High-temperature quantum anomalous Hall regime in a MnBi₂Te₄/Bi₂Te₃ superlattice

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The quantum anomalous Hall effect¹–³ is a fundamental transport response of a topological insulator in zero magnetic field. Its physical origin is a result of an intrinsically inverted electronic band structure and ferromagnetism⁴, and its most important manifestation is the dissipationless flow of chiral charge currents at the edges of the system⁵, a property that has the potential to transform future quantum electronics⁶,⁷. Here, we report a Berry-curvature-driven⁸–¹⁰ anomalous Hall regime at temperatures of several Kelvin in the magnetic topological bulk crystals in which Mn ions self-organize into a period-ordered MnBi₂Te₄/Bi₂Te₃ superlattice. Robust ferromagnetism of the MnBi₂Te₄ monolayers opens a surface gap⁵,⁶, and when the Fermi level is tuned to be within this gap, the anomalous Hall conductance reaches an $e^2/h$ quantization plateau, which is a clear indication of chiral transport through the edge states. The quantization in this regime is not obstructed by the bulk conduction channels and therefore should be present in a broad family of topological magnets.

In the quantum Hall effect (a quantized version of the conventional Hall effect), the Hall conductance arises from a transverse voltage generated by a longitudinal electric current and has a profound topological origin¹¹. Consequently, it takes on quantized values that are dependent only on the fundamental constants $h$ (Planck’s constant) and $e$ (electron charge). When a two-dimensional (2D) electron system is placed under a high magnetic field, electron orbits are quantized into Landau levels, which results in an insulating state with dissipationless chiral currents flowing around the edges. Distinct from the quantum Hall effect is the quantized anomalous Hall (QAH) effect¹, which occurs in zero field and without Landau quantization. Instead, it requires time-reversal symmetry to be broken and the presence of a topologically non-trivial band structure with the Fermi level inside the gap in the electronic band structure. In a 2D QAH insulator, the Hall conductance is $G_y = Ce^2/h$, where $C$ is an integer topological invariant called the Chern number⁴,¹¹ that originates from the Berry curvature⁶ in momentum space.

Among the many systems proposed to host QAH¹²,¹³, magnetic topological insulators¹⁴, which are formed by an intricate interplay between spin-orbit interactions¹⁵ and induced intrinsic magnetism, appeared to be the most promising. Indeed, the first experimental demonstration of QAH was in Cr-doped ($x = 0.15$) ultrathin epitaxial films of (Bi,Sb)$_2$Te$_3$ (ref. ¹⁶). The out-of-plane ferromagnetism that is induced by doping with Cr breaks time-reversal symmetry and opens a small (5–10 meV) Dirac mass gap¹⁷ in the topological surface states, and the Fermi level is then fine-tuned into this gap by electrostatic gating. Quantization was observed in a limited (5–10 nm) thickness range¹⁷, with the lower thickness limit fundamentally set by hybridization of the top and bottom surface states. The upper thickness limit depends on the top and bottom ‘asymmetry’ and on the nature of magnetic order¹⁷. It is widely acknowledged that the ubiquitous magnetic doping inhomogeneities and Bi–Sb alloying disorders¹⁸ set the magnetization $M(T)$ in such films to be non-mean-field-like, with the onset of anomalous Hall conductance $G_y(T)$ in the low sub-Kelvin temperature range¹⁹–²¹, which is below the ferromagnetic Curie temperature $T_C ≈ 30 K$ by orders of magnitude. It has been shown that subsurface modulation doping²² can raise the temperature at which anomalous Hall conductance appears to near 2 K, but so far QAH has been observed only in ultrathin films under the most restrictive materials engineering constraints.

Here, we report the discovery of a much higher-temperature QAH regime in a low-disorder bulk crystal of a topological material, in which the crystalline order and the magnetic structure (Fig. 1a–c and Supplementary Fig. 1) are self-organized into a well-ordered topological superlattice. The quantization is observed when the Fermi level aligns with the Dirac mass gap. This occurs within an energy window in the conduction band (the ‘Q-window’) in which the bulk contribution to the anomalous Hall conductance is zero, a regime that was not previously accessible in thin films. We note that an analogous superlattice structure was proposed for a magnetic Weyl semimetal²³, in which the Hall conductivity per topological layer is quantized and chiral edge states can emerge in three dimensions.

The system is nominally a canonical topological insulator Bi$_2$Te$_3$, with a small (2% in total) population of Mn ions, grown as an $n$-type semiconductor (an electron conductor)²⁴ (see Methods). A crystal structure composed of quintuple layers (QLs) of undoped Bi$_2$Te$_3$ (ref. ²⁵) is modified from a disordered magnetic impurity system into a nearly periodic alternating sequence of QLs (Bi$_2$Te$_3$) and septuple layers (SLs) of atoms, with the MnBi$_2$Te$_4$ crystal structure in each SL²⁶. Mn$^{2+}$ ions are predominantly incorporated into a monolayer within each SL (Fig. 1a–c), with only sparse amounts in QLs, and the SLs are roughly separated by four QLs (Fig. 1a and Supplementary Fig. 1). The magnetization $M$, which is mainly set by the SLs, aligns out of the plane, and the ferromagnetic...
Fig. 1 | Quantized Hall conductance in a magnetic superlattice MnBi2Te4/Bi2Te3. a, Periodic sequence of Bi2Te3 (QLs) and MnBi2Te4 (SLs). b, Enlargement of a. Mn monolayers within SLs imaged by high-resolution scanning transmission electron microscopy (left) and X-ray elemental analysis (right) (see Supplementary Information). c, The crystal structure of MnBi2Te4. A single SL atomic block (outlined in orange) corresponds to the indicated layer in Supplementary Information). d, Illustration of the out-of-plane magnetization M, with one-dimensional chiral channels on the edges of a three-dimensional topological insulator. Both surfaces contribute to the Hall conductance \( G_{\text{xy}} = 0.5e^2/h \) with the same sign, resulting in a total \( G_x = \pm e^2/h \). e, M(T) (red circles) measured in a -100 Oe field. \( G_{\text{AHE}}(T) \) (blue squares) appears at \( T_c \lessgtr 13 \) K and begins to approach the quantized value \( e^2/h \) at around 7 K when cooled. f, The \( G_{\text{AHE}}(T) \) hysteresis loop for sample S1 at 1.9 K with a coercive field \( H_c \approx 50 \) mT and the zero-field half-the-loop height \( G_0 = e^2/h \). The corresponding longitudinal conductance \( G_{\text{xx}}(H) \) (see Supplementary Information section C for reciprocal conductance-to-resistance tensor conversion).

The electronic band structure of our self-organized magnetic superlattice is very different from that of dilute randomly doped or undoped Bi2Te3 (ref. 26). Recent density functional theory (DFT) calculations of the surface bands in thin MnBi2Te4/Bi2Te3 (SL/QL) heterostructures and bilayers, without accounting for the ever-present vacancies, predict a much enlarged Dirac gap of approximately 70 meV located roughly in the middle of the bulk gap. A large Dirac gap has been reported in a ferromagnetic SL/QL stack in an angularly resolved photoemission spectroscopy (ARPES) study4. It is consistent with a recent observation of QAH in ultrathin flakes of pure MnBi2Te4, in which the net out-of-plane moment is set by an odd number of layers28.

To reach the quantum anomalous Hall regime, the Fermi level should be positioned within the surface Dirac gap. However, the as-grown crystals are initially strongly \( p \)-type semiconductors. Therefore, to tune the surface band structure to quantization, we use a two-step vacancy engineering process (see Supplementary Figs. 4,5), a technique that we have demonstrated previously29 in a variety of topological insulators. First, we irradiate the crystal with high-energy (2.5 MeV) electrons (see Methods) to create a uniform distribution of predominantly donor vacancies, which makes the material less \( n \)-type (Supplementary Fig. 5a). Next, thermal annealing at a series of annealing temperatures is used to shift the surface bands upward relative to the bulk bands. The measurement of the

Fig. 2 | Tuning MnBi2Te4/Bi2Te3 superlattice to quantization. a, Vacancy redistribution by the thermal annealing process establishes a vacancy density gradient, with surface regions (blue shade) progressively denuded of vacancies. The annealing temperature increases in the direction of the arrow, with colours representing different \( T_c \). The vacancy profile is consistent with the sample thickness \( t \gg L_D \), with Debye screening length \( L_D \approx 2 \) nm. b, The vacancy gradient induces subsurface band bending, and 2DEG states form in the confined depleted regions (see text).Bulk valence bands are marked BVB. Here, \( E_g \) is a bulk gap, the red dashed line indicates the position of the Fermi level at quantization, and vertical grey dashed line delineates the band-bending region. c, A step-by-step illustration of the positions of surface bands (blue) relative to the bulk bands (grey) as created by the thermal annealing process. AnEssential at (Q) will occur when \( E_g \) and the Dirac gap \( \Delta \) align within the Q-window. \( E_g^1 \) is \( E_g \) at quantization. d, \( G_x(H) \) measured at 1.9 K for different annealing temperatures \( T_c \), RT, room temperature. Initially, the normal Hall conductance \( G_{\text{xx}}(H) \) is of \( n \)-type and nearly field linear in the range shown. It decreases monotonically with \( T_c \) until it reaches an apparent plateau shown at the \(-1.5T \) field in e, prior to the eventual conversion to \( p \)-type (negative \( G_{\text{xx}} \) shaded area). Inset in e, Optical image of the van der Pauw contact geometry used. f, Voltage gating at Q reveals ambipolar behaviour in Hall resistance \( R_H \) (blue circles) superimposed on the bulk background. \( R_H \) (white circles) shows a minimum expected in a chiral state. g, \( G_{\text{AHE}}(H) \) hysteresis loops at different values of \( T_c \). Quantization is reached at \( T_c = 92 \) K. h, \( G_{\text{AHE}}(red) \) and \( G_{\text{AHE}}(blue) \) as a function of \( T_c \). A dip in \( G_x(T_c) \) corresponds to a reduced density of states (DOS) within \( E_{\text{p}} \) (blue shaded region in d). The dip is slightly shifted from the \( G_{\text{AHE}}(H) \) to \( G_0 \), owing to a misalignment of the DOS minimum and the Dirac gap \( \Delta \). The schematic steps in e and h are labelled as in i. DOS of the \( n \)-type bulk bands (blue) and surface \( n \)-type (green) and \( p \)-type (red) bands at Q. The minimum in the bulk DOS is within \( E_{\text{p}} \) and the location of the Dirac gap \( \Delta \) is also indicated.
low-temperature longitudinal and Hall conductance between each step, allows us to fine tune the process and track the evolution of the band structure. The vacancy-driven band structure modification we use here to tune the system to quantization is supported by the DFT calculations that incorporate vacancies (Supplementary Fig. 6 and Supplementary Information section B) and is consistent with the band structure obtained from ARPES (Supplementary Fig. 7).

The process of annealing promotes charged vacancy diffusion to the surfaces\(^3\) and has two distinct elements: (1) the carrier concentration within the bulk layer is reduced (the Fermi level, \(E_F\), moves down), and (2) a vacancy gradient (Fig. 2a) is established, which causes subsurface bulk bands to bend (Fig. 2b). Quantum confinement in the vacancy depleted regions results in the formation of subsurface 2D electron gas (2DEG) quantum-well-like states\(^3\). To better illustrate this process, the bulk and surface bands are shown separately and the movement of \(E_F\) (down) and the surface bands (up) relative to the bulk conduction band (BCB) minimum are indicated for four steps during the annealing process (Fig. 2c). Initially (Fig. 2c, step 1), the Dirac bands and 2DEG bands are aligned with the bulk bands, and the Fermi level is within the BCB at position \(E_F^0\). When annealed, the Fermi level shifts slowly downward, while at the same time the surface bands rapidly shift upward relative to...
the bulk bands. Thus, we can expect that, as a result of the annealing process, the Fermi level will first pass into the 2DEG separation \( E_D \) (Fig. 2c, step 2), then into the surface Dirac gap \( \Delta \) (Fig. 2c, step 3), and finally into the 2DEG and Dirac valence bands (Fig. 2c, step 4), all while the bulk remains \( n \)-type. This will lead to the superposition of the quantized surface anomalous Hall response on conventional \( n \)-type conduction.

The magnetotransport data are shown for a sample (sample S1) irradiated with a 3.1 \( \text{cm}^{-2} \) electron dose and annealed at a series of temperatures \( T_a \) (Fig. 2d–h). The Hall conductance \( G_{yx} = G^N_{yx} + G_{yx}^{\text{AHE}} \) (Fig. 2d) reflects the sum of the normal bulk response \( G^N_{yx} \), which is linear in an applied field, and the anomalous surface response \( G_{yx}^{\text{AHE}} \), which shows a hysteretic step response around zero field. Consistent with the formation of a vacancy density gradient, the initial linear slope is large and negative (\( n \)-type). Upon annealing, it first decreases with increasing \( T_a \), then increases again when \( T_a \) is in the 70–90°C range, and then decreases again for higher \( T_a \), eventually converting to a positive (\( p \)-type) slope (see also Supplementary Fig. 5). The overall downward trend with an apparent plateau at intermediate annealing temperatures is seen clearly when the Hall conductance at \(-1.5 \text{T}\) is plotted as a function of \( T_a \) (Fig. 2e). The onset of the plateau in \( G_{yx} \) corresponds to the point at which the Fermi level enters the 2DEG separation gap (point 2 in Fig. 2c,e). This is confirmed by the measurement of the gating response (Fig. 2f). The ambipolar behavior centered near zero voltage is direct evidence for type conversion\(^{2,21} \), and the minimum in \( R_x \) points to the presence of chiral edge channels on the gapped surfaces\(^{21} \) coexisting with 2DEG conductance channels.

The anomalous Hall conductance \( G_{yx}^{\text{AHE}} \) is obtained by the subtraction of the linear normal Hall conductance \( G^N_{yx} \) from \( G_{yx} \) (Fig. 2g). With increasing \( T_a \), the height of the \( G_{yx}^{\text{AHE}} \) loop first decreases, reaches a minimum half-height quantized value \( G_0 = 1.001 \text{e}^2/\text{h} \) and then increases again. Notably, the quantized value of \( G_{yx}^{\text{AHE}} = G_0 \), when \( E_{F} \) and \( \Delta \) align, coincides with the edge of the plateau in \( G_{yx} \) (Fig. 2c,e, point 3). A plot of \( G_{yx}^{\text{AHE}} \) and the longitudinal conductivity...
Fig. 4 | Gating dependence of Berry-curvature-driven QAH. a, The bulk contribution to $G_x^{\text{AHE}}$ from Fig. 3b added to the contribution from the surface states (SSs) (red) for SSs upshifted to the edge of BCB but still aligned with $E_F$ (indicated by the vertical dashed line). The structure in the total $G_x^{\text{AHE}}$ that arises from the bulk closely emulates that seen in samples S3 (blue squares) and S5 (green squares) once the temperature range is rescaled by $\Delta T_{\varDelta}$. The plateau is not observed when $E_F$ is outside the $Q$-window (grey circles). The right vertical scale units are the same as on the left. Inset, model calculation under finite surface voltage (Supplementary Information section D) confirms the existence of the $e^2/h$ plateau and reproduces the behaviour of $G_x$ outside the $Q$-window. In our calculation, $G_x$ is obtained by the integration of the Berry curvature $\Omega^+ \omega$ with the Fermi-Dirac distribution $f(\epsilon_a - \epsilon_g)$ over the Brillouin zone of the $n$ bands corresponding to our superlattice. b, $G_x(V_g)$ (here at $-14$ T field) exhibits a quasi-plateau akin to that obtained by annealing. Lower inset, a schematic of the surface and bulk states on the quasi-plateau. Upper inset, a three-dimensional contour plot of $\delta G_{\varDelta} = G_{\varDelta}(V_g) - G_{\varDelta}(\nu_g = 0)$, a measure of the Dirac gap $\Delta$, which widens with applied magnetic field (see also Supplementary Fig. 13). d, The subtraction of the high-field bulk contribution $G_x^B$ from the total $G_x$ gives $\Delta G_{\varDelta}(V_g) \approx G_0 = e^2/h$ (dashed line).

$G_x$ versus $T_{\varDelta}$ (Fig. 2h) shows that the minimum in $G_x$ slightly precedes quantization of $G_x$. This indicates that the minimum in the 2DEG density of states is slightly above the Dirac gap (Fig. 2i). Such behaviour is seen in different samples (see Supplementary Fig. 8).

At quantization, $G_x = gG_0$ is rather high ($g \sim 1,000$) in our system, in contrast to a previously reported QAH regime\(^1\) in which $G_x$ approaches zero. Below, we show with a simple summation of the 2D surface and bulk conductance channels how they are separable in 2D, which makes possible the detection of quantization in the surface Hall (transverse) channel when the Fermi level is tuned into the surface gap, even in the presence of a finite longitudinal conductance channel.

The total conductance is a sum of the surface and bulk contributions, so that $G_x = G_x^S + G_x^B$. At quantization, when $E_F$ aligns with the Dirac mass gap, the conductance matrices are given by

\[
G_x^S = \frac{\pi}{\hbar} \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix} \quad \text{and} \quad G_x^B = \frac{\pi}{\hbar} \begin{pmatrix} \eta & 0 \\ 0 & \eta \end{pmatrix},
\]

which satisfy $G_{xx}^S \to 0$ (dissipation-free chiral edge conductance) and $G_{xx}^B = G_0$ (quantized Hall conductance, QAH) and enforce the experimental result that in the quantized regime there is minimal contribution to $G_x$ from the bulk (see Fig. 3b,c). The total resistance becomes small, consistent with our experiments (see Supplementary Information sections C and D). Next, we show that there indeed exists a $Q$-window in which the bulk contribution to $G_x$ is zero.

The ease of tunability of the Fermi level and Dirac mass gap rests on the delicate balance between the thickness of the sample and the irradiation dose (Supplementary Fig. 9). The $T_{\varDelta}$ range, $\Delta T_{\varDelta}$ needed to reach quantization (in the putative $Q$-window) is larger in thicker samples and at lower electron irradiation. This is shown for three representative samples in Fig. 3a. The bulk contribution to AHE is deduced by the subtraction of a presumed quantized surface state contribution $G_{xx}^S$ (see Supplementary Information sections C and D) from the $G_{xx}^{\text{AHE}}$ data (for example, Fig. 3b, sample S1), which
reveals the existence of the Q-window in the samples shown. A consistent presence of the Q-window is further seen in the samples that cannot be tuned to quantization. This is shown for sample S4, for which within our window \( G_{yx} \) is always less than \( G_0 \) (Fig. 3e,f). For this sample, the surface bands are initially already in the BCB. When thermally tuned, the vacancy distribution upshifts the gapped surface Dirac cone even deeper into the BCB, so that only the tail of \( G_{yx}^{\text{MHE}} \) is left within the regime in which \( G_{yx}^{\text{MHE}} \approx 0 \). When the Fermi level resides in the \( G_{yx}^{\text{MHE}} \approx 0 \) window, \( G_{yx} \) is much smaller than \( G_0 \), as neither the bulk nor surface contributes significantly (Fig. 3c,d). In this sample, the Hall slope switches from negative to positive (a transition from an n-type to p-type conduction) at low magnetic fields while remaining negative at high fields (Fig. 3e). Clearly, this is not an ambipolar conversion across charge neutrality point, CNP (the high-field negative slope of \( G_{yx} \) indicates that bulk is still n-type when the conversion is observed); instead, it is when p-type surface carriers compensate for n-type bulk (Fig. 3d). Here, \( E_F \) is still within the BCB, but \( G_{yx} \) becomes very small (less than 0.04 \( G_0 \)), which confirms that the bulk contribution within the window is indeed negligible.

To further test this regime, we add the bulk contribution deduced from sample S1 to that from the surface states to obtain the total \( G_{yx}^{\text{MHE}} \) for when the surface states are at the edge of the Q-window and \( E_F \) is still aligned with \( \Delta \). This is the case for samples S3 and S5 (see Supplementary Fig. 10), for which the initial \( G_{yx} \) is relatively large (greater than 2\( G_0 \)) and both extrinsic and intrinsic contributions to \( G_{yx} \) are possible. As \( E_F \) enters the Q-window, \( G_{yx}^{\text{MHE}} \) quickly drops to \( G_0 \) and remains close to \( e^2/h \) on a plateau before it exits the Q-window when the sample is further annealed. Indeed, the total deduced \( G_{yx}^{\text{MHE}} \) closely mimics the observed bulk features once the energy range is rescaled by \( \Delta T_e \) (see Fig. 4a). Here, once the surface bands are relocated to the vicinity of the Q-window, we can explore and fine-tune \( G_{yx} \) by electrostatic gating. \( G_{yx} \) versus gate voltage \( V_g \) at zero magnetic field displays a true \( e^2/h \) plateau, which is completely absent when the gating range is shifted beyond the Q-window (Fig. 4b). In a finite field, \( G_{yx} (V_g) \) exhibits a quasi-plateau (Fig. 4c and Supplementary Fig. 11), similar to that observed under thermal tuning, which reflects the field dependence of the surface gap (Fig. 4d,e and Supplementary Fig. 12).

The fundamental reason for the existence of the Q-window is that Berry curvature of the bulk bands is concentrated around the small gaps (‘avoided crossings’) opened by spin-orbit coupling\(^7\) and small gaps (‘avoided crossings’) opened by spin-orbit coupling\(^7\) and 4d,e and Supplementary Fig. 12). The fundamental reason for the existence of the Q-window is that Berry curvature of the bulk bands is concentrated around the small gaps (‘avoided crossings’) opened by spin-orbit coupling\(^7\) and small gaps (‘avoided crossings’) opened by spin-orbit coupling\(^7\) and
27. Otrokov, M. et al. Highly-ordered wide bandgap materials for quantized anomalous Hall and magnetoelectric effects. *2D Mater.* 4, 025082 (2017).
28. Deng, Y. et al. Quantum anomalous Hall effect in intrinsic magnetic topological insulator MnBi2Te4. *Science* 367, 895–900 (2020).
29. Zhao, L. et al. Stable topological insulators achieved using high energy electron beams. *Nat. Commun.* 7, 10957 (2016).
30. Hu, S. M. *Atomic Diffusion in Semiconductors* (ed. Shaw, D.) 217 (Springer, 1973).
31. Bahramy, M. S. et al. Emergent quantum confinement at topological insulator surfaces. *Nat. Commun.* 3, 1159 (2012).
32. Checkelsky, J. et al. Trajectory of the anomalous Hall effect towards the quantized state in a ferromagnetic topological insulator. *Nat. Phys.* 10, 731–736 (2014).
33. Xu, Y., Miotkowski, I. & Chen, Y. Quantum transport of two-species Dirac fermions in dual-gated three-dimensional topological insulators. *Nat. Commun.* 7, 11434 (2016).
34. Chen, T. et al. High-mobility Sm-doped Bi2Se3 ferromagnetic topological insulators and robust exchange coupling. *Adv. Mater.* 27, 4823–4829 (2015).
35. Yue, C. et al. Symmetry-enforced chiral hinge states and surface quantum anomalous Hall effect in the magnetic axion insulator Bi1−xSmxSe3. *Nat. Phys.* 15, 577–581 (2019).

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Methods

Crystal growth and structural characterization. Crystals of the intrinsic MnBi$_x$Te$_{3-x}$ superlattice were grown by the vertical Bridgman method following the two-step technique. In the first step, ground (20–100 mesh) high-purity (99.999%) components, bismuth (Bi), tellurium (Te) and manganese (Mn), were weighted according to the formula Mn$_x$Bi$_{3-x}$Te$_{3}$ and loaded into the double-wall quartz ampoules to avoid depressurization during the cooling process. The ampoules were evacuated to 10$^{-6}$ Torr, sealed and loaded into a vertical furnace, and heated to 900 K, where they remained for 48 h to achieve better homogenization. Afterwards, the furnace was cooled down to 550 K (at a rate of 60 K h$^{-1}$), annealed for 24 h, and then switched off and cooled down to room temperature (300 K). In the second step, the synthesized material was ground again, loaded into the ampoules for the Bridgman growth, evacuated to 10$^{-6}$ Torr, and sealed. To create the seed crystal, ampoules with a small diameter (1.5–2.0 mm) along the tip in the lower end were used. Two ingots obtained from the first (synthesis) step were used to fill the Bridgman growth ampule. To obtain a homogenized solution, the material was heated to 1,073 K and rotated along the ampule axis for 5 d in the hot part of the furnace. The samples were then moved down from the hot part at the speed of 2 mm d$^{-1}$. The temperature in the lower part of the furnace was kept at 873 K. This procedure resulted in n-type superlattice crystals with average sizes of 50 mm length and 14 mm diameter. It should be noted that the single step process can result in randomly Mn-doped p-type crystals$^{36}$ or in p-type crystals that are Mn-doped Bi$_x$Te$_{3-x}$ with well-separated MnBi$_x$Te$_{3}$ layers (see Supplementary Figs. 4.5). We remark that the well-known band structure of Bi$_2$Te$_3$-type crystals$^{36}$ or in...

Electron irradiation. Electron irradiations were carried out in a National Electrostatics Pelletron-type electrostatic accelerator configured with a low-temperature target maintained at 20 K in a chamber filled with liquid hydrogen fed from a close-cycle refrigerator$^{29}$. All irradiations were performed with samples kept at 20 K, below the mobility threshold of the interstitials that tend to be more mobile than vacancies. This ensured the stability of all charges introduced by the irradiation process. The beam current density, typically 2 μA on a 0.2 cm$^2$ surface, allowed modifications of the carrier concentration on the order of 10$^{10}$ cm$^{-3}$. We note that in randomly Mn-doped (2%) Bi$_x$Te$_{3}$ crystals, irradiation induces the expected p-to-n conversion (Supplementary Figs. 4a,b). However, the anomalous Hall effect in this case is null on the p-type side (Supplementary Fig. 4c) and is small throughout the conversion (see also ref. $^{17}$), although the magnetization is still carrier independent and large (Supplementary Fig. 4d). This observation brings into focus the critical role played by the band structure and signals the importance of Berry curvature effects.

Magnetic and transport measurements. Direct current magnetization measurements of the single crystal samples were performed in the Superconducting Quantum Interference Device (SQUID) magnetic property measurement system. Transport measurements were performed in a 14 T Quantum Design Physical property measurement system in 1 Torr (at low temperature) of He gas on many samples, each subjected to the same annealing protocol. The samples were annealed in situ starting from a temperature of 330 K and reaching up to 400 K. The annealing temperature ramp rate was 7 K min$^{-1}$ with the annealing time typically ~1 h. For $T_\alpha > 400$ K, samples were annealed ex situ in a vacuum furnace. Crystals were mechanically exfoliated onto 300 nm SiO$_2$/Si++ wafers, which typically resulted in micron-size crystals with thicknesses less than ~400 nm, as determined by the atomic force microscope. Electrical contacts in the van der Pauw configuration were lithographically patterned and a sputtered Au metallurgy was used (Fig. 2d). The van der Pauw resistance measurements were carried out using a custom-configured system; the current direction was reversed to minimize thermal electromotive force (see Supplementary Information section A for further sample characterization).

Data availability
Source data are available for this paper. All other data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.

References
36. Hor, Y. et al. Development of ferromagnetism in the doped topological insulator Bi$_{1-x}$Mn$_x$Te$_3$. Phys. Rev. B 81, 195203 (2010).
37. Checkelsky, J. G., Ye, J., Onose, Y., Iwasa, Y. & Tokura, Y. Dirac-fermion mediated ferromagnetism in a topological insulator. Nat. Phys. 8, 729–733 (2012).

Acknowledgements
We wish to acknowledge A. Millis for helpful insights and J. Hone for critical reading of the manuscript. This work was supported by the US National Science Foundation grants DMR-1420634 (Columbia-CCNY MRSEC) and HRD-1547830, and by the Polish National Science Center grant 2016/21/B/ST3/02565. Irradiations on SIRIUS platform were supported by EMIR&A under project 18-9136. Computational support was provided by Virginia Tech Advanced Research for Computing and the San Diego Supercomputer Center under DMR-06009N.

Author contributions
Experiments were designed by L.K.-E. and H.D. Device fabrication and the transport and magnetic measurements were performed by H.D. Structural and elemental characterization of the crystals grown by I.V.F. was done by K.S. and J.B. A.W. and J.S. characterized samples by ferromagnetic resonance. Electron irradiations were conducted by M.K. with the assistance of Z.C. and H.D. ARPES studies were performed by T.H. and L.P. K.P. calculated the DFT band structure. A.B.G. and J.C. calculated AHE conductance from the Berry curvature. Data analysis was done by H.D. and L.K.-E. L.K.-E. wrote the manuscript with the input from H.D.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information is available for this paper at https://doi.org/10.1038/s41567-020-0998-2.
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