Magnetic topological insulator in MnBi$_6$Te$_{10}$ with zero-field ferromagnetic state

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Magnetic topological insulator with nontrivial topological electronic structure and broken time-reveal symmetry exhibits various exotic topological quantum phenomena, like quantum anomalous Hall effect (QAHE) and axion insulator state. The realization of such exotic phenomena at high temperature is one of central topics in this area. Here, combined with experimental measurements and theoretical calculations, we reveal that MnBi$_6$Te$_{10}$ is a strong TI with Dirac surface states at paramagnetic state and it evolves into an antiferromagnetic TI below 10.8 K. More importantly, the ferromagnetic axion insulator state emerges once spins polarized at field as low as 0.1 T, accompanied with saturated anomalous Hall resistivity up to 10 K. At 2 K, such state is preserved even external field down to zero. Furthermore, our calculations indicate that the few-layer ferromagnetic MnBi$_6$Te$_{10}$ have a non-zero Chern number. These outstanding features suggest MnBi$_6$Te$_{10}$ is a promising system to realize axion electrodynamics effects and high-temperature QAHE.

Topological insulators (TIs) have induced tremendous interests not only for demonstrating a novel classification approach of matters based on the topological electronic structures but also because of manifesting many of exotic phenomena protected by time-reversal symmetry, such as gapless helical states with Dirac-like dispersion on the surface or edge, and quantum spin Hall effect in two-dimensional (2D) TIs [1][2]. When introducing magnetism into TIs, the time-reversal-symmetry breaking brings even richer novel quantum effects [3][4]. A paradigm is the quantum anomalous Hall effect (QAHE), an integer QHE at zero external magnetic field, appearing in 2D Chern insulator with a non-zero Chern number. After theoretically predicted in honeycomb lattice with staggered magnetic fluxes [5], the experimental realization of QAHE has been sought for years, until it was observed in ferromagnetic (FM) TIs induced by magnetic dopants [6][7][8]. But the prefect QAHE in FM TI thin films can only be observed at temperature region (below 2 K) much lower than Curie temperature (a few tens of Kelvin) [6][7][8][9]. In addition, the harsh experimental conditions like precisely controlled composition and homogenized extrinsic magnetic dopants also make the exploration of related emergent phenomena and potential technological applications become unattainable.

Intrinsic magnetic TIs with periodic magnetic lattice avoid the inhomogeneity of extrinsic magnetic dopants and provide a possible route to realize high-temperature
QAHE without stringent requirements of material fabrication techniques. MnBi$_2$Te$_4$ is the first practical example. At zero field, MnBi$_2$Te$_4$ is an antiferromagnetic (AFM) TI [10][11][12][13][14][15][16][20], hosting possible AFM axion insulator state which is a promising platform for axion electrodynamics in condensed matter [2][16][17][18][19]. Importantly, down to the 2D limit, the high-field quantized Hall effect probably rooting in the topologically protected chiral edge states of Chern insulator and the transition from axion to Chern insulator has been predicted and demonstrated in thin MnBi$_2$Te$_4$ flakes [20][22][23][24]. However, the high field is required to fully polarize the spin orientation in few-layer MnBi$_2$Te$_4$ in order to realize the quantization of Hall conductance. Hence, to obtain magnetic TIs with (nearly) zero-field FM state is highly desirable.

Structurally, MnBi$_2$Te$_4$ is the first member ($m = 1, n = 0$) of the ternary homologous series of (MnBi$_2$Te$_4$)$_m$(Bi$_2$Te$_3$)$_n$ with stacking $m$ [Te-Bi-Te-Mn-Te-Bi-Te] septuple layers ([MnBi$_2$Te$_4$] SLs) and $n$ [Te-Bi-Te-Bi-Te] quintuple layers ([Bi$_2$Te$_3$] QLs) alternately along the $c$ axis [25][26]. These SLs or QLs are coupled through weak van der Waals force. Intuitively, inserting the QLs between the SLs should weaken the interlayer AFM interaction, leading to lower spin-flip-transition field $H_{sf}$. This has been confirmed in recently discovered MnBi$_4$Te$_7$ ($m = 1, n = 1$) with AFM TI ground state and significantly low $H_{sf}$ (~0.22 T) [25][26][27]. But bulk MnBi$_4$Te$_7$ still exhibits the absence of zero-field FM state at high temperature and the quick decrease of anomalous Hall conductance above 2 K [25][26]. In this work, we demonstrate that MnBi$_6$Te$_{10}$, the third member of the homologous series of (MnBi$_2$Te$_4$)$_m$(Bi$_2$Te$_3$)$_n$ ($m = 1$ and $n = 2$) (Fig. 1a), is a strong TI with Dirac surface states in band gap at paramagnetic state and becomes an AFM TI at low temperature. More importantly, the FM axion insulator state appears at field as low as 0.1 T. Such FM state with saturated anomalous Hall resistivity persists at zero field and 2 K, and still observable even up to 10 K at 0.1 T. Thus, it is significantly superior than all of other known (MnBi$_2$Te$_4$)$_m$(Bi$_2$Te$_3$)$_n$ materials, in favor of the realization of (nearly) zero-field QAHE in 2D limit.

All of peaks in XRD pattern (Fig. 1b) can be well indexed by the (00l) reflections of MnBi$_6$Te$_{10}$ [28], indicating that the surface of crystal is parallel to the $ab$ plane, consistent with the morphology of crystal (inset of Fig. 1b). The refined $c$-axial lattice parameter is about 102.19(2) Å. It is close to the reported value (~
increases \( T \), i.e., entering spin curve AFM configuration (intralayer FM and interlayer AFM interactions) in values of \( \mu = \frac{5}{2} \), MnBi and \( ( \) temperature modified high suggests the MnBi along the \( c \) axis. As shown in Fig. 1f, \( ( \) \( ( \) respectively, reflecting isotropic magnetic interaction above \( T_N \). Both positive values of \( \theta \) strongly suggest the in-plane FM interaction and therefore the A-type AFM configuration (intralayer FM and interlayer AFM interactions) in MnBi\(_6\)Te\(_{10}\).

As shown in Fig. 1f, at 2 K, the linear shape of initial magnetizations \( M(\mu_0H) \) curve below 0.12 T for \( H//c \) is consistent with the AFM ground state of MnBi\(_6\)Te\(_{10}\). A spin-flip transition occurs with hysteresis at around 0.13 T and quickly saturates at 0.2 T, i.e., entering a polarized FM state. The \( H_{sf} \) is much lower than MnBi\(_2\)Te\(_4\) (3.5 T - 8 T) \([12][13]\) but comparable to MnBi\(_4\)Te\(_7\) (0.15 T - 0.22 T) \([25][26]\), while the saturation moment 3.2 \( \mu_B/\text{Mn} \) is very close to MnBi\(_4\)Te\(_7\) (3.1 \( \mu_B/\text{Mn} \) \([25][26]\) and slightly smaller than MnBi\(_2\)Te\(_4\) (3.7 \( \mu_B/\text{Mn} \) \([12][13]\). For \( H//ab \), the \( M(\mu_0H) \) curve increases linearly with field and saturates when \( \mu_0H > 1.5 \) T without significant

101.985(8) \( \text{Å} \) \([28]\) and much larger than those in MnBi\(_2\)Te\(_4\) (\(~ 40.888(2) - 40.932(2) \text{Å}\) \([12][13][29]\) and MnBi\(_4\)Te\(_7\) (\(~ 23.815(3) \text{Å}\) \([28]\). The measured actual atomic ratio of Mn : Bi : Te is 0.73(7) : 6.70(2) : 10.00 when setting the content of Te as 10. Deviating from the stoichiometric ratio implies the possible existence of Mn-Bi antisite defects in this material, similar to MnBi\(_2\)Te\(_4\)\([12]\).

Temperature dependence of the magnetic susceptibility \( \chi(T) \) at \( \mu_0H = 5 \) mT for \( H//c \) shows a distinct cusp at around 11 K, in contrast to the saturating behavior of \( \chi(T) \) for \( H//ab \). As observed in MnBi\(_2\)Te\(_4\) \([12][13]\) and MnBi\(_4\)Te\(_7\) \([25][26]\), it suggests the occurrence of an antiferromagnetic (AFM) order at \( T_N = 10.8 \) K with spin direction along the \( c \) axis. The lower \( T_N \) in MnBi\(_6\)Te\(_{10}\) than MnBi\(_2\)Te\(_4\) (~ 25 K) \([12][13]\) and MnBi\(_4\)Te\(_7\) (~ 12 K) \([25][26]\) could be ascribed to the weaker AFM interaction along the \( c \) direction. At high field (\( \mu_0H = 1 \) T), the suppression of AFM transition and the increase of \( \chi(T) \) when lowering temperature for both field directions (Fig. 1c and d) suggests the emergence of field-induced FM state. For both field directions, the high-temperature \( \chi(T) \) curves (20 K - 300 K) at 1 T can be fitted very well using the modified Curie-Weiss law:

\[
\chi(T) = \chi_0 + \frac{C}{T - \theta}
\]

where \( \chi_0 \) is a temperature-independent term, \( C \) is the Curie constant and \( \theta \) is the Weiss temperature (Fig. 1e). The obtained effective moment \( \mu_{\text{eff}} \) is 5.30(6) and 5.33(6) \( \mu_B/\text{Mn} \) for \( H//c \) and \( H//ab \), respectively, close to those in MnBi\(_2\)Te\(_4\) (~ 5.3 - 5.9 \( \mu_B/\text{Mn} \) \([12][29]\) and MnBi\(_4\)Te\(_7\) (~ 5.3 \( \mu_B/\text{Mn} \) \([25][26]\). It confirms the high spin state of Mn\(^{2+}\) (spin-only \( S = \frac{5}{2} \), \( \mu_{\text{eff}} = 5.92 \mu_B \)) in MnBi\(_6\)Te\(_{10}\). The fitted \( \theta \) is 12.3(4) and 11.4(3) K for \( H//c \) and \( H//ab \), respectively, reflecting isotropic magnetic interaction above \( T_N \). Both positive values of \( \theta \) strongly suggest the in-plane FM interaction and therefore the A-type AFM configuration (intralayer FM and interlayer AFM interactions) in MnBi\(_6\)Te\(_{10}\).
hysteresis. Above observations confirm that the magnetic easy axis is along the c axis, consistent with the \( M(T) \) results. The almost same saturation moments for two filed directions imply that the spin orientation can be polarized easily by an external field irrespective of the crystal orientations.

The zero-field in-plane longitudinal resistivity \( \rho_{xx}(T) \) exhibits a metallic behavior when \( T > 20 \text{ K} \) (Fig. 1g). With further cooling, \( \rho_{xx}(T) \) shows a cusp centered at 11.5 K (inset of Fig. 1g), possibly caused by the enhanced electron scattering from spin fluctuations. Then below 11.5 K, a sharp decrease of \( \rho_{xx}(T) \) reflects the weakened spin-disorder scattering due to an onset of long-range AFM magnetic order. With increasing c-axial fields, the cusp is suppressed and becomes a smooth drop at 1 T (Supplementary Fig. 1). All of these are consistent with the A-type AFM state at zero field and polarized FM state at high field as shown in Fig. 1f.

Low-field magnetotransport properties further demonstrate that bulk MnBi\(_6\)Te\(_{10}\) crystals can exhibit anomalous Hall effect. As illustrated in Fig. 2a, for the \( M(\mu_0 H) \) loop at \( T = 2 \text{ K} \), the polarized FM state can be persevered at zero field and decreases at \( |\mu_0 H| = 0.01 \text{ T} \). This is remarkably different from MnBi\(_2\)Te\(_4\) in which the FM state can only appears when \( \mu_0 H > 3.5 \text{ T} \) [10][13], and also superior to MnBi\(_4\)Te\(_7\) where the FM state becomes to be weakened either at finite positive field [25] or when the field is just crossing zero [26]. There are two plateaus in the \( M(\mu_0 H) \) curve near \( \pm 0.1 \text{ T} \), suggesting MnBi\(_6\)Te\(_{10}\) enters the AFM state between the FM states. The non-zero moments of plateaus imply that there may be some residual FM layers in the AFM state.

The Hall resistivity \( \rho_{xy}(\mu_0 H) \) exhibits perfectly linear behavior with negative slope at high-field region (0.3 T - 3 T) in the whole measuring temperature range (2 K - 300K) (inset of Fig. 2b and Supplementary Fig. 2). It clear indicates that a single \( n \)-type carrier is dominant in MnBi\(_6\)Te\(_{10}\), in agreement with the ARPES results. The normal Hall coefficient \( R_0 \) is evaluated by linear fit of high-field \( \rho_{xy}(\mu_0 H) \) (0.3 T - 3 T) (Supplementary Fig. 2) and the apparent carrier density \( n_a \) is deduced using the single carrier model \( n_a = 1/e|e|\rho_{xy}R_0 \) (Fig. 2g). It is in the range of \( 4\times10^{20} - 6\times10^{20} \text{ cm}^{-3} \), close to those in MnBi\(_2\)Te\(_4\) and MnBi\(_4\)Te\(_7\) [25][26][29]. The anomalous Hall resistivity \( \rho_{xy}^A(\mu_0 H) \) can be obtained after subtracting the normal Hall resistivity \( \rho_{xy}^N(\mu_0 H) \) (= \( R_0\mu_0 H \)) from \( \rho_{xy}(\mu_0 H) \). As shown in Fig. 2b and a, the \( \rho_{xy}^A(\mu_0 H) \) and \( M(\mu_0 H) \) at 2 K
exhibits striking similarity, i.e., a significant hysteresis loop with a saturation behavior when \(\mu_0 H > 0.2\) T. Importantly, the saturated \(\rho_{xy}^A(\mu_0 H)\) still can be observed when the field approaches zero, thus, unlike MnBi\(_2\)Te\(_4\) [10][29], the zero-field AHE is truly realized in MnBi\(_6\)Te\(_{10}\). Moreover, when the field direction is just reversed, there are spike-like peak in \(\rho_{xy}^A(\mu_0 H)\) curve in contrast to the \(M(\mu_0 H)\) curve. It could be due to the real-space topological Hall effect (THE), originating from non-coplanar spin texture with non-zero scalar spin chirality when the spins flip from the polarized FM state to the AFM state [35]. Such THE has been observed in pyrochlore Nd\(_2\)Mo\(_2\)O\(_7\) and SrRuO\(_3\)/SrIrO\(_3\) multilayer films etc [33][34]. The \(\rho_{xx}(\mu_0 H)\) at 2 K also has a clear response to the evolution of magnetization (Fig. 2c) and the butterfly shape of \(\rho_{xy}(\mu_0 H)\) is well consistent with the hysteresis of \(M(\mu_0 H)\) loop (Fig. 2a). The \(\rho_{xx}(\mu_0 H)\) changes slowly with field in the AFM and FM states in contrast to the significant changes during the spin-flip process. Moreover, the kinks in the \(\rho_{xx}(\mu_0 H)\) curve at \(\sim 0.01\) T could reflect the influence of possible spin texture on longitudinal resistivity.

At higher temperatures, although the zero-field \(\rho_{xy}^A(\mu_0 H)\) decreases to nearly zero quickly (Fig. 2d), similar to \(M(\mu_0 H)\) curves (Supplementary Fig. 3), the saturated values of \(\rho_{xy}^A(\mu_0 H = 0.1\) T) are almost unchanged even \(T\) is close to \(T_N (= 10.8\) K) (Fig. 2d), distinctly different from MnBi\(_4\)Te\(_7\) where the saturated value of \(\rho_{xy}^A(\mu_0 H)\) diminishes rapidly when \(T > 5\) K [26]. When \(T > T_N\), the \(\rho_{xx}(\mu_0 H)\) decreases quickly and becomes almost zero at about 14 K (Fig. 2d and f). For \(\rho_{xx}(\mu_0 H)\), besides the sharp change at the spin-flip field while \(T \leq 10\) K, there is a plateau area between -0.03 T and +0.03 T, correspondingly the \(M(\mu_0 H)\) (Supplementary Fig. 3) and \(\rho_{xy}^A(\mu_0 H)\) become very small. Actually, at 2 K, such plateaus also appear but shift to the field ranges of \(\pm (0.03 - 0.13)\) T (Fig. 2c). Because the AFM state should appear in these field regions (Fig. 2a and Supplementary Fig. 3), it strongly suggests that MnBi\(_6\)Te\(_{10}\) could host AFM axion insulator state, as observed in 6 SLs of MnBi\(_2\)Te\(_4\) when field below 3 T [24]. On the other hand, in the polarized FM state, the decrease of \(\rho_{xx}(\mu_0 H)\) with increasing field, i.e., negative magnetoresistance (MR), can be partially ascribed to the suppression of spin-disorder scattering. The negative MR becomes positive one at higher fields or temperatures (Supplementary Fig. 2), possibly due to the dominance of normal positive orbital MR.

In order to illuminate the topological properties of MnBi\(_6\)Te\(_{10}\), first-principles calculations were carried out. At high temperature, the magnetic order is absent. Here,
an “open core” treatment of Mn 3d electrons has been used to treat them as core states. The corresponding band structure with SOC is presented in Fig. 3a, which shows there is a gap at $E_F$. Based on the Fu-Kane parity criterion, the computed $Z_2$ indices are 1(000), which indicate that it's a strong TI in the PM state at high temperatures. In contrast to the high-temperature PM state, the AFM state emerges at low temperature, and there are two Mn atoms with different spin orientation in an AFM unit cell. In the AFM configuration, the spatial inversion symmetry $P$ (with the origin located at a Mn atom) is preserved. Even though the TRS $\Theta$ is broken, the two kinds of Mn atoms can be related by a half translation of the lattice vector (i.e. $T_{1/2} = [\vec{a} + \vec{b} + \vec{c}] / 2$) in the $z$-direction, where $\vec{a}$, $\vec{b}$ and $\vec{c}$ are primitive lattice vectors, respectively. In other words, the anti-unitary symmetry $S = \Theta T_{1/2}$ is respected. In the band structure of the AFM state, all the bands are doubly-degenerate due to the presence of the combined anti-unitary symmetry with the condition $(PS)^2 = -1$ for every $k$-point. In the $k_z = 0$ plane, there is an AFM $Z_2$ invariant ($\nu_{AFM0}$) due to $S^2 = -1$. In the presence of inversion symmetry $P$, the AFM $Z_2$ can be further simplified as follows:

$$(-1)^{\nu_{AFM0}} \equiv \prod_{i=1}^{4} \prod_{n}^{n_{occ}/2} \xi_{2n}(K_i)$$

Explicitly, $K_1 = (0, 0, 0)$; $K_2 = (0.5, -0.5, 0)$; $K_3 = (0.5, 0, -0.5)$; $K_4 = (0, 0.5, -0.5)$, where $(u, v, w)$ are given in units of primitive reciprocal lattice vectors. $K_2$, $K_3$, $K_4$ are related by $C_{3z}$. The parity eigenvalues for occupied Kramers pairs of bands are given in Table I. The obtained $\nu_{AFM0} = 1$ suggests that it's an AFM TI with a clear bulk gap in Fig. 3b, resulting the gapless surface states on the side surfaces where the anti-unitary symmetry $S$ is preserved.

Most interestingly, the FM state can be stable even though the external magnetic field decreases to zero at the temperature $T = 2$ K. Thus, we calculated the band structure with the FM configuration, as presented in Fig. 3c. The gap is read to be 0.15 eV. In the FM state preserving inversion symmetry, the parity-based invariant is defined by [36][37]:

$$Z_4 = \sum_{\alpha=1}^{8} \sum_{n=1}^{n_{occ}} \frac{1 + \xi_n(\Lambda_{\alpha})}{2} \mod 4$$

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Based on the computed parity eigenvalues of occupied states at eight inversion-symmetry-invariant momenta (only four of them are distinct) in Table I, the obtained $Z_4 = 2$ indicates that the FM MnBi$_6$Te$_{10}$ is an axion insulator with $\theta = \pi$. Here, the coefficient $\theta$ is defined in the field theory description of the topological magnetoelectric (TME) effect $S_\theta = \frac{\theta e^2}{4\pi^2} \int dt d^3 x E \cdot B$ with $E$ and $B$ electromagnetic fields [4][17]. Moreover, we have performed the calculations for the FM slab structures of different layers. The results show that two, three and four trilayers composed of one [MnBi$_2$Te$_4$] SL and two [Bi$_2$Te$_3$] QLs are intrinsic Chern insulators. The electronic structure of the four-trilayer slab is presented in Fig. 3d, with an energy gap of 20 meV. Its Chern number is obtained to be -1 from the Wilsonloop calculations in the inset of Fig. 3d (See more results in Supplementary Fig. 4). As a result, the QAHE can be expected in the few-layer films of MnBi$_6$Te$_{10}$ with no external field.

To confirm the topological properties of MnBi$_6$Te$_{10}$, we have used angle-resolved photoemission spectroscopy (ARPES) to measure the electronic structures on the (001) surface. The ARPES results are summarized in Fig. 4. The data show that the MnBi$_6$Te$_{10}$ samples are electron doped, in agreement with the negative Hall coefficient. The conduction bands lie above -0.2 eV and the valence bands lie below -0.4 eV, forming a band gap of around 0.2 eV, which is consistent with the band calculation in Fig. 3. The most remarkable feature in the ARPES data is the existence of Dirac surface states within the band gap. The Dirac point is located at -0.29 eV in the middle of the band gap. Fig. 4a and b shows that the band dispersions of the Dirac surface states and bulk states do not change with photon energy. As shown in the band calculation in Fig. 3, the bulk states are quite two-dimensional due to weak interlayer coupling.

We also observed the Dirac surface states using laser ARPES with a photon energy of 7 eV, as shown in Fig. 4c-f. The band dispersions near the Dirac point can be determined in the laser ARPES data. As shown in Fig. 4e and 4f, the lower branch of the Dirac surface states has a “Λ” shape, while the upper branch has a shallow “W” shape, which is obviously different from the linear band crossing in MnBi$_2$Te$_4$ and MnBi$_4$Te$_7$ [14][29][30][31][32]. Furthermore, we observed an energy gap at the Dirac point in the PM phase of MnBi$_2$Te$_4$ and MnBi$_4$Te$_7$ [31], whereas the Dirac surface
The Dirac surface states of MnBi_{6}Te_{10} appear gapless. The observation of the Dirac surface states provides strong experimental evidence for the nontrivial topology of MnBi_{6}Te_{10}.

In summary, we have demonstrated that the A-type antiferromagnet MnBi_{6}Te_{10} is an AFM TI, which is converted into a FM axion insulator state under the field as low as 0.1 T, leading to an AHE without any external field at 2 K and constant anomalous Hall resistivity up to 10 K. These conditions are easily accessible in many experimental studies. MnBi_{6}Te_{10} provides a very promising platform to realize the QAHE as well as other exotic topological quantum effects at high temperature and (nearly) zero field.

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METHODS

Sample synthesis, structural and composition characterizations. Single crystals of \( \text{MnBi}_6\text{Te}_{10} \) were grown by the self-flux method. The high-purity Mn (piece), Bi (shot) and Te (shot) were put into corundum crucibles and sealed into quartz tubes with a ratio of Mn : Bi : Te = 1 : 11.7 : 18.55 (MnTe : Bi\(_2\)Te\(_3\) = 1 : 5.85). The tube was heated to 1223 K at a rate of 40 K/h and held there for 12 h, then cooled to close to 858 K at a rate of 10 K/h. The flux was removed by centrifugation, and shiny crystals with typical size about 1×1×0.1 mm\(^3\) can be obtained. The single crystal X-ray diffraction (XRD) pattern was performed using a Bruker D8 X-ray diffractometer with Cu \( K_\alpha \) radiation (\( \lambda = 0.15418 \) nm) at room temperature. The elemental analysis was performed using energy-dispersive X-ray (EDX) spectroscopy analysis in a FEI Nano 450 scanning electron microscope (SEM).

Transport and magnetization characterizations. Magnetization and electrical transport measurements were performed in Quantum Design PPMS-14T and MPMS3. Both longitudinal and Hall electrical resistivity were measured simultaneously in a standard five-probe configuration. In order to effectively get rid of the influence of voltage probe misalignment, we measured both resistivity at positive and negative fields. The final longitudinal and Hall resistivity were obtained by symmetrizing and antisymmetrizing raw data.

Angle-resolved photoemission spectroscopy. Synchrotron ARPES measurements on \( \text{MnBi}_6\text{Te}_{10} \) were performed at the “Bloch” beamline, MAXIV, Sweden, with a Scienta DA30 analyzer. High-resolution ARPES measurements on \( \text{MnBi}_6\text{Te}_{10} \) were performed using the 7-eV laser ARPES at the Institute of Solid Physics, University of Tokyo with a Scienta R4000 analyzer.

Theoretical calculations. The calculations were performed using the full-potential linearized-augmented plane-waves (FP-LAPW) method as implemented in the WIEN2K package [38]. The exchange-correlation in the parametrization of Perdew, Burke and Ernzerhof within GGA was applied in the calculations [39]. SOC was included as a second variational step self-consistently. The radii of the muffin-tin sphere \( R_{MT} \) were 2.5 bohrs for Mn, Bi and Te. The truncation of the modulus of the reciprocal lattice vector \( K_{\text{max}} \) was set to \( R_{MT}K_{\text{max}} = 7 \). The \( k \)-point sampling grid of the BZ was \( 12 \times 12 \times 12 \) for paramagnetic and ferromagnetic states, while that was \( 6 \times 6 \times 6 \).
for the calculations for the antiferromagnetic states. The experimental lattice parameters were employed in Ref. 28. The atomic position optimization was carried out until the maximal force on each ion becomes less than 0.01 eV Å\(^{-1}\). In order to simulate the high-temperature PM state, an “open core” treatment of 3\(d\) electrons was used in effect regarding these as core electrons [40]. In the FM (or AFM) calculations, the GGA + Hubbard-\(U\) (GGA + \(U\)) method was adopted, and the exchange parameter \(U = 4\) eV was applied to the Mn 3\(d\) states. vdW corrections were included by the DFT-D3 method in the calculations for the slab structures.

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Author contributions

H.-C.L. supervised the project. S.-Y.G. and H.L. performed ARPES measurements with the assistance of J.A., H.F., W.-H.F., P.Z., T.Q., H.D., T.K. and S.S.; S.-M.N., Y.-T.Q. and Z.-J.W. performed \textit{ab initio} calculations; S.-J.T., C.-S.G. and Y.F. synthesized the single crystals and carried out structural characterization, transport and magnetization measurements; H.-C.L., S.-Y.G. and T.Q. analysed the
experimental data and plotted the figures; S.-J.T., Z.-J.W., T.Q. and H.-C.L. wrote the manuscript based on discussion with all the authors.

**Competing interests**

The authors declare no competing financial interests.

**Data and materials availability**

All data needed to evaluate the conclusions in the paper are present in the paper and the supplementary materials.
Figure 1 | Crystal structure, magnetic and transport properties of MnBi$_6$Te$_{10}$. 

- **a.** Crystal structure of MnBi$_6$Te$_{10}$ with trigonal tetradymite-type structure (space group $R$-3m).
- **b.** XRD pattern of a MnBi$_6$Te$_{10}$ single crystal. Inset: photo of a typical MnBi$_6$Te$_{10}$ crystal on 1 mm-grid paper.
- **c** and **d.** Temperature dependence of magnetic susceptibility $\chi(T)$ at magnetic fields of 5 mT and 1 T along the $c$ axis and $ab$ plane, respectively.
- **e.** High-temperature $\chi(T)$ and inverted $1/(\chi(T) - \chi_0(T))$ curves with the Curie-Weiss fittings (red solid lines) for both field directions.
- **f.** Isothermal $M(H)$ curves for $H//c$ and $H//ab$ at $T = 2$ K.
- **g.** Temperature dependence of in-plane resistivity $\rho_{xx}(T)$ between 2 - 300 K at zero field. Inset: enlarged part of $\rho_{xx}(T)$ at low-temperature region.
Figure 2 | Magnetotransport properties of MnBi$_6$Te$_{10}$. a-c, Field dependence of isothermal magnetization $M(\mu_0H)$, anomalous Hall resistivity $\rho_{xy}^A(\mu_0H)$ and longitudinal resistivity $\rho_{xx}(\mu_0H)$ at $T = 2$ K and $\mu_0H$ up to $\pm 0.3$ T. d and e, $\rho_{xy}^A(\mu_0H)$ and $\rho_{xx}(\mu_0H)$ at various temperatures with $-0.3 \leq \mu_0H \leq 0.3$ T. f, Temperature dependence of $\rho_{xy}^A(\mu_0H = 0.1$ T). The blue dashed line denotes the $T_N$. g, Temperature dependence of calculated apparent carrier density $n_a$ using the formula $n_a = 1/|e|R_0$, where the ordinary Hall coefficient $R_0$ is obtained from the linear fit of high-field $\rho_{xy}(\mu_0H)$ curve ($\mu_0H = 0.3$ T - 3 T).
Figure 3 | Band structure and topological properties of MnBi$_6$Te$_{10}$ based on first-principles calculations. The band structures for different magnetic configurations along high symmetry lines. The high-symmetry $k$-points are labeled in the corresponding conventional Brillouin zones. Band structure a is for the PM state of MnBi$_6$Te$_{10}$ at high temperature. b and c are for the AFM and FM states, respectively. The electronic structure of the FM slab of four trilayers with an energy gap of 20 meV is shown in d, and the corresponding Wilson loop calculations show that it has a nontrivial Chern number ($C = -1$).
Figure 4 | Dirac SSs on the (001) surface of MnBi₆Te₁₀.  

- **a**, ARPES intensity maps along the cut through $\Gamma$ measured from $h\nu = 21$ eV to 25 eV at 47 K.  
- **b**, Intensity map of the ARPES data at $E_F$ along $\Gamma - \bar{K}$ measured in a range of $h\nu$ from 21 to 25 eV.  
- **c**, Three-dimensional intensity plot of the ARPES data around $\Gamma$ measured with the 7-eV laser at 40 K.  
- **d**, ARPES intensity maps along the cut through $\Gamma$ measured with the 7-eV laser at 40 K. The Dirac point at -0.292 eV.  
- **e**, EDCs around the Dirac point of the data in d. The red curve indicates the EDC at $\Gamma$.  
- **f**, Zoom-in ARPES intensity maps (top) and the second derivative with respect to energy (bottom) near the Dirac point in d.
| $K_1$    |   (0, 0, 0)   | (0.5, -0.5, 0) | (0.5, 0, -0.5) | (0, 0.5, -0.5) |
|----------|--------------|---------------|---------------|----------------|
| AFM      | 36, 29       | 35, 30        | 35, 30        | 35, 30         |
| $\Lambda_\alpha$ |   (0, 0, 0)  | (0.5, 0, 0)  | (0.5, 0.5, 0) | (0.5, 0.5, 0.5) |
| PM       | 16, 14       | 15, 15        | 15, 15        | 15, 15         |
| FM       | 37, 28       | 35, 30        | 35, 30        | 35, 30         |

**Table 1** | The number of occupied bands of odd and even parity at the corresponding TRIM points. $(u, v, w)$ are the coordinates of $k$-point with respect to the primitive reciprocal vectors. "$p$, $q$" indicate the numbers of (the numbers of pairs of) even-parity and odd-parity occupied bands for the FM state (for the PM and AFM states), respectively. The total numbers of occupied bands for the PM, FM and AFM states are 60, 75, and 130, respectively.
Supplementary Figures

**Supplementary Figure 1.** Temperature dependence of $\rho_{xx}(T)$ at $\mu_0H = 0, 0.1$ and $1$ T for $H//c$. 
Supplementary Figure 2. a and b, Field dependence of $\rho_{xy}(\mu_0 H)$ and magnetoresistance MR ($= [\rho_{xx}(\mu_0 H) - \rho_{xx}(0)]/\rho_{xx}(\mu_0 H)$) at various temperatures with field up to 3 T. c, Temperature dependence of fitted normal Hall coefficient $R_0$ obtained from the linear fit of high-field $\rho_{xy}(\mu_0 H)$ curve ($\mu_0 H = 0.3$ T - 3 T).
Supplementary Figure 3. Isothermal $M(\mu_0 H)$ loops at various temperatures for $H//c$. 
Supplementary Figure 4. The band structures of the ferromagnetic slab of one trilayer a, two trilayers b and three layers c. The corresponding Chern number is computed by the Wilson-loop method. The winding of the 1D Wilson-loop total phase indicates the nontrivial Chern number in the system.

We have performed the calculations for the slabs in the structures of different layers, where the basic unit is the trilayer of the Bi$_2$Te$_3$-MnBi$_2$Te$_4$-Bi$_2$Te$_3$ structure. The band structures of the FM slabs of different trilayers are given in the left panels of Fig. 4. Accordingly, the Wilson-loop calculations of the Chern numbers are presented in the right panels of Fig. 4. The results show that one trilayer is a trivial insulating phase, while the two trilayer and three trilayer slabs are both Chern insulators with the FM configuration, which suggest that the quantized QAH can be realized in the few-trilayer film.