive. Remarkably, their coexistence in the same phase has been reported in several frustrated magnetic systems [4–11] but still lacks a suitable explanation.

To explore this phenomenon, we focus on the $\text{FeTe}_2\text{O}_5\text{Br}$ multiferroic, in which the magnetic exchange network consists of alternating Fe^{3+} ($S = 5/2$) spin chains coupled by weaker frustrated interactions within the bc layers [12] (Fig. 1). The magnetic order at 5 K, i.e., well below the Néel temperature $T_N = 10.5$ K, was described as an IC collinear amplitude-modulated (AMOD) structure, with magnetic vector $\mathbf{q} = (\frac{1}{2}, 0.463, 0)$ [13]. In such a state, only part of the total Fe magnetic moment at each site contributes to LRO, while its counterpart (at each site) is disordered and may fluctuate. Generally, on cooling the ordered component fully develops, which manifests either as a “squaring” of the AMOD structure [14] or as a consecutive transition, where a perpendicular ordered component develops leading to a circular helix [15]. Since $\text{FeTe}_2\text{O}_5\text{Br}$ exhibits no subsequent transition down to 300 mK [16], it is possible that PSD and LRO coexist.

In this Letter, we report on a combined study of spherical neutron polarimetry (SNP) and neutron diffraction, which reveals that the IC AMOD magnetic structure

persists to the lowest accessible temperatures ($T/T_N \sim 1/175$). This is consistent with muon spin relaxation (μSR) measurements at $T/T_N \sim 1/200$, which in addition to static LRO signify the presence of PSD. The coexistence of LRO and PSD is supported by specific-heat data, indicating gapped as well as gapless magnetic excitations. Our study suggests that this is intrinsic to AMOD magnetic

FIG. 1 (color online). Top: Magnetic structure in $\text{FeTe}_2\text{O}_5\text{Br}$ at 2 K. Light (orange) and dark (blue) spheres are Fe_1 and Fe_2 magnetic ions, respectively. The shades (colors) of the magnetic moments (arrows) are different for the eight magnetically inequivalent sites Fe_{mn} . The amplitude of the moment changes with its orientation within the ellipsoidal envelope. Black and gray lines denote the dominant intrachain and the weaker interchain exchange interactions, respectively. Bottom: IC amplitude modulation of the Fe_{11} magnetic moment along the b axis.

structures. It offers a well-defined framework and a coherent explanation for the coexistence of LRO and PSD that has been missing for the known cases [11].

We begin by a reinvestigation of the $\text{FeTe}_2\text{O}_5\text{Br}$ magnetic ground state, combining single-crystal neutron diffraction and SNP, which was proven before to be invaluable for determination of complex magnetic structures [17]. The experiments were performed at 5 and 2 K, respectively, on high-quality single crystals [13] with an average size of $15 \times 8 \times 2 \text{ mm}^3$ at the Swiss Neutron Spallation Source, Paul Scherrer Institute (PSI), Switzerland. For SNP the MuPAD device on the triple axis spectrometer TASP ($\lambda = 3.2 \text{ \AA}$) was used, while the diffraction experiment employed the single-crystal diffractometer TriCS ($\lambda = 2.317 \text{ \AA}$). Polarization matrices were measured for three different crystal orientations [18], accessing for the first time [13,19] also $hkl \pm \mathbf{q}$ ($l \neq 0$) magnetic reflections.

The general magnetic structure model, in which the magnetic order breaks all crystallographic symmetry operations [13,19], dictates the magnetic moment at a particular Fe site to follow an elliptical helix with the pitch along the magnetic $\mathbf{q}$ vector:

$$\mathbf{S}_{mn}(\mathbf{r}_i) = \mathbf{S}_{0mn}^{\text{Re}} \cos(\mathbf{q} \cdot \mathbf{r}_i - \psi_{mn}) + \mathbf{S}_{0mn}^{\text{Im}} \sin(\mathbf{q} \cdot \mathbf{r}_i - \psi_{mn}). \quad (1)$$

Here, vector $\mathbf{r}_i$ defines the origin of the i th cell, $m = 1, 2$ identifies the crystallographically inequivalent Fe sites, and $n = 1-4$ denotes the four Fe positions within the crystallographic unit cell (see the caption of Table I). The complex vector $\mathbf{S}_{0mn}$ is determined by its real and imaginary components $\mathbf{S}_{0mn}^{\text{Re}}$ and $\mathbf{S}_{0mn}^{\text{Im}}$, respectively, defining the amplitude and the orientation of the magnetic moments, while ψ_{mn} denotes a phase shift. We assume the same moment $\mathbf{S}_{0mn} \equiv \mathbf{S}_{0m}$ for all crystallographically equivalent Fe sites. The reliability of the refined magnetic structure is ensured by simultaneous refinement of all SNP and integrated magnetic-peak intensities data [18]. The best solution (Fig. 1 and Table I) with the dominant component pointing $\sim 45^\circ$ away from a towards the $-b$ axis agrees

TABLE I. Parameters of the best magnetic structure model at 2 K for two independent magnetic atoms (Fe_1 and Fe_2), and eight magnetic phases ψ_{mn} , i.e., one for each of the magnetic Fe_{mn} atoms in the unit cell ($m = 1, 2, n = 1-4$). The sites $\text{Fe}_{12} - \text{Fe}_{14}$ are obtained from Fe_{11} [0.1184(6), $-0.001(1)$, 0.9734(7)] and $\text{Fe}_{22} - \text{Fe}_{24}$ from Fe_{21} [0.9377(6), 0.2953(1), 0.8562(6)] by symmetry elements i , 2_{1y} , and $2_{1y}i$, respectively. The orientation of the moments is given in the a^*bc coordinate system.

$s = \text{Re, Im}$	Fe_1^{Re}	Fe_1^{Im}	Fe_2^{Re}	Fe_2^{Im}	n	ψ_{1n}	ψ_{2n}
$S_{0x}^s/ S_0^s $	0.70	0.71	0.64	0.76	1	0.00	0.00
$S_{0y}^s/ S_0^s $	0.70	-0.67	0.76	-0.61	2	0.04	0.93
$S_{0z}^s/ S_0^s $	-0.14	0.20	-0.10	0.21	3	0.17	0.20
$ S_{0m}^s / S_0^s $	0.39	0.96	0.35	1.00	4	0.21	0.15

with the previously proposed simplified collinear IC AMOD structure [13]. In addition, a small perpendicular component is identified, resulting in an overall elongated-elliptical cycloid ($|S_0^{\text{Re}}|/|S_0^{\text{Im}}| \sim 0.37$), with its normal canted $\sim 15^\circ$ away from the c axis. The new data thus reveal a magnetic structure that combines AMOD and helical properties. The quality of the new refinement reflects in the reduced $\chi^2 = 9.6$ of the polarization matrices being notably decreased with respect to its value in the collinear AMOD (14.7) and the circular helical structures (14.9) [18]. A small misfit [Figs. 2(a) and 2(b)] inevitably originates from a weak nuclear-magnetic-interference term [18].

At $T \rightarrow 0$, the ordered component of the magnetic moments in the elliptical IC AMOD structure is expected to grow at the expense of the disordered one. This can reflect either as a change of the magnetic-reflection intensities, if circular helix is formed, or, alternatively, as additional magnetic reflections with propagation vectors $3\mathbf{q}$, $5\mathbf{q}$, ... and intensities $1/9$, $1/25$, ... of the first-order magnetic reflections in the case of "squaring" [14]. To detect such changes, single crystal neutron diffraction was performed at 60(3) mK ($T/T_N \sim 1/175$), where we recorded broad k scans of several $hkl \pm \mathbf{q}$ pairs, i.e., intersecting the positions of $hkl \pm n\mathbf{q}$, $n = 3, 5, \dots$, reflections. Surprisingly, we find no significant difference between 60 mK and 2 K data as well as no trace of higher harmonics [Fig. 2(c)], which implies that both the AMOD ordered component and its disordered counterpart are still present at the lowest accessible temperatures.

Since the neutron diffraction experiments probe only static magnetism, we employed a local-probe μSR technique, which is extremely sensitive to internal magnetic fields and can distinguish between fluctuating and static

FIG. 2 (color online). Refinement quality for (a) polarization matrices and (b) integrated intensities. (c) Top: k scans at 60(3) mK and 2 K across $(3.5 - 0.5 \pm \delta 0)$ reflections, witnessing the absence of higher harmonics, expected to occur at marked positions. Bottom: The difference between the two scans which match within the error bar.

magnetism, as well as between LRO and static magnetic disorder [20]. The μ SR experiments were performed on the MUSR instrument at the ISIS facility, Rutherford Appleton Laboratory, United Kingdom, on the same high-purity powder samples as used in our earlier study [18,21]. All the data in the following are shown with properly subtracted background signal ($\sim 15\%$) [18]. Preliminary results indicated that muons stop at several inequivalent positions [21]. However, to test the magnetic structure model, these must be precisely determined. New measurements were therefore first performed in the paramagnetic state, at 50 K [Fig. 3(a)], where weak static nuclear magnetic fields are expected to govern the μ^+ spin relaxation [20]. Since these fields can be exactly calculated from the crystal structure, they are essential for identification of the muon stopping sites, as demonstrated below. In the case of a single muon stopping site, in a paramagnetic powder sample the muon polarization in zero applied magnetic field (ZF) is given by the Gaussian Kubo-Toyabe relaxation function $G_{KT}(t, \Delta) = \frac{1}{3} + \frac{2}{3}[1 - (\Delta t)^2] \exp[-(\Delta t)^2/2]$ multiplied by the exponential function $\exp[-\lambda_L t]$. The former accounts for a static Gaussian nuclear field distribution with the width Δ/γ_μ ($\gamma_\mu = 2\pi \times 135.5$ MHz/T), whereas the latter describes a weak dynamical relaxation due to fast electronic fluctuations [20]. The resulting function has a single dip, which is removed only by a fast dynamical relaxation, i.e., $\lambda_L \geq \Delta$. Our ZF data exhibit a more complex behavior [Fig. 3(a)], as they require the two-component model

$$G(t) = [G_{KT}(t, \Delta_A) + G_{KT}(t, \Delta_B)] \exp[-\lambda_L t]. \quad (2)$$

This model is supported by measurements in longitudinal applied magnetic fields, where decoupling of the muon relaxation from nuclear-magnetic fields occurs in two steps at ~ 0.5 and ~ 2 mT [Fig. 3(a)], respectively. Simultaneous fit of all 50 K μ SR data to the two-component model, extended for the case of longitudinal applied magnetic fields [20] [Fig. 3(a)], yields the relative occupancies of the two muon stopping sites of 83(3)% and 17(3)% with corresponding $\Delta_A/\gamma_\mu = 0.064(5)$ mT and $\Delta_B/\gamma_\mu = 0.304(5)$ mT, whereas $\lambda_L = 0.039(3) \mu\text{s}^{-1}$.

Since μ^+ are positively charged particles and are generally expected to stop at the electrostatic-potential minima, we calculated the electrostatic potential in the $\text{FeTe}_2\text{O}_5\text{Br}$ unit cell by using density functional theory [18]. This way several possible stopping sites were identified [Fig. 3(b)], for which dipolar nuclear-magnetic field distributions were calculated [18]. The dominant Δ_A/γ_μ is found to be in excellent agreement with nuclear field distributions calculated at two local electrostatic-potential minima $P_1 = (0.59, 0.12, 0.43)$ and $P_2 = (0.25, 0.15, 0.90)$ [Fig. 3(b)]. These are thus assigned as the prime muon stopping sites. On the contrary, Δ_B/γ_μ does not agree with calculated distribution at any electrostatic-potential minima [18], so the third (least occupied) muon stopping site remains unassigned.

Identification of the P_1 and P_2 stopping sites allows us to calculate local dipolar magnetic fields from the ordered Fe moments and thus to double check the magnetic structure determined by neutrons. We computed [18] normalized field distributions $D_{ij}(B)$ at four inequivalent sites ($j = 1-4$) [Wyckoff position 4(e)] for each P_i ($i = 1, 2$) felt by muons stopping in random unit cells [insets in Fig. 3(c)]. In Fig. 3(c), the ZF μ SR data collected at 1.6 K at PSI [21] are shown together with the fit (dashed line) to the corresponding model

$$G(t) = \frac{1}{3} \exp[-(\lambda_L t)^\alpha] + \frac{2}{3} \sum_{i,j} \left[A_i \exp(-\lambda_i t) \int_0^\infty D_{ij}(B) \cos(\gamma_\mu B t) dB \right]. \quad (3)$$

In this expression the first term, commonly called the “1/3 tail” [20], describes muons in a powder sample, whose initial polarization is parallel to the internal magnetic field and therefore changes only due to fluctuations of this field. This term (dynamical relaxation rate λ_L and stretch exponent α) is determined from long-time decay measurements presented in the next paragraph. Now we focus on the second term, which depicts oscillations due to internal fields induced by the LRO magnetic order. These oscillations are damped (λ_i) by spin fluctuations and/or static relaxation resulting from a distribution of muon stopping

FIG. 3 (color online). (a) Longitudinal-field μ SR measurements on powder sample $\text{FeTe}_2\text{O}_5\text{Br}$ at 50 K (symbols) and corresponding fits (lines)—see the text. (b) Electrostatic potential at $b = 0.15$, i.e., intersecting the most pronounced minima, including P_1 and P_2 . (c) Zero-field μ SR measurement at 1.6 K with corresponding two- (dashed line) and three-component (solid line) fits given by the magnetic structure (see the text). Insets: Calculated field distributions $D_{ij}(B)$ at P_i ($i = 1, 2$) sites for the four magnetically inequivalent positions j within the unit cell.

positions around P_i [18], with occupancy A_i ($\sum_i A_i = 1$). The fit yields $\lambda_1 = 0.60(5) \mu\text{s}^{-1}$, $\lambda_2 = 50(8) \mu\text{s}^{-1}$, and $A_2/A_1 = 1.4(1)$. Most importantly, our model excellently accounts for the experimental oscillation frequencies of the muon polarization determined by $D_{ij}(B)$ (no adjustable parameters) and thus firmly affirms the elongated-elliptical IC AMOD state. The small discrepancy [Fig. 3(c)] is most likely due to the neglected 17(3)% of muons, with unknown stopping position (Δ_B). Indeed, adding a third component with $A_3 = 17\%$ and its decay approximated by $\exp(-\lambda_3 t)$ leads to a perfect agreement with the experiment [Fig. 3(c)]. The improved model yields $\lambda_3 = 11(1) \mu\text{s}^{-1}$, while parameters λ_1 , λ_2 , and A_2/A_1 stay within the error bars of the two-component model. The agreement of the neutron and the μSR results proves that μSR probes the intrinsic magnetic properties and can thus provide an invaluable insight into spin dynamics of the $\text{FeTe}_2\text{O}_5\text{Br}$ system.

In the case of completely static ($\lambda_L = 0$) local magnetic fields, the $1/3$ tail persists in the $t \rightarrow \infty$ limit [Eq. (3)]. Since preliminary ZF μSR data implied its decay [21], we extended these measurements to longer times and very low temperatures [Fig. 4(a)]. The new data confirm the decay of the $1/3$ tail and clearly show its persistence down to the lowest accessible temperature of 53(3) mK, i.e., $T/T_N \sim 1/200$. Since this decay can only be of dynamical origin, it unambiguously proves that muons, experiencing static magnetic fields due to the IC AMOD order, also experience local-field fluctuations, i.e., revealing the coexistence of PSD and LRO at $T \rightarrow 0$.

FIG. 4 (color online). (a) Muon depolarization (symbols) at low temperatures, indicating the presence of PSD, and corresponding fits (lines) to the stretched-exponential function $\exp[-(\lambda_L t)^{0.3}]$. Inset: Relaxation rate distribution at 53 mK determined from fit (see the text). (b) Extracted relaxation rates, showing linear temperature dependence (solid line). For comparison, activation behavior (dashed line) for gapped ($\epsilon_g = 11.5$ K) magnon excitations is shown. (c) Temperature dependence of the magnetic contribution to the specific heat, $c_{\text{mag}}(T)$, together with corresponding fits (see the text for details).

To obtain deeper insight into PSD, we focus on the dynamic part of the μSR signal, i.e., muon polarization for $t > 0.5 \mu\text{s}$, where the second term in Eq. (3) is already relaxed [Fig. 3(b)]. These data are fitted with $G(t) = \frac{1}{3} \exp[-(\lambda_L t)^\alpha]$ [Fig. 4(a)], where α accounts for the dynamical relaxation rate distribution $\rho_{\lambda_L}(\nu)$, related to the stretched-exponential function by the Laplace transform $\exp[-(\lambda_L t)^\alpha] = \int_0^\infty \rho_{\lambda_L}(\nu) \exp[-(\nu t)] d\nu$ [22]. A small $\alpha = 0.30(2)$, found temperature independent below T_N , indicates a very broad $\rho_{\lambda_L}(\nu)$ [inset in Fig. 4(a)], which most likely reflects a distribution and dynamical nature of the disordered parts of the magnetic moments in the AMOD state. Additionally, $\rho_{\lambda_L}(\nu)$ can be broadened, because three different muon stopping sites are present. The obtained λ_L [Fig. 4(b)] shows a linear temperature dependence and, most importantly, converges to a finite zero-temperature value $\lambda_L^0 = 0.012(2) \mu\text{s}^{-1}$, characteristic of PSD. We note that, in spite of the small λ_L^0 , the broad $\rho_{\lambda_L}(\nu)$ spans far into the μSR time window ($\nu > 0.1 \mu\text{s}^{-1}$), as evident from substantial experimental decay of the $1/3$ tail at early times [Fig. 4(a)].

In LRO antiferromagnets, $\lambda_L(T)$ is expected to follow either T^n dependence with $n > 2$ for $T \gg \epsilon_g$ or $\exp(-\epsilon_g/T)$ dependence for $T \ll \epsilon_g$ [11,23–25]. Here ϵ_g is the magnon energy gap. Considering $\epsilon_g = 11.5$ K, as determined in the recent antiferromagnetic-resonance study [12], $\lambda_L(T)$ should change exponentially with temperature in the inspected temperature range, which is clearly not the case [Fig. 4(b)]. Hence, $\lambda_L(T)$ signifies magnetic excitations, which are different from the usual magnon modes in LRO states. Finite relaxation at $T \rightarrow 0$ is typically found in gapless spin liquids where it is ascribed to quantum spin fluctuations [2,3,26,27]. In $\text{FeTe}_2\text{O}_5\text{Br}$, these most probably originate from the disordered component of the magnetic moment at each Fe site, which naturally accompanies the ordered component in an IC AMOD structure. The dual nature of the ground state is confirmed by the magnetic contribution $c_{\text{mag}}(T)$ to specific heat [16], which is proportional to T^3 below T_N and exhibits an additional broad hump around 3 K [Fig. 4(c)]. The former implies three-dimensional antiferromagnetic gapless excitations [5,28–30], while the latter reveals an additional thermally activated $\exp(-\epsilon_g/T)$ term due to the gapped magnetic excitations [29].

Finally, we point out similar spin dynamics in the magnetic ground state of volborthite [25], where spin-density-wave-like modulation [25] and IC spin correlations [31] were found. Moreover, recent calculations [32] showed that volborthite, like $\text{FeTe}_2\text{O}_5\text{Br}$ [12], has to be treated as a frustrated coupled-spin-chain system. This suggests that such systems are keen to form an IC AMOD magnetic ground state, which offers a sound phenomenological explanation of the coexistence of PSD and LRO.

In conclusion, we have found that in $\text{FeTe}_2\text{O}_5\text{Br}$ the elongated-elliptical IC AMOD magnetic structure persists

down to $T/T_N \sim 1/200$, as a result of frustrated chain topology. In this LRO state, fluctuations of the remaining disordered spin component at each magnetic site are intrinsic, and they manifest as PSD at $T \rightarrow 0$. Similar observations in volborthite suggest that IC AMOD magnetic order is a natural habitat for PSD and can serve as a model structure inherently encompassing the intriguing coexistence of PSD and LRO. This conjecture could be tested on linarite, for which frustrated chains were recently reported to induce a LRO AMOD structure [33]. Further in-depth theoretical investigations are required to account for PSD in IC AMOD structures on the microscopic level.

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