

quantization with increasing temperature [18]. As a result, the QAHE is stable only below $T_{\text{QAH}} = 20$ mK.

The intrinsic magnetic topological insulators $(\text{MnBi}_2\text{Te}_4)(\text{Bi}_2\text{Te}_3)_n$ (MBT_n , $n = 0 - 4$), whose functional constituents are $(\text{MnBi}_2\text{Te}_4)$ septuple layers (SLs) with the central sheet of FM-ordered Mn atoms, separated by n Bi_2Te_3 QLs, offer several advantages. Whereas for QL termination, angle-resolved photoemission spectroscopy (ARPES) measurements on MBT_1 and MBT_2 yield similar results to $(\text{Bi,Sb})_2\text{Te}_3$, without a discernible DP, there is a DP within the bulk gap for the SL termination [19]. Also, the topmost sheet of the ferromagnetically arranged Mn moments should strongly couple with the topological surface states (TSS), albeit a thorough spectroscopic investigation of the surface magnetism is still pending. As a result, a much higher $T_{\text{QAH}} = 1.4$ K is achieved in MnBi_2Te_4 [20]. This is despite the fact that MnBi_2Te_4 is suboptimal due to its antiferromagnetic (AFM) order ($T_N = 24$ K) and a complex layer-number dependence of the quantization effects, with an odd number of SLs required to realize the QAHE [20].

Yet, the potential of the other MBT_n for a further substantial increase of T_{QAH} is strong: Increasing n weakens the interlayer AFM coupling so that FM properties gradually develop. Indeed, most studies report a complex metamagnetic behavior in MnBi_4Te_7 and $\text{MnBi}_6\text{Te}_{10}$ [21–27], but a clear FM state only for $n \geq 3$ [22, 23, 28]. Already metamagnetic MnBi_4Te_7 hosts the QAHE up to several degree kelvin in the bulk regime [29]. This experimental realization of a QAHE device out of a bulk crystal required technically challenging but feasible efforts to attain the charge neutrality condition [29]. Consequently, envisioning this clear, technical realization path once an appropriate bulk crystal exists, we focus here on the next obvious step, namely to strengthen the FM properties of MBT_n . Our synthesis endeavors culminate in the robust FM order in $\text{MnBi}_6\text{Te}_{10}$ (i.e. already for $n = 2$), an intrinsic magnetic TI [19] and QAHE candidate [30].

We confirm the FM state both in the bulk and on the surface of $\text{MnBi}_6\text{Te}_{10}$ crystals by using bulk-sensitive superconducting quantum interference device (SQUID) magnetometry and surface-sensitive x-ray magnetic circular dichroism (XMCD). The clear FM characteristics seemingly contradict the weak AFM coupling anticipated by our density functional theory (DFT) calculations for the atomically ordered compound. This disagreement is resolved by including the experimentally determined Mn substoichiometry and Mn/Bi site intermixing into account. Our calculations pinpoint that the magnetic coupling can be tuned towards ferromagnetism by appropriate intermixing already in MnBi_4Te_7 and even MnBi_2Te_4 . Considering the intermixing patterns in our $\text{MnBi}_6\text{Te}_{10}$ samples and those reported showing no ferromagnetism, we rationalize their differing magnetic behavior. Our results demonstrate that carefully engineered intermixing can accomplish a robust FM order and, there-

fore, is the key towards enhanced QAHE properties in the MBT_n family of intrinsic magnetic topological insulators.

II. RESULTS

A. Crystal growth and structure refinement

$\text{MnBi}_6\text{Te}_{10}$ crystals were grown by slow crystallization from a melt (see Sec. IV). Besides $\text{MnBi}_6\text{Te}_{10}$, the obtained ingot contained admixtures of Bi_2Te_3 and MnTe_2 (see Fig. S1 in the Supporting Information). Observing side phases fully agrees with our earlier studies of $\text{MnBi}_6\text{Te}_{10}$ melting and decomposition by differential scanning calorimetry [31]. Their occurrence can be related to crystal growth being a competitive process between $\text{MnBi}_6\text{Te}_{10}$, $\text{MnBi}_8\text{Te}_{13}$ and Bi_2Te_3 , all having nearly the same crystallization temperatures.

A series of EDX (energy-dispersive x-ray spectroscopy) point measurements on individual crystals extracted from the ingot demonstrated a compositional range between Mn: 5.0, Bi: 36.6, Te: 58.4 and Mn: 4.2, Bi: 37.1, Te: 58.7 (in at. %). Our samples were thus consistently more Mn-deficient than expected from the nominal chemical formula $\text{MnBi}_6\text{Te}_{10}$ of the atomically ordered material (Mn: 5.9, Bi: 35.3; Te: 58.8). Again, this echoes our earlier published single-crystal structure refinement of $\text{Mn}_{0.73(4)}\text{Bi}_{6.18(2)}\text{Te}_{10}$ by x-ray diffraction [31], where we systematically showed that Mn-substoichiometry is determined by the Mn/Bi intermixing. Both features are also present in $\text{Mn}_{0.85}\text{Bi}_{2.10}\text{Te}_4$ [32] and $\text{Mn}_{0.75}\text{Bi}_{4.17}\text{Te}_7$ [26]. To facilitate perception, we denote our samples as $\text{MnBi}_6\text{Te}_{10}$ in the following text, keeping in mind that they are in fact substoichiometric.

The present study was performed on four individual Mn-deficient $\text{MnBi}_6\text{Te}_{10}$ crystals (denoted as Sample #1 – #4 henceforward; for their chemical compositions (EDX) see Fig. S2). Powder x-ray diffraction (PXRD) measurements, which required grinding the crystals to a homogeneous powder, were conducted after all other measurements had been finalized, in order to elucidate the underlying intermixing phenomenon. We confirmed that all four samples exhibit the crystal lattice of $\text{MnBi}_6\text{Te}_{10}$ with a sequence of one SL and two QLs (Fig. 1a) plus notable cation antisite disorder. $\text{MnBi}_6\text{Te}_{10}$ constituted the main phase as per Rietveld method and we established a firm link between the Mn content as found by EDX and the underlying crystal lattice of $\text{MnBi}_6\text{Te}_{10}$ in our samples.

This approach is exemplified on Sample #2 (see Fig. 1b and more procedural details in the Supporting Information sec. I). We confirmed that sample #2 was $\text{Mn}_{1-x}\text{Bi}_{6+x}\text{Te}_{10}$ ($x \approx 0.20 - 0.25$) which crystallized in the rhombohedral space group $R\bar{3}m$ (No. 166) with the unit cell lattice parameters $a = 4.36778(8)$ Å and $c = 101.8326(6)$ Å. To stabilize a further Rietveld refinement, the EDX compositions (e.g. $\text{Mn}_{0.76}\text{Bi}_{6.24}\text{Te}_{10}$