

Depth-resolved magnetic order in superconducting topological insulator/FeTe thin film heterostructures

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(Received 13 June 2025; accepted 4 September 2025; published 14 October 2025)

The search for chiral topological superconductivity in magnetic topological insulator (TI)-FeTe heterostructures is a key frontier in condensed matter physics, with potential applications in topological quantum computing. The combination of ferromagnetism, superconductivity, and topologically nontrivial surface states brings together the key elements required for chiral Majorana physics. In this work, we examine the interplay between magnetism and superconductivity at the interfaces between FeTe and a series of Te-based TI overlayers. In both Te/FeTe and superconducting MnBi₂Te₄/FeTe, any interfacial suppression of antiferromagnetism must affect at most a few nanometers. On the other hand, (Bi, Sb)₂Te₃/FeTe layers exhibit near-total suppression of antiferromagnetic ordering. Ferromagnetic Cr_x(Bi, Sb)_{2-*x*}Te₃ (CBST)/FeTe bilayers exhibit net magnetization in both CBST and FeTe layers, with evidence of interactions between superconductivity and ferromagnetism. These observations identify magnetic TI/FeTe interfaces as an exceptionally robust platform to realize chiral topological superconductivity.

DOI: 10.1103/15fx-3cr2

I. INTRODUCTION

The discovery of interface-induced superconductivity in heterostructures of FeTe and the topological insulator (TI) Bi₂Te₃ revealed a promising candidate for topological superconductivity [1]. Cr-doping of the TI to yield ferromagnetic Cr_x(Bi, Sb)_{2-*x*}Te₃ (CBST)/FeTe bilayers completes the prerequisites for chiral topological superconductivity: time-reversal symmetry breaking, topologically nontrivial surface states, and superconductivity [2–4]. Superconductivity also emerges at the interface between FeTe and the antiferromagnetic TI MnBi₂Te₄ (MBT), indicating compatibility with a range of magnetic states [5].

FeTe belongs to the same iron chalcogenide family as superconducting FeSe, but is a nonsuperconducting antiferromagnet (AFM) [6,7]. Selenium-doping FeTe yields superconductivity alongside suppression of the AFM order [6–12]. It has therefore been suggested that AFM suppression in FeTe plays a key role in the superconducting state, either through competing magnetic interactions or band-bending and charge transfer [13–15], but this question remains debated. There is significant overlap between the superconducting and AFM phases in bulk FeTe_{1-*x*}Se_{*x*} [7,16–18]. However, excess Fe content, which incorporates into interstitial sites, can both inhibit superconductivity and promote AFM ordering [19–21], so that the low superconducting volume fraction in the overlap region of *x* cannot exclude phase segregation.

This question is further complicated by the dimensionality change when moving from bulk crystals to thin film heterostructures. In FeTe_{1-*x*}Se_{*x*} (*x* < 0.1) nanoribbons studied with scanning tunneling microscopy (STM), superconductivity and AFM order were spatially separated, with superconducting edge states at which the AFM order is suppressed, and a nonsuperconducting AFM interior [22]. On the other hand, for monolayer FeTe on a Bi₂Te₃ crystal, spin-polarized STM indicated the coexistence of a superconducting

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TABLE I. Table of properties for each heterostructure studied. Properties listed for individual constituent materials represent the properties of the material in thin film form, but not in the heterostructure.

Bilayer System	BST/FeTe on SrTiO ₃ (001)	CBST/FeTe on SrTiO ₃ (001)	MBT/FeTe on SrTiO ₃ (001)	Te/FeTe on SrTiO ₃ (001)
Overlayer	(Bi, Sb) ₂ Te ₃	Cr _x (Bi, Sb) _{2-x} Te ₃	MnBi ₂ Te ₄	Te
Overlayer properties	Nonmagnetic topological insulator	Ferromagnetic topological insulator	Antiferromagnetic $T_N = 24$ K [24,25] topological insulator	Nonmagnetic, topologically trivial
Bilayer properties	Superconducting $T_{c,\text{onset}} \approx 12.5$ K $T_{c,0} \approx 10$ K	Superconducting $T_{c,\text{onset}} \approx 12.5$ K $T_{c,0} \approx 10$ K	Superconducting $T_{c,\text{onset}} \approx 11$ K $T_{c,0} \approx 9$ K	Nonsuperconducting
FeTe AFM	Suppressed	Suppressed	Not suppressed	Not suppressed

gap with bi-collinear AFM [23]. Indeed, there is evidence that in the TI-based systems, the superconductivity results from competition between AFM interactions in the FeTe and ferromagnetic interactions mediated by the topological surface state [13]. In this interpretation, it is these interfacial magnetic interactions, not the FeTe magnetic order, which are central to the emergent superconducting state in FeTe-based heterostructures.

Both AFM order and Fe stoichiometry in thin films are challenging to probe, so their role in superconducting TI/FeTe heterostructures remains a mystery. Here, we address these questions using thin film neutron diffraction, low-energy muon spin relaxation (LE- μ SR) spectroscopy, and polarized neutron reflectometry (PNR). We study four different FeTe-based heterostructures – (Bi, Sb)₂Te₃ (BST)/FeTe, CBST/FeTe, MBT/FeTe, and, as a control, Te/FeTe bilayers. BST is a nonmagnetic TI, while CBST and MBT are the prototypical magnetic TIs [24,26–29]. The electrical transport characteristics of these systems have been reported in our prior studies [2,5,13], with onset temperatures ($T_{c,\text{onset}}$) between 11 K and 12.5 K and zero-resistivity ($T_{c,0}$) between 9 K and 10 K. Representative temperature-dependent longitudinal resistance curves are shown in the Supplemental Material [30–33]. Table I summarizes the characteristics of the bilayers studied, while their crystal structures are shown in Figs. 1(a)–1(c).

In this work, we observe superconductivity both in systems where the AFM is nearly suppressed (BST/FeTe, CBST/FeTe) as well as those with nearly homogeneous AFM order (MBT/FeTe). LE- μ SR indicates that any AFM suppression in superconducting MBT/FeTe must affect a layer less than 2 nm thick, while such a layer in nonsuperconducting Te/FeTe bilayers is similarly limited to less than 3 nm in thickness, complicating the relationship between AFM and superconductivity in FeTe. Furthermore, we observe unexpected magnetic order in BST/FeTe and MBT/FeTe as well as a net magnetization within the FeTe. PNR provides direct evidence that the superconducting region is surrounded by ferromagnetic order in CBST/FeTe. With reported thicknesses of the superconducting region ranging from 6 nm–11 nm [2,5], our observations reveal interfacial superconductivity overlapping and coexisting with both AFM and ferromagnetism.

II. MAGNETIC ORDER IN FeTe UNDERLAYERS

To determine the magnetic order of the FeTe layers, we performed neutron diffraction measurements on superconducting CBST (15 nm)/FeTe (30 nm), superconducting MBT (10 nm)/FeTe (22 nm), and nonsuperconducting Te (15 nm)/FeTe (30 nm). Structural peaks for the SrTiO₃ (001) substrate and FeTe films are shown in the Supplemental Material [33]. Magnetic peaks should be temperature-dependent, appearing only in the ordered state. Figure 1 shows cuts along the [L0L] direction through the FeTe ($\frac{1}{2}, 0, \frac{1}{2}$) reflection, which is the most intense magnetic diffraction peak associated with bi-collinear AFM order in bulk FeTe [34].

There is no evidence of magnetic diffraction peaks in CBST/FeTe down to 1.5 K [Figs. 1(d) and 1(e)]. On the other hand, both MBT/FeTe [Figs. 1(f) and 1(g)] and Te/FeTe [Figs. 1(h) and 1(i)] exhibit statistically significant ($\frac{1}{2}, 0, \frac{1}{2}$) peaks, appearing at $T = 15$ K but not at $T = 90$ K, above the Néel temperature of bulk FeTe [7]. The ratio of MBT/FeTe and Te/FeTe peak intensities scale precisely as expected with the FeTe thicknesses of the two samples, as discussed further in the Supplemental Material [33]. We searched for other magnetic diffraction peaks, such as an incommensurate helimagnetic state [34], but no evidence of such reflections was found. Therefore, neutron diffraction supports suppressed AFM order in CBST/FeTe and long-range bi-collinear AFM order in both MBT/FeTe and Te/FeTe.

Magnetic interactions at the TI/FeTe interface are thought to be central to the superconducting state, but may differ from the rest of the film. We therefore use LE- μ SR to obtain depth-resolved magnetic information. By varying the kinetic energies of implanted muons (1 keV–5 keV), we tune the muon implantation depth. LE- μ SR probes the local magnetic field at each implantation site, where each muon (μ^+) spin precesses about the local magnetic field before decaying to yield a positron. The direction of positron emission is asymmetric with respect to the muon spin direction at the moment of decay. Thus, applying a weak transverse magnetic field – perpendicular to the initial muon polarization – yields a time-dependent directional asymmetry, which can be described as $A(t) = A_0 e^{-\lambda t} \cos(\omega_L t)$. This allows the extraction of the muon depolarization rate λ , which, along with the precession frequency ω_L , describes the distribution of local

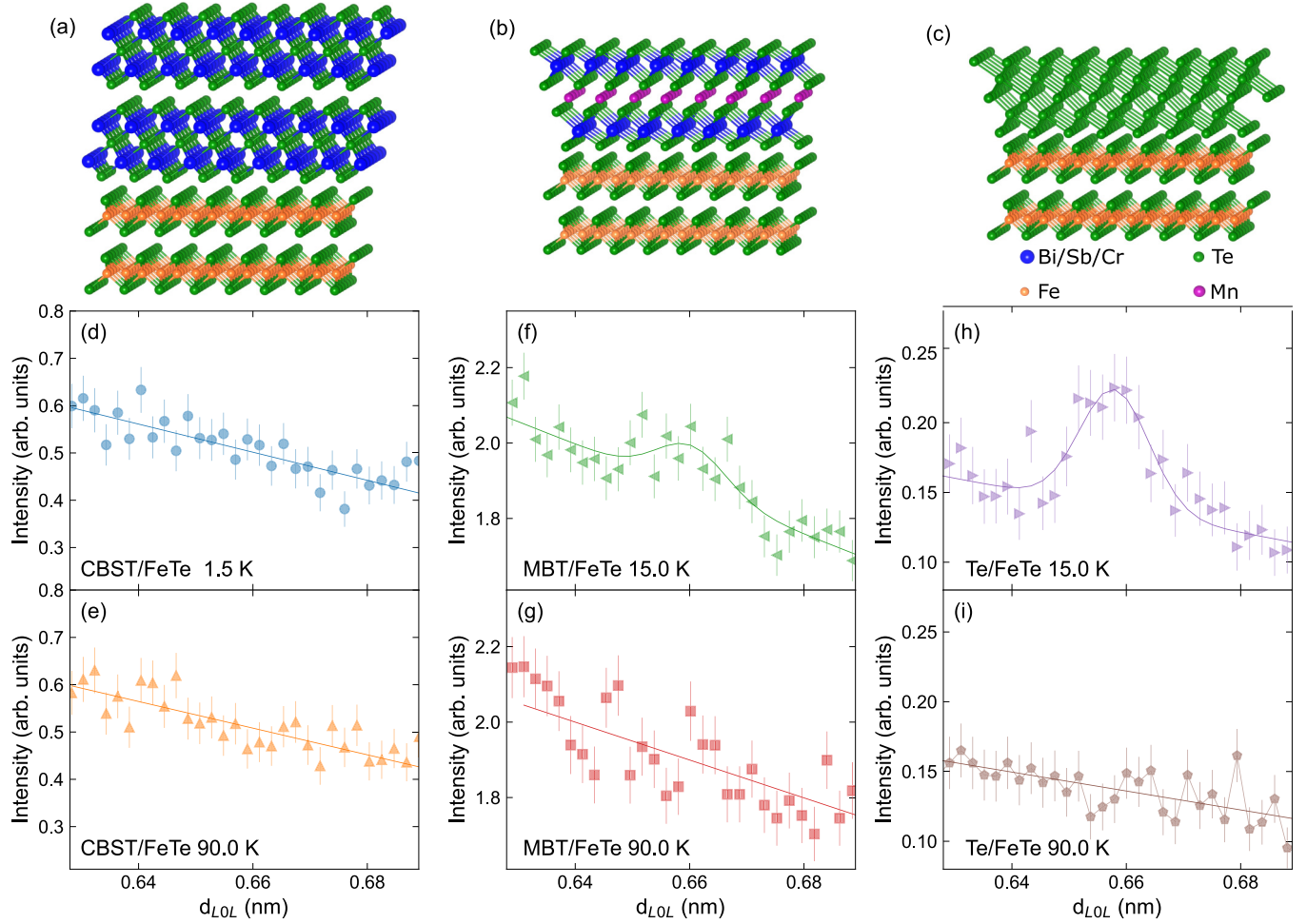


FIG. 1. (a)–(c) Schematics [36] of the (a) ferromagnetic CBST/FeTe, (b) AFM MBT/FeTe, and (c) nonsuperconducting Te/FeTe bilayer heterostructures. (d)–(e) A cut through the neutron diffraction data along the (LOL) direction of a representative CBST (15 nm)/FeTe (30 nm) sample at (d) 1.5 K and (e) 90 K. Cuts are taken at the expected position of the bulk-like ($\frac{1}{2}$ 0 $\frac{1}{2}$) film reflection. Similar cuts are shown for (f)–(g) an MBT (10 nm) / FeTe (22 nm) sample at (f) 15 K and (g) 90 K, and for (h)–(i) a Te (15 nm) / FeTe (30 nm) sample at (h) 15 K and (i) 90 K. All diffraction curves are plotted alongside fits to the data—a linear background plus a Gaussian peak if the fitted area was statistically significant. Error bars represent ± 1 standard deviation.

magnetic fields B . By tracking the temperature-dependence of the directional asymmetry $A_0(T)$ and normalizing it to a nonmagnetic high-temperature baseline, the fraction of muons which stop in a magnetically ordered film region (F_M) may be determined. Combining F_M with TRIM.SP simulations of the muon stopping depth distributions [35], we can probe the layer-dependent magnetically ordered volume fraction.

Figure 2 compares LE- μ SR data from BST (15 nm)/FeTe (30 nm), MBT (10 nm)/FeTe (22 nm), and Te (15 nm)/FeTe (30 nm) bilayers. The implantation profile simulations for the BST/FeTe [Fig. 2(a)] and MBT/FeTe [Fig. 2(b)] samples show that each implantation energy probes multiple regions of the heterostructure. While the lowest energies are more sensitive to the TI layers, at 3 keV we primarily probe FeTe for both samples, with minimal contribution from the SrTiO₃ substrate [Fig. 2(c)]. The 3 keV data [Fig. 2(d)] reveal increased F_M in Te/FeTe and MBT/FeTe upon cooling from 80 K to 40 K. Based on the halfway point of this transition, we estimate an FeTe T_N of 55 K–60 K for both bilayers, corresponding to an approximate composition of Fe_{1.10}Te [34]. This stoichiometry

matches the bi-collinear AFM order observed *via* neutron diffraction, though it is near the Fe_{1.11}Te Lifshitz point above which Fe_{1+x}Te is helimagnetic [34].

The dashed lines in Fig. 2(d) represent the upper bound of F_M based on the stopping profiles, calculated by assuming that all potentially magnetic layers (FeTe, MBT) are 100% magnetically ordered. In MBT/FeTe, any drop from this maximum could be explained by AFM suppression in either the FeTe or in the MBT. The assumption that all AFM suppression occurs in a region of FeTe at the interface imposes an upper limit on the thickness of this layer. We find that $7.2\% \pm 0.7\%$ (MBT/FeTe) or $7.0\% \pm 0.7\%$ (Te/FeTe) of 2 keV muons are implanted in a nonmagnetic region, limiting any nonmagnetic interfacial FeTe layer to thicknesses of approximately 2 nm and 3 nm, respectively. Thus, the FeTe in both MBT/FeTe and Te/FeTe is close to fully magnetically ordered, with nearly identical nonmagnetic volume fractions for both superconducting and nonsuperconducting bilayers. We conclude that suppressing the AFM is not sufficient to establish superconductivity.

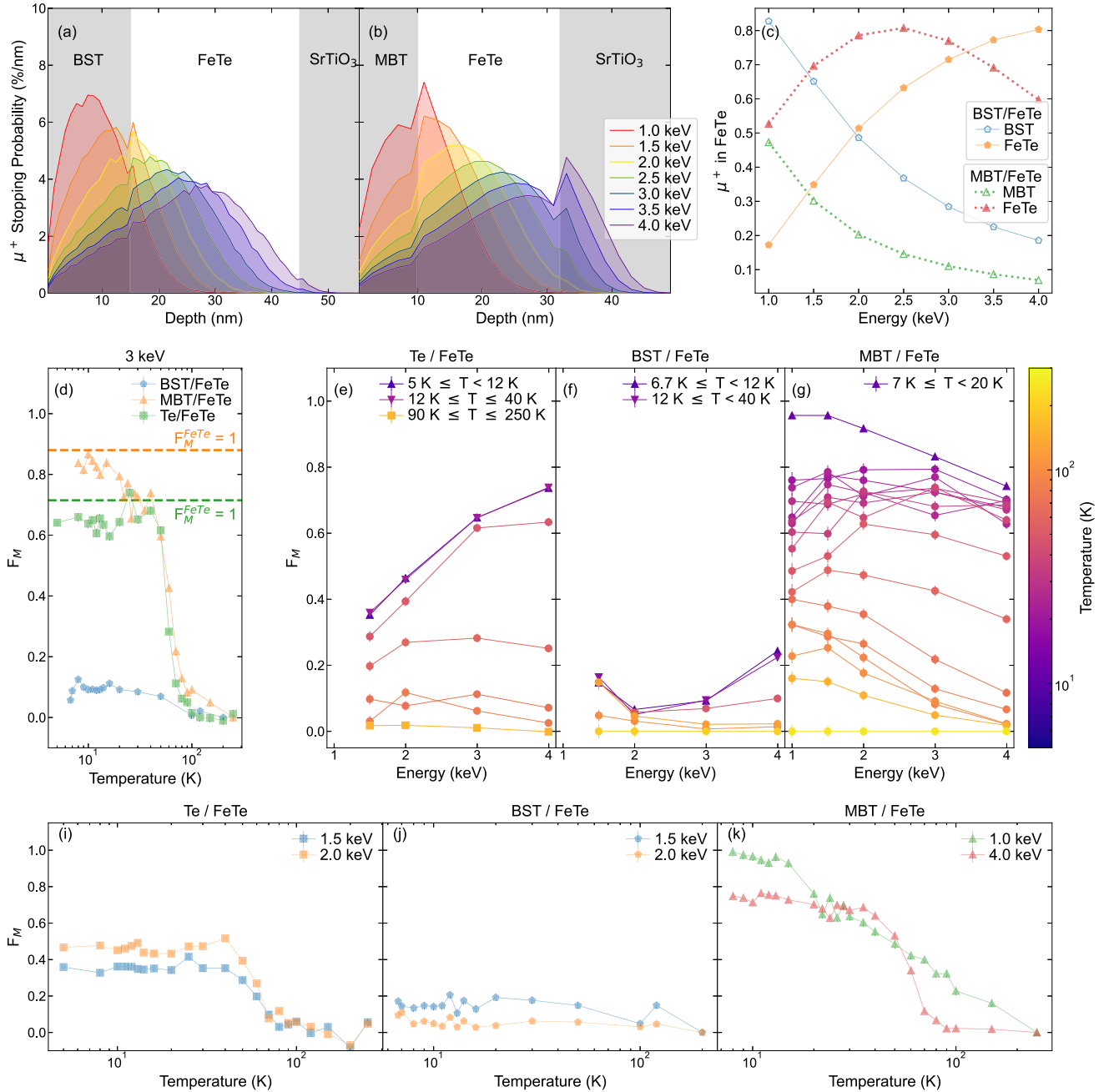


FIG. 2. (a) Simulated muon stopping probability as a function of depth expressed as the % of implanted muons expected to stop per nm for a given bin for the BST (15 nm)/FeTe (30 nm) bilayer. (b) Muon stopping probability for the MBT (10 nm)/FeTe (22 nm) bilayer. (c) The calculated fraction of muons stopping in BST, MBT, and FeTe layers vs. implantation energy. (d) F_M as a function of temperature for BST/FeTe, MBT/FeTe, and Te/FeTe at 3 keV. Dashed lines indicated the expected value for F_M if all magnetic layers are 100% ordered. (e) F_M vs implantation energy and temperature for Te (15 nm) / FeTe (30 nm), (f) BST (15 nm) / FeTe (30 nm), and (g) MBT (10 nm)/FeTe (22 nm) bilayers. Note that some temperature regimes, indicated in the legend, have been averaged if little or no temperature dependence was noted. The full unbinned dataset is plotted in the Supplemental Material [33]. (h) F_M vs temperature for Te (15 nm)/FeTe (30 nm) at 1.5 keV and 2.0 keV. (i) F_M vs temperature for BST (15 nm)/FeTe (30 nm) at 1.5 keV and 2.0 keV. (j) F_M vs temperature for MBT (10 nm)/FeTe (22 nm) at 1.0 keV and 4.0 keV. Error bars represent ± 1 standard deviation.

In BST/FeTe samples, the F_M temperature dependence is much weaker. LE- μ SR requires that the AFM volume fraction of FeTe must be suppressed by 80% – 90%, supporting

the interpretation of neutron diffraction from CBST/FeTe. Superconductivity is therefore clearly compatible with AFM suppression throughout the majority of the FeTe layer.

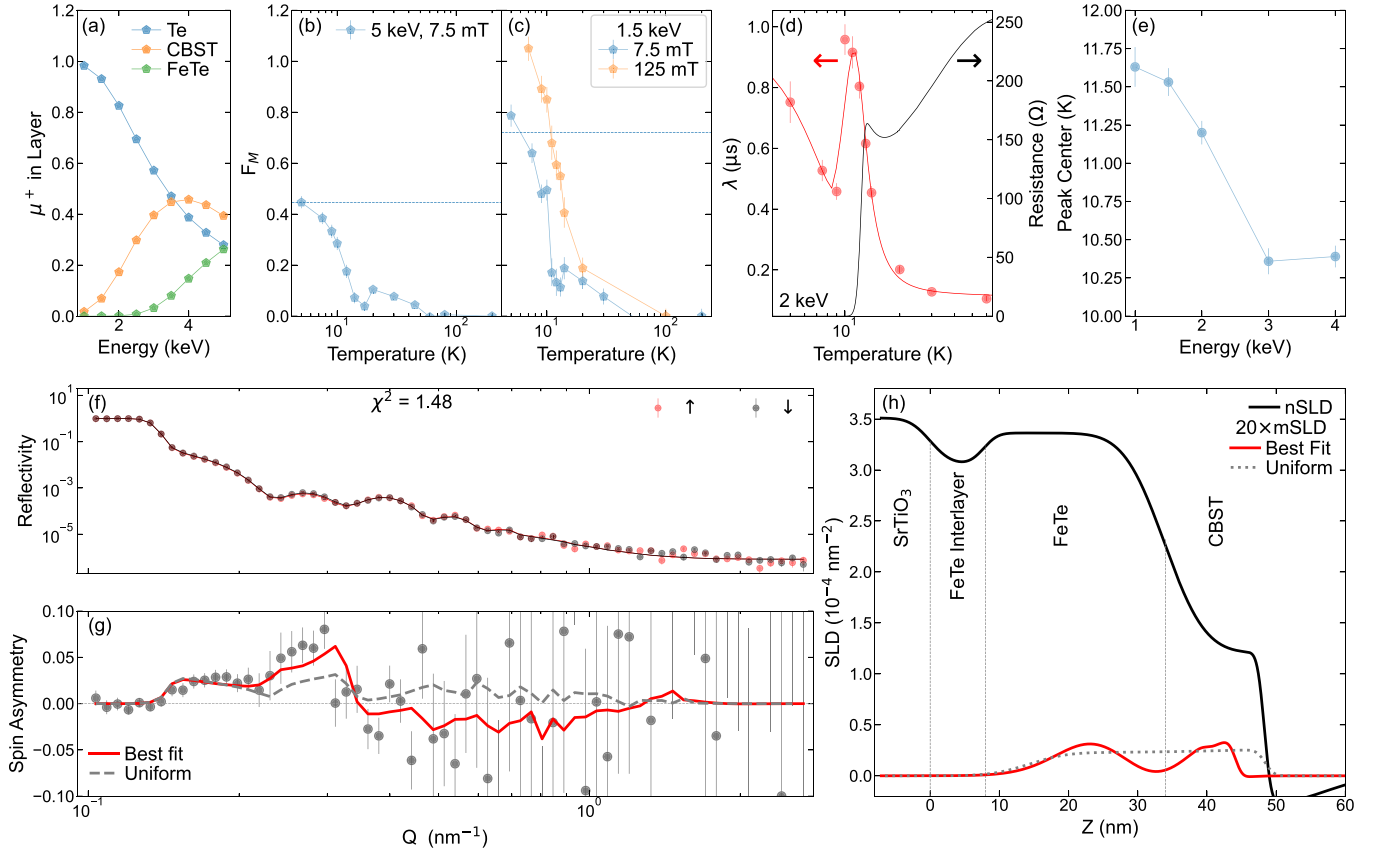


FIG. 3. (a) Muon fraction stopping in each layer of Te (25 nm)/CBST (15 nm)/FeTe (10 nm) vs implantation energy. (b) F_M vs temperature for 5 keV muons in Te (25 nm)/CBST (15 nm)/FeTe (10 nm), with a dashed line showing the expected value for 100% ferromagnetically ordered CBST and 20% ordered FeTe. Measured in a transverse field of 7.5 mT (c) F_M vs temperature for CBST (15 nm)/FeTe (30 nm) at an implantation energy of 1.5 keV, with a dashed line showing the expected value for 100% ferromagnetically ordered CBST and 20% ordered FeTe. Measured in transverse fields of 7.5 mT and 125 mT. (d) Muon depolarization rate (red circles) vs temperature alongside fit to the data using a Lorentzian peak and a magnetic order parameter. Data taken at 2 keV and 7.5 mT. The black line is a typical resistance vs temperature curve for CBST/FeTe bilayers. (e) Temperature of the λ peak vs muon implantation depth. (f) PNR measured for the CBST (15 nm)/FeTe (30 nm) sample under a 1 T applied magnetic field at 4 K. Solid lines represent theoretical fits to the data. (g) Spin asymmetry calculated from the measured data. The dashed gray line is the spin asymmetry of a fit assuming uniform magnetization in each layer, while the solid red line allows for an interfacial zero-magnetization region. (h) nSLD and mSLD profiles corresponding to the fitted curves in (f) and (g). Error bars represent ± 1 standard deviation.

III. MAGNETIC ORDER IN TI OVERLAYERS

We next examine magnetic order within the TI overlayer. Figures 2(e), 2(f), and 2(g) show the energy and temperature-dependent F_M for Te/FeTe, BST/FeTe, and MBT/FeTe, respectively. Below 100 K, 1.5 keV muons implanted into BST/FeTe yield a higher F_M , 0.16 ± 0.01 , than 2.0 keV muons, 0.058 ± 0.007 . This unexpected behavior is highlighted in Figs. 2(i) and 2(j), where Te/FeTe displays the expected trend in which higher energies are more sensitive to the FeTe and consequently exhibit higher F_M . This trend is reversed in BST/FeTe, implying the emergence of magnetic order in the BST.

For MBT/FeTe, the 1 keV, 1.5 keV, and 2 keV implantation energies exhibit F_M increases upon cooling from 250 K to 90 K. This feature is well above T_N for either MBT or FeTe, and weakens at higher implantation energies. These trends are further emphasized in Fig. 2(k), which compares $F_M(T)$ at 1 keV and 4 keV for MBT/FeTe.

A promising explanation for emergent magnetic order in BST and MBT overlayers is Fe-doping. Excess Fe in $\text{FeTe}_{1-x}\text{Se}_x$ has long been thought to suppress superconductivity and strengthen AFM order [19–21]. Fe incorporation into TIs is also reported to yield a net magnetization, so that the diffusion of excess Fe from the $\text{Fe}_{1.1}\text{Te}$ into the BST or MBT could simultaneously stabilize dilute magnetism in the BST and MBT while promoting interfacial superconductivity [37–39].

All LE- μ SR data presented thus far have focused on nonferromagnetic TI layers. With measurements on undoped BST/FeTe as a baseline, we evaluate the magnetic ordering in the ferromagnetic Cr-doped TI system. We performed LE- μ SR measurements on trilayer Te (25 nm)/CBST (15 nm)/FeTe (10 nm) and bilayer CBST (15 nm)/FeTe (30 nm). A layer-by-layer breakdown of the expected muon locations for the trilayer samples is shown as a function of implantation energy in Fig. 3(a). The CBST (15 nm)/FeTe (30 nm) implantation profiles closely track those already shown for BST (15 nm)/FeTe (30 nm).

Figure 3(b) shows F_M for the Te/CBST/FeTe trilayer samples under an applied transverse field of 7.5 mT. These samples exhibit a sharp F_M increase below the superconducting T_c . The horizontal dashed line represents the expected F_M from 100% ferromagnetically ordered CBST and 20% AFM ordered FeTe, as suggested by the measurements of BST/FeTe. The 5 K data matches the calculated value, indicating fully ordered CBST below T_c . Figure 3(c) similarly shows F_M for 1.5 keV muons in the CBST/FeTe bilayers at 7.5 mT, where the calculated F_M value (horizontal dashed line) agrees with the 5 K data within a 95% confidence interval.

In Figs. 3(b) and 3(c), F_M dips by approximately 0.05 near T_c before increasing upon further cooling. Commensurate with this dip in F_M , there is a peak in the muon depolarization rate, shown in Fig. 3(d). This peak is absent from the BST/FeTe, MBT/FeTe, and Te/FeTe data, as well as examples previously reported LE- μ SR on CBST films [40]. Fitting the peak with a Lorentzian function, we plot the peak temperature vs muon implantation depth and find a shift to lower temperatures for muons probing deeper into the FeTe. We speculate that the F_M dip and λ peak originate in the direct spatial overlap between the ferromagnetism and unconventional superconductivity. This interpretation is supported by the lack of these features in BST/FeTe bilayers.

In 125 mT applied fields, F_M increases, reaching $F_M \approx 1.0$ [Fig. 3(c)]. That is, all muons stopping in either CBST or FeTe are in close proximity to large static magnetic fields. The LE- μ SR data, therefore, require field-induced magnetic order in the FeTe in the superconducting state. AFM order in CBST/FeTe is substantially suppressed and would not be expected to exhibit a significant field response. Instead, we propose the polarization of excess interstitial Fe, leading to the emergence of a net magnetization in the FeTe at high field. Such behavior explains magnetometry reported in Bi₂Te₃ (9 nm)/FeTe (140 nm) by Q. L. He *et al.* [41], who found an unexpected enhancement of the net magnetization below the superconducting T_c , similar to the lineshapes shown in Figs. 3(b)–3(c).

IV. MAGNETISM AT THE INTERFACE

The emergence of a net magnetization from the polarization of interstitial Fe presents an opportunity to probe the deeper origins of superconductivity in TI/FeTe bilayers. Small interfacial changes in Fe-stoichiometry are challenging to detect and quantify through electron microscopy, especially if a thin layer of interfacial FeTe distributes excess Fe throughout the TI cap. Precisely determining the magnetization distribution within the FeTe layer may provide evidence for interfacial Fe depletion in the form of a layer with reduced magnetization.

Therefore, we performed PNR measurements on CBST (15 nm)/FeTe (30 nm). PNR is sensitive to the depth profile of the composition and density of the sample through the nuclear scattering length density (nSLD) and the net in-plane magnetization through the magnetic scattering length density (mSLD), both of which are determined through theoretical fits to the data. Measurements were performed in a 1 T applied magnetic field at 4 K and 13 K. The 4 K spin-dependent

neutron reflectivities are shown in Fig. 3(f), while the spin asymmetry, defined as the difference between the spin-up and spin-down neutron reflectivities normalized by their sum, is shown in Fig. 3(g). Solid lines represent fits to the data. The best-fit nSLD and mSLD depth profiles for the 4 K data are shown in Fig. 3(h). The fitted thicknesses match the target values almost exactly, and there is a region of the FeTe film with reduced nSLD near the FeTe/SrTiO₃ interface. This lower nSLD region is consistent with a discontinuous film and has a thickness of $7.9 \text{ nm} \pm 0.3 \text{ nm}$, similar to the reported minimum thickness necessary to achieve the zero-resistance state [2].

At 13 K (see Supplemental Material [33]), the spin splitting is vanishingly small, and, consequently, the net magnetizations of the CBST and FeTe layers are on the edge of statistical significance. The data are best fit by a net magnetization of $12.4 \text{ kA/m} \pm 5.8 \text{ kA/m}$ in CBST and $4 \text{ kA/m} \pm 2 \text{ kA/m}$ in FeTe. Below T_c , the spin splitting grows, so that the 4 K data are best fit by $6.3 \text{ kA/m} \pm 2.6 \text{ kA/m}$ within the CBST layer and $9.5 \text{ kA/m} \pm 1.9 \text{ kA/m}$ within the FeTe. At both temperatures, the magnetization within a region near the FeTe/CBST interface is effectively zero.

To examine the sensitivity of PNR to this interface region with zero net magnetization, we fit the data using an alternative model, which imposed a uniform magnetization within the CBST layer and within the FeTe layer. Thus, this “uniform” model does not allow an interfacial layer of suppressed magnetization. The best-fit resulting from the “uniform” model is shown as gray dashed lines in Fig. 3(g), while the associated mSLD profile is shown as a gray dotted line in Fig. 3(h). The “uniform” model is unable to reproduce the spin asymmetry between $Q = 0.2 \text{ nm}^{-1}$ and $Q = 0.3 \text{ nm}^{-1}$, demonstrating that an interfacial region of mSLD = 0 is needed to describe the data. Further, attempts to fit the data without a magnetized region of the FeTe layer, shown in Fig. S16, increase χ^2 to 1.73.

V. DISCUSSION AND CONCLUSIONS

Though our LE- μ SR measurements cannot distinguish between different types of magnetic order, when combined with our understanding of ferromagnetism and AFM in these structures from PNR and neutron diffraction, respectively, the energy dependence of magnetic volume fraction F_M yields a depth-resolved picture of the magnetic order, shown schematically in Fig. 4. As discussed further in the Supplemental Material [33], We employ a toy model in which any deviation from the maximum possible F_M is presumed to originate in an AFM dead layer at the top of the FeTe. The simulated fraction of implanted muons with depth is then used to estimate the thickness of this dead layer, placing upper limits on the suppression of magnetic order in the FeTe layers. For Te/FeTe, this implies a non-AFM region which is $2.8 \text{ nm} \pm 0.2 \text{ nm}$ thick. This is thicker than any possible AFM dead layer in MBT/FeTe, where the toy model, which presumes $F_M = 1$ within the MBT layer, limits the thickness of any AFM dead layer in FeTe to at most 2 nm. We emphasize that the AFM suppression in both Te/FeTe and MBT/FeTe affects a very small volume fraction, such that we cannot precisely locate it at the interface with the present data. Trace regions of

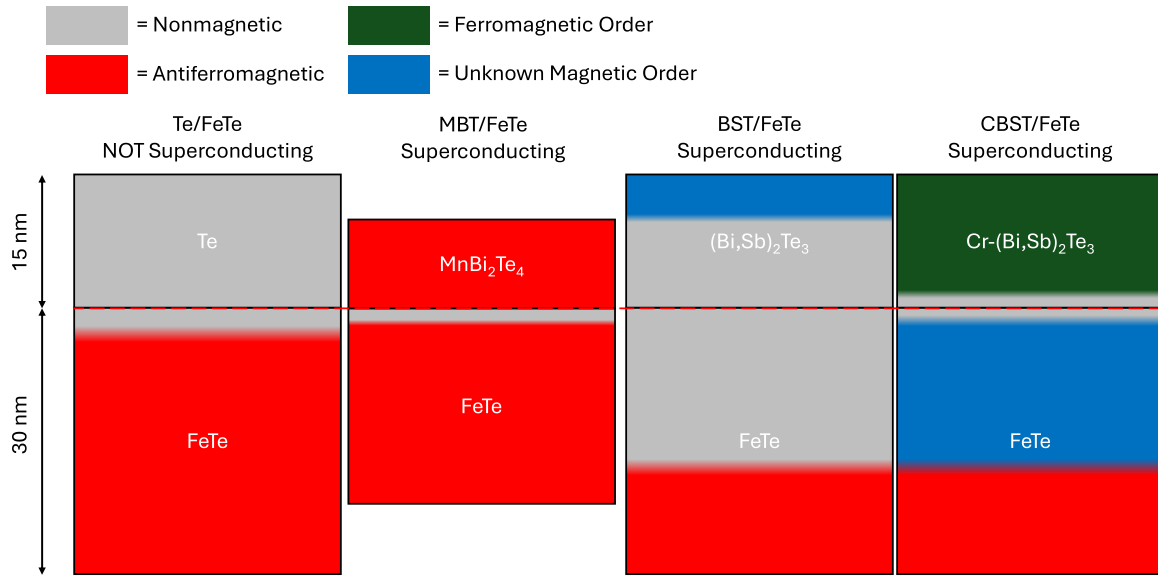


FIG. 4. Schematic depth profiles of magnetic order in Te/FeTe, MBT/FeTe, BST/FeTe, and CBST/FeTe, as estimated using PNR and LE- μ SR measurements. Nonmagnetic regions are shown in grey, antiferromagnetic regions in red, and ferromagnetic regions in green. While the LE- μ SR of the BST/FeTe sample requires some magnetic order in the BST layer, the specific nature remains uncertain, marked in blue. Similarly, while field-polarized Fe moments are the most obvious explanation for the high-field FeTe magnetization in the CBST/FeTe, a definitive identification is not possible at this time (blue).

non-AFM FeTe could plausibly be spread throughout the layer in either or both samples. The slight AFM suppression in Te/FeTe, therefore, does not provide evidence of Fe-diffusion across the interface.

On the other hand, BST/FeTe samples exhibit suppressed FeTe AFM, with the F_M energy-dependence best reproduced by an 18 nm thick AFM dead layer at the TI/FeTe interface (see Supplemental Material [33] for more details). Thus, superconductivity coexists with either nearly homogeneous (MBT/FeTe) or substantially suppressed (BST/FeTe and CBST/FeTe) AFM. At the same time, the AFM depth profile of nonsuperconducting Te/FeTe is nearly indistinguishable from superconducting MBT/FeTe. These observations raise the question of whether AFM suppression is necessary for superconductivity or merely a byproduct of other interface effects, such as modified Fe-stoichiometry or interactions mediated by the TI surface state [13,20].

The PNR profiles highlight the importance of Fe-stoichiometry in TI/FeTe bilayers. On the FeTe side of the interface, the emergence of a net magnetization at high magnetic field, which increases dramatically below T_c , is attested by both the PNR and LE- μ SR of CBST/FeTe. We speculate that Fe interstitials known to reside in the FeTe van der Waals gap become spin polarized in high-field, yielding a net magnetization and depolarizing muons which stop nearby. This picture is supported by x-ray magnetic circular dichroism measurements (see Supplemental Fig. S19) of the Fe $L_{3,2}$ -edge, which reveal a vanishingly small net magnetization at 200 K in an applied field of 6 T, which cannot be associated with the lower-temperature AFM order. We posit that the region of near-zero net magnetization at the CBST/FeTe interface represents the depletion of interstitial Fe through diffusion into the TI overlayer, as depicted in Fig. 4. This would also explain the unexpected magnetic orders in

BST and MBT overlayers observed *via* LE- μ SR. In fact, properly describing the F_M energy-dependence in BST/FeTe requires a 5 nm thick region of magnetic order at the BST surface (see Fig. 4 and the Supplemental Material [33]). We speculate that Fe from the FeTe diffuses across the interface towards the BST surface, where it forms magnetically ordered clusters.

This work raises questions regarding the underlying mechanism stabilizing superconductivity in TI/FeTe bilayers. AFM order in MBT/FeTe is indistinguishable from that of nonsuperconducting Te/FeTe, with both being relatively uniform. On the other hand, BST/FeTe and CBST/FeTe exhibit substantial AFM suppression. Thus, AFM suppression may be an incidental outcome of changing Fe-stoichiometry, rather than the sole requirement for superconductivity. Further support can be found in a recent report of superconductivity in CrTe₂/FeTe bilayers, where a similar van der Waals gap is present without a topological surface state [42]. Regardless of the underlying origin, previous works have estimated thicknesses of 6 nm–11 nm [1,2] for the superconducting region. This thickness exceeds the maximum estimated width of any nonmagnetic regions in our CBST/FeTe and MBT/FeTe bilayers, such that spatial overlap between the emergent superconductivity and static magnetic order is exceedingly likely. Further, there is evidence of unusual interactions between the superconductivity and magnetism, including enhancement of the net FeTe magnetization below T_c and a combination of transitory F_M suppression with a peak in λ . This is consistent with previously reported transport measurements, which observe an upper critical field exceeding the Pauli limit, suggesting unconventional superconductivity, which is highly tolerant of magnetic fields [2]. Our study demonstrates that the TI/FeTe interface is an incredibly robust system for entwining topological electronic states, superconductivity, and

magnetism to satisfy the key requirements for chiral Majorana physics.

VI. EXPERIMENTAL METHODS

A. Sample synthesis

The magnetic CBST/FeTe and undoped BST/FeTe heterostructures used in this work were grown in two separate commercial MBE chambers (ScientaOmicron), each with a vacuum better than 2×10^{-8} Pa. The insulating 0.5 mm SrTiO₃ (100) substrates used for MBE growth are first soaked in hot deionized water (approximately 80 °C) for 1.5 h and then put in an approximately 4.5% HCl solution for 1 h. After that, we anneal these substrates at 974 °C for 3 h in a tube furnace with flowing oxygen. Through these treatments, the SrTiO₃ surface is passivated and reconstructed, making it suitable for MBE growth of CBST/FeTe and BST/FeTe heterostructures. Next, the heat-treated SrTiO₃ (100) substrates are loaded into the MBE chamber and outgassed at 600 °C for 1 h. High-purity Bi (99.9999%), Sb (99.9999%), Cr (99.999%), Fe (99.995%), and Te (99.9999%) are evaporated from Knudsen effusion cells. The growth temperatures are 340 °C for the FeTe and 210 °C for the CBST and BST layers. The growth rate is 0.4 unit cells per minute for the FeTe layer and 0.2 quintuple layers per minute for the CBST and BST layers. Both FeTe and CBST/BST layers are grown in a Te-rich environment to prevent Te deficiency in these heterostructure samples. The MBE growth is monitored using reflection high-energy electron diffraction (RHEED) patterns.

For MBT/FeTe samples, the growth temperatures are 340 °C for the FeTe and 270 °C for the MBT layers. The growth rate is 0.3 unit cells per minute for the FeTe layer and 0.2 septuple layers per minute for the MBT layer. Both are calibrated by measuring the heterostructure thickness using scanning transmission electron microscopy and atomic force microscopy measurements. The Mn effusion cell purity is (99.9998%). Representative structural characterization and microscopy images have been previously published in [2,5,13].

B. Neutron diffraction

Neutron diffraction measurements were performed using the time of flight Laue diffractometer WISH at the ISIS Neutron and Muon Source [43]. The samples were mounted on an aluminum strip and oriented such that the FeTe *b*-axis was vertical, giving access to the H0L scattering plane. The FeTe orientation was then chosen such that the {L0L} direction was in the optimal flux condition for the $(\frac{1}{2}, 0, \frac{1}{2})$ reflection. The measurements were performed at 1.5 K, 15 K, and 90 K with the help of an Oxford instrument ⁴He cryostat. The raw data were analyzed with the Mantid software [44].

C. Low-energy muon spin relaxation

The LE- μ SR measurements were performed using the Low-Energy Muon Facility (LEM) at the Swiss Muon Source, Paul Scherrer Institute PSI, Switzerland [45,46]. All experiments were performed using two identical 10×10 mm² samples of the target heterostructure, except for the undoped

BST system, in which case only a single piece was used and the incident beam area was reduced accordingly. Samples were mounted with silver paint on a nickel-coated aluminum plate to minimize background effects. The samples were mounted in a helium flow cryostat (CryoVac, Konti), which is capable of maintaining the sample stage temperature within ± 0.1 K of the target temperature. Fully polarized muons are accelerated to variable energies to implant them at different depths. Implantation profiles modeled using TRIM.SP are used to select energies probing different depths [45]. For our heterostructures, the LE- μ SR measurements utilize all or a subset of the following implantation energies: approximately 1.0 keV, 1.5 keV, 2.0 keV, 3.0 keV, 4.0 keV, and 5 keV. The mean implantation depths are presented for the various heterostructures in Figure S4 of the Supplemental Material [33]. The beam transport settings are set to 12.0 kV. Upon implantation into the sample, the muon polarization is oriented within the film plane, and transverse magnetic fields of 7.5 mT and 125 mT are applied perpendicular to the surface of the sample. Measurements are performed upon cooling with multiple energies at each temperature. Before subsequent cooling, the sample is heated to the maximum measurement temperature (at least 200 K) and the magnet is degaussed.

Muons decay to positrons preferentially along their polarization axis, and these positrons are detected using a set of four scintillation detectors. These detectors are positioned to the left, above, to the right, and below the beam axis. For each muon, the time between implantation and decay is recorded, with a time resolution of approximately 5 ns. The number of positrons detected at each position *i* is given by

$$N_i(t) = B_i + N_0 e^{-(t/\tau_\mu)} (1 + A_i(t)) \quad (1)$$

where $\tau_\mu = 2.2 \mu\text{s}$ is the muon lifetime, B_i is the background signal at each detector. In the chosen weak transverse magnetic field experimental setup used for these experiments, the asymmetry $A_i(t)$ is the oscillatory signal caused by the muon spin precession. The time spectra for each detector between 0.1 μs to 10 μs are simultaneously fitted using the musrfit software [47] to the following functional form

$$A_i(t) = A_0 e^{-\lambda t} \cos(B\gamma_\mu t + \phi_i) \quad (2)$$

where B is the average magnetic field experienced by the muon and λ is the depolarization rate, corresponding to the width of the field distribution or characterizing spin fluctuations. By subtracting the signal from reflected muons [48], the resulting asymmetry can be used to calculate the magnetically ordered volume fraction:

$$F_M(E, T) = 1 - \frac{A_0(E, T)}{A_0(E, \text{“high T”})} \quad (3)$$

We note here that there are some signs of a hysteretic temperature dependence in the LE- μ SR data. Specifically, while the vast majority of data were taken while cooling the samples, a few data points were taken during warming due to unavoidable time constraints in the allocated beamtime. This yielded the single largest outlier in the measurement, at 120 K on the BST (15 nm)/ FeTe (30 nm) sample. In this case, the

120 K F_M value does not lie between the 100 K and 200 K data, instead matching the 50 K measurement. We cannot at this time speculate regarding the origin of this hysteresis.

Some of these measurements were previously included in the Supplemental Material [33] sections of references [2] and [5]. Specifically, the μ SR measurements on MBT/FeTe are shown in Fig. 2, and on CBST/FeTe in Figs. 3(c) and 3(d).

D. Polarized neutron reflectometry

Polarized neutron reflectometry measurements were performed using the SHARAKU (BL17) [49] instrument with a horizontal scattering geometry installed at the Materials and Life Science Experimental Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC). A pulsed neutron beam was obtained by the spallation reaction at the mercury target, where 3 GeV energy proton beam with a repetition rate of 25 Hz was injected. The neutron beam has a white spectrum, which is analyzed by time-of-flight (TOF) from 0.010 s to 0.040 s, which means from wavelength $\lambda = 0.24$ nm to 0.88 nm. [50] The neutron spins were polarized parallel or antiparallel to the 1 T applied magnetic field, and the spin-dependent reflectivities were measured as a function of the momentum transfer (Q) along the film normal direction. The data was collected by a two-dimensional position-sensitive detector using a Multi-Wire Proportional Counter (MWPC). [51] The SHARAKU uses the event data recording method as a standard control system “IROHA” and data acquisition (DAQ) one [52], and the data conversion and polarization correction from event data to momentum transfer Q was done by software “Utsusemi” [53].

Preliminary PNR was performed at the ISIS facility (UK) on the POLREF beamline, and this data was used to inform future sample growth and design the final PNR experiments.

Data were fit using the Refl1D software package, using the DREAM Markov-Chain Monte Carlo algorithm, which is implemented in the BUMPS Python package for model optimization and uncertainty estimation.

E. Magnetic x-ray spectroscopy

Magnetic x-ray absorption spectroscopy was performed using the I06 beamline at the Diamond Light Source. Measurements were performed on a nonsuperconducting SrTiO₃/FeTe (28 nm) / Te (3 nm) sample in fluorescence yield. Samples were field-cooled to 200 K in a 6 T magnetic field applied along the film normal. The measurement geometry was normal incidence, *i.e.*, with the x-ray propagation vector along the film normal direction.

Preliminary x-ray measurements were performed at beamline 4.0.2 of the Advanced Light Source.

ACKNOWLEDGMENTS

The work done at Penn State is supported by the DOE grant (DE-SC0023113) and the Penn State MRSEC for Nanoscale Science (DMR-2011839). CZC acknowledges the support from the Gordon and Betty Moore Foundation’s EPiQS Initiative (Grant No. GBMF9063 to CZC).

The neutron experiments on beamline SHARAKU (BL17) at the Materials and Life Science Experimental Facility of the J-PARC were performed under proposal numbers 2023B0108. The authors acknowledge the Science and Technology Facility Council (STFC) for the provision of beam time on the WISH diffractometer at the ISIS facility (UK). The LE- μ SR experiments were performed at the Swiss Muon Source $S\mu S$, Paul Scherrer Institute, Villigen, Switzerland. The raw data of the neutron diffraction data can be found at [54]. Preliminary PNR was performed at the ISIS facility (UK) on the POLREF beamline, and this data was used to inform future sample growth and design the final PNR experiments. The raw data for that PNR work can be found at [55]. Research performed in part at the NIST Center for Nanoscale Science and Technology. The magnetic x-ray spectroscopy experiment was performed at beamline I06 of Diamond Light Source under Proposal No. MM42224. This research used resources of the Advanced Light Source, which is a DOE Office of Science User Facility under contract no. DE-AC02-05CH11231. Certain commercial equipment, instruments, software, or materials are identified in this paper in order to specify the experimental procedure adequately. Such identifications are not intended to imply recommendation or endorsement by NIST, nor are they intended to imply that the materials or equipment identified are necessarily the best available for the purpose. Unless otherwise noted, NIST work was funded solely by the United States Government. We thank Prof. T. Hesjedal and Dr. Nina-Juliane Steinke for assistance in comparing our LE- μ SR data with that of their CBST films.

DATA AVAILABILITY

Some of the data that support the findings of this article are openly available [54–56]. Other data supporting the conclusions of this work are not publicly available upon publication because it is not technically feasible, and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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