Massive Dirac Fermion at the Surface of the van der Waals Antiferromagnet MnBi$_2$Te$_4$

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The layered van der Waals compound MnBi$_2$Te$_4$ has been predicted to combine the band ordering of archetypical topological insulators like Bi$_2$Te$_3$ with the magnetism of Mn, making this material a viable candidate for the realization of various magnetic topological states. We have systematically investigated the surface electronic structure of MnBi$_2$Te$_4(0001)$ single crystals by use of angle-resolved photoelectron spectroscopy (ARPES) experiments. Combining photon-energy-dependent and spin-resolved measurements we establish the presence of a single massive Dirac fermion at the MnBi$_2$Te$_4(0001)$ surface, both, in the low-temperature antiferromagnetic phase and in the paramagnetic phase. Our experimental results establish the coexistence of a topological surface state and magnetic order in MnBi$_2$Te$_4$.

The hallmark of a topological insulator is a single spin-polarized Dirac cone at the surface which is protected by time-reversal-symmetry and originates from a band inversion in the bulk [1, 2]. Notably, breaking time-reversal symmetry by magnetic order does not necessarily destroy the non-trivial topology but instead may drive the system into another topological phase. One example is the quantum anomalous Hall (QAH) state that has been observed in magnetically doped topological insulators [11]. The QAH state, in turn, may form the basis for yet more exotic electronic states, such as axion insulators [4, 5] and chiral Majorana fermions [6]. Another example is the antiferromagnetic topological insulator state which is protected by a combination of time-reversal and lattice translational symmetries [7].

Magnetic order in a topological insulator has mainly been achieved by doping with 3d impurities [3, 9], which however inevitably gives rise to increased disorder. By contrast, the layered van der Waals material MnBi$_2$Te$_4$ [8, 10] has recently been proposed to realize an intrinsic magnetic topological insulator [11–14], i.e. a compound that features magnetic order and a topologically non-trivial bulk band structure at the same time. MnBi$_2$Te$_4$ is isostructural to the known topological insulators GeBi$_2$Te$_4$ [15, 17] and PbBi$_2$Te$_4$ [18], but the substitution of Ge or Pb by Mn introduces local magnetic moments.
which order antiferromagnetically below $\sim 24$ K [11,[19,[21]. The resulting interplay of magnetic order and topology in a single compound has been proposed as a promising platform for the realization of a variety of magnetic topological states [11,[14,[22].

In this work we present a systematic investigation of the surface electronic structure in MnBi$_2$Te$_4$(0001) single crystals employing angle-resolved photoelectron spectroscopy (ARPES), spin-resolved ARPES and resonant photoemission at the Mn $L$-edge. Our measurements reveal a surface state in the bulk band gap that forms a spin-polarized massive Dirac cone in agreement with recent theoretical predictions [11,[13]. The large mass-induced gap of $\sim 100$ meV is robust across the T$_N$ = 24 K [11,[19,[21]. Furthermore, they indicate an out-of-plane orientation of the magnetic moments with ferromagnetic intralayer coupling and antiferromagnetic interlayer coupling. This is in agreement with first-principles calculations [11,[13] and fully supported by our XMCD and XMLD measurements [see Fig. S1 in the supplemental material].

Figure 1(a) presents an ARPES data set for MnBi$_2$Te$_4$(0001) around the $\Gamma$-point, showing a massive Dirac state with an energy splitting of ca. 100 meV between the upper and the lower Dirac cone (UDC and LDC) [T = 80 K, $h\nu$ = 21.5 eV], (b) Crystal structure of MnBi$_2$Te$_4$ along one unit cell (black lines) according to Ref. [8]. The black arrow indicates the van der Waals-gap between two septuple layers as the natural cleavage plane. (c) X-ray photoemission spectrum showing core level lines of Mn, Bi and Te.

The ARPES measurements were performed at the MAESTRO [Fig. 1(a), 2(a)-(e), Fig. S2] and Merlin [Fig. 4(c)] endstations of the Advanced Light Source (USA) as well as at beamline I05 of the Diamond Light Source (UK) [Fig. 3(a)-(b)]. The energy resolution of the ARPES experiments was better than 20 meV. Spin-resolved ARPES measurements were performed at the APE beamline of the Elettra synchrotron by use of a Scienta DA30 hemispherical analyser combined with a spin polarimeter based on very low-energy electron diffraction (Sherman function $S = 0.3$). We acquired XMCD and XMLD data in total electron yield (TEY) mode at the BOREAS beamline of the ALBA Synchrotron (Spain). Resonant soft X-ray photoemission data and Mn 2$p$ core-level spectra were collected at the ASPHERE III endstation at beamline P04 of PETRA III (Germany) with an energy resolution of ca. 25 meV. Platelet-like MnBi$_2$Te$_4$ single crystals were obtained via an optimized crystal-growth procedure as described in Ref. [19]. MnBi$_2$Te$_4$ crystallizes in a trigonal lattice (the ordered GeAs$_2$Te$_4$ structure type) with septuple [Te-Bi-Te-Mn-Te-Bi-Te] layers stacked in the ABC fashion [8]. The septuple layers are separated by a van der Waals gap, as shown in Fig. 1(b).

Figure 1(a), including the size of the splitting of $\sim 100$ meV at $\Gamma$, is in good agreement with previous first-principles theory for antiferromagnetic MnBi$_2$Te$_4$(0001) [11,[13] and for a single, ferromagnetic MnBi$_2$Te$_4$ septuple layer on Bi$_2$Te$_3$(0001) [25]. These calculations predict the presence of a spin-polarized Dirac surface state with a gap in the magnetically ordered state.

A more detailed analysis of the electronic band structure is presented in Figs. 2(a)-(c). Besides the UDC, which shows a narrow line shape typical for surface states, we observe
more diffuse features close to the Fermi level which we attribute to the bottom of the bulk conduction band (BCB). One BCB feature is centered at the Γ-point in between the UDC and LDC directions which form a flower-like pattern in the Fermi surface and partly overlap with the UDC. This characteristic shape of the conduction band minimum poses an important constraint for theoretical approaches. We note that similar pockets at finite wave vectors along ΓM direction which form a flower-like pattern in the Fermi surface and partly overlap with the UDC. This characteristic shape of the conduction band minimum poses an important constraint for theoretical approaches. We note that similar pockets at finite wave vectors along ΓM direction which form a flower-like pattern in the Fermi surface and partly overlap with the UDC. This characteristic shape of the conduction band minimum poses an important constraint for theoretical approaches.

Collectively, these results provide strong experimental evidence for a spin-polarized massive Dirac state at the surface of MnBi2Te4(0001). To study the origin of the splitting of the Dirac cone, we compare ARPES data sets obtained below and above the antiferromagnetic ordering temperature $T_N = 24$ K.

A key property of the Dirac surface state in a topological insulator is a high degree of spin polarization. Our spin-resolved ARPES data in Fig. 2(g)–(h) indeed confirm a spin polarization of the UDC. As sketched in Fig. 2(f), the spin-polarized data were obtained at wave vectors along $k_x$ with a spin-quantization axis along $y$, i.e. in-plane and perpendicular to the probed wave vector along $k_z$. The reversal of the spin polarization of the UDC at $\pm k_x$ indicates a helical spin configuration. All data sets except for (d)-(e) were acquired with $h\nu = 21.5$ eV and at $T = 80$ K.

FIG. 2: (color online) (a) ARPES constant energy contours for MnBi2Te4(0001). At the Fermi level a hexagonal warping of the UDC and a flower-like pattern of the bulk conduction band (BCB) is observed. (b)-(c) ARPES data sets along the ΓM- and ΓK-directions. (d)-(e) Photon-energy-dependent data for the UDC and LDC ($T = 18$ K). (f) Schematic of the experimental geometry for the spin-resolved measurements. (g)-(h) Spin-resolved momentum- and energy distribution curves obtained for a spin-quantization axis along $y$, i.e. in-plane and perpendicular to the probed wave vector along $k_z$. The reversal of the spin polarization of the UDC at $\pm k_x$ indicates a helical spin configuration. All data sets except for (d)-(e) were acquired with $h\nu = 21.5$ eV and at $T = 80$ K.
in Fig. 3. Apparently, the dispersion shows no pronounced changes when passing from the antiferromagnetic into the paramagnetic state. The EDC taken at the \( \bar{\Gamma} \) point suggest a slight shift of both the UDC and LDC to higher binding energy for lower \( T \), but there is no significant change in the size of the splitting above \( T_N \).

The lack of a temperature dependence up to substantially above \( T_N \) indicates that the splitting of the Dirac cone does not originate from long-range magnetic order with out-of-plane magnetization, as was expected based on previous calculations for MnBi\(_2\)Te\(_4\) [12–13, 25, 28] and other materials [29, 32]. However, the paramagnetic state has not been considered in these works. Despite this discrepancy we note that the size of the measured splitting is of similar magnitude as those predicted theoretically for magnetically ordered MnBi\(_2\)Te\(_4\) [11–13]. This suggests that the coupling strength between the surface state and the magnetic moments of Mn is generally sufficient to induce a splitting of up to 100 meV as observed experimentally. We note that also in other systems a gap in the Dirac cone was observed in the paramagnetic regime, including Fe- and Mn-doped Bi\(_2\)Se\(_3\) [9, 33, 34]. Similar findings were also obtained for TlBi(Se\(_x\)Te\(_{1-x}\))\(_2\) [35].

Overall, our finding of a massive Dirac cone on the MnBi\(_2\)Te\(_4\)(0001) surface is in agreement with several first-principles theory studies, which unanimously predict that the bulk band structure of MnBi\(_2\)Te\(_4\) [11–13] and also MnBi\(_2\)Se\(_4\) [36, 37] is topologically non-trivial. A potential concern here is that an accurate description of the localized Mn 3\(d\) orbitals is not readily expected in calculations based on density functional theory (DFT) and could play an essential role [38].

To assess the character of the Mn 3\(d\) states we have carried out resonant photoemission experiments at the \( L_{2,3} \) absorption edge [Fig. 4]. The measured Mn density of states (DOS) shows a state at ca. 3.8 eV and another one at ca. 0.8 eV that is attributed to hybridization between Mn 3\(d\) and Te 5\(p\) orbitals near the top of the valence band, which is in rather good agreement with DFT [8]. This result suggests that these calculations capture the energetics of the Mn 3\(d\) states reasonably well and thus substantiates the topological classification.

The persistence of the gap above \( T_N \), however, calls for further theoretical investigation. In general, the protection of the Dirac point in a topological insulator is based on the absence of backscattering due to destructive interference between scattering channels related by time-reversal symmetry. A recent theory shows that, if this destructive interference is perturbed, strong impurity scattering may induce gap features at the Dirac point of similar size as observed here for MnBi\(_2\)Te\(_4\) [39]. Notably, single magnetic Mn impurities placed in the topological insulator Bi\(_2\)Te\(_3\) have been found to constitute strong scattering centers for the topological surface state [40].

FIG. 3: (color online) (a)-(b) Temperature-dependent ARPES data for MnBi\(_2\)Te\(_4\)(0001) below and above \( T_N \) measured with \( h\nu = 21.5 \) eV along \( \bar{\Gamma}\bar{M} \). (c) Energy distribution curves (EDC) at \( k_{||} = 0 \).

FIG. 4: (color online) (a) Valence band spectra for MnBi\(_2\)Te\(_4\)(0001) obtained by resonant excitation at photon energies \( h\nu \) near the Mn 2\(p\) → 3\(d\) absorption edge. The inset shows the Mn \( L_3 \) absorption edge with colored arrows indicating the corresponding excitation energies. (b) Difference of the spectra measured under on- (light blue) and off-resonant (black) conditions, showing the contribution of the Mn 3\(d\) states to the valence band. (c) Angle-resolved valence band spectrum measured with \( h\nu = 79 \) eV.
Therefore, based on our results we anticipate that the local magnetic moments in the Mn layers in MnBi$_2$Te$_4$ may allow for backscattering processes and thereby destroy the topological protection of the Dirac point. A rather local nature of the Mn states in MnBi$_2$Te$_4$ is supported by the measured 3$d$ DOS in Fig. [3] that resembles the one of single Mn impurities in Sb$_2$Te$_3$ [41].

In summary, our experiments unveil a topologically nontrivial surface electronic structure of the antiferromagnet MnBi$_2$Te$_4$(0001). In particular, MnBi$_2$Te$_4$ provides a first instance of a material that features a massive Dirac surface state and relies neither on doping nor alloying. Our results could provide new pathways to exploit the interplay of antiferromagnetism and topological edge states in spintronic device concepts based on the emerging material class of van der Waals magnets [42].

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