Sample growth

The films investigated in this work were prepared by molecular beam epitaxy (MBE) using a Scienta Omicron Lab-10 chamber and standard effusion cells. The cells are charged with elemental Te and stoichiometric Bi$_2$Te$_3$ and MnTe. MnTe powder was prepared by annealing a stoichiometric mixture of the elements in a sealed quartz ampule at 993 K for 3 days. Prior to the heating, the elements were ball-milled and pressed into a pellet to enhance homogenization. Commercially purchased Mn chips (Aldrich, 99%) were reduced before the synthesis to get rid of oxide contamination. Phase-purity of the obtained MnTe was confirmed by powder X-ray diffraction (Philips X-PERT, Cu-Kα).

Freshly cleaved BaF$_2$(111) (Korth Kristalle GmbH) and epi-ready BaF$_2$(111) substrates (Crystal GmbH) were used to grow the Bi$_2$Te$_3$ epitaxial layers that hosts the single MnBi$_2$Te$_4$ layer. The freshly cleaved slices were loaded in the ultra-high vacuum system (UVH) and pre-heated to 400 °C for 1h, while the epi-ready substrates were pre-heated up to 650 °C for 3h. Due to the very small lattice mismatch (0.04 %), BaF$_2$(111) substrates provide an excellent platform to host Bi$_2$Te$_3$(0001) epitaxial films. Barium fluoride also presents a large band gap (8 eV) and is transparent from the near Ultra-Violet to the far Infra-Red, facilitating electrical transport investigations and optical measurements [1]. The Bi$_2$Te$_3$ epitaxial buffer was grown using a stoichiometric Bi$_2$Te$_3$ effusion cell and an extra Te cell, to compensate the loss of Te during the growth. The beam equivalent pressure (BEP) was measured before and after the growth by a retractable Bayard-Alpert ion gauge. By carefully choosing the growth parameters, high-quality Bi$_2$Te$_3$ epitaxial layers can be obtained [2].

The growth parameters directly impact the structural and electrical properties of the bismuth telluride epitaxial layers. At elevated substrate temperatures, Te$_{2+}$ vacancies are more likely to be formed during epitaxy, resulting in n-type doping [3]. Figure S1(a) shows a 3D representation measured by ARPES of the electronic bandstructure of a 6 QLs thick Bi$_2$Te$_3$ epitaxial film prepared with Φ = 0, i.e., no extra Te supply. A cut along high-symmetry directions Γ-M and Γ-K is shown in Figure S1(b,c), respectively. The M-shaped Bi$_2$Te$_3$ bulk valence band (BVB) can be clearly observed as well as the bulk conduction band (BCB), extending from the Fermi level down to binding energies of 60 meV. The occupied BCB indicates that the sample is n-type doped. The measured electronic band gap is around 170 meV. At the Fermi level the TSS present a hexagonal warping, characteristic of the compound Bi$_2$Te$_3$ [4]. Figure S1(d) presents ARPES spectra acquired near the Γ point of Bi$_2$Te$_3$ epitaxial layers prepared with Φ = 1 and substrate temperatures of: (i) $T_{SUB} = 245$ °C; (ii) $T_{SUB} = 255$ °C and (iii) $T_{SUB} = 265$ °C. Despite a clear decrease of the line-width of the measured electronic dispersion on raising the substrate temperature, indicating improved crystalline quality, all samples present the Fermi level located inside the bulk band gap. By carefully choosing the extra Te supply at the given growth conditions, the formation of tellurium vacancies can be suppressed, resulting in an intrinsically bulk insulating bismuth telluride epitaxial layer [2].

The bismuth telluride buffer layer was prepared with Φ = 1 films and with substrate temperature $T_{SUB}$ kept at 265 °C, in order to obtain high-quality layers (Figure S1(d)). A growth rate of 1.0 Å/min was used for the growth of a Bi$_2$Te$_3$ film, as depicted in
FIG. S1: (a) 3D-representation of the surface state bandstructure of a high quality Bi$_2$Te$_3$ film, grown without additional Te offer. (b,c) spectra along $\Gamma$-$M$ and $\Gamma$-$K$ high symmetry directions. The growth without additional Te-offer leads to a n-doping of the thin film populating the conduction band. (d) i-iii Growth series for rising substrate temperatures (245, 255, 265°C). The growth mechanism allows to reach a high sample quality, while keeping the sample in an insulating state.

Figure S2(a).

FIG. S2: (a) Growth of Bi$_2$Te$_3$ epitaxial layer using stoichiometric Bi$_2$Te$_3$ cell and an extra Te source, with $\Phi = 1$. (b) Deposition of one monolayer of MnTe. (c) Annealing at higher temperature under Te atmosphere. After the growth of the high-quality epitaxial Bi$_2$Te$_3$ film, one layer of MnTe was deposited, as depicted in Figure S2(b). The growth was performed inside the same chamber by using a stoichiometric MnTe effusion cell. A deposition rate of approximately 0.4 Å/min was used, which was calibrated using the growth rate of bulk MnBi$_2$Te$_4$ in the same setup [5]. After the deposition of the MnTe layer, a 10 min annealing at $T_{SUB} = 280$ °C and under Te atmosphere was performed, as illustrated in Figure S2(c). The Te atmosphere was kept while cooling down to $T_{SUB} = 265$ °C.

Figure S3 shows the reflection high energy electron diffraction (RHEED) images acquired during the growth along the [10$ar{1}$0] azimuth. All RHEED patterns were acquired with an energy of 15 keV. Figure S3(a) shows the streaky RHEED pattern obtained right after the growth of 7 QLs of Bi$_2$Te$_3$ on a freshly cleaved (111) BaF$_2$ substrate. After the growth of a high quality Bi$_2$Te$_3$ epitaxial film, a MnTe monolayer is deposited on the surface. The RHEED pattern measured after the deposition of MnTe, shown in Figure S3(b), also presents elongated streaks, evidencing a flat surface. However, the intensity of the rods and the transmitted spot is reduced due to increased scattering at the surface. After annealing the sample at $T_{SUB} = 280$ °C for 10 min under Te atmosphere, a higher intensity is observed indicating that a flat and smooth surface is recovered, as shown in Figure S3(c).

Figure S4 shows two low energy electron diffraction (LEED) images acquired from a single MnBi$_3$Te$_4$ SL at 58 and 63 eV, respectively. Bright spots with very low background intensity are observed, indicating a highly oriented surface. Pseudo-hexagonal patterns, characteristic for the three-fold symmetry of the (0001) surface, are visible in both images.
FIG. S3: Reflection High Energy Electron Diffraction (RHEED) images acquired from: (a) 7 QLs-thick Bi$_2$Te$_3$ epitaxial film. (b) After the deposition of one monolayer of MnTe on top. (c) After annealing at elevated temperatures, transforming the top-most Bi$_2$Te$_3$ QL into a MnBi$_2$Te$_4$ SL.

FIG. S4: Low Energy Electron Diffraction (LEED) images acquired after the growth of single MnBi$_2$Te$_4$ SL on top of 6 Bi$_2$Te$_3$ QLs.

In order to perform ex-situ measurements and avoid surface contamination and oxidation, the samples were covered by a protective capping layer. For surface sensitive measurements, in UHV conditions, the samples were covered by a 50-nm thick Te capping layer. The capping layer was mechanically removed prior to the surface measurements [6]. Especially for the case of single layer samples, it is important to mention that the mechanical removal of the protective capping layer is an easy and clean method to expose pristine surfaces, while annealing of the sample with the objective to thermally desorb the protective capping layers inside UHV chambers can result in the diffusion of adsorbed impurities, e.g. oxygen, resulting in the contamination of the sample surface [7]. For transport measurements, a 35-nm thick BaF$_2$ protective capping layer was deposited. As barium fluoride is highly insulating, the transport occurs only through the epitaxial layers. To penetrate through the capping layer and contact the topological insulator thin films while preventing its oxidation, a small circular trench was first etched in the film by focused ion beam (FIB). The resulting trench was then coated with conducting Pt deposited in-situ by FIB. Finally, contacts were painted with silver paint on the Pt rings for the transport experiments.

STEM measurement

Cross-sectional 50 nm-thick lamellas for scanning transmission electron microscopy (STEM) were prepared at the Wilhelm Conrad Roentgen Research Center for Complex Material System (RCCM) using a FEI Helios Nanolab Dual Beam equipment with Ga+ ion beam milling. STEM images were acquired using an uncorrected FEI Titan 80-300 equipment.

X-ray diffraction analysis

High resolution x-ray diffraction (XRD) and x-ray reflectivity (XRR) measurements were performed using a Bruker D8-Discover high resolution X-ray diffractometer equipped with a Göbel mirror and a Ge (220) monochromator. Cu K$_\alpha$ radiation ($\lambda$ = 1.5406 Å) was used for the measurements.

As depicted in Figure 1c in the main text, XRD measurements combined with a mathematical model were used as an integral characterization method for the structure of the sample. The measurements were conducted in a setup for thin film analysis where $\omega - 2\theta$-scans along the [0001] surface direction of the heterostructure were acquired. The full range scan of an heterostructure covered with Te capping layer is depicted in Figure S5a. The curve indicates a high-crystalline quality of the thin film, with clear interference fringes around the main diffraction peaks. The diffraction curve clearly shows the contributions of the Bi$_2$Te$_3$ epitaxial layer [8] as well as the peaks originating from the BaF$_2$ substrate and the Te capping layer. Especially for the Bi$_2$Te$_3$ (0006),(00015) and (00018) diffraction peaks, an asymmetry in the peak shape is identifiable. Figure S5b shows a complementary XRR analysis in grazing incidence. A modelling of the XRR-data was done using the software GenX [9] with a model consisting of a BaF$_2$ substrate, a Bi$_2$Te$_3$ buffer, 1 SL of MnBi$_2$Te$_4$ and a Te capping layer. STEM analysis on the same sample (see. Fig. 1(e), main text) has confirmed this structure and yielded a thickness of approximately 6 QL for the Bi$_2$Te$_3$ buffer layer. The XRR fit yields an average Bi$_2$Te$_3$ thickness of 64.7 Å, which is in excellent agreement with the direct
observations made by STEM.

FIG. S5: (a) Full range XRD scan of a SL MnBi₂Te₄ on 6 QLs of Bi₂Te₃ together with a simulation consisting of a SL terminated Bi₂Te₃ buffer with height variation (distribution shown in the inset). (b) X-ray reflectivity scan of the sample together with a simulation. The XRR simulation was conducted for a structure with a 64.7 Å Bi₂Te₃ buffer terminated by a single septuple layer of MnBi₂Te₄ and a 334 Å Te capping layer. (c) XRD simulations for various structures with a single SL termination. The surface modification can be seen as an asymmetry contribution in the Bi₂Te₃ (0006) diffraction peak. (d) XRD simulations of the (0006) diffraction peak for a two SLs termination. Compared to the single SL termination, the two SL-terminated rather manifests as an peak splitting with a flat plateau-like shape.

To gain a deeper understanding of the correlation between the shape of the diffraction peaks and the sample structure, a state-of-the-art XRD modelling [8, 10] was employed. Diffraction patterns were calculated for heterostructures with thickness of the Bi₂Te₃ epitaxial layer varying between 4 and 7 QLs and terminated in 0, 1 or 2 MnBi₂Te₄ SLs. Subsequently assuming a lateral variation in epilayer height, a linear combination of calculated curves was used to simulate the experimental measurement. As seen in Fig. S5a, a good agreement is reached assuming a distribution of 5, 6 and 7 Bi₂Te₃ QLs, each terminated with only one single MnBi₂Te₄ SL. Fig. S5c and S5d show the calculations around the (0006) Bi₂Te₃ diffraction peak for several thickness of the Bi₂Te₃ epitaxial film with a single (c) and a double MnBi₂Te₄ layer (d) termination. An increased thickness of the Bi₂Te₃ hosting film leads to a more narrow peak-shape, a closer spacing of the interference fringes and, due to the shrinking relative volume of the surface layer, also to a decay of the asymmetry contribution in both cases. While the fingerprint of a single MnBi₂Te₄ layer termination can typically be seen as a right-leaning of the peak, a double layer termination will lead to a more plateau-like shape or even a clear peak splitting. This XRD analysis confirms the extent of the local structure observed by STEM as covering most of the sample area.

Transport and Contact Preparation

Transport measurements were performed in a AMI cryostat equipped with a 13T magnet, using standard lock-in techniques. Contacts were applied on the thin-films by a focused ion beam. The AHE signal shown in Fig. ??(c) was obtained by removing a linear high-field slope (normal Hall effect) from the raw Hall effect (see SI, Fig. S6).

Epitaxial films with a thin (3u.c.) Bi₂Te₃ layer covered with one septuple layer of MnBi₂Te₄, as described in the main text, were investigated. The films were then capped with a ~ 35nm layer of BaF₂ to prevent the oxidation of the septuple layer. This layer however prevents direct electrical contact on the film with silver paint. In order to achieve electrical contact, we used a focused-ion-beam (FIB)-based method (see Fig.S6a). The films were first locally sputtered with a Ga-FIB to free access to the metallic layer. In a second step, the sputtered trenches were filled with platinum using the FIB in metal deposition mode. Since the contact between Pt and thin film would occur on the perimeter of the side of the trenches, a ring-shape was chosen for the trenches in order to maximise the contact perimeter. This technic allowed to achieve reliable ohmic contacts on films below 5 nm thickness without using lithography technics. The Pt ring contacts were
FIG. S6: Transport measurements and contacting of Bi$_2$Te$_3$/MnBi$_2$Te$_4$ heterostructure. (a), FIB-based contacting strategy. A hole is first cut into the capping layer using FIB. The hole is subsequently filled in-situ with FIB-deposited platinum. (b) Optical picture of the sample for transport presented in the main text. The Pt ring contacts are highlighted in the inset. The measurement geometry of current and Hall measurements is schematically presented. (c) Measured Hall effect, after subtraction of a small longitudinal component, corresponding to the data shown in Figure 2 of the main text. The curves are vertically shifted for clarity.

afterward contacted with silver paint. For the sample investigated in the main text, six contacts were taken in FIB with a pseudo Hall bar geometry to allow Hall measurements (see Fig.S6b). The anomalous Hall data shown in the main text Figure 2 are obtained by removing from the raw transverse resistance, first a field-symmetric component (corresponding to a residual longitudinal resistivity), and second a field-linear component (corresponding to the normal Hall effect). The transverse resistance after removal of a small longitudinal component is shown in Fig.S6c. From the high field slope $a$ of the Hall effect, one can extract a surface carrier density $n_{2D}$ or bulk $n_{3D}$ as $n_{2D} = 1 / (a e)$ and $n_{3D} = 1 / (a t e)$, where $t = 4.35$nm is the film thickness and $e$ is the electron charge. We obtain $n_{3D} \approx 3.10^{14}$cm$^{-2}$ and 3D equivalent $n_{3D} \approx 6.10^{20}$cm$^{-3}$, showing a strong doping of the thin film. This high bulk doping value further corroborates why no quantum anomalous Hall effect is observed in this sample, which is far away from undoped insulating behavior. From this carrier density a low mobility $\mu = 35$cm$^2$/V.s can be calculated. Contrary to the magnetic properties which can be unambiguously attributed to the MnBi$_2$Te$_4$ SL, so that the respective contributions of each layer and of the surface states can not be disentangled.

**Magnetic Anisotropy and Scaling**

The XAS and XMCD measurements were conducted at the HECTOR endstation of the BOREAS beamline (bl-29) at the ALBA Synchrotron in Barcelona - Spain [11]. The system yields a base-pressure < $1 \times 10^{-10}$ mbar and hosts a vector magnet reaching up to 6T. Measurements were conducted in total electron yield mode and normalized by a reference gold mesh in the beam path. To confirm the out of plane anisotropy of the MnBi$_2$Te$_4$ SL, we have conducted XMCD measurements in grazing incidence on the sample previously magnetized out of plane. Due to geometrical limitations typically present in XMCD measurements, the grazing incidence angle was chosen to be $70^\circ$ away from the surface normal. As seen in Fig. S7c), the XMCD signal is still finite, but represents only 26 % of the strength measured out of plane. Given the incidence angle of $70^\circ$, this is well in line with a pure out of plane magnetisation of the Mn sub layer. To characterize the in-plane magnetisation behaviour, we have further conducted a hysteresis scan on the Mn L$_3$ edge, by monitoring the asymmetry of the L$_3$ peak maximum normalized to the pre-edge region. Compared to the hysteresis acquired in normal incidence (Fig. 2b, main text), the strength of the asymmetry accounts for only 50 % of the normal incidence value and the hysteresis is overlayed by a strong paramagnetic slope. This again confirms the strong out of plane anisotropy of the MnBi$_2$Te$_4$ SL and renders it a well suited magnetic extension for breaking TRS at the surface of the Bi$_2$Te$_3$ 3D-TI. The discrepancies in coercive field resulting from the different probes as shown in Fig. 2b,c (main text) are known to occur for similar material systems, see e.g. differing results in [12, 13]. They can most probably be attributed to the fundamentally different nature of the probing process and the surrounding experimental conditions. In any case, the results found in transport experiments can be deemed as the ones relevant for a technological applicability.
FIG. S7: Anisotropy Analysis of the sample. (a) XAS Spectrum taken in gracing incidence (GI) geometry (60° off normal) exhibiting a similar lineshape than the normal incidence measurements [Fig. 2(a) main text]. (b) Definition of the experimental geometry. The dashed coordinate system corresponds to a sample in GI. (c) XMCD Spectrum in remanence and gracing incidence geometry. The strength of the GI XMCD signal corresponds to approx. 26% of the normal incidence XMCD. (d) GI Hysteresis loop showing a strong paramagnetic contribution of the in-plane magnetisation.

As a further characterisation of the magnetic behaviour, we use the XMCD temperature series as shown in Fig. 2d (main text) to determine the critical scaling behaviour of the remanent magnetisation. As XMCD poses a direct probe of the magnetic polarisation of a sample, the strength of the XMCD signal in remanent conditions is expected to scale according to a general law of

\[ A_{XMCMD}(T) = A_0 \left( 1 - \frac{T}{T_c} \right)^\beta = A_0 \tau^{\beta} \]  

with the Curie temperature \( T_c \), the saturation amplitude \( A_0 \) and the critical exponent \( \beta \), where the latter is a characteristic of the underlying magnetic universality class. In Fig. S8a we show the normalized remanent XMCD amplitude over the reduced temperature \( \tau = 1 - T/T_c \) for a set of Curie temperatures \( T_c \). We determine a critical temperature of 14.89 K from the decay of remanent magnetisation, confirmed by the double logarithmic representation deviating from a linear behaviour for values higher and lower than our result. Next we determine the critical coefficient \( \beta \) by a fit with the determined critical temperature, as shown in Fig. S8b to \( \beta = 0.484 \). The fitting was restricted to temperatures higher than 8 K, to minimize perturbations by a the potential temperature-uncertainty at the lowest temperatures. The sample temperature was thoroughly calibrated by the use of a temperature sensor at the sample position under the same external conditions.

As apparent from the linear representation shown in Fig. S8c, also showing the extrapolated critical behaviour to the lowest temperatures, the sample magnetisation is following the power law over the entire temperature range without systematic deviations again confirming a high sample quality and hinting towards negligible surface degradation during the measurements. The suitability of XMCD for the extraction of critical exponents due to the direct proportionality of the signal amplitude and the element specific magnetic moment, has already been worked out in previous works (see SI of [14]).

Systematics of the Photoemission Results

Photoemission and ARPES data was acquired at the 1\(^{3}\)-endstation of the Bessy II synchrotron in Berlin. The endstation yields a base temperature of approximately 1 K and a energy resolution of 1 meV for beamline and the Scienta-Omicron R8000 photoelectron analyzer, separately. A pristine surface was exposed by mechanically removing the Te capping layer [6], inside the measurement chamber at a base pressure < 5 \times 10^{-10} \text{mbar} prior to the measurements. All photoemission data presented in this work was extracted from detailed 2D Fermi surface maps to exclude artifacts stemming from thermal sample drift or similar effects.

Especially for the bulk-compound MnBi\(_2\)Te\(_4\) and its related derivatives, it is well known, that the surface states exhibit a quite peculiar photon energy dependence and can vanish completely, if the chosen photon energy is too high [15]. In this regard we have studied the \( h\nu \)-dependence of the spectral intensity within the accessible range at the 1\(^{3}\) end-station. The evolution seen
FIG. S8: (a) Normalized remanent XMCD amplitude over reduced temperature $\tau$ for a set of assumed critical temperatures $T_c$. In close vicinity to the Curie temperature of $T_c = 14.89\,\text{K}$ the curve adapts a linear behaviour in the double logarithmic representation. (b) Amplitude over the reduced temperature for the determined Curie temperature together with a critical exponent fit resulting in a critical exponent of $\beta = 0.484$. (c) Linear representation of the data shown in (b) over the sample temperature indicating a good fit over the entire temperature range. The region of extrapolation is indicated by the dashed line.

in Fig. S9a-h, very well traces a monotonous decay in intensity of the surface state compared the valence band spectrum. The surface state is best seen at $h\nu = 12\,\text{eV}$ and decays rapidly in intensity for higher photon energies. While in the range between 15 and 18 eV the V- and M-shaped valence band states are still well visible, at $h\nu = 22\,\text{eV}$ the spectral weight has nearly completely transferred to the bright feature located at $\approx 1.5\,\text{eV}$. For higher photon energies the intensity oscillates back and forth between the valence band states, but never again towards the surface state. Moreover, with rising photon energy no movement of the TSS in binding energy is recognisable, well in line with the 2D nature of the heterostructure. All spectra were acquired at the base temperature of the system (1.5 K).

While in bulk MnBi$_2$Te$_4$ the TSS has been found to disperse alongside a hole-like valence and an electron-like conduction band, the case for the heterostructure is significantly more clear [15, 16]. The TSS can here be identified as a single feature in the bulk bandgap connecting valence and conduction band.

FIG. S9: $h\nu$-series, showing a monotonous decay in the surface state intensity with increasing $h\nu$. Photon energies from a-h are 12, 13, 14, 15, 16, 17, 18, 22 eV.

Figure S10a shows ARPES results acquired on a heterostructure sample directly after the sample growth and without the use of any surface protection techniques. The spectra were acquired using monochromatized (He- and Xe-) plasma discharge sources
providing photon energies of 21.2 eV and 8.4 eV, respectively. The high agreement between the surface band structure depicted in Fig. S10d and the structure seen after Te decapping (Fig. 3 of the main text) confirms the suitability of the mechanical removal capping technique [6], even for surface modified samples.

The spectrum in panel S10a ($h\nu = 21.2$ eV) shows the TSS symmetrically dispersing inside of the bulk band gap, as well as the valence- and conduction-band states. The indirect bulk bandgap can be estimated to be $> 180$ meV from the photoemission data. The full 2-dimensional dispersion of the surface band structure is depicted in Fig. S10b as a set of stacked iso-energy surfaces. Both the conduction, as well as the valence band structure are star shaped and reflect the pseudo-hexagonal symmetry of the crystalline structure. Figure S10b shows DFT calculations adapted from a recent publication of a similar structure (a 3 QLs-thick Bi$_2$Te$_3$ buffer terminated by MnBi$_2$Te$_4$ SL’s on both surfaces) [17]. The calculations very well reflect the iso-energy surfaces measured on our heterostructure, especially also showing the very eccentric warping of the states.

As presented in Fig. S10d, using lower photon energies ($h\nu = 8.4$ eV) significantly changes the contrast between TSS and valence band structure allowing for a clear visualisation of the TSS, which is well in line with the photon energy series presented in Fig. S9. The TSS exhibits the same warped structure, as seen in Fig. S10a. Given the large spot size of the light source and the low photon energies of the emitted electrons, the spectra in S10d carry slight distortions of experimental origin.

FIG. S10: (a) In-situ photoemission spectrum of a heterostructure sample acquired with a He I-\(\alpha\) light source in the $\Gamma$M high symmetry direction. (b) Stacked iso-energy surfaces through the 2D Brillouin zone, showing the highly eccentric surface state dispersion. (c) DFT calculations of the Iso-energy contours as published in [17]. (d) In-situ photoemission spectra along the $\Gamma$M and $\Gamma$K high symmetry directions acquired with a Xe light source. The low photon energies strongly highlight the dispersion of the TSS.

To further disentangle the TSS especially from conduction band contributions, we have employed dichroism measurements facilitating circular and linear dichroism. Fig. S11a shows the sum of circular plus and minus polarisation, while the difference is depicted in S11b. The spectra were all extracted from k-space maps and corrected for sample drift before subtraction. As apparent from the difference spectrum depicted in S11b, the TSS shows a uniform dichroism with different sign the two branches, switching at the Dirac point. Compared to the case present e.g. in the related compound MnBi$_6$Te$_{10}$ [18], here the TSS is clearly separate from the conduction band spectrum and no additional bands dispersing in the bandgap are visible. The linear dichroism was investigated by anti-symmetrizing the data acquired in p-polarized geometry and confirms the observations made from the circular dichroism. The measurement geometry is shown in Fig. S11e, with the detector slit perpendicular to the plane of measurement, therefore imaging the circular dichorism in the plane of incidence.

**Photomission simulations**

While ARPES represents a very direct probe on changes in the electronic structure of our sample, especially for narrow features in k-space, disentangling these changes from the experimental conditions can be non-trivial at times. To enhance our understanding of the nature of the magnetic gap opening, we simulate our photoemission-spectra using model calculations based on our experimental data. From the k-resolved dispersion of the TSS as depicted in Fig. 3c (main text), we estimate a group velocity of $v_g = 5.110(56)$ m s$^{-1}$ for the TSS close to the Dirac point. Based on these experimental parameters, the Dirac cone can be
FIG. S11: (a) Photoemission spectrum at the $\Gamma$-point of the bandstructure. The spectrum shows the addition of $c^+$ and $c^-$ polarization. (b) Circular dichroism of the spectrum shown in (a), clearly highlighting the TSS with a sign change at the Dirac point. (c) Spectrum at the $\Gamma$-point, perpendicular to the analyzer slit direction, showing a clear linear dichroism. (d) Anti-symmetrized spectrum as shown in (c), highlighting the linear dichroism. (e) Experimental geometry at the 1-cubed end-station.

approximately written as

$$E_{\pm}(k) = E_D \pm \sqrt{\left(\hbar v_g k\right)^2 + \frac{E_g^2}{2}}$$

with $E_D$ as the Dirac point energy and $E_g$ the size of the magnetic gap [19]. Our photoemission intensity is subsequently simulated by calculating a spectral function for both upper and lower cone on an evenly spaced grid of $k$-points. The spectral function of our toy model can be written as

$$A_{k_{0,i}}(\omega, k) = A_i \frac{\Delta_{\epsilon}}{(\omega - \epsilon_{k_{0,i}})^2 + \Delta_{\epsilon}^2}$$

with $\Delta_{\epsilon}$ as the natural linewidth in energy, $i$ denoting the respective state and $A_i$ as an amplitude used to compensate for experimental cross-section effects. The spectral function as well as the linewidth parameters were adopted from a model applied to the closely related material system MnBi$_6$Te$_{10}$ (See Suppl. of [18]). To adapt the spectral function to the experimental conditions, the spectra were subsequently convolved with a 2-dimensional gaussian lineshape incorporating the effects of the finite experimental resolution. Two spectra reflecting the experimental conditions are depicted in Fig. S12a,b for a gap of $E_g = 35$ meV and 0 respectively.

Fig. S12c shows the EDC’s as also presented in Fig. 3a (main text) well below and above the critical temperature of the heterostructure. To further elucidate the effect of finite resolution Fig. S12d shows simulated EDC’s for different assumed $k$-resolutions. The simulations show an apparent increase in gap size, solely caused by resolution effects on the TSS dispersion, therefore making a careful treatment of the experimental data necessary. For our data we have observed a apparent gap size of $\sim 50$ meV which can be referred to a magnetic hybridization gap of $\sim 35$ meV, taking the experimental resolution into account. Overall we have achieved a good agreement between our toy-model simulation and the experimental EDC’s, showing that the main effect of a magnetic gap in the TSS is located in a small energy region around the Dirac point.

In Fig. S12e we show a simulation of the temperature dependent gap opening across $T_c$. Here, the experimental parameters were adapted to the data at highest and lowest temperature and all datasets were subsequently fitted with the gap-size, scaling and background as the only free parameters, keeping all other parameters fixed. The data is adapted with a slight asymmetry between upper and lower cone to account for the cross-section effects as apparent from the experimental data (also Fig. 3b, main text). The fitted simulations excellently reflect key features of the experimentally observed temperature dependence. For low temperatures a clear double peak feature is visible slowly merging to an asymmetric plateau-like shape when approaching $T_c$. With the closing of the gap at $T_c$ the spectral weight distribution reflects a single peak feature with a slight asymmetry of the peak-flanks due to the differing cross-section effects. Fig. S12f depicts the amplitude of the gap size (red balls) together with
amplitude of the remanent XMCD and the estimated critical power law as seen from Fig. S8, following a law of

$$E_g = \left(1 - \frac{T}{T_c}\right)^\beta$$

(4)

and a critical exponent of $\beta = 0.484$. All Data was normalized to converge to 1 at hypothetical zero temperature. Among all depicted curves a striking accordance can be found strongly hinting towards a direct proportionality of the Dirac point gap opening and the magnetism present at the sample surface.

FIG. S12: (a,b) Photoemission simulations using a TSS toy model adapted to the data as depicted in Fig. 3(b,c) main text.(c) Simulations (streaked) of the EDC close to the Dirac point of the band structure together with experimental results (dotted) as shown in Fig. 3(a), main text for temperatures below (blue) and above (red) $T_c$. The simulations show a good agreement between experimental EDC’S and simulations. (d) Simulations for a gapped and ungapped TSS, depicting the influence of a finite k-resolution to lead to an increase in the effective gap size. (e) Simulations adapted and fitted to the experimental Data for temperatures of 1.5, 5, 9, 12, 15, 17 and 19K. (f) Comparison of the relative gap-size, XMCD-amplitude and critical power law as determined in Fig. S8.

* Electronic address: celso.fornari@physik.uni-wuerzburg.de
† Electronic address: Hendrik.Bentmann@physik.uni-wuerzburg.de


