limited thermodynamic stability [31, 32] and, thus, offer very narrow growth temperature windows, their degrees of intermixing appear to be far less dramatic than in $MnSb_2Te_4$ and, therefore, more challenging to trace experimentally. Less substitutional disorder than in the MST_n (on average) may be a blessing when it comes to optimizing an MBT_n material's system for the QAHE device fabrication [29].

In summary, the prominent ferromagnetic characteristics of our sample, with a rather large T_c, and a substantial ordered, out-of-plane moment both in the bulk and at the surface, categorizes $MnBi_6Te_{10}$ as a particularly interesting candidate for the realization of a high-temperature QAH material [20, 25, 29, 37]. Moreover, a monolayer of ferromagnetic $MnBi_6Te_{10}$ appears as a perspective candidate for magnetic extension [30, 76] and proximity setups, since an FM $MnBi_6Te_{10}$ slab was predicted to exhibit QAHE [77].

IV. METHODS

A. Crystal growth and characterization

Pre-synthesized, phase-pure MnTe and Bi_2Te_3 powders were mixed in a ratio 0.85 : 2 at. %, pelletized and placed in an evacuated quartz tube. This was inserted at $T = 923\,\mathrm{K}$ into a preheated two-zone tube furnace with temperature control via external thermocouples (Reetz GmbH). The ampule was subsequently cooled down to 858 K at a rate of 1 K/hour, tempered for 14 days and then quenched in water. Platelet-like $MnBi_6Te_{10}$ crystals (lateral size up to 1 mm) were mechanically separated from the obtained ingot.

Powder x-ray diffraction data were collected on an X'Pert Pro diffractometer (PANalytical) with Bragg-Brentano geometry (featuring variable divergence slits) operating with a curved Ge(111) monochromator and Cu-Kα_1 radiation ($\lambda = 154.056\,\mathrm{pm}$). The phase composition of the polycrystalline ingot and individual crystals was estimated by Le Bail or Rietveld methods in JANA2006 [78]. The preferred orientation of the crystallites was described by March-Dollase corrections, the roughness for the Bragg–Brentano geometry was accounted for by the Suorti method.

Scanning electron microscopy (SEM) was performed using a SU8020 (Hitachi) equipped with a X-MaxN (Oxford) Silicon Drift Detector (SDD) at $U_a = 2-5\,\mathrm{kV}$. The composition of selected single crystals was determined by semi-quantitative energy dispersive x-ray analysis at $20\,\mathrm{kV}$ acceleration voltage.

B. Bulk magnetometry measurements

Field and temperature dependent magnetization studies were performed using a Quantum Design superconducting quantum interference device (SQUID) magnetometer equipped with a vibrating sample magnetometer (VSM) option (MPMS3). Our magnetization data on samples $\#1 - \#4$ are normalized to the real compositions determined via EDX. To obtain the absolute magnetization M per Mn atom, a precise knowledge of the sample mass is important. Samples #1 and #4 have an approximately 10 times smaller mass than sample #2, increasing the error of M. Nevertheless, the data for all four samples agree well with each other (Fig. S4). Furthermore, we here refer to the projection of the total magnetic moment onto the z-axis ($\mathbf{H}||\mathbf{z}$), which is the quantity obtained from the SQUID magnetometry measurements, as $m_{\mathrm{SQ}}^{\mathrm{tot}}$.

A setup made of two half-cylindrical quartz rods fixed with a small quantity of GE varnish to the main quartz VSM sample holder was designed to ensure an alignment of the crystals such that the external magnetic field was applied perpendicular to the crystal surface. Note that this setup, however, results in a rather temperature-independent (at not too low temperature) but non-negligible background contribution to the magnetic susceptibility, hindering a reliable extraction of the Curie-Weiss constant θ_{CW} and the temperature independent susceptibility χ_0 for our low-mass samples $MnBi_6Te_{10}$.

C. Bulk DFT (GGA+U) calculations

Fully relativistic DFT calculations based on the Generalized Gradient Approximation + U were performed with the parametrization of Perdew, Burke, and Ernzerf [38], using the full localized limit for the double-counting correction with $U = U_{dd}$ (the latter as obtained in Sec. II E) and $J = \left(F_{dd}^{(2)} + F_{dd}^{(4)}\right)/14$, with $F_{dd}^{(2)}$ and $F_{dd}^{(4)}$ the Slater integrals for the initial states presented in Table I. The spin–orbit coupling was included in the four-component formalism as implemented in FPLO. The total energy difference between the FM and A-type AFM configurations was computed using for Brillouin zone integrations a linear tetrahedron method. For $MnBi_6Te_{10}$, we use a mesh of the Brillouin zone having $14 \times 14 \times 14$ subdivisions. The magnetic anisotropy energy was calculated in the AFM state based on a mesh having $10 \times 10 \times 10$ subdivisions. The Mn $3d$ occupancy and the spin projection presented in the main text correspond to the gross projections. For the calculations of $MnBi_4Te_7$ based on a $2 \times 1 \times 2$ supercell, we use a mesh with $6 \times 12 \times 2$ subdivisions.

D. XAS and XMCD measurements

The XAS and XMCD measurements were performed using the high-field cryomagnet end station HECTOR of the BOREAS beamline at the ALBA synchrotron radiation facility [79] and at the high-field diffractometer at the UE46 PGM-1 beamline, BESSY II [80]. The single crystals were glued with conducting silver epoxy onto Cu