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# High-temperature quantum anomalous Hall regime in a $\text{MnBi}_2\text{Te}_4/\text{Bi}_2\text{Te}_3$ superlattice

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**The quantum anomalous Hall effect [1, 2] is a fundamental transport response of a topologically non-trivial system in zero magnetic field. Its physical origin relies on the intrinsically inverted electronic band structure and ferromagnetism [3], and its most consequential manifestation is the dissipationless flow of chiral charge currents at the edges [4] that can potentially transform future quantum electronics [5, 6]. Here we report a Berry-curvature-driven [4, 7] anomalous Hall regime at temperatures of several Kelvin in the magnetic topological bulk crystals where Mn ions self-organize into a period-ordered  $\text{MnBi}_2\text{Te}_4/\text{Bi}_2\text{Te}_3$  superlattice. Robust ferromagnetism of the  $\text{MnBi}_2\text{Te}_4$  monolayers opens a surface gap [8–10], and when the Fermi level is tuned to be within this gap, the anomalous Hall conductance reaches an  $e^2/h$  quantization plateau — a clear indication of chiral transport through the edge states. The quantization in this new regime is not obstructed by the bulk conduction channels and thus should be present in a broad family of topological magnets.**

In the quantum Hall (QH) effect — a quantized version of the conventional (normal) Hall effect — the Hall conductance, arising from a transverse voltage generated by the longitudinal current, has a profound topological origin [11]. Consequently, it takes on quantized values dependent only on fundamental constants:  $h$  (Planck’s constant) and  $e$  (electron charge). When a two-dimensional (2D) electron system is under high magnetic field, electron orbits are quantized into Landau levels, engendering an insulating state with dissipationless chiral currents flowing around the edges. Distinct from QH is the quantized anomalous Hall (QAH) effect [1], which occurs in zero field without Landau quantization. Instead, it requires breaking time-reversal symmetry and a topologically nontrivial bandstructure with the Fermi level inside the bandgap. In a 2D QAH insulator, the Hall conductance is  $G_{yx} = Ce^2/h$ , where  $C$  is an integer topological invariant called the Chern number [4, 11] originating from the Berry curvature [7] in momentum space.

Among diverse systems proposed to host QAH [2, 3, 12] magnetic topological insulators [3], formed by an intricate interplay between spin-orbit interactions [13] and induced intrinsic magnetism, appeared most promising. Indeed, the first experimental realization of QAH was in Cr-doped ( $x = 0.15$ ) ultrathin epitaxial films of  $(\text{Bi,Sb})_{2-x}\text{Te}_3$  [14]. The out-of-plane ferromagnetism induced by doping with Cr (or V) breaks time-reversal symmetry and opens a small (5 – 10 meV) Dirac mass gap [15] 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 range [16], with the lower limit fundamentally set by hybridization of the top and bottom surface states. The upper limit depends on the top and bottom ‘asymmetry’, and on the nature of magnetic order [17]. It is widely acknowledged that the ubiquitous magnetic doping inhomogeneities and Bi-Sb alloying disorders [18] set the magnetization  $M(T)$  in such films to be non-mean-field-like, with the onset of anomalous Hall conductance  $G_{yx}(T)$  in the low sub-Kelvin range [19, 20], downward-lagging the ferromagnetic Curie temperature  $T_C \approx 30$  K by orders of magnitude. Sub-surface modulation doping [21] helped raise the temperature to the vicinity of  $\lesssim 2$  K, but thus far QAH has been realized only in ultrathin films under most restrictive materials engineering constraints.

Here we report a discovery of a much higher-temperature QAH regime in a low-disorder bulk crystals of a topological material, where the crystalline order and the magnetic structure (Figs. 1a-c, and S1) 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 (‘Q-window’) in which bulk contribution to the anomalous Hall conductance is null — a regime not previously considered or accessed in thin films. We note that analogous superlattice structure was proposed for a magnetic Weyl semimetal [22], where Hall conductivity per topological layer is quantized and chiral edge states can emerge in 3D.

The system is nominally a canonical topological insulator (TI)  $\text{Bi}_2\text{Te}_3$  with a small (2at% in total) population of Mn ions, grown as  $n$ -type (electron conductor) [23], see Methods. The quintuple-layered (QL) crystal structure of undoped  $\text{Bi}_2\text{Te}_3$  [24] is modified from a disordered magnetic impurity system into a nearly-periodic alternating sequence of QLs ( $\text{Bi}_2\text{Te}_3$ ) and septuple layers (SL) of atoms, each SL with the  $\text{MnBi}_2\text{Te}_4$  crystal structure [25].  $\text{Mn}^{2+}$  ions are pre-

dominantly incorporated into a monolayer within each SL (Figs. 1a-c), with only sparse amounts populating QLs and with SLs roughly separated by four QLs (Figs. 1a and S1). The magnetization  $M$ , set mainly by SLs, aligns out-of-plane and the ferromagnetic  $T_C \cong 13\text{ K}$  (Figs. 1e, S2a, and S2b) does not depend on carrier density or type.  $\text{Mn}^{2+}$  within QLs are spin-polarized by the intimate proximity to SLs (Fig. S3).  $M(T)$  of our crystals sharply rises just below  $T_C$  (Fig. 1e), in a remarkably close correspondence with the temperature dependence of anomalous Hall conductance  $G_{yx}^{AHE}(T)$  (Fig. 1e), which also onsets at  $T_C$ . The approach to quantized value  $e^2/h$  on cooling, shown here for a thick (300 nm) crystal (sample S1), is evident at temperatures of several Kelvin. A nearly-square zero-field quantized  $G_{yx}^{AHE}(H)$  hysteresis loop and the corresponding change in the longitudinal magnetoconductance  $G_{xx}^{AHE}(H)$  are shown in Figs. 1f, g.

The electronic bandstructure of our self-organized magnetic superlattice is very different from that of dilute randomly-doped or undoped  $\text{Bi}_2\text{Te}_3$  [26]. Recent density functional theory (DFT) calculations [8, 27] of the surface bands in thin  $\text{MnBi}_2\text{Te}_4/\text{Bi}_2\text{Te}_3$  (SL/QL) heterostructures and bilayers, without accounting for the ever present vacancies, predict a much enlarged ( $\gtrsim 70\text{ meV}$ ) Dirac gap located roughly in the middle of the bulk gap. Large Dirac gap has been reported in a ferromagnetic SL/QL stack in angularly resolved photoemission spectroscopy (ARPES) study [8]. It is consistent with a recent observation of QAH in ultrathin flakes of pure  $\text{MnBi}_2\text{Te}_4$ , where the net out-of-plane moment is set by *odd* number of layers [28].

In order 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 *n*-type. Therefore, to tune the surface bandstructure to quantization we utilize a two-step vacancy engineering process (see Figs. S4 and S5) — a technique we have demonstrated previously [29] in a variety of TIs. First, irradiation with high-energy (2.5 MeV energy) electrons is used (see Methods) to create a uniform distribution of (predominantly donor) vacancies, making the material less *n*-type (Fig. S5a). Next, thermal annealing is used to shift the surface bands upward relative to the bulk bands. By annealing at a series of temperatures and measuring the low-temperature longitudinal and Hall conductance between each step, we are able to fine-tune the process and track the evolution of the bandstructure. The vacancy-driven bandstructure modification we employ here to tune the system to quantization is supported by the DFT calculations that incorporate vacancies (Fig. S6 and Supplementary Information, Section B) and is consistent with ARPES (Fig. S7).

Figures 2a-2c schematically depict bandstructure tuning by thermal annealing. Annealing promotes charged vacancy diffusion to the surfaces [30], with two distinct elements in this process: (1) carrier concentration within the bulk is reduced (Fermi level,  $E_F$ , moves down), and (2) vacancy gradient (Fig. 2a) is established, causing subsurface band bending of bulk bands, as illustrated in Fig. 2b. Quantum confinement in the vacancy depleted regions results in formation of subsurface 2D electron gas (2DEG) quantum-well-like states [31]. To better visualize this process, we sketch in Fig. 2c the bulk and surface bands separately and illustrate the movement of  $E_F$  (down) and the surface bands (up) relative to the bulk conduction band (BCB) minimum at four steps during the annealing process. Initially (step 1) the Dirac bands (blue lines) and 2DEG bands (grey lines) are aligned with the bulk bands (grey shaded areas), and the Fermi level is within the BCB at position

$E_F^0$ . Upon annealing, 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 with annealing, the Fermi level will first pass into the 2DEG separation  $E_{2D}$  (point 2), then into the surface Dirac gap  $\Delta$  (point 3), and finally into the 2DEG and Dirac valence bands (point 4), all while the bulk remains  $n$ -type. This will lead to quantization of the surface anomalous Hall response superposed on conventional  $n$ -type conduction.

Figures 2d-h show magnetotransport data for a sample (sample S1) irradiated with a  $3.1 \text{ C/cm}^2$  electron dose, and annealed at a series of temperatures  $T_a$ . The Hall conductance  $G_{yx} = G_{yx}^N + G_{yx}^{AHE}$  (Fig. 2d) reflects the sum of the normal bulk response  $G_{yx}^N$ , which is linear in applied field, and the anomalous surface response  $G_{yx}^{AHE}$ , which shows a hysteretic step response around zero field. Consistent with the vacancy diffusion picture, the initial linear slope is large and negative ( $n$ -type). Upon annealing, it first decreases with increasing  $T_a$ , remains roughly constant when  $T_a$  is in the  $70 - 90^\circ\text{C}$  range, and then decreases again for higher  $T_a$ , finally converting to a positive ( $p$ -type) slope (also see Fig. S5). The overall downward trend with an apparent plateau at intermediate annealing temperatures is seen clearly by plotting the Hall conductance at  $-1.5 \text{ T}$  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 Figs. 2c and 2e). This is confirmed by measuring the gating response (Fig. 2e). The ambipolar behavior centered near zero voltage is direct evidence for type conversion [32, 33], while the minimum in  $R_{xx}$  points to the presence of chiral edge channels on the gapped surfaces [2, 3] coexisting with 2DEG conductance channels.

Figure 2g shows the anomalous Hall conductance  $G_{yx}^{AHE}$ , obtained by subtracting the linear normal Hall conductance  $G_{yx}^N$  from  $G_{yx}$ . With increasing  $T_a$ , the height of the  $G_{yx}^{AHE}$  loop first decreases, reaches a minimum half-height quantized value  $G_0 = 1.001e^2/h$  and then increases again. Notably, the quantized value of  $G_{yx}^{AHE} = G_0$ , when  $E_F$  and  $\Delta$  align, coincides with the edge of the plateau in  $G_{yx}$  (point 3 in Figs. 2c and 2e). Plotting  $G_{yx}^{AHE}$  and the longitudinal conductivity  $G_{xx}$  vs.  $T_a$  (Fig. 2h) shows that the minimum in  $G_{xx}$  slightly precedes quantization of  $G_{yx}$ . This indicates that the minimum in the 2DEG density of states is slightly above the Dirac gap (Fig. 2i). Such behavior is seen in different samples, see Fig. S8.

Remarkably, contrary to previously reported QAH regime where  $G_{xx} \rightarrow 0$ , in our system at quantization  $G_{xx} = \eta G_0$  is rather high ( $\eta \sim 1000$ ). Here we illustrate with a simple summation of the 2D surface and bulk conductance channels how they are separable in 2D, which makes possible to detect quantization in the surface Hall (transverse) channel by tuning the Fermi level 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, i.e.  $G_Q^{tot} = G_Q^{SS} + G_Q^B$ . At quantization, when  $E_F$  aligns with the Dirac mass gap, the conductance matrices are given by  $G_Q^{SS} = \frac{e^2}{h} \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix}$  and  $G_Q^B = \frac{e^2}{h} \begin{pmatrix} \eta & 0 \\ 0 & \eta \end{pmatrix}$ , which satisfy  $G_{xx}^{SS} \rightarrow 0$  (dissipation-free chiral edge conductance) and  $G_{yx}^{SS} \rightarrow G_0$  (quantized Hall conductance, QAH) and enforce the experimental result that in the quantized regime there is minimal contribution to  $G_{yx}$  from the

bulk (see Figs. 3b,c below). The total resistance becomes small, consistent with our experiments (see Supplementary Information, Sections C and D). Next we will show that, indeed, there exists a quantization window (‘Q-window’) in which the bulk contribution to  $G_{yx}$  is zero.

The ease of tunability of the Fermi level and Dirac mass gap rests on the delicate balance between thickness and irradiation dose (Fig. S9). The  $T_a$  range,  $\Delta T_a$ , needed to reach quantization — in the putative Q-window — is larger in thicker samples and lower electron irradiation. This is shown for three representative samples in Fig. 3a. The bulk contribution to AHE is deduced by subtracting a presumed quantized surface state contribution  $G_{yx}^{SS}$  (see Supplementary Information, Sections C and D) from the  $G_{yx}^{AHE}$  data (e.g. for sample S1 in Fig. 3b), revealing the existence of the Q-window in the samples shown. A consistent presence of the Q-window is further seen in the samples that can not be tuned to quantization. This is illustrated for sample S4, where within our window  $G_{yx}$  is always less than  $G_0$  (Figs. 3e,f). For this sample the surface bands are initially already in the BCB. With thermal tuning, the vacancy distribution upshifts the gapped surface Dirac cone even deeper into the BCB, so that only the tail of  $G_{yx}^{SS}$  is left within the regime where  $G_{yx}^B \cong 0$ . When the Fermi level resides in the  $G_{yx}^B \cong 0$  window, then  $G_{yx}$  is much much smaller than  $G_0$ , as neither the bulk nor surface contribute significantly (Figs. 3c,d). In this sample, the Hall slope switches from negative to positive ( $n$ -to- $p$  conduction type transition) at low magnetic fields while remaining negative at high fields (Figs. 3e). This is clearly not an ambipolar conversion across CNP — the high-field negative slope of  $G_{yx}$  indicates that bulk is still  $n$ -type when the conversion is observed — it is when  $p$ -type surface carriers compensate  $n$ -type bulk (Fig. 3d). Here,  $E_F$  is still within BCB, but  $G_{yx}$  becomes tiny ( $\leq 0.04 G_0$ ), confirming 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  $G_{xy}^{AHE}$  for when the surface states are at the edge of the Q-window and  $E_F$  still aligned with  $\Delta$ . This is the case of samples S3 and S5 (see Fig. S10), where the initial  $G_{yx}$  is relatively large ( $> 2 G_0$  and both extrinsic and intrinsic contributions [4] to  $G_{yx}$  are possible). As  $E_F$  enters the Q-window,  $G_{xy}^{AHE}$  quickly drops to  $G_0$  and remains at  $\cong e^2/h$  on a plateau before exiting the Q-window on further annealing. Indeed, the total deduced  $G_{yx}^{AHE}$  closely mimics the observed bulk features once the energy range is rescaled by  $\Delta T_a$  (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.  $G_{yx}$  vs. 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 (Figs. 4c and Fig. S11) — similar to that observed under thermal tuning — reflecting the field dependence of the surface gap (Figs. 4d,e, and S12)).

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], quickly becoming zero away from them. Thus, when the surface Dirac cone resides within the bulk conduction band, under tunable bandstructure conditions, the Hall conductance  $\sigma_{yx}$  will settle on a plateau to a great precision around its quantized value. As a proof-

of-principle, a simple model calculation (Supplementary Information, Section D) of  $\sigma_{yx}^{AHE} = \frac{e^2}{h} \int_{BZ} \frac{d^2k}{(2\pi)^2} \sum_n \Omega_{k_x k_y}^{nk} f(\epsilon_{nk} - \epsilon_F)$  captures the key observed behaviors, namely the presence of the  $e^2/h$  plateau as well as the anomalous Hall conductance trend outside the Q-window (inset in Fig. 4b). Crucially, this happens without fine-tuning when there is an energy window where the 2D bulk  $G_{yx} = 0$ , which allows  $G_{yx}$  to attain the quantized surface value of  $e^2/h$  (Figs. S13-S14).

The Q-window uncovered here is robust and eminently tunable — it can be controlled on-demand by optimizing the initial (nonmagnetic) TI bandstructure so that the Dirac point is well within the bulk gap. The 3D intrinsic superlattices can be grown with other magnetic elements; replacing Mn with e.g. Sm may increase  $T_C$  by a factor of several [34], and there is also an intriguing possibility that such system can support a higher order topological state [35], in which, in addition to surface QAH, chiral hinge channels could emerge on the side edges. The layered van der Waals structure of the superlattice naturally lends itself to superflat exfoliated interfaces, advantageous for novel device structures and as a platform for building QAH-superconductor systems for transport detection of Majorana modes.

## Methods

Crystal growth and structural characterization. Crystals of intrinsic  $\text{MnBi}_2\text{Te}_4/\text{Bi}_2\text{Te}_3$  superlattice were grown by the vertical Bridgman method following the two-step technique. In the first step ground (20-100 mesh) high purity components (99.999%), bismuth (Bi), tellurium (Te) and manganese (Mn), were weighted according to the formula  $\text{Mn}_x\text{Bi}_{2-x}\text{Te}_3$  and loaded into the double-wall quartz ampules to avoid depressurization during the cooling process. The ampules were evacuated to  $10^{-6}$  Torr, sealed and loaded into a vertical furnace, heated to 900 K, where they remained for 48 h to achieve better homogenization. Afterwards, the furnace was cooled down to 550 K (at 60 K per hour), annealed for 24 hours, and the furnace was then switched off and cooled down to room temperature. In the second step, the synthesised material was ground again, loaded into the ampules for the Bridgman growth, evacuated to  $10^{-6}$  Torr, and sealed. To create the seed crystal, ampules 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 1073 K and rotated along the ampule axis for five days in the hot part of the furnace. The samples were then moved down from the hot part at the speed of 2 mm per day. 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 the  $p$ -type crystals which are Mn-doped  $\text{Bi}_2\text{Te}_3$  with well separated  $\text{MnBi}_2\text{Te}_4$  layers (see Figs. S4, S5). We remark that the well-known bandstructure of  $\text{Bi}_2\text{Te}_3$  [26] is not an obvious candidate for QAH, since the Dirac point, nestled in the bulk valence bands, would be largely unaffected by a random distribution of Mn in the dilute limit [36].

Electron irradiation. Electron irradiations were carried out in a NEC Pelletron-type electrostatic accelerator at the Laboratoire de Physique des Solides at École Polytechnique, Palaiseau, 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 which tend to be more mobile than vacancies – this ensured the stability of all charges introduced by the irradiation process. The beam current density, typically  $2 \mu\text{A}$  on a  $0.2 \text{ cm}^2$ , allowed modifications of carrier concentration on the order of  $10^{20} \text{ cm}^{-3}$ . We note that in *randomly* Mn-doped (2at%)  $\text{Bi}_2\text{Te}_3$  crystals, the irradiation induces the expected *p*-to-*n* conversion (Figs. S4a, b). However, the anomalous Hall effect in this case is null on the *p*-type side (Fig. S4c) and small all through the conversion (see also Ref. [37]), even though the magnetization is still carrier independent and large (Fig. S4d). This observation brings into focus the critical role played by the bandstructure and signals the importance of Berry curvature effects.

Magnetic and transport measurements. dc Magnetization measurements of the single crystal samples were performed in the Superconducting Quantum Interference Device (SQUID) magnetic property measurement system (MPMS). Transport measurements were performed in a 14 Tesla Quantum Design Physical property measurement system (PPMS) 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 330 K to 400 K. The annealing temperature ramp rate was 7 K per minute with the annealing time typically  $\sim 1$  hour. For  $T_a > 400\text{K}$ , samples were annealed *ex situ* in a vacuum furnace. Crystals were mechanically exfoliated onto 300 nm  $\text{SiO}_2/\text{Si}^{+++}$  wafers, typically resulting in micron-size crystals with thicknesses less than  $\sim 400$  nm, as determined by the atomic force microscope (AFM). Electrical contacts in the van der Pauw (vdP) configuration were lithographically patterned and a sputtered Au metallurgy was used (Fig. 2d). The vdP dc measurements were carried out using a custom-configured system; reversing current direction was employed to minimize thermal *emf*. For further sample characterization see Supplementary Section A.

### **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.

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### **Author Contributions**

Experiments were designed by L.K.-E. and H.D.. Device fabrication, transport, and magnetic measurements were performed by H.D.. Structural (TEM) and elemental characterization of the crystals grown by I.V.F. was done by K.S. and J.S.. A.W. characterized samples by ferromagnetic resonance (FMR). Electron irradiations at LSI were conducted by M.K. with the assistance of Z.C. and H.D. ARPES studies were performed by T. H. and L.P. DFT bandstructure was calculated by K.P., 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 xxxxxx Correspondence and requests for materials should be addressed to L. K.-E.

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## FIGURE LEGENDS

**Figure 1 | Quantized Hall conductance in a magnetic superlattice MnBi<sub>2</sub>Te<sub>4</sub>/Bi<sub>2</sub>Te<sub>3</sub>.** **a**, Periodic sequence of Bi<sub>2</sub>Te<sub>3</sub> (QLs) and MnBi<sub>2</sub>Te<sub>4</sub> (SLs). **b**, Zoom of **(a)**. Mn monolayers within SLs imaged by high-resolution scanning TEM (*left*) and EDX (*right*), see Supplementary file. **c**, The crystal structure of MnBi<sub>2</sub>Te<sub>4</sub>. Single SL atomic block (orange box) corresponds the indicated layer in **(b)**. **d**, Illustration of the out-of-plane magnetization  $M$ , with 1D chiral channels on the edges of a 3D TI. Both surfaces contribute to the Hall conductance  $G_{yx} = 0.5 e^2/h$  with the same sign, resulting in a total  $G_{yx} = \pm e^2/h$ . **e**,  $M(T)$  (red) measured in a -100 Oe field.  $G_{yx}^{AHE}(T)$  (blue) onsets at  $T_C \cong 13$  K, and begins to approach the quantized value  $e^2/h$  from about 7 K on cooling. **f**,  $G_{yx}^{AHE}(H)$  hysteresis loop at 1.9 K with a coercive field  $H_c \approx 50$  mT and the zero-field  $1/2$  the loop height  $G_0 = e^2/h$ . **g**, The corresponding longitudinal conductance  $G_{xx}^{AHE}(H)$ . For reciprocal conductance-to-resistance tensor conversion see Supplementary Information, Section C.

**Figure 2 | Tuning MnBi<sub>2</sub>Te<sub>4</sub>/Bi<sub>2</sub>Te<sub>3</sub> superlattice to quantization.** **a**, Vacancy redistribution by thermal annealing establishes vacancy density gradient, with surface regions (blue shade) progressively denuded of vacancies. Vacancy profile is consistent with the sample thickness  $t \gg L_D$ , with Debye screening length  $L_D \sim 2$  nm. **b**, Vacancy gradient induces subsurface band bending, with 2DEG states forming in the confined depleted regions (see text). **c**, Step by step illustration of the positions of surface bands relative to the bulk bands by thermal annealing. Quantization (Q) will realize when  $E_F$  and the Dirac gap  $\Delta$  align within the Q-window.  $E_F^Q$  is  $E_F$  at quantization. Bulk valence bands are marked BVB. **d**,  $G_{yx}(H)$  for different annealing temperatures  $T_a$ . Initially, the normal Hall conductance  $G_{yx}^N(H)$  is  $n$ -type and nearly field-linear in the range shown. It monotonically decreases with  $T_a$  until it hits a snag on an apparent plateau in **(e)**, prior to the eventual conversion to  $p$ -type (negative  $G_{yx}$ ). *Inset*: Optical image of van der Pauw contact geometry used. **f**, Voltage gating at Q reveals ambipolar behavior in Hall resistance  $R_{yx}$  riding on the bulk background.  $R_{xx}$  shows a minimum expected in a chiral state. **g**,  $G_{yx}^{AHE}(H)$  hysteresis loops at different  $T_a$ . Quantization is reached at  $T_a = 92^\circ\text{C}$ . **h**,  $G_{yx}^{AHE}$  and  $G_{xx}$  as a function of  $T_a$ . A dip in  $G_{xx}(T_a)$  corresponds to a reduced density of states (DOS) within  $E_{2D}$  (blue shade), see **(i)**. The dip is slightly shifted from the  $G_{yx}^{AHE} \cong G_0$  owing to a misalignment of DOS minimum and the Dirac gap  $\Delta$ . The schematic steps in **(e)** and **(h)** are labeled as in **(c)**. **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  $E_{2D}$ .

**Figure 3 | Quantization window for a finite  $n$ -type  $G_{xx}$ .** **a**,  $G_{yx}^{AHE}$  vs. the annealing temperature interval ( $\Delta T_a$ ) needed to arrive at quantization (Q), normalized to  $T_a$  at Q and to samples' thickness  $t$  for representative samples S1 ( $t = 300$  nm), S2 ( $t = 70$  nm) and S3 ( $t = 101$  nm) with irradiation doses  $\phi$  of 3.1, 0.75 and 0.5 C/cm<sup>2</sup> respectively. Samples S3 and S5 (see Fig. S10) both have a larger initial  $G_{yx}$  deriving from the bulk. Sample S6 is nearly identical to S2 (see Fig. S8). *Inset*: Bandstructure cartoon at Q. **b**,  $G_{yx}^{AHE} = G_0$  at Q has a minimum since the contribution to

Hall conductance from the (3D) bulk (grey shade) drops precipitously near Q and the contribution from the subsurface 2DEG (purple shade) is shown to vanish within a Q-window within BCB, see text. Both contributions add to the intrinsic Berry contribution from the surface states (SS), which peaks at  $G_0$ . The quantization is observed within this Q-window when the surface gap  $\Delta$  is aligned with  $E_F$  (vertical dash), here illustrated for sample S1. **c**, When the initial location of  $\Delta$  is deep in the BCB above the Q-window but  $E_F$  is still within it, only a tail ( $\ll G_0$ ) of SS contributes to  $G_{yx}^{AHE}$ . Red arrows indicate the direction of the SS upshift (from red dash to solid). **d**, DOS sketch illustrates how  $n$ -type bulk carriers ( $G_{xx} \neq 0$ ) can be compensated by the  $p$ -type surface carriers. **e**,  $G_{yx}(H)$  of a non-quantized sample S4 ( $t = 300$  nm,  $\phi \cong 0.5$  C/cm<sup>2</sup>). The  $n$ -to- $p$  transition (negative-to-positive  $G_{yx}(H)$  slope) is seen only at low magnetic fields, indicating that the bulk is still  $n$ -type. **f**, The surface  $n$ -to- $p$  conversion in sample S4, where the Q-window and the surface  $\Delta$  do not align and the quantization is not observed. It demonstrates that in the Q-window the contribution from the bulk is indeed very small ( $\leq 0.04 G_0$ ). Labels **1** and **2** correspond to  $T_a = 48^\circ\text{C}$  and  $127^\circ\text{C}$  respectively.

**Figure 4 | Gating dependence of Berry-curvature-driven QAH.** **a**, The bulk contribution to  $G_{yx}^{AHE}$  from Fig. 3B added to the SS contribution (red) for SS upshifted to the edge of BCB but still aligned with  $E_F$ . The structure in the total  $G_{yx}^{AHE}$  arising from the bulk closely emulates that seen in samples S3 (blue squares) and S5 (green squares) once the temperature range is rescaled by  $\Delta T_a$ . The SS plateau is clearly revealed under thermal annealing. **b**, Quantization plateau in  $G_{yx}(V_g)$  at zero field when  $E_F$  in sample S3 is aligned with  $\Delta$  (red circles). The plateau is not observed when  $E_F$  is outside the Q-window (grey circles). *Inset*: Model calculation under finite surface voltage (Supplementary Information, Section **D**) confirms the existence of the  $e^2/h$  plateau and reproduces the behavior of  $G_{yx}$  outside the Q-window. In our calculation  $G_{yx}$  is obtained by integrating the Berry curvature  $\Omega^{nk}$  with the Fermi-Dirac distribution  $f(\epsilon_{nk} - \epsilon_F)$  over the Brillouin zone (BZ) of the  $n$  bands corresponding to our superlattice. **c**,  $G_{yx}(V_g)$  exhibits a quasi-plateau akin to that obtained by annealing. *Lower inset*: Cartoon of the surface and bulk states on the quasi-plateau. *Upper inset*: 3D contour plot of  $\frac{dG_{yx}(V_g, H)}{dV_g}$  — a measure of the Dirac gap  $\Delta$  which widens with applied magnetic field (see also Fig. S13). **d**, Subtracting the high-field bulk contribution  $G_{yx}^B$  from the total  $G_{yx}$ , gives  $\Delta G_{yx}(V_g) \cong G_0 = e^2/h$ .









