Large magnetic gap at the Dirac point in $\text{Bi}_2\text{Te}_3/$Mn$\text{Bi}_2\text{Te}_4$ heterostructures

Magnetically doped topological insulators enable the quantum anomalous Hall effect (QAHE), which provides quantized edge states for lossless charge-transport applications\textsuperscript{1–8}. The edge states are hosted by a magnetic energy gap at the Dirac point\textsuperscript{2}, but hitherto all attempts to observe this gap directly have been unsuccessful. Observing the gap is considered to be essential to overcoming the limitations of the QAHE, which so far occurs only at temperatures that are one to two orders of magnitude below the ferromagnetic Curie temperature, $T_c$ (ref. \textsuperscript{3}). Here we use low-temperature photoelectron spectroscopy to unambiguously reveal the magnetic gap of Mn-doped $\text{Bi}_2\text{Te}_3$, which displays ferromagnetic out-of-plane spin texture and opens up only below $T_c$. Surprisingly, our analysis reveals large gap sizes at 1 kelvin of up to 90 millielectronvolts, which is five times larger than theoretically predicted\textsuperscript{9}. Using multiscale analysis we show that this enhancement is due to a remarkable structure modification induced by Mn doping: instead of a disordered impurity system, a self-organized alternating sequence of Mn$\text{Bi}_2\text{Te}_4$ septuple and $\text{Bi}_2\text{Te}_3$ quintuple layers is formed. This enhances the wavefunction overlap and size of the magnetic gap\textsuperscript{10}. Mn-doped $\text{Bi}_2\text{Se}_3$ (ref. \textsuperscript{11}) and Mn-doped $\text{Sb}_2\text{Te}_3$ form similar heterostructures, but for $\text{Bi}_2\text{Se}_3$ only a nonmagnetic gap is formed and the magnetization is in the surface plane. This is explained by the smaller spin–orbit interaction by comparison with Mn-doped $\text{Bi}_2\text{Te}_3$. Our findings provide insights that will be crucial in pushing lossless transport in topological insulators towards room-temperature applications.

The QAHE was first demonstrated in chromium-doped tetradymite topological insulators\textsuperscript{2,3}. Subsequently, replacing chromium with vanadium was a successful strategy for achieving precise quantization with vanishing longitudinal resistance\textsuperscript{6,7}. The effect occurs because of a modification of the band inversion in the ferromagnetic state. Exchange splitting and spin–orbit coupling lead to a release of the inversion of one of the spin sub-bands\textsuperscript{2}. This should manifest itself as a magnetic gap that opens up at the Dirac point when the system is cooled below the Curie temperature. So far, however, direct observation of this gap has remained elusive and no clear correlation with ferromagnetism has been established.

Angle-resolved photoemission spectroscopy (ARPES) is the method of choice for the direct observation of the magnetic gap. Nevertheless, the situation has been confusing: large gaps of 0.05–0.2 eV were first reported for Mn-doped $\text{Bi}_2\text{Se}_3$ (refs. \textsuperscript{12,13}), but were later shown not to be of magnetic origin\textsuperscript{14}. Such gaps, however, did not appear when magnetic impurities were deposited on the surface of $\text{Bi}_2\text{Se}_3$ (refs. \textsuperscript{14–16}). At low temperatures, a mobility gap of 32 meV was inferred from scanning tunnelling Landau level spectroscopy of V-doped $\text{Sb}_2\text{Te}_3$ (ref. \textsuperscript{17}), but scanning tunnelling spectroscopy (STS) did not show a gap in this system\textsuperscript{16}. STS did reveal gaps of 20–100 meV in Cr-doped (Bi, Sb)$_2\text{Te}_3$ (ref. \textsuperscript{18}), but the temperature dependence was not investigated. In fact, a similar gap of around 75 meV was found for Cr-doped $\text{Bi}_2\text{Se}_3$ even at room temperature\textsuperscript{19}. This suggests a nonmagnetic origin for these effects, because the ferromagnetic $T_c$ is well below 50 K in all of these systems.

Interestingly, the configuration of the magnetic dopants is also contradictory. For isovalent magnetic doping, it has been predicted that
Bi$_2$Se$_3$, Bi$_2$Te$_3$, and Sb$_2$Te$_3$ will form a QAHE state, which should thus occur when Bi or Sb are substituted by Cr or Fe, but not when the substituents are Ti or V, owing to their metallicity. Moreover, nonisovalent magnetic dopants turn out to have surprisingly little effect on carrier concentration: that is, Mn-doped Bi$_2$Se$_3$ and Bi$_2$Te$_3$ always remain n-type, even though divalent Mn replacing trivalent Bi should act as a strong acceptor.

To resolve these issues, we present a comprehensive study of Mn-doped Bi$_2$Te$_3$ and Bi$_2$Se$_3$ that unequivocally reveals a large magnetic exchange splitting at the Dirac point of Bi$_2$Te$_3$. This splitting vanishes above the Curie temperature, which is clear-curt evidence for its magnetic origin. No increase in the gap size is observed for Mn-doped Bi$_2$Se$_3$ at temperatures down to 1 K. Through a multiscale structural analysis, we reveal that the actual lattice structure is very different to the anticipated random impurity system, as Mn doping induces the formation of self-organized heterostructures. This turns out to be crucial for obtaining large magnetic gaps.

**Bandgap, spin texture and magnetism**

Figure 1a–g shows the ARPES dispersions of Mn-doped Bi$_2$Te$_3$ and Bi$_2$Se$_3$ measured above and below the ferromagnetic phase transition ($T_c = 10$ K and 6 K, respectively). For Mn-doped Bi$_2$Te$_3$, the photoemission spectrum recorded at $h\nu = 50$ eV at the centre of the surface Brillouin zone shows an intensity maximum at a binding energy of 0.3 eV from the bulk valence band, while the Dirac point of the topological surface state (TSS) contributes a smaller peak at around 0.2 eV. On cooling from 20 K through $T_c$ down to 1 K, the low energy flank of the peak develops a pronounced shoulder, forming a plateau at around 0.2 eV (Fig. 1a–c). Assuming that the single component for the topological surface state (TSS) contributes a smaller peak at around 0.2 eV. On cooling from 20 K through $T_c$ down to 1 K, the low energy flank of the peak develops a pronounced shoulder, forming a plateau at around 0.2 eV (Fig. 1a–c). Assuming that the single component for the topological surface state (TSS) contributes a smaller peak at around 0.2 eV. On cooling from 20 K through $T_c$ down to 1 K, the low energy flank of the peak develops a pronounced shoulder, forming a plateau at around 0.2 eV (Fig. 1a–c). Assuming that the single component for the topological surface state (TSS) contributes a smaller peak at around 0.2 eV. On cooling from 20 K through $T_c$ down to 1 K, the low energy flank of the peak develops a pronounced shoulder, forming a plateau at around 0.2 eV (Fig. 1a–c). Assuming that the single component for the topological surface state (TSS) contributes a smaller peak at around 0.2 eV.
oriented field ($M'$) leads to the opposite spin polarization (Fig. 1h, lower panel). This unambiguously proves that the out-of-plane spin polarization below $T_C$ is due to ferromagnetic ordering of the system. Figure IJ shows that, away from the Dirac point, the characteristic helical in-plane spin texture of the parent Bi$_2$Te$_3$ is preserved, such that an overall spin texture as displayed in Fig. 1k is formed.

Our measurement of the gap probes the exchange splitting of $p$ electrons of the host material, which ferromagnetically couple to the localized magnetic moments of the Mn ions. The magnitude of the gap thus depends on the exchange coupling $J$, and the magnetization, $M$, along the surface normal direction. Indeed, the gap size nicely follows the temperature dependence of the perpendicular magnetization, $M_\perp$ (Fig. 1k, blue crosses). This clearly demonstrates the direct correlation between the gap and ferromagnetism in the system. In Fig. 1e–g we show that such temperature dependence is not observed for Bi$_2$Se$_3$ with a similar Mn concentration of 6%, where instead a large gap of roughly 200 meV exists at all temperatures from 1 K to 300 K (ref. 14).

In particular, the gap size does not increase when cooling down to 1 K, the expected periodic sequence of Te–Bi–Te–Bi–Te quintuple layers. This unambiguously proves that the out-of-plane spin polarization below $T_C$, whereas it is negligible in Mn-doped Bi$_2$Se$_3$ (Fig. 2d). This perpendicular anisotropy in Mn-doped Bi$_2$Te$_3$ is precisely the pre-condition for the magnetic bandgap opening and the QAHE, whereas an in-plane magnetization as observed for Mn-doped Bi$_2$Se$_3$ merely shifts the Dirac cone in momentum parallel to the surface.

**Multiscale structure analysis**

To clarify how Mn is actually incorporated into Bi$_2$Te$_3$ and Bi$_2$Se$_3$, we carried out a multiscale structure analysis for both systems. Figure 3a shows Mn-doped Bi$_2$Te$_3$ in high-resolution scanning transmission electron microscopy (HRSTEM). Strikingly, we observe the emergence of a new structure composed of septuple and quintuple layers, instead of the expected periodic sequence of Te–Bi–Te–Bi–Te quintuple layers. The septuple layers consist of the sequence Te–Bi–Te–Mn–Te–Bi–Te, where the Mn atoms predominantly occupy the centre of the septuple. This self-organized heterostructure formation does not exist for stoichiometric Bi$_2$Te$_3$ and Bi$_2$Se$_3$ (Extended Data Fig. 4a) and obviously disagrees with the commonly held notion of substitutional Mn incorporation. Figure 3b and Extended Data Fig. 4b show that the same Mn-induced septuple/quintuple heterostructure formation also occurs in Bi$_2$Se$_3$, in agreement with recent observations. Thus, it is a universal mechanism in both material systems. Moreover, we identify this structure as the explanation for the surprisingly small effect of Mn doping on the carrier concentration and Fermi level of the system. Mn in MnBi$_3$Te$_4$ is electrically neutral because each septuple...
The images were recorded along the [1100] zone axes ([110] and [1210] zone axes (b)). Owing to the atomic-number contrast, the heavy atoms (Bi) appear brighter in the septuple layers (SLs) inserted between Bi$_2$Te$_3$ (Bi$_2$Se$_3$) quintuple layers (QLs). As a result, the septuple layers appear darker in the overview images because of incorporation of the lighter Mn atoms. The Mn concentration was 10% in a, and locally 9% and on average 6% in b, according to X-ray diffraction measurements (Fig. 4). c–f, Spectroscopic determination of the Mn-incorporation sites in Mn-doped Bi$_2$Te$_3$ (c, e) and Bi$_2$Se$_3$ (d, f) by X-ray absorption spectroscopy (XANES in c, d, XANES in e, f) at the Mn K-edge. Experimental data (symbols) are compared with simulations (solid lines) performed for different Mn-incorporation sites in the septuple and quintuple layers, in the van der Waals (vdW) gap or on Te (Se) antisites (see Extended Data Fig. 6).

To obtain element-specific information on the Mn-incorporation sites, we carried out X-ray absorption near-edge spectroscopy (XANES) at the Mn K-edge. Experimental data (symbols) are compared with simulations (solid lines) performed for different Mn-incorporation sites in the septuple and quintuple layers, in the van der Waals (vdW) gap or on Te (Se) antisites (see Extended Data Fig. 6).

Fig. 3 | Structure analysis by STEM and X-ray absorption spectroscopy. a, b, STEM cross-sections of Mn-doped Bi$_2$Te$_3$ (a) and Bi$_2$Se$_3$ (b), revealing the formation of layered heterostructures consisting of MnBi$_2$Te$_4$ (MnBi$_2$Se$_4$) septuple layers (SLs) inserted between Bi$_2$Te$_3$ (Bi$_2$Se$_3$) quintuple layers (QLs). The images were recorded along the [1100] zone axes ([110] and [1210] zone axes (b)). Owing to the atomic-number contrast, the heavy atoms (Bi) appear brighter in the septuple layers (SLs) inserted between Bi$_2$Te$_3$ (Bi$_2$Se$_3$) quintuple layers (QLs). As a result, the septuple layers appear darker in the overview images because of incorporation of the lighter Mn atoms. The Mn concentration was 10% in a, and locally 9% and on average 6% in b, according to X-ray diffraction measurements (Fig. 4). c–f, Spectroscopic determination of the Mn-incorporation sites in Mn-doped Bi$_2$Te$_3$ (c, e) and Bi$_2$Se$_3$ (d, f) by X-ray absorption spectroscopy (XANES in c, d, XANES in e, f) at the Mn K-edge. Experimental data (symbols) are compared with simulations (solid lines) performed for different Mn-incorporation sites in the septuple and quintuple layers, in the van der Waals (vdW) gap or on Te (Se) antisites (see Extended Data Fig. 6).
The electronic structure of transition-metal impurities in Bi$_2$Te$_3$ and Bi$_2$Se$_3$ has been studied extensively through density functional theory (DFT) calculations\textsuperscript{32-35}. For Mn in Bi$_2$Se$_3$, a nonmagnetic bandgap of the measured size (200 meV) does not appear in any DFT calculation. Although in principle, depending on orbital symmetry, small gaps of around 4 meV might open even for an in-plane magnetization\textsuperscript{36}, this is obviously much less than what we observe experimentally. The only prediction of a nonmagnetic gap of the magnitude seen in our experiments is from calculations that assume an on-site Coulomb interaction, $U$, at the impurity site\textsuperscript{37}. On the one hand, Mn forms more substitutional sites in Bi$_2$Te$_3$ than in Bi$_2$Se$_3$. They will lead to a larger Coulomb $U$ than for Mn in the centre of the septuple layer, where Mn 3d levels can delocalize in the plane. The size of $U$, also termed the impurity strength\textsuperscript{38}, indeed affects the nonmagnetic gap: comparing Mn with In doping for Bi$_2$Se$_3$, we find that to reach the same gap size as for 8% Mn, only 2% In is required\textsuperscript{39}. On the other hand, the effect of impurities on the nonmagnetic gap decreases with higher spin–orbit interaction in the host material\textsuperscript{40}, so that Bi$_2$Te$_3$ is less susceptible to a nonmagnetic gap opening than Bi$_2$Se$_3$.

To explain the marked difference in the magnetic anisotropy of Mn-doped Bi$_2$Te$_3$ and Bi$_2$Se$_3$, we calculated the magnetocrystalline anisotropy for the Bi$_2$Y$_x$MnBi$_2$Y$_z$ (Y = Te, Se) heterostructures (see Methods section ‘DFT calculation of magnetic anisotropy’). In agreement with recent model calculations\textsuperscript{40}, we find that the strong magnetocrystalline anisotropy favours out-of-plane magnetization in the telluride. In the selenide, however, because of the reduced spin–orbit interaction the magnetocrystalline anisotropy is 3.5 times smaller and practically cancelled by the shape anisotropy. Thus, the higher spin–orbit interaction in the telluride heterostructures turns the magnetization out of the plane and enables the magnetic gap to form at the Dirac point.

**Discussion**

The electronic structure of transition-metal impurities in Bi$_2$Te$_3$ and Bi$_2$Se$_3$ has been studied extensively through density functional theory (DFT) calculations\textsuperscript{32-35}. For Mn in Bi$_2$Se$_3$, a nonmagnetic bandgap of the measured size (200 meV) does not appear in any DFT calculation. Although in principle, depending on orbital symmetry, small gaps of around 4 meV might open even for an in-plane magnetization\textsuperscript{36}, this is obviously much less than what we observe experimentally. The only prediction of a nonmagnetic gap of the magnitude seen in our experiments is from calculations that assume an on-site Coulomb interaction, $U$, at the impurity site\textsuperscript{37}. On the one hand, Mn forms more substitutional sites in Bi$_2$Te$_3$ than in Bi$_2$Se$_3$. They will lead to a larger Coulomb $U$ than for Mn in the centre of the septuple layer, where Mn 3d levels can delocalize in the plane. The size of $U$, also termed the impurity strength\textsuperscript{38}, indeed affects the nonmagnetic gap: comparing Mn with In doping for Bi$_2$Se$_3$, we find that to reach the same gap size as for 8% Mn, only 2% In is required\textsuperscript{39}. On the other hand, the effect of impurities on the nonmagnetic gap decreases with higher spin–orbit interaction in the host material\textsuperscript{40}, so that Bi$_2$Te$_3$ is less susceptible to a nonmagnetic gap opening than Bi$_2$Se$_3$.

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Finally, our magnetic gap size of 90 meV for Mn-doped Bi₂Te₃ is five times larger than that predicted theoretically for substitutional Mn. This huge enhancement arises from the naturally formed heterostructure and the enhanced wavefunction overlap of the TSS with the Mn atoms in the MnBi₂Te₃ septuple layer, which supports large magnetic gaps of 38–87 meV, as predicted⁵⁹. Mn-doped Sb₂Te₃ displays the same heterostructure formation and out-of-plane magnetic anisotropy as Bi₂Te₃ (Extended Data Fig. 10), and because it is p-type, the Fermi level can be tuned into the magnetic gap by alloying of these systems. This demonstrates the great potential of such structures for stabilizing edge transport in QAHE devices. Theory also suggests that the nontrivial topology is retained in the heterostructures⁶⁰. In accordance with the persistence of the Dirac cone surface state and out-of-plane spin texture seen in our ARPES experiments. Therefore, Mn-based topological insulator heterostructures might not only boost edge transport in QAHE devices, but also facilitate the realization of new topological phases such as the axion insulator state³⁰,³¹ and the chiral Majorana fermion²⁴.

Online content
Any methods, additional references, Nature Research summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41586-019-1826-7.

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Methods

Sample growth
Mn-doped Bi$_2$Te$_3$, Bi$_2$Se$_3$, and Sb$_2$Te$_3$ layers were grown by molecular beam epitaxy (MBE) on BaF$_2$(111) substrates using a Riber 1000 and a Varian GEN II MBE system. Compound Bi$_2$Te$_3$, Bi$_2$Se$_3$, and Sb$_2$Te$_3$ as well as elemental sources for Mn, Te and Se were used for control of stoichiometry and composition. For Pb-, Sn- and Ge-doped Bi$_2$Te$_3$, we used additional PbTe, SnTe and GeTe sources. Deposition was carried out at a growth temperature of 330 °C for Bi$_2$Te$_3$ and Sb$_2$Te$_3$ and 360 °C for Bi$_2$Se$_3$ to obtain perfect two-dimensional growth independently of dopant concentrations. This was verified by in situ reflection high-energy electron diffraction (RHEED; Extended Data Fig. 9). Details of growth procedures have been reported previously$^{14,20}$. Our notation of x% Mn in Bi$_2$Te$_3$ refers to a nominal composition of (Bi$_{1-x}$Mn$_x$)$_2$Te$_3$. All layers exhibited n-type conduction with electron concentrations of the order of a few times $10^{20}$ cm$^{-3}$ (see Extended Data Fig. 5c), except for Mn-doped Sb$_2$Te$_3$ which is p-type with a hole concentration of a few times $10^{20}$ cm$^{-3}$. Immediately after growth, samples used for ARPES were capped in situ with amorphous Se and Te capping layers at room temperature to protect the surface against oxidation. This cap was removed just before the ARPES experiments by in situ sputtering and annealing.

ARPES
Photoemission experiments were performed with the ARPES-1$^\text{st}$ end station at the UE112-PGM2b undulator beam line of the BESSY II synchrotron radiation source. The lowest reachable temperature is 1 K. The experimental geometry has the following characteristics: with the central axis of the analyser lens and the polar rotation axis of the sample defined as the x and z axes of a spherical coordinate system, the photons impinge the sample under an azimuthal angle $\phi$ of 45° and a polar angle of 84°. The light polarization is horizontal (along the x axis). The entrance slit of the hemispherical analyser is placed parallel to the z axis. The measurements at $h\nu = 50$ eV were performed with an energy resolution of 10 meV. The temperature-dependent leading-edge shifts in Fig. 1c and Extended Data Fig. 1a are obtained by approximating the photoemission intensity in the indicated ranges with a line. The slope of this line is constrained to be identical for the low- and high-temperature spectra within the same section. Our notation $\Delta$ refers to the full gap.

The measurement of the magnetic exchange splitting at the Dirac point through the temperature dependence in ARPES is in a certain sense analogous to the case of gadolinium metal. In both cases the magnetic coupling of low-dimensional magnetic moments (3d in Mn, 4f in Gd) is mediated by itinerant electrons ($5p$ in Te, $5d$ in Gd) which can be probed by ARPES. At the Dirac point, electronic states of the $5p$ character are probed. In Gd, the $5d$ band probed by ARPES splits by 0.85 eV when the temperature is lowered from 1.02 $T_C$ to 0.27 $T_C$ ($T_C = 293$ K for bulk Gd)$^{35}$. Below the ferromagnetic transition $T_C$ the anomalous Hall contribution is absent.

Spin-resolved ARPES
Spin-resolved ARPES was measured at the RGBL2 end station at the U125/2 undulator beamline of BESSY II. It comprises a Scienta R4000 hemispherical analyser with two Mott-type spin polarimeters operated at 26 kV (ref. $^{14}$). The lowest temperature is 6.5 K. Light is incident under an azimuthal angle of 45° and a polar angle of 90° (Extended Data Fig. 2a). The light polarization is horizontal (along the z axis). The spin polarimeter detects the out-of-plane and one in-plane component of the spin polarization. The measured in-plane projection is tangential to the Dirac cone and perpendicular to the analyser entrance slit and electron momentum. The out-of-plane component lies within the electron emission plane and is parallel to the sample normal along the z–direction. The angular resolution was 0.75° and the energy resolution of the measurement at a photon energy of 30 eV was set to 45 meV. We note that the measurement of the spin splitting is not limited by the energy resolution because the spin-up and spin-down channels count independently of each other. An example of this is the exchange splitting of a Ni(111) surface state, measured by spin-resolved inverse photoemission as 18 ± 3 meV at an energy resolution of 300–400 meV (ref. $^{30}$).

Transport measurements
Temperature-dependent transport measurements were performed in van der Pauw geometry with out-of-plane magnetic fields ranging from 0 T to 2 T and temperatures down to 1 K using a Cryogenic mini cryogen-free system. Extended Data Fig. 5 shows the complete data set (Hall resistance plotted against magnetic field, as well as carrier concentration, $n$, and carrier mobility, $\mu$, plotted against temperature) for the Mn-doped Bi$_2$Te$_3$ and Bi$_2$Se$_3$ films with respectively 6% and 5% Mn. Below the ferromagnetic transition $T_C$ (10 K and 6 K, respectively), the Hall resistance comprises the contribution from the ordinary Hall effect (proportional to 1/$n$) and the anomalous Hall effect (proportional to the magnetization, $M$). Because the latter is proportional to the perpendicular magnetization, the anomalous Hall contribution is minute for Mn-doped Bi$_2$Se$_3$ films, for which the magnetization vector is nearly parallel to the film plane. Above $T_C$ the anomalous Hall contribution is absent.

Magnetic characterization
The magnetic properties were determined by measuring magnetization, $M$, as a function of the applied external field, $H$, and as a function of temperature, $T$ (ranging from 2 K to 300 K), using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design MPMS-XL5). We determined the Curie temperature from the $M(T)$ curves as exemplified in Extended Data Fig. 3. Note that we do not observe an enhancement of $T_C$ at the surface probed by XMCD$^{40}$. The magnetic field was applied either parallel (out-of-plane) or perpendicular (in-plane) to the c axis of the films. The diamagnetic contribution of the BaF$_2$(111) substrate was determined from the slope of the $M(H)$ curve recorded at 300 K in high magnetic fields, and was subtracted from the raw data. Identical sample pieces were used for in-plane and out-of-plane measurements. The sample size was 4 × 4 mm$^2$.

Scanning transmission electron microscopy
Atomic resolution HRSTEM images were obtained with a FEI Titan G2 60–300 STEM equipped with a Cs probe corrector and a FEI Titan 60–300 Themis equipped with a Cs image corrector, which were operated at 300 keV. The HRSTEM data were recorded with a HAADF detector and the images processed using a Wiener filter for noise minimization$^{43}$. Thin cross-sectional lamellae from Mn-doped Bi$_2$Te$_3$ and Bi$_2$Se$_3$ films with Mn concentrations of 10% and 6%, respectively, were prepared by focused ion beam (FIB) milling (ZEISS Crossbeam XB 1540 and FEI Helios NanoLab 660) along two different crystallographic directions of the BaF$_2$(111) substrate ([111] and [011], respectively). Owing to the epitaxial relationship between the films and the BaF$_2$(111) substrate, this yields [110] and [110] zone axes with respect to the Bi$_2$Te$_3$ and Bi$_2$Se$_3$ layers. Pre-characterization of the lamellae and overview STEM images were obtained with a JEOI JEM-2200FS STEM operated at 200 keV. Shortly before the HRSTEM images were taken, the lamellae were additionally thinned to remove the amorphous surface layers and damaged regions caused by the initial FIB preparation using a Fischione 1040 NanoMill. To map the element distribution (Extended Data Fig. 4c), we carried out energy-dispersive X-ray analysis using a Bruker Super-X detector.

XANES and EXAFS measurements and simulations
The XANES and EXAFS spectra at the Mn K-edge were recorded at, respectively, the ID21 and BM23 beamlines of the European Synchrotron Radiation Facility in total fluorescence yield$^{67}$. The isotropic XANES spectrum was derived from a weighted average of two XANES spectra recorded with two orthogonal linear polarizations parallel
and perpendicular to the c axis of the film. The resulting X-ray linear dichroism spectra corroborate the findings of XANES and EXAFS with regard to Mn incorporation.

For XANES simulations, we used the FDMNES code\textsuperscript{38} with a multiple scattering approach on a muffin-tin potential, for a supercell comprising the nominal bulk Bi\textsubscript{2}Te\textsubscript{3} or Bi\textsubscript{2}Se\textsubscript{3} lattices with a Mn atom replacing one Bi atom within the quintuple layers (substitutional Mn), and with Mn incorporated in the central layers of the septuples (Extended Data Fig. 6). Note that placing Mn as an octahedral interstitial within the van der Waals (vdW) gap leads to somewhat similar results as with Mn in the central position of the septuple layer, while tetrahedral Mn interstitials were in lesser agreement with experiment (Fig. 3c–f). Concerning the pre-edge features in the XANES data, it is known that the FDMNES code using the multiple scattering formalism has difficulties in reproducing the 3d–4p hybridized states at the pre-edge feature well; nevertheless the main absorption features are well reproduced and we draw conclusions only from that spectral region.

We used identical input geometries for the EXAFS and XANES simulations. We measured EXAFS spectra for a series of Mn-doped Bi\textsubscript{2}Te\textsubscript{3} and Bi\textsubscript{2}Se\textsubscript{3} samples with Mn concentrations ranging from 4% to 13%. We fitted the EXAFS data at the Mn K-edge using the FEFF9 code, assuming Mn at different lattice sites. The spectra with a model of Mn atoms in the centre position of the septuple layer are shown in Extended Data Fig. 7. The first coordination shell includes six anion atoms in the octahedral environment. We note again that the substitutional position and the interstitial position in the van der Waals gap have octahedral coordination. The distances of the nearest neighbours in the first coordination shell derived from the fits are listed in Extended Data Fig. 7e. We note that Mn atoms substituting Bi, Te(1) or Se(1) atoms in the quintuple layers (using the notation of Extended Data Fig. 6) have two different neighbours with different distances.

**X-ray diffraction and simulation with random stacking model**

We determined the crystal structure using symmetric X-ray diffraction scans and reciprocal space maps in the vicinity of the (101.20) reciprocal lattice point. The measurements were performed using a Rigaku SmartLab diffractometer with a copper X-ray tube and channel-cut Ge(220) monochromator. Symmetric scans along the [000.1] reciprocal space direction (c axis) were fitted with a modified one-dimensional paracrystal model\textsuperscript{39}, in which random sequences of Bi\textsubscript{2}Te\textsubscript{3} (or Sb\textsubscript{2}Te\textsubscript{3} or Bi\textsubscript{2}Se\textsubscript{3}) quintuple segments alternate with MnBi\textsubscript{2}Te\textsubscript{3} (or MnBi\textsubscript{2}Se\textsubscript{3}) quintuple segments along the c axis. For the samples doped with X = Pb, Sn or Ge (Fig. 4e), the septuples consist of XBi\textsubscript{2}Te\textsubscript{3}. For the quintuples, the spacings of atomic planes were set to the nominal values of Bi\textsubscript{2}Te\textsubscript{3} and Bi\textsubscript{2}Se\textsubscript{3}, and for the septuples the distance of the Mn (or Pb, Sn or Ge) plane to the nearest neighbour Te (or Se) was set to correspond to the nearest-neighbour distances determined by EXAFS.

The random sequences of quintuple and septuple layers are generated using the following assumptions. First, the length of the individual quintuple segments, N\textsubscript{qt}, is given by the gamma distribution with a certain mean value, ⟨N\textsubscript{qt}⟩, and a r.m.s. deviation, σ (r.m.s.d.); we show its relative value, σ/⟨N\textsubscript{qt}⟩. The septuple segments always consist of only one single septuple layer—that is, septuple layers are not positioned next to each other. Note that we carried out additional test fits assuming a variable length of septuple segments, but the best fits tend to the result with just one septuple layer embedded in the blocks of quintuple layers. Thus, we fixed the length of the septuple segments to one, in order to keep the number of fitting parameters as small as possible.

Second, we set the distances of the individual atomic planes in the quintuples to the values of the pure Bi\textsubscript{2}Y\textsubscript{3} phases (Y = Te, Se), as we described previously\textsuperscript{38}. For the septuples we have set the distances of the next Te or Se anion sites to the Mn in the central planes to correspond to the distances determined by EXAFS. The total thickness of the septuple equals approximately 4/3 of d\textsubscript{qt}; that is, d\textsubscript{sq} = 4/3d\textsubscript{qt}.

This relation is almost exactly satisfied for Bi\textsubscript{2}Te\textsubscript{3}, whereas for Bi\textsubscript{2}Se\textsubscript{3} d\textsubscript{sq} = 1.02 × 4/3d\textsubscript{qt}.

For the random sequences generated in this way, we calculated the XRD diffraction spectra using the one-dimensional paracrystal model described before\textsuperscript{39}, and compared the spectra to the experimental data. Examples of simulated profiles with various parameter values are shown in Extended Data Fig. 8a–d. Extended Data Fig. 8a, b depict the influence of the average number of quintuples, ⟨N\textsubscript{qt}⟩, between the septuples for a fixed disorder, that is, σ/⟨N\textsubscript{qt}⟩ = 0.5. High values of ⟨N\textsubscript{qt}⟩ correspond to an almost pure Bi\textsubscript{2}Y\textsubscript{3} lattice with just few septuple present in the stack. Such a system corresponds to samples with low Mn doping. Smaller values of ⟨N\textsubscript{qt}⟩ lead to a multilayer system, in which additional satellite diffraction peaks appear. The limiting case of ⟨N\textsubscript{qt}⟩ = 1, which is on average one quintuple alternating with one septuple, corresponds to the top blue line, with the peak positions corresponding to an average periodicity P = d\textsubscript{qt} + d\textsubscript{sq} along the growth direction. Extended Data Fig. 8c, d show the influence of the randomness (r.m.s.) on the diffraction spectra for a constant ⟨N\textsubscript{qt}⟩ of 5. A small r.m.s. corresponds to a periodic multilayer of quintuple and septuple segments, with corresponding sharp superlattice maxima, while larger r.m.s. values correspond to a disordered system with accordingly smeared profiles. For the samples doped with X = Pb, Sn or Ge (Fig. 4e), the fitted paracrystal parameters are listed in Extended Data Fig. 8g.

We note that the relation d\textsubscript{sq} = 4/3d\textsubscript{qt} has quite an important consequence for the diffraction spectra, because there is a single periodicity that is common to both quintuples and septuples. The simulated and experimental diffraction profiles shown in Fig. 4 have sharp peaks corresponding to such a periodicity independently of the statistical ordering of quintuple and septuple segments. The corresponding peaks appear at the positions (000.9) and (000.18) of the Bi\textsubscript{2}Y\textsubscript{3} structure. The average interplanar distance in the septuples is smaller than in the quintuples, because the septuple has 7/5 = 1.4 more atomic planes but is only thicker by a factor of 4/3 = 1.33.

We have also determined the Mn-concentration dependence of the in-plane lattice parameter a of the Mn-doped Bi\textsubscript{2}Y\textsubscript{3} layers from asymmetric reciprocal space maps recorded in the vicinity of the (101.20) reciprocal lattice point of the Bi\textsubscript{2}Y\textsubscript{3} structure. These results, as well as those for the average interplanar distance in the c-axis direction, ⟨d⟩, are plotted in Extended Data Fig. 8e, f. In Mn-doped Bi\textsubscript{2}Te\textsubscript{3}, we observe a decrease in both lattice parameters with increasing Mn content. This can be explained by the fact that a higher concentration of septuple layers leads to a smaller average interplanar distance, while for Mn-doped Bi\textsubscript{2}Se\textsubscript{3} the Mn content has less influence on ⟨d⟩ owing to the smaller number of septuple layers formed. In fact, in Bi\textsubscript{2}Se\textsubscript{3} we do not observe any concentration dependence of the interplanar distance ⟨d⟩ up to Mn concentrations of 8%. This is in agreement with the finding from X-ray diffraction that, for low Mn contents in Bi\textsubscript{2}Se\textsubscript{3}, only a very low number of septuple layers (Fig. 4) is present. Mn atoms also cause a small shrinking of the in-plane lattice parameter a for both Bi\textsubscript{2}Te\textsubscript{3} and Bi\textsubscript{2}Se\textsubscript{3} (Extended Data Fig. 8e, f).

**DFT calculation of magnetic anisotropy**

To get a reliable value for the magnetic anisotropy of Mn-doped Bi\textsubscript{2}Y\textsubscript{3} (Y = Te, Se) with septuple/quintuple layer structure, we carry out ab initio calculations using the well-established full-potential linearized augmented plane wave (FLAPW) method, as implemented in the WIEN2k code\textsuperscript{40}. Our calculations are based on the local density approximation. Here we used experimental lattice parameters for the multilayer system with alternating septuple/quintuple layers, which corresponds to an Mn concentration of 8%. One part of the magnetic anisotropy energy, known as magnetocrystalline anisotropy (MCA), arises from spin–orbit coupling; the other part, the so-called shape anisotropy, \( \mathcal{E}_{\text{shape}} \), comes from the magnetic dipole–dipole interaction of the individual magnetic moments. It is well known that a sufficiently dense mesh in the Brillouin zone is important for \( k \)-space integration.
On the basis of this insight, we used a 45 × 45 × 7 Monkhorst–Pack grid in the full Brillouin zone. In our benchmark calculation for FePt, we showed that in addition to the convergence of k-points, it is important to incorporate all FLAPW eigenfunctions when spin–orbit coupling is included as an additional term to the scalar-relativistic Hamiltonian, the so-called second variational step. This basis set is controlled by the energy parameters \( E_{\text{min}} \) and \( E_{\text{max}} \). To achieve a high accuracy, we set \( E_{\text{min}} \) to be -10 Ry and \( E_{\text{max}} \) to be 5 Ry. To obtain a stable value for the MCA, the energy parameters, \( E_{\ell} \), used for calculating radial wavefunctions, \( (r, E_{\ell}) \), are determined very precisely—that is, to better than 0.1 mRy.

As the WIEN2k code solves the Dirac equation in an approximate way, it is necessary to check the reliability of these sensitive calculations. For cross-checking we used the multiple-scattering KKR Green function method as implemented in the SPRKKR code. This scheme solves the proper Dirac equation; hence the relativistic effects are fully included in SPRKKR, unlike with the second variational step.

Obtaining the MCA energy by subtracting the total energies is computationally very costly. The need for self-consistent calculations for two magnetization directions can be avoided if one relies on the magnetic force theorem. In this approach the MCA energy is calculated using a frozen spin-dependent potential. The MCA energy is then obtained by subtracting the band energies, \( E_{\text{band}} \), is calculated using classical electromagnetic theory. Good agreement has been found between WIEN2k and SPRKKR codes for the MCA energy calculation. For the sake of simplicity we present only the WIEN2k results (Extended Data Fig. 3g).

Ge-, Sn- and Pb-doped Bi, Te, and Mn-doped Sb, Te

Natural formation of quintuple/septuple layer heterostructures is a generic and universal feature in Bi- and Sb-based tetradymite chalcopyride topological insulators doped with elements that prefer a 2 + oxidation state. This is demonstrated by a series of complementary thin film samples consisting of Bi, Te, doped with Ge, Sn, and Pb, as well as of Sb, Te, doped with Mn, using similar growth conditions to those described above. In all cases, two-dimensional growth was observed under the given growth conditions, as shown by the RHEED patterns recorded in situ during growth and depicted in Extended Data Fig. 9a-f. The structure of the films was analysed by X-ray diffraction, and the measured diffraction spectra were fitted with the same random stacking paracrystal model described in the section ‘X-ray diffraction and simulation with random stacking’, assuming that the additional doping element \( X = \{\text{Pb, Ge, Sn, Mn}\} \) induces the same type of septuple layer formation in which an additional \( X \)Te double layer is inserted. As shown in Fig. 4e, in all cases an excellent fit of the diffraction spectra is obtained with our paracrystal structure model. Moreover, for the same concentration of dopant elements, the average number of quintuple layers, \( \langle N_{\text{Ql}} \rangle \), between these additionally inserted septuple layers and its r.m.s. variation, \( \sigma_{\text{Ql}} \), derived from the model fits (see Extended Data Fig. 9g) turn out to be nearly the same for each dopant element.

Data availability

The data sets generated and analysed here are available from the corresponding authors on reasonable request.

Code availability

The code for the paracrystal model is available from the corresponding authors upon request. The electronic structure codes Wien2K and SPR-KKR and X-ray absorption fine structure codes FDMNES and FEFF9 can be downloaded after the corresponding licence requirements given on the respective webpages are fulfilled.

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Acknowledgements

ARPES experiments were performed at BESSY II of Helmholtz-Zentrum Berlin and the EXAFS and XANES experiments at the European Synchrotron Radiation Facility. We thank B. Henne, F. Wilhelm and A. Rogalev for support with XANES and EXAFS measurements; W. Grafener and G. Hesser for TEM sample preparation; V. Holy for advice on the paracrystal model, and G. Bihlmayer and A. Ernst for helpful discussions. This project was supported by the Austrian Science Fund (FWF, project P30969-N27 and P28185-N27), the Austrian Federal Ministry for Digital and Economic Affairs, the National Foundation for Research, Technology and Development in the frame of the Christian Doppler Laboratory for Nanoscience Phase Transformations; the Deutsche Forschungsgemeinschaft (grants SP 1668, SFB 1143 project C4, SFB 1277 project A2), the Central European Institute of Technology (CEITEC) Nano research infrastructure (ID LM2015040), MEYS CR, 2016-2019) and Computational and Experimental Design of Advanced Materials with New Functionalities (CEDAMNF; grant CZ.02.01.01/0.0/00.015_0030003580 of the Czech Ministry of Education, Youth and Sports), the Impuls- und Vernetzungsfonds der Helmholtz-Gemeinschaft (Virtual Institute New States of Matter and their Excitations and Helmholtz-Russia Joint Research Group no. HRSF-0067), and the European Union Horizon 2020 programme (grant 823717-ESTEEM3).

Author contributions

Samples were grown by S.W., H.S., V.V.V. and G.S. X-ray analysis was carried out by S.W., H.S., G.S., J.R. and O.C. O.C. performed paracrystal modelling and magnetotransport measurements. XANES and EXAFS measurements were made by A.N., O.C., G.S. and the simulations by O.C., A.N., J.R. and J. Minár. SQUID was carried out by A.N., and HR-STEM by M.A., H.G., S.W., G.S., O.C. and J. Michalička. DFT calculations were done by S.A.K., J. Minár and H.E. ARPES was carried out by E.D.L.R. and P.S.M., and spin-resolved ARPES by J.S.-B., F.F. and A.V. The work was coordinated by G.S., O.C. and J.R. The manuscript was written by O.R. and O.C. with input from all authors.

Competing interests

The authors declare no competing interests.

Additional information

Supplementary information is available for this paper at https://doi.org/10.1038/s41586-019-1826-7.

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Peer review information

Nature thanks Alexander Balatsky, Jack Fuhruly and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

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Extended Data Fig. 1 | ARPES measurements. a, Data for an additional Mn-doped Bi$_2$Te$_3$ film with Mn concentration of 10%. The normal emission spectra shown on the left, recorded at 1 K and 30 K, show a substantial redistribution of spectral weight around the binding energy of approximately 180 meV when crossing the ferromagnetic transition at $T_C = 12$ K. The shifts in the regions marked S1 and S2, shown on the right on a magnified scale, are of similar magnitude to that seen for the 6% Mn-doped case in Fig. 1. The shifts marked by arrows are compatible with a 100 meV gap opening at the Dirac point. ARPES was measured with p-polarized light and $h\nu = 50$ eV. b, c, ARPES measurements, showing that the gap in 6%-Mn-doped Bi$_2$Se$_3$ is independent of temperature. b, ARPES $E(k)$ map recorded at 1 K, with the angle-dependent binding energies of the upper Dirac cone and bulk valence band indicated with blue and orange lines, obtained from Lorentzian fits to energy-distribution curves. c, Temperature dependence of the binding energies of the upper Dirac cone minimum (blue circles), the bulk valence band at $\Gamma$ (orange squares) and their difference (black diamonds). The ferromagnetic Curie temperature is 6 K as obtained by SQUID. This analysis represents an alternative to that in Fig. 1g, which was based on fits of the upper and lower Dirac cones. In both cases, the data do not provide any indication of a relative or absolute shift of the band edges, or of a gap of the order seen in Mn-doped Bi$_2$Te$_3$, when crossing the ferromagnetic transition temperature.
Extended Data Fig. 2 | Spin-resolved ARPES of Mn-doped Bi$_2$Te$_3$.

a, Geometry of the spin-resolved ARPES experiments, including the magnetization directions indicated by $M^+$ and $M^-$. Hor. Pol., horizontal light polarization.

b, Plot of the fit results from spin-up and spin-down spectra of the topological surface state (TSS) at the Dirac point ($E_D$), and determination of the magnetic exchange splitting, $\Delta = 56 \pm 4$ meV, at 6.5 K. c, d, Fit to the spin-resolved spectra at 6.5 K, including a transition from the bulk conduction band (BCB).

e, Demonstration that the spin polarization reverses when the magnetization, $M$, is reversed. This reversal was achieved by field cooling in an applied field of 10 mT. f, Temperature-dependent magnetization, $M(T)$, measured by SQUID on a reference sample that was identical to that used to determine the magnetic field necessary for field cooling and magnetization reversal.

g, h, Before the spin-resolved ARPES measurement, the reversible temperature-dependent broadening in ARPES was verified by cooling from 14 K to 6.5 K and warming up again to 14 K. At a photon energy of 30 eV, most of the intensity near the Fermi energy stems from the bulk conduction band. i, Reversible broadening of the energy-dispersion curves upon cooling from above to below $T_C$ and warming up again.

j, TSS gap at the Dirac point $\Delta$ derived by ARPES and spin-resolved ARPES (red squares), plotted against temperature, together with the temperature-dependent out-of-plane magnetization (blue crosses), showing that the magnetic exchange splitting at the Dirac point $\Delta$ faithfully follows the magnetization perpendicular to the sample surface. Data at 1K and 20 K are from Fig. 1c, d. Data from spin-resolved photoemission (at 6.5 K and 300 K) have the smallest error. Data for 14 K were derived from i, taking the spin-resolved data from 6.5 K as a reference point.
Extended Data Fig. 3 | Magnetic properties of Mn-doped Bi$_2$Se$_3$ and Bi$_2$Te$_3$.

**a, d.** Temperature-dependent magnetization, $M(T)$, used to determine the ferromagnetic Curie temperature, $T_C$. Mn-doped Bi$_2$Se$_3$ (a) and Bi$_2$Te$_3$ (d). The magnetization was measured after field cooling (FC) at 10 mT. As indicated, below $T_C$, the magnetization of the samples rises steeply by more than two orders of magnitude. **b, e.** In-plane (ip) versus out-of-plane (oop) hysteresis loops at 300 K and 2 K, showing the absence of ferromagnetism at room temperature. **c, f.** Magnetization versus applied field, $M(H)$, for samples with different Mn concentrations, $x_{\text{Mn}}$, as indicated. For all Mn-doped Bi$_2$Se$_3$ films, the easy axis of magnetization is found to be in plane, whereas for all Mn-doped Bi$_2$Te$_3$ films it is perpendicular to the surface. The insets show the Curie temperature, $T_C$, plotted against Mn concentration. For all measurements, the diamagnetic contribution of the substrate measured at 300 K was subtracted. **g.** Magnetocrystalline anisotropy energy, $E_{\text{MCA}}$, obtained through DFT for Bi$_2$Se$_3$/MnBi$_2$Se$_4$ and Bi$_2$Te$_3$/MnBi$_2$Te$_4$ by subtracting band energies for two orientations of the magnetization. Shown are magnetocrystalline anisotropy $E_{\text{MCA}} = E_{\text{MCA}}^\parallel - E_{\text{MCA}}^\perp$, shape anisotropy $E_{\text{shape}}$, and total magnetic anisotropy $E_{\text{aniso}} = E_{\text{MCA}} + E_{\text{shape}}$. 

| Material          | $E_{\text{MCA}}$ (meV) | $E_{\text{shape}}$ (meV) | $E_{\text{aniso}}$ (meV) |
|-------------------|-------------------------|---------------------------|-------------------------|
| Mn-doped Bi$_2$Se$_3$ | 0.1087                  | -0.0875                   | 0.0212                  |
| Mn-doped Bi$_2$Te$_3$ | 0.3546                  | -0.0699                   | 0.2847                  |
Extended Data Fig. 4 | STEM of pure and Mn-doped Bi₅Se₃ and EDX maps of Mn-doped Bi₂Te₃.

**a, b.** Comparison of HR-STEM HAADF cross-sections of Bi₅Se₃ (a) and Mn-doped Bi₅Se₃ (x_Mn = 6%) (b) films grown under identical growth conditions, showing the high structural perfection and that additional septuple layers are formed only with Mn doping, whereas the pure Bi₅Se₃ film consists only of quintuple layers. These STEM cross-sections were recorded along two different zone axes.

**c.** Atomic-layer-resolved distribution of the Bi, Te and Mn atoms of a Mn-doped Bi₂Te₃ film (x_Mn = 10%) obtained by STEM-EDX mapping. The Mn atoms are predominantly incorporated in the centre of the septuple layers and to a lesser extent in the outer layers of the septuple units. No Mn is seen in the van der Waals gaps. Note that in this sample, because of the higher Mn concentration, two subsequent septuples are observed in the STEM cross-section.
Extended Data Fig. 5 | Anomalous Hall effect. Data for Mn-doped Bi$_2$Te$_3$ and Bi$_2$Se$_3$ with respectively 6% and 8% Mn. a, b, Raw data for Hall resistance as a function of magnetic field applied perpendicularly to the surface, measured at different temperatures above and below $T_c$ as indicated. c, d, Temperature dependence of the carrier concentration and Hall mobility. Note that above $T_c$ the contribution of the anomalous Hall effect to the Hall voltage is negligible and therefore does not affect the carrier concentration and mobility measurements.
Extended Data Fig. 6 | Possible Mn-incorporation sites in Bi$_2$Te$_3$ and Bi$_2$Se$_3$.

**a**. Structure models for, left to right: Mn in the centre of septuple layers; Mn substituting for Bi in quintuple layers; and interstitial Mn in the van der Waals gaps on octahedral sites and tetrahedral sites.

**b**. Nominal nearest-neighbour (NN) distances of Mn atoms located in various positions as derived by EXAFS analysis, including also possible Mn on Te (Se) antisites in the quintuples. Index '1' refers to Te (Se) sites next to the van der Waals gaps, index '2' to those in the centre of the quintuple.
Extended Data Fig. 7 | Fits of EXAFS oscillations for different amounts of Mn doping. a, b, Bi₂Te₃; c, d, Bi₂Se₃. Experimental data points are represented by the red or blue circles; black lines denote fitted curves (unlike the simulations in Fig. 3). a, c are plotted with respect to the wavevector k and the background-subtracted EXAFS absorption χ(k) is k-weighted; b, d show the magnitude, that is, the absolute value of χ(R) after Fourier transformation. e, EXAFS-fitted nearest-neighbor distances of Mn atoms in Bi₂Te₃ and Bi₂Se₃ as a function of Mn concentration.
Extended Data Fig. 8 | Simulated diffraction patterns. 

a–d, Varying paracrystal parameters of the septuple/quintuple heterostructures of: 

a, c, Mn-doped Bi₂Te₃, and b, d, Mn-doped Bi₂Se₃. 

a, b depict the influence of different average numbers of quintuples, \( <N_{QL}> \), between the septuples with a fixed relative r.m.s. deviation (r.m.s.d.) of its distribution, set to 0.5. At high \( <N_{QL}> \), the system approaches the pure Bi₂Y₃ (Y = Te, Se) phase. 

c, d show simulations for different r.m.s.d. of the statistical distribution of the number of quintuples between the septuples but constant average separation, \( <N_{QL}> = 5 \). The limit of r.m.s.d. = 0 corresponds to a perfectly periodic multilayer of five quintuples alternating with one septuple; the additional maxima are the resulting superlattice satellite peaks. A larger r.m.s.d. means a more disordered multilayer. 

Dashed lines are plotted at positions of the (000.\( \ell \)) peaks of the pure Bi₂Y₃ structure. 

e shows the average vertical (0001) lattice plane spacing \( \langle d \rangle \) as a function of Mn concentration determined by the fit of the experimental diffraction spectra presented in Fig. 4a,b, and panel f the corresponding in-plane lattice constants determined from reciprocal space maps around the (101.20) reciprocal lattice point.
Extended Data Fig. 9 | RHEED. a–f, Comparison of RHEED patterns for: 

- a, Mn-doped Bi$_2$Te$_3$;
- b, Mn-doped Bi$_2$Se$_3$;
- c, Mn-doped Sb$_2$Te$_3$;
- d, Pb-doped Bi$_2$Te$_3$;
- e, Sn-doped Bi$_2$Te$_3$; and
- f, Ge-doped Bi$_2$Te$_3$, recorded during epitaxial growth, showing perfect two-dimensional growth in all cases. The layer thickness was 200 nm and the dopant concentration was around 10% in all cases. The corresponding X-ray diffraction curves of the samples are shown in Fig. 4, and the values derived from the fits using the septuple/quintuple paracrystal stacking model are listed in g. $(N_{QL})$ is the average number of quintuples between consecutive $X$Bi$_2$Te$_4$ septuple layers; $\sigma_{QL}$ is the relative r.m.s.d. of the distribution; and %SL is the occupancy of Mn sites in the septuple layers.

| Material system     | $<N_{QL}>$ | $\sigma_{QL}$ | %SL |
|---------------------|------------|---------------|-----|
| Pb-doped Bi$_2$Te$_3$ | 2.1        | 1.3           | 14  |
| Sn-doped Bi$_2$Te$_3$ | 2.6        | 1.5           | 12  |
| Ge-doped Bi$_2$Te$_3$ | 3.0        | 1.5           | 10  |
| Mn-doped Bi$_2$Te$_3$ | 2.3        | 1.8           | 14  |
| Mn-doped Sb$_2$Te$_3$ | 3.5        | 1.7           | 11  |
Extended Data Fig. 10 | Electronic and magnetic properties of Mn-doped Sb$_2$Te$_3$. a, Angle-resolved photoemission spectrum recorded at a temperature of 38 K and a photon energy of 23 eV photon energy, showing p-type behaviour and that the Fermi level, $E_F$, is close to the Dirac point, the latter being only slightly above the top of the valence band. b, In-plane and out-of-plane hysteresis curves, $M(H)$, recorded through SQUID at 2 K (blue) and 300 K (red), showing the same perpendicular magnetic anisotropy with easy axis normal to the surface as for Mn-doped Bi$_2$Te$_3$ (Fig. 2 and Extended Data Fig. 3).