Topological Electronic Structure and Intrinsic Magnetization in MnBi$_4$Te$_7$: A Bi$_2$Te$_3$ Derivative with a Periodic Mn Sublattice

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Combinations of nontrivial band topology and long-range magnetic order hold promise for realizations of novel spintronic phenomena, such as the quantum anomalous Hall effect and the topological magnetoelectric effect. Following theoretical advances, material candidates are emerging. Yet, so far a compound that combines a band-inverted electronic structure with an intrinsic net magnetization remains unrealized. MnBi$_2$Te$_3$ has been established as the first antiferromagnetic topological insulator and constitutes the progenitor of a modular (Bi$_2$Te$_3$)$_n$(MnBi$_2$Te$_4$) series. Here, for $n = 1$, we confirm a nonstoichiometric composition proximate to MnBi$_4$Te$_7$. We establish an antiferromagnetic state below 13 K followed by a state with a net magnetization and ferromagnetic-like hysteresis below 5 K. Angle-resolved photoemission experiments and density-functional calculations reveal a topologically nontrivial surface state on the MnBi$_4$Te$_7$ (0001) surface, analogous to the nonmagnetic parent compound Bi$_2$Te$_3$. Our results establish MnBi$_4$Te$_7$ as the first band-inverted compound with intrinsic net magnetization providing a versatile platform for the realization of magnetic topological states of matter.

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I. INTRODUCTION

Soon after the discovery of topological insulators (TIs) a decade ago [1], the role of magnetism and its potential to modify the electronic topology emerged as a central issue in the field of topological materials. Magnetic degrees of freedom provide a powerful means of tuning the decisive characteristic of any topological system: its symmetry. By now it is recognized that the interplay between magnetic order and electronic topology offers a rich playground for the realization of exotic topological states of matter, such as the quantum anomalous Hall state [2,3], the axion insulator state [4–6], and magnetic Weyl and nodal-line semimetals [7–11], enabling in turn different routes to spintronic applications [12–14].

The nontrivial topology in paradigmatic TIs like Bi$_2$Te$_3$ is a result of band inversion driven by strong spin-orbit interaction [15,16]. Until recently, the interplay with magnetism in topological insulators has been mostly explored by extrinsic methods, such as doping a known TI with magnetic impurities [15] or interfacing it with ferromagnets [17]. Numerous derivatives of Bi$_2$Te$_3$ doped by transition metals have been explored over the last

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years [2,3,18], but the magnetically active atoms did not form a periodic crystal sublattice. Initiated by works on epitaxial MnBi2Se4 layers [19,20], the compound MnBi2Te4 has recently arisen as the first derivative of Bi2Te3 that hosts structurally and magnetically ordered Mn atoms on well-defined crystallographic sites [21–24]. The emergence of an antiferromagnetic TI state in MnBi2Te4 is now being broadly scrutinized by theoretical and experimental methods [6,23,25–30]. At the same time, MnBi2Te4 provides the first example of an intrinsic magnetic TI [23,28,30,31].

In this joint experimental and theoretical work we establish another ternary manganese-bismuth telluride, MnBi2Te3, i.e., the \((n = 1)\) member of a modular \((\text{Bi}_2\text{Te}_3)_n(\text{MnBi}_2\text{Te}_4)\) series, as the first instance of a compound that features both an inverted electronic band structure and an intrinsic net magnetization. Moreover, we observe several competing magnetic states in MnBi2Te3, which, in combination with the presence of topological surface states, could provide a versatile platform for tunability between different topological regimes.

II. CRYSTAL GROWTH AND STRUCTURE

Recent systematic, synthetic explorations of the Mn-Bi-Te system have revealed new compounds that are ordered \((\text{Bi}_2\text{Te}_3)_n(\text{MnBi}_2\text{Te}_4)\) \((n = 1, 2)\) modular stackings of quintuple \((\text{Bi}_2\text{Te}_3)\) and septuple \((\text{MnBi}_2\text{Te}_4)\) blocks [Fig. 1(a)] [30,31]. In this section, we report a new crystal-growth protocol for \(n = 1\), assisted by our preceding thermochemical studies [31], and the structure elucidation by single-crystal x-ray diffraction (SCXRD). In Ref. [31] we developed robust synthetic protocols for phase-pure powders of \(n = 1\) and \(n = 2\) members based on differential scanning calorimetry. For all members of the \((\text{Bi}_2\text{Te}_3)\) \((\text{MnBi}_2\text{Te}_4)\) \((n = 0, 1, 2)\) series we found an ubiquitous deviation from the idealized compositions [24,31]. Henceforward, the title compound is denoted as Mn147, keeping in mind its nonstoichiometry, and the MnBi2Te4 compound is denoted as Mn124.

As shown first in Ref. [31], Mn147 is thermodynamically stable in a high-temperature interval well above room temperature. Whereas Mn124 melts at 600/5 °C [24], the melting point of Mn147 is 590/5 °C [31] and, thus, offers a very narrow window above the crystallization point of \(\text{Bi}_2\text{Te}_3\) (586 °C), in which Mn147 can be grown from a melt. Crystal growth is thus very challenging, and our experiments show evidence that polycrystals grown outside the determined temperature window exhibit stacking variants of both Mn124 and Mn147. Mn147 is not thermodynamically stable at room temperature, but can be obtained as a metastable product by quenching from 585 °C [31]. Based on these findings, we have established an optimized crystal-growth technique for Mn147: millimeter-sized platelets [Fig. 1(b)] can be obtained by a stepwise slow cooling of a heterogeneous \(\text{MnTe}_3(\text{Bi}_2\text{Te}_3(\square))\) melt and long-term annealing at a precisely controlled temperature of 585 °C, followed by rapid water quenching (see the Appendix for further details). These high-quality crystals enable the following studies of the crystal structure, the magnetic order, the transport, and the surface electronic structure (Secs. III and IV). The compositions of all crystals used for physical property measurements are verified by energy-dispersive x-ray spectroscopy (EDX).

Our current structure elucidation by SCXRD confirms cationic nonstoichiometry in Mn147 in full accordance with our previous data on Mn147 powders [31]. The disorder manifests itself in \(\text{Mn}^{2+}/\text{Bi}^{3+}\) antisite defects and related cationic vacancies (\(\square\)) in the septuple \((\text{Mn}_{1-x}\square_{x/3}\text{Bi}_{2x/3}\text{Te}_4)\) blocks, predominantly in the \(1a\) site in the middle of the septuple block (Supplemental Material, Tables S1, S2, Note 1 [32]). Interestingly, the \((\text{Bi}_2\text{Te}_3)\) block appears unaffected by cationic intermixing (Table S3). These mixed occupancies of the cationic positions and Mn vacancies result in a nonstoichiometric composition \(\text{Mn}_{0.75(3)}\text{Bi}_{4.17(3)}\text{Te}_7\) as refined from a SCXRD experiment. This stoichiometry slightly deviates

**FIG. 1.** (a) Crystal structure of \(\text{Mn}_{1-x}\text{Bi}_{2x/3}\text{Te}_7\) (Ge\(\text{Bi}_2\text{Te}_7\) structure type) with alternating \((\text{Bi}_2\text{Te}_3)\) and \((\text{Mn}_{1-x}\square_{x/3}\text{Bi}_{2x/3}\text{Te}_4)\) blocks. Mn atoms are shown in green; Bi in blue; Te in orange. (b) As-grown crystals with the experimental composition (EDX) \(\text{Mn}_{0.8(1)}\text{Bi}_{4.3(1)}\text{Te}_7\).
from the one previously determined for polycrystalline powders, \( \text{Mn}_{0.85(3)}\text{Bi}_{4.10(2)}\text{Te}_7 \) [31]; thus, indicating that a homogeneity range \( 0.15 \leq x \leq 0.25 \) may exist for the \( \text{Mn}_{1-x}\square_x\text{Bi}_{4+2x/3}\text{Te}_7 \) phase.

The cationic disorder, however, does not alter the trigonal lattice symmetry of \( \text{Mn}_{147} \) (space group \( P\bar{3}m1 \); the \( \text{GeBi}_4\text{Te}_7 \) structure type [33]), neither inhibits long-range magnetic order. A similar intrinsic phenomenon has been reported for isostructural [33], and structurally [24] and compositionally related [34,35] compounds. In contrast to some of them, we find no indications of massive stacking faults in our crystals by x-ray or electron diffraction methods [31].

III. MAGNETIC PROPERTIES

In this section, we analyze the bulk and surface magnetic properties of \( \text{Mn}_{147} \) crystals, based on which we discuss the topological electronic properties in Sec. IV. Electrical resistivity \( (\rho_{xx}) \) measurements as a function of temperature \( (T) \) reveal a metallic behavior [see Fig. 2(a)]. Focusing on the most salient features of the data, a clear upturn anomaly is visible at 13 K, which is reminiscent of the typical signature of magnetic ordering in itinerant materials [36]. The upturn indicates enhanced fluctuations causing electron scattering, which is strongly reduced in the ordered phase, in which a steep decrease of \( \rho_{xx} \) occurs. Upon lowering the temperature, a jumplike drop at about 5 K reveals further reduction of scattering, possibly related to a rearrangement of the magnetic structure.

Indeed, measurements of the magnetization \( (M) \) in an external field \( (H) \) on a \( \text{Mn}_{0.82(7)}\text{Bi}_{4.2(1)}\text{Te}_{7.00(5)} \) crystal (as determined by EDX) as a function of temperature show an antiferromagnetic phase transition at \( T_N \approx 13 \) K [Fig. 2(b)]. For \( H \perp ab \) and small fields, such as \( H = 200 \) Oe, a ferromagnetic-like increase occurs upon further cooling. This is followed by a splitting of field-cooled (FC) and zero field-cooled (ZFC) curves at around 7 K, as well as a kink

![Figure 2](https://example.com/figure2.png)

**FIG. 2.** Magnetic and transport properties of Mn147. (a) In-plane electrical resistivity as a function of the temperature. (b),(c) Normalized magnetization as a function of temperature for fields applied both perpendicular and parallel to the \( ab \) directions. Open and filled symbols correspond to ZFC and FC protocols, respectively. (d),(e) Hall resistivity and magnetization as a function of the field applied perpendicular to the \( ab \) planes. (f) Magnetization as a function of the field applied parallel to the \( ab \) planes. The hysteretic behavior for temperatures \( T \leq 5 \) K indicates ferromagnetic (intraplane) interactions.
and a peak at about 5 K, respectively, the latter coinciding with the jump in the resistivity. Both features are rapidly suppressed by applying an external magnetic field.

In the magnetically ordered phase, interesting metamagnetic behavior occurs for $H \perp ab$ as is evidenced by the magnetization curves [Fig. 2(e)]. For example, at $T = 10$ K a spin-flop-like feature is observed, and for $T < 7$ K dominating hysteresis typical for ferromagnets is apparent. This complex magnetic phenomenology is also reproduced by the Hall resistivity $\rho_{xy}$ [Fig. 2(d)]. For $T < T_N$, the system exhibits an anomalous Hall effect tracking the observed metamagnetic behavior. For $T \leq 7$ K the data reveal an additional metamagnetic transition in the low-field region $\mu_0 H \leq 300$ Oe. This observation is confirmed by a close inspection of the magnetization data. At 2.5 K a large hysteresis associated with global ferromagnetism is present. Above $T_N$, the anomalous contribution disappears, and only a standard component persists with a negative sign consistent with an $n$-type conduction [inset in Fig. 2(d)].

The magnetic anisotropy of the compound is examined via additional measurements for $H \parallel ab$ [Figs. 2(c) and 2(f)]. The magnetic moment values in the ordered state are much lower in this case. At lower fields both an antiferromagnetic transition and a ZFC-FC splitting are observed, but the suppression of these features occurs at higher fields than
for \( H \perp ab \). For the \( H || ab \) direction just below \( T_N \) the magnetization increases almost linearly with the applied magnetic field as expected for an antiferromagnet. On top of that, a spin reorientation at lower temperatures is indicated by an increase of \( M/H \) below ca. 10 K and a ferromagnetic net magnetization is clearly present. Apparently, the field necessary to observe such a feature can be sample dependent, as is evident from a comparison of our results with recent reports [37–39]. This finding may be associated with slight differences in the Mn content due to Mn/Bi intermixing and Mn vacancies, which, however, have no influence on the lattice symmetry. Notably, all reports are in line regarding the behavior of magnetization as a function of the magnetic field in both directions. The results show that the system is yet not fully saturated at \( \mu_0 H = 5 \) T as indicated by a small slope at higher fields [inset in Fig. 2(e)]. High-field experiments are necessary to gain a better insight into the details of the magnetic phase diagram.

The Curie-Weiss fitting of the magnetization high-temperature data in both directions yields positive values of the Curie-Weiss temperature: \( \theta_{CW}^{ab} = 13.7(5) \) K and \( \theta_{CW}^c = 14.7(5) \) K, thus, confirming the predominantly ferromagnetic character of the largest (intraplane) exchange interaction (see Supplemental Material, Note 2 and Fig. S1 [32]). In addition, the estimated effective magnetic moments, given the homogeneity range (0.15 \( \leq x \leq 0.25 \)), fall into the ranges 5.2 \( \mu_B \leq \mu_{eff}^b \leq 5.6 \mu_B \) and 5.1 \( \mu_B \leq \mu_{eff}^c \leq 5.5 \mu_B \), where \( \mu_B \) is the Bohr magneton, which suggest the manganese(II) high-spin configuration \( S = \frac{5}{2} \).

The microscopic nature of the different magnetic states as a function of field and temperature is a matter of debate and requires further elucidation.

The observed high-frequency electron spin resonance (ESR) signal of Mn147 [Figs. 3(a)–3(c)] is almost isotropic above \( T \sim 30 \) K and follows a typical Mn(II)-ion paramagnetic resonance condition \( h\nu = g \mu_B \mu_0 H_0 |m_z^c - (m_z^b \pm 1)| \) with the g factor very close to 2. Here, \( h \) is the Planck constant, and \( m_z^c \) is the projection of the spin on the quantization (magnetic field) axis. Importantly, below \( T \sim 30 \) K an energy gap \( \Delta \) develops in the ESR response, and the resonance condition is modified to \( \nu = \Delta + g \mu_B \mu_0 H_0/h \). The measured linear dependence of \( \nu \) vs \( \mu_0 H_0 \) for \( \mu_0 H \parallel ab \) [Fig. 3(a)] is typical for the wave vector \( q = 0 \) spin wave excitation (ferromagnetic resonance—FMR) in an easy-axis-type ferromagnetically ordered material, where \( \Delta \) represents the magnetic anisotropy gap. Considering the smallness of \( \Delta \) as compared with the applied magnetic fields, such linearity is incompatible with the resonance response of an ordered collinear antiferromagnet in this field regime [40,41]. The opening of the excitation gap \( \Delta \) at \( T \leq 30 \) K and its gradual increase [Fig. 3(b)] shows evidence of significant ferromagnetic spin correlations on the time scale of ESR (10\(^{-11}\) s) unrelated to 3D antiferromagnetic ordering which sets in at \( T_N = 13 \) K. Therefore, given the pronounced low dimensionality of the system, it is likely that the Mn-containing blocks are inherently ferromagnetic and give rise to a typical FMR signal in strong fields. At the same time, the application of a magnetic field suppresses the expected much weaker interlayer antiferromagnetic coupling responsible for the 3D long-range order at \( T_N \) in zero and small fields, whereas a paramagnetic state with strong intraplane ferromagnetic correlations—denoted in the following as correlated paramagnet (CPM)—persists up to temperatures of the order 30 K.

To complement the magnetic characterization, we carried out x-ray magnetic circular (XMCD) and linear dichroism (XMLD) experiments at the Mn \( L_{2,3} \) absorption edge in total electron yield mode (TEY) with a typical probing depth of a few nm [42]. The XMCD data collected at \( T = 2 \) K provide evidence for a substantial remnant net magnetization of the Mn ions along the surface normal [Fig. 3(d)], in sharp contrast to our previous observations for Mn124 with antiferromagnetic order [24]. This confirms that the spontaneous ferromagnetic polarization observed in the bulk magnetization data below \( T \sim 5 \) K extends up to the surface layers. A sizable XMLD signal in grazing light incidence and its absence in normal incidence further confirm the remnant out-of-plane magnetization [Fig. 3(e)] in agreement with the ESR results. The XMLD signal gradually diminishes with increasing temperature, confirming its magnetic origin and indicating the transition into the paramagnetic regime, in line with our bulk magnetization results.

By density-functional calculations (DFT) we consider various possible magnetic structures for the ordered MnBi\(_4\)Te\(_7\) model (see Supplemental Material, Note 3 [32]). The Mn atoms are found in the high-spin Mn(II) configuration, in agreement with the high-temperature magnetization measurements and similar to Mn124 [24]. Our calculations show that the magnetic moments within the Mn layers prefer intraplane ferromagnetic order with an out-of-plane spin configuration. The first-neighbor coupling is estimated as \(-0.09 \) meV/\( \mu_B^2 \), which is very close to the value reported for Mn124 [23]. Furthermore, we find that antiferromagnetic ordering between the Mn layers (AFM1 state) results in a smaller total energy than the ferromagnetic ordering (FM state). The energy difference is, however, only about 0.5 meV/Mn atom, which is an order of magnitude smaller than in Mn124 [22]. Moreover, this value is very close to the magnetic anisotropy energy, which yields about 0.5 meV/Mn atom in favor of the easy-axis configuration. These estimates corroborate a scenario with competing magnetic states differing slightly in energy, and, hence, a more complex magnetic response shown by Mn147, as compared to Mn124.

**IV. ELECTRONIC STRUCTURE AND TOPOLOGICAL SURFACE STATE**

Having established the crystal structure and the magnetic properties of Mn147, we will now discuss its electronic...
structure based on DFT and angle-resolved photoemission (ARPES) experiments. The structural resemblance between Mn147 and Bi$_3$Te$_3$ opens the question, to what extent Mn147 inherits properties from the (Bi$_3$Te$_3$) building blocks. We begin our theoretical analysis of the topology of the electronic structure with an auxiliary calculation without spin polarization (see Supplemental Material, Note 4 [32]), which shows that, in the absence of magnetism, the system would be both a strong topological insulator and a topological crystalline insulator, just like Bi$_3$Te$_3$ [43].

The influence of magnetism on the topological properties is first examined for the band-inversion phenomena. Figures 4(a) and 4(b) show the band structure of Mn147 for the FM and AFM1 (layerwise AFM) order, respectively (see Fig. S3 [32]). The symbol size is proportional to the overlap between the corresponding Bloch states and the indicated orbitals. For reference, Fig. S3f shows the well-known case of Bi$_3$Te$_3$, whose nontrivial topology originates in the inversion between Bi $p_z$ and Te $p_z$ orbitals of opposite parity [15,16,32]. The same orbitals contribute to the band inversion in Mn147 with the difference that the inverted bands are spin-polarized in the presence of a ferromagnetic component. Namely, the occupied Bi states form two bands [B1 and B3 in Fig. 4(a)] of predominantly opposite spin.

Naturally, in the AFM1 configuration the spin polarization in each band is compensated, i.e., each band is spin degenerate. In this phase, the system realizes a $Z_2$ antiferromagnetic topological insulator (AFMTI), protected by a combination of time-reversal symmetry and translation along the $c$ axis. This case is analogous to the recently established AFMTI state in Mn124 [23]. The $Z_2$ topology is analyzed in Fig. 4(c) based on a Wannier charge center (WCC) in the $k_z = 0$ plane. An arbitrary horizontal line crosses the WCC an odd number of times, and thus the $k_z = 0$ plane behaves as a quantum spin Hall insulator [44]. This ensures the existence of gapless surface states on side surfaces parallel to the $c$ axis.

A natural question is to what extent the observed structure tendencies to Mn/Bi intermixing and to Mn vacancies affect the electronic structure. To address this point, we performed supercell calculations for the compositions Mn$_{0.75}$Bi$_{1.25}$Te$_7$ and Mn$_{0.50}$Bi$_{1.25}$Te$_7$ (lifting up the restriction of electron neutrality used in the SCXRD refinements). Our calculations show that the fundamental gap remains open, suggesting that, in the present amounts, Mn/Bi intermixing and Mn vacancies do not affect the nontrivial topology of the material, but rather only shift the chemical potential (see Supplemental Material, Note 7 [32]).

The existence of a topologically nontrivial surface state is confirmed by the calculated (0001) surface spectral density in Fig. 4(d) for a quintuple-layer (QL) termination. As expected for the AFM1 state with an intraplane ferromagnetic configuration the surface spectral density shows a gaplike feature at the $\Gamma$ point [23]. We find a similar situation in our surface calculations for the case of a septuple-layer (SL) termination (Fig. S8) and also for a FM magnetic configuration (Fig. S5).

To experimentally support the nontrivial topology of the electronic structure predicted by our calculations, we conducted ARPES measurements on the natural cleaving (0001) surface of Mn147 (Fig. 5). The overview band structures [Fig. 5(a), Supplemental Material Figs. S9d, S9e, Note 6 [32]] bear clear resemblance to previous ARPES experiments for the topological insulator Bi$_3$Te$_3$ [15,45]. Most importantly, we likewise find a state with a V-shaped dispersion in the bulk gap between the conduction and valence band states (BCB and BVB), near the Fermi level $E_F$ [Figs. 5(b) and 5(g)]. Systematic photon-energy-dependent measurements confirm the surface character of this state [Fig. 5(d)]. By comparison to our density-functional calculations in Fig. 4(d) we identify it as a topologically nontrivial surface state (TSS).

![Fig. 4. Band inversion phenomena in Mn147 (GGA + U + SOC). (a) Band structure in the ferromagnetic configuration. The symbol size in each $k$ point and band is proportional to the overlap between the corresponding Bloch state and the indicated orbitals. For reference, Fig. S3f shows the well-known case of Bi$_3$Te$_3$, whose nontrivial topology originates in the inversion between Bi $p_z$ and Te $p_z$ orbitals of opposite parity [15,16,32]. The same orbitals contribute to the band inversion in Mn147 with the difference that the inverted bands are spin-polarized in the presence of a ferromagnetic component. Namely, the occupied Bi states form two bands [B1 and B3 in Fig. 4(a)] of predominantly opposite spin.](image-url)
The observation of conduction band states below \( E_F \) is in line with our transport measurements, although the prominent feature BCB shows a markedly 2D character and possibly arises from band bending, as commonly found for \( \text{Bi}_2\text{Te}_3 \) \[46\] and \( \text{Bi}_2\text{Se}_3 \) \[47\]. At higher binding energies in the valence band, we observe additional surface states SS1 and SS2 \[Fig. S9(d), S9(e)\] that are similar to those previously detected for \( \text{Bi}_2\text{Te}_3 \) \[45\]. These states are highly surface localized well within a single \( \text{Bi}_2\text{Te}_3 \) QL \[45\], suggesting that our results in Figs. 5(a)–4(f) represent a surface terminated by a \( \text{Bi}_2\text{Te}_3 \) QL. This is supported by our calculations in Fig. S7a \[32\] for \( \text{Bi}_2\text{Te}_3 \)-terminated Mn147, where similar surface states are found. Measurements on a single (0001) surface also revealed areas with a different well-defined band structure \[Fig. 5(g), Fig. S10\], which we tentatively attribute to the second possible surface termination by a \( \text{MnBi}_2\text{Te}_4 \) SL. The reduced data quality for this SL termination may arise from the higher defect density in the SL than in the QL as evidenced by our x-ray diffraction results in Sec. II. Nevertheless, we observe qualitatively similar features in ARPES as for the QL termination. Both terminations accommodate a dispersionless feature at a binding energy near 3.8 eV, which can be attributed to the Mn 3d states \[Fig. 5(a); Supplemental Material Fig. S10\], as confirmed by resonant photoemission measurements at the Mn \( L \) edge (Fig. S11).

Unlike for \( \text{Bi}_2\text{Te}_3 \), our measurements for Mn147 in the AFM1 state suggest the presence of a finite separation between the TSS and the BVB. This gaplike feature shows a subtle photon-energy dependence arising mainly from changes in the spectral appearance of the BVB maximum, as exemplified by the three data sets in Figs. 5(b), 5(c), and 5(f). The latter consists of at least two different features within a narrow energy range that exhibit complex \( h\nu \)-dependent intensity variations and possibly arise from a coexistence of surface- and bulk-derived states. Measurements at \( T = 80 \) K \[Fig. 5(e)\] do not show strong changes in the spectra for \( h\nu = 52 \) eV. However, towards higher temperatures we observe an increased spectral-weight filling of the gaplike feature, suggesting its mitigation with increasing temperature (Fig. S12).

A comprehensive picture of the detailed spectral-weight behavior of the TSS near the Dirac point is yet to emerge. Gaplike features, even in the paramagnetic regime, are also found in different magnetically doped TIs \[48,49\], in Mn124 \[23,25\] and, very recently, also in Mn147 \[37,39,50,51\]. We expect our detailed discussion of the \( h\nu \) dependency to be a useful ingredient, which, e.g., indicates a gaplike feature considerably smaller than the one observed in \[37\]. Additionally, other reports have found a vanishing gap in Mn147 \[52–54\] and argued about a surface magnetic structure possibly different from that in

![FIG. 5. Electronic structure of the Mn147(0001) surface as measured by ARPES. (a) Overview data set of the valence band structure obtained at \( T = 8 \) K showing characteristic surface states SS1 and SS2 and a feature related to Mn 3d states (cf. Figs. S9–S12 \[32\]). (b),(c),(f) High-resolution data sets of the electronic structure near \( E_F \) obtained at different photon energies and a temperature of \( T = 8 \) K, showing a topological surface state (TSS) in the gap between conduction and valence-band derived states (BCB and BVB). (d) Photon-energy dependence of the ARPES intensity at \( E_F \) \((T = 8 \) K). (e) Same as in (f), but for \( T = 80 \) K. (g) Same as in (f), but for a septuple layer (SL) termination. All other data sets in Fig. 4 are assigned to a quintuple layer (QL) termination. The ARPES data sets in Fig. 5 are measured along the \( \Gamma-M \) high-symmetry direction.](image-url)
V. MAGNETIC TOPOLOGICAL PHASES

Motivated by the experimental observations of a TSS due to band inversion and of the competition between different magnetic phases, we outline, based on our calculations, a topological phase diagram as a function of temperature and magnetic field. Figure 6 sketches the phases theoretically explored in the ordered MnBi$_4$Te$_7$ model. Below the Neél temperature, our calculations predict that the antiferromagnetic phase (AFM1) hosts a $Z_2$ topological insulating phase protected by a combination of the time-reversal and translation symmetries along the polar axis. In the lowest-temperature regime the experiments reveal a phase with net magnetization at zero magnetic field [see Figs. 2(e) and 2(f)]. To mimic this regime we consider a collinear FM state which, interestingly, also features a nontrivial topology (see Supplemental Material, Note 5 [32]). Namely, this phase realizes a topological crystalline insulator (TCI) tunable by the magnetization orientation. Specifically, the crystal structure presents three mirror planes that contain the polar axis and are related by $2\pi/3$ rotations. When the magnetization points perpendicular to one of these planes, it preserves the corresponding reflection symmetry. The calculated mirror Chern number in such a magnetic configuration equals $-1$. For other magnetization orientations, as in particular out-of-plane orientations, the topological protection at the (0001) surface is lifted and the corresponding surface state is gapped (see Fig. S5 [32]). In addition, as shown in Fig. 4(a), doped samples can be of interest, since a ferromagnetic component splits the double degeneracy of the bulk bands and opens up the possibilities of Weyl physics. Indeed, Weyl nodes are revealed only 24 meV above the gap, very close to the bottom of the conduction bands [see the arrow in Fig. 4(a) and Fig. S6].

VI. CONCLUSION

We present a comprehensive study of structural, magnetic, and electronic properties of the Bi$_2$Te$_3$-derivative Mn$_{0.25(3)}$Bi$_{4.17(3)}$Te$_7$, i.e., the $(n = 1)$-member Mn147 of the modular (Bi$_2$Te$_3$)$_n$(MnBi$_2$Te$_4$) series. Our results indicate that Mn147 realizes an intrinsic magnetic topological insulator, similar to the recently established first antiferromagnetic topological insulator MnBi$_2$Te$_4$ for $n = 0$ [23]. Unlike for MnBi$_2$Te$_4$, Mn147 develops a strong out-of-plane ferromagnetic component at low temperatures. In this regime Mn147 realizes the first instance of a compound that features both an intrinsic net magnetization and a topologically nontrivial surface state originating from a band inversion. In the thin-film limit these properties could facilitate the realization of the quantum anomalous Hall effect in an intrinsic material, as recently reported for Mn124 where, however, it requires large external fields due to the robust antiferromagnetism [55,56]. Moreover, our calculations show how the complex magnetic phases of Mn147, that we observe experimentally, may facilitate the tunability between different topological regimes, including antiferromagnetic topological and topological crystalline insulator states.

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APPENDIX: METHODS

In this appendix we provide further information on the different methods used in this work.

1. Crystal growth

First indications for the existence of Mn$_{1-x}\frac{4}{x}$Bi$_{4+2x/3}$Te$_7$ (0.15 $\leq x \leq 0.25$) and its composition were obtained in our previously published differential scanning calorimetry experiments [24]. Attempts to synthesize a phase-pure powder of Mn$_{0.85}$Bi$_{4.10}$Te$_7$ following the synthetic route described in Ref. [24], namely by long-term annealing of a stoichiometric mixture of Bi$_2$Te$_3$ and $\alpha$-MnTe at subsolidus temperature, lead to considerable amounts (up to 15 wt %) of MnBi$_2$Te$_4$ admixtures. A phase-pure ingot of Mn$_{0.85}$Bi$_{4.10}$Te$_7$ was synthesized by annealing at 590°C for 3 days, subsequent slow cooling to 585°C and, finally, annealing for 1 day followed by rapid quenching in water. Worth noting is that powders with the idealized MnBi$_2$Te$_4$ composition that were prepared by this route contained impurities, suggesting that this composition lies outside the homogeneity range. High-quality single crystals of Mn$_{1-x}\frac{4}{x}$Bi$_{4+2x/3}$Te$_7$ were grown by slow cooling (-1 K/h) of a melt from 650°C down to 585°C (above the solidification point of Bi$_2$Te$_3$), followed by annealing for 10 days and rapid quenching. Platelet-like strongly intergrown crystals were mechanically extracted from the obtained ingots. Their compositions were controlled by EDX analysis.

2. X-ray diffraction and energy-dispersive x-ray spectroscopy

Single-crystal x-ray diffraction data were collected on a four-circle Kappa APEX II CCD diffractometer (Bruker) with a graphite(002) monochromator and a CCD detector at $T = 296(1)$ K. Mo-K$_\alpha$ radiation ($\lambda = 71.073$ pm) was used. A numerical absorption correction based on an optimized crystal description was applied [57], and the initial structure solution was performed in JANA2006 [58]. The structure was refined in SHELXL against $F^2$ [59]. For further details on the crystal structure investigations of Mn$_{0.75(3)}$Bi$_{4.17(3)}$Te$_7$, see Ref. [60].

Powder x-ray diffraction data were measured using an X-Pert Pro diffractometer (PAAnalytical) with Bragg-Brentano geometry or a Huber G670 diffractometer with an integrated imaging plate detector and read-out system. Both machines operate with a curved Ge(111) monochromator and Cu-K$_\alpha$ radiation ($\lambda = 1.54.06$ pm). Variable divergence slits were used on the X-Pert Pro equipment to keep the illuminated sample area constant. The graphics of the structures were developed with the DIAMOND software [61].

Energy dispersive x-ray spectra (EDX) were collected on a scanning electron microscope Hitachi SU8020 using an Oxford Silicon Drift X-MaxN detector at an acceleration voltage of 20 kV and 100 s accumulation time. The EDX analysis was performed using the $P/B$-ZAF standardless method (where $Z =$ atomic number correction factor, $A =$ absorption correction factor, $F =$ fluorescence factor, and $P/B =$ peak to background model). Experimentally determined compositions (EDX) fall into a range from Mn$_{0.7(1)}$Bi$_{4.4(1)}$Te$_7$ to Mn$_{0.8(1)}$Bi$_{4.3(1)}$Te$_7$.

3. Angle-resolved photoelectron spectroscopy

ARPES measurements on the (0001) surface of cleaved crystals in a temperature range range between 8 and 80 K were carried out at the high-resolution-branch of beamline i05 at the Diamond Light Source, U.K., using $p$-polarized photons with energies between $h\nu = 20$ and 90 eV and energy resolution <10 meV [Fig. 5 and S10]. The spot size of the photon beam was ca. 30 $\mu$m. Supplementary ARPES experiments were performed at the Microscopic and Electronic Structure Observatory (MAESTRO) at beamline 7 of the Advanced Light Source (ALS) (Fig. S9) and at the LE branch of APE beamline at the Elettra synchrotron (Fig. S11). All measurements were performed in ultrahigh vacuum of lower than 10$^{-10}$ mbar. Supplementary core-level photoemission data were acquired at the ASPHERE III end station at beamline P04 of PETRA III (DESY, Hamburg) (Fig. S11).

4. Electron spin resonance measurements

ESR experiments were performed on a single crystal with a homemade ESR setup in the microwave frequency range $\nu =$ 75–300 GHz, in the temperature range $T =$ 4–35 K, and in magnetic fields up to $\mu_0H_0 =$ 16 T.

5. X-ray magnetic circular and linear dichroism

XMCD and XMLD measurements on the (0001) surface of cleaved crystals were carried out in total electron yield (TEY) mode at the BOREAS beamline of the ALBA synchrotron [62].
6. Density-functional calculations

Fully relativistic density functional theory (DFT) calculations were performed using the Perdew-Burke-Ernzerhof implementation [63] of the generalized gradient approximation (GGA) and treating the spin-orbit coupling in the 4-spinor formalism, as implemented in FPLO-18 [64]. For results presented in the main text, the experimental crystal structure based on a fully ordered MnBi$_4$Te$_7$ was used in our calculations. Namely, the cationic intermixing and cation deficiency were neglected and the stoichiometric limit MnBi$_4$Te$_7$ was considered. Effects of these sorts of defects were studied by supercell calculations (see Supplemental Material, Note 7 [32]). For the ordered model calculations, a linear tetrahedron method with a mesh of $16 \times 16 \times 2$ subdivisions (or $16 \times 16 \times 1$ in the AFM1 state) in the full Brillouin zone was used. GGA + $U$ calculations were performed using the atomic limit implementation of the double-counting correction and fixing $J = 1$ eV. The value of $U$ affects the resulting bulk gap and determines at which energies the spectral weight associated with Mn $d$ states is placed. We find that the position of the Mn 3$d$ states measured with core-level spectroscopy (see Fig. S11 [32]) is best described by a moderate value of $U \sim 2$ eV (see Fig. S3c [32]). This value renders a bulk gap of $\sim 75$ meV. In MnBi$_4$Te$_7$, higher values of $U$ have been used aiming to reproduce the experimental estimation of the gap [23]. The difficulty in finding a single value of $U$ that correctly accounts for all experimental results suggests that a quantitative comparison in these materials may necessitate the usage of exchange and correlation functionals beyond GGA.

7. Magnetization and transport

The dc magnetization measurements were performed in a superconducting quantum interference device (SQUID) vibrating sample magnetometer (VSM) from Quantum Design for 1.8 K up to room temperature and in magnetic fields up to 7 T. Zero-field-cooled (ZFC) and field-cooled (FC) magnetization curves were recorded upon warming.

The transport properties were performed in the standard four-wire configuration using a homemade probe inserted in a He-bath cryostat by Oxford Instruments, endowed with a 15/17 T magnet.
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