High-Chern-number and high-temperature quantum Hall effect without Landau levels

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ABSTRACT

The quantum Hall effect (QHE) with quantized Hall resistance of $h/νe^2$ started the research on topological quantum states and laid the foundation of topology in physics. Since then, Haldane proposed the QHE without Landau levels, showing nonzero Chern number $|C| = 1$, which has been experimentally observed at relatively low temperatures. For emerging physics and low-power-consumption electronics, the key issues are how to increase the working temperature and realize high Chern numbers ($C > 1$). Here, we report the experimental discovery of high-Chern-number QHE ($C = 2$) without Landau levels and $C = 1$ Chern insulator state displaying a nearly quantized Hall resistance plateau above the Néel temperature in MnBi2Te4 devices. Our observations provide a new perspective on topological matter and open new avenues for exploration of exotic topological quantum states and topological phase transitions at higher temperatures.

Keywords: quantum Hall effect without Landau levels, Chern insulator, topological matter, topological quantum states, high Chern number, high temperature

INTRODUCTION

The quantum Hall effect (QHE) with quantized Hall resistance plateaus of height $h/νe^2$ was first observed in two-dimensional (2D) electron systems in 1980 [1]. Here, $h$ is Planck’s constant, $ν$ is Landau filling factor and $e$ is electron charge. The QHE in 2D electron systems with high mobility is originated from the formation of Landau levels (LLs) under strong external magnetic field. Subsequently, the exact quantization was explained by Laughlin based on gauge invariance and was later related to a topological invariance of the energy bands, which is characterized by Chern number $C$ [2–5]. A nonzero Chern number distinguishes the QHE systems from vac- 

uum with number distinguishes the QHE systems from vacuum with zero Chern number $C = 0$ [2,3]. The discovery of QHE introduces the concept of topology into condensed matter physics and is extremely important to physical sciences and technologies. However, the rigorous conditions of ultrahigh mobility, ultralow temperature and strong external magnetic field limit the deep exploration and wide applications of QHE.

Theoretical proposals based on the intrinsic band structure of 2D systems open up new opportunities.

In 1988, Haldane theoretically proposed a time-reversal symmetry (TRS) breaking 2D condensed-matter lattice model with quantized Hall conductance of $e^2/h$ in the absence of an external magnetic field [6]. This indicates that QHE can be realized without the formation of LLs. The QHE induced by spontaneous magnetization in such insulators is called quantum anomalous Hall effect (QAHE), and such insulators are called Chern insulators. An alternative mechanism of realizing QAHE through localization of band electrons was later proposed in 2003 [7]. The emergence of topological insulators (TIs) in which strong spin-orbit coupling (SOC) gives rise to topological band structures provides a new system for the investigation of QHE without strong external magnetic field. The QAHE with quantized Hall conductance of $e^2/h$ was predicted to occur in magnetic TIs by doping transition metal elements (Cr or V) into time-reversal-invariant TIs Bi2Te3, Bi2Se3 and Sb2Te3 [8]. In 2013, the QAHE with quantized Hall conductance of $e^2/h$ was experimentally observed in thin films of...
RESULTS

High-Chern-number Chern insulator states

Figure 1b shows an optical image of the MnBi$_2$Te$_4$ device (s6) with Hall bar geometry. Atomic force microscope measurements were carried out to determine the thickness of s6 (Fig. S1f). The line profile reveals a thickness of 13.4 ± 0.4 nm, corresponding to 10-SL. The temperature dependence of longitudinal resistance $R_{xx}$ is shown in Fig. 1c, in which a sharp resistance peak gives the $T_N$ at around 22 K.

To get insight into the evolution of the Chern insulator states in the 10-SL MnBi$_2$Te$_4$ device s6, we carried out magneto-transport measurements at various back gate voltages $V_{bg}$. Figure 1d and e displays the gate-dependent magneto-transport properties of s6 under perpendicular magnetic field at $T = 2$ K. Two sharp transitions at around 3 T and 5 T can be clearly observed on both $R_{xx}$ and $R_{yx}$ in Fig. 1d and e. These two transitions may mark the beginning and ending of the spin-flipping process. With further application of a perpendicular magnetic field, the sample is supposed to enter the perfectly aligned FM state [19].

The well-quantized Hall resistance plateau with height of 0.99 $h/2e^2$ is detected at $-15$ T by applying a $V_{bg} = -17$ V, accompanied by a longitudinal resistance as small as 0.004 $h/2e^2$ as shown in Fig. 1d and e. The quantized Hall resistance plateau almost does not change when further tuning $V_{bg}$ to $-58$ V (within the tolerance of the substrate), which can be clearly observed in Fig. 1f. Besides, the Hall resistance plateau deviates from the quantized value when $V_{bg}$ is above $-5$ V. The well-quantized Hall resistance plateau and nearly vanishing longitudinal resistance are characteristics of high-Chern-number QHE without LLs contributed by dissipationless chiral edge states and indicate a well-defined Chern insulator state with $C = 2$.

In the absence of a magnetic field, MnBi$_2$Te$_4$ bulk is an AFM TI, whose side surfaces are gapless and (111) surfaces are intrinsically gapped by exchange interactions [11,12,21]. The gapped surface states are characterized by a quantized Berry phase of $\pi$ and can display the novel half-quantum Hall effect [23,24]. Due to the AFM nature of the bulk, Hall conductance or topological Chern number of MnBi$_2$Te$_4$ (111) films is dictated by the surface states, which depend critically on the film thickness. For even- and odd-layer films, the two surfaces (on the top and bottom) display half-integer Hall conductance of opposite and identical signs, leading to $C = 0$ and 1, respectively [11]. Obviously, one would never obtain high Chern number $C > 1$ in AFM MnBi$_2$Te$_4$. 

However, in the above-mentioned QHE systems without LLs, only a Hall resistance plateau with $C = 1$ can be obtained by coupling topological surface states with magnetism. High-Chern-number QHE without LLs has never been observed experimentally. Besides, the requirement of ultralow temperatures limits the study of QHE without LLs. Efforts on high-Chern-number and high-temperature QHE without LLs are still highly desired for exploring emergent physics and low-power-consumption electronics [10].

Here we report the first experimental discovery of the high-Chern-number QHE without LLs above 10 K and $C = 1$ QHE without LLs above the Néel temperature ($T_N$) in MnBi$_2$Te$_4$ devices. We show that when modulated into the insulating regime by a small back gate voltage, the nine-layer and ten-layer MnBi$_2$Te$_4$ devices can be driven to Chern insulator with $C = 2$ at moderate perpendicular magnetic field. Quantized Hall resistance $h/2e^2$ accompanied by vanishing longitudinal resistance with the temperature as high as 13 K is observed in the ten-layer device. When reducing the thickness of the devices down to eight layer and seven layer, a quantized Hall resistance plateau $h/e^2$ is detected at a temperature much higher than the Néel temperature of the devices. This quantization temperature is the highest record in systems showing QHE without LLs. Our discoveries break new ground in the exploration of topological quantum states and provide a platform for potential applications in related low-consumption electronics.

MnBi$_2$Te$_4$ is a layered material which can be viewed as a layer of Bi$_2$Te$_3$ TI intercalated with an additional Mn-Te layer [11–20]. This material exhibits ferromagnetic (FM) order within septuple layer (SL) and anti-ferromagnetic (AFM) order between neighboring SLs with an out-of-plane easy axis [11], as displayed in Fig. 1a. By tuning the magnetic structure through thickness or magnetic field, exotic topological states, such as type-I topological Weyl semimetal (WSM) in 3D, Chern insulator in 2D and higher-order topological Móbius insulator, can be realized in MnBi$_2$Te$_4$ [21,22]. In this work, the MnBi$_2$Te$_4$ flakes were mechanically exfoliated from high-quality MnBi$_2$Te$_4$ single crystals. These flakes were then transferred to 300 nm-thick SiO$_2$/Si substrates and the standard e-beam lithography followed by e-beam evaporation was used to fabricate electrodes. The doped Si served as the back gate and a back gate voltage applied between Si and the sample could modulate the sample into insulating regime. The magnetic field is perpendicular to the samples throughout the text.
However, when MnBi$_2$Te$_4$ is driven from AFM to FM states by external magnetic field, physical properties of the material change dramatically. While the interlayer coupling is restricted by the $PT$ (combination of inversion and time-reversal) symmetry in AFM MnBi$_2$Te$_4$ [11,21], it gets greatly enhanced in the FM state by $PT$ symmetry breaking, which generates more dispersive bands along the $\Gamma-Z$ direction than the AFM state (Fig. S9). Remarkably, the magnetic transition results in a topological phase transition from an AFM TI to a ferromagnetic Weyl semimetal in the bulk [11,12], leading to a physical scenario in which Chern insulators with $C > 1$ are designed [21,25–27]. Figure 1g shows the schematic FM order and electronic structure of the $C=2$ Chern insulator state with two chiral edge states across the band gap.

Figure 2 shows the temperature evolution of the high-Chern-number QHE without LLs with the $V_{bg} = -19$ V. As the temperature increases to 13 K, the height of the Hall resistance plateau stays above $0.97 \frac{h}{2e^2}$ and $R_{xx}$ remains below $0.026 \frac{h}{2e^2}$. With the temperature further increasing to 15 K, the value of the Hall resistance plateau reduces to $0.964 \frac{h}{2e^2}$ and $R_{xx}$ increases to $0.032 \frac{h}{2e^2}$. This working temperature of the high-Chern-number QHE without LLs is much higher than liquid helium temperature, which shows potential application of QHE in low-dissipation electronics. Furthermore, the high-Chern-number QHE
The height of Hall resistance plateau can reach 0.97 $R_h/e^2$ at 13 K.

**DISCUSSION**

A fundamental question is whether the observed quantized Hall resistance plateau is caused by Landau level quantization, as the ordinary QHE with LLs can also give rise to quantized Hall resistance plateaus and vanishing $R_{xx}$. We estimate the mobility values of our devices according to the slope of Hall resistance near zero magnetic field [18]. The mobility ranges from 100 to 300 cm$^2$ V$^{-1}$ s$^{-1}$, which are typically below the critical value for formation of LLs up to 15 T [28]. To further exclude the possibility of QHE with LLs, we performed controlled measurements by changing the carrier type. In general, the Chern number in ordinary QHE corresponds to the occupancy of LLs and the sign of the Chern number will change once the carrier type is switching. However, as shown in Fig. S6c and d, the carrier type in the device s4 (7-SL) with $C = 1$ is tuned from p to n when increasing the back gate voltage from 0 V to 99.5 V, while the sign of the Chern number does not change. Furthermore, for the $C = 2$ devices, the quantized $R_{yx}$ plateau in device s5 with n-type carriers (Fig. 1d) and s7 with p-type carriers (Fig. S4a) have the same sign. These observations unambiguously demonstrate that the observed quantized Hall resistance plateau has noth-
The nearly quantized Hall resistance plateau can stay at a temperature up to 45 K (Hall resistance plateau of 0.904 $\hbar/e^2$). (f) $B$-$T$ phase diagram of s3. The AFM state disappears at $T_\text{N} \sim 22.5$ K and the well-defined quantization can stay till 30 K (Hall resistance plateau of 0.967 $\hbar/e^2$).

\[
\sigma_{xy} = \frac{c_0}{2\pi} \int_{-\pi}^{\pi} \sigma_{xy}(k_z) \, dk_z = \left| \tilde{k}_W \right| e^2 / h, 
\]

where $c_0$ is the out-of-plane thickness of each SL, and $\tilde{k}_W = \left| k_W \right| c_0 / \pi$. For an N-layer FM thin film, its electronic states can be viewed as quantum-well states and possess a finite band gap due to quantum confinement. Generally, $\sigma_{xy}$ of thin films would grow with film thickness, as its ideal bulk contribution is $N \left| k_W \right| e^2 / h$. On the other hand, $\sigma_{xy}$ of 2D gapped films must take quantized values $C(N) e^2 / h$ as topologically required. Therefore, for thick films with minor surface effects, the thickness-dependent Chern number $C(N)$ would change discretely by 1 for every $\Delta N = 1 / \left| k_W \right|$, implying that high Chern number is feasible by increasing film thickness. The discrete increase of Chern number with increasing film thickness is a generic feature of ferromagnetic Weyl semimetals, which can also be understood by the topological band inversion picture as discussed in Methods.

The above physical picture is confirmed by the first-principles study, which gives $\tilde{k}_W = 0.256 \approx 1/4$ for the bulk and shows that $C(N)$ indeed increases by 1 for every $\Delta N = 4$ (Fig. 4b). Note that it is theoretically challenging to accurately predict $C(N)$, since the predicted $\tilde{k}_W$ depends sensitively on the exchange-correlational functional and the lattice structure. Based on the mBJ functional [29], we systematically tested the influence of lattice parameter $c_0$ on band structure and $C(N)$ (Fig. S9), and finally decided to use the experimental value $c_0 = 13.6$ Å. As shown in Fig. 4b, the 9-SL film is a high-Chern-number band insulator with $C = 2$. Compared to the AFM films studied before [11], band structure of the FM film displays much more pronounced quantum confinement effects, as visualized by significant band splitting between quantum well states (Fig. 4c). A quantum confinement induced gap $\sim 5$ meV is located at the $\Gamma$ point. The edge-state calculation reveals that there exist two chiral gapless edge channels within the gap (Fig. 4d), which confirms $C = 2$. Therefore, first-principles calculations indicate that high-Chern-number band insulators can be realized in the FM Weyl semimetal MnBi$_2$Te$_4$ by means of quantum confinement.

The theory suggests that the topological Chern number is tunable by controlling film thickness of FM MnBi$_2$Te$_4$. By reducing the film thickness to 7-SL, the Chern number decreases to $C = 1$, as found experimentally. The 8-SL is the marginal
CONCLUSION

In summary, we discovered high-Chern-number QHE ($C = 2$) without LLs showing two sets of dissipationless chiral edge states above 10 K and $C = 1$ Chern insulator state above the Néel temperature, which is also the highest temperature for QHE without LLs. Our findings open a new path for exploring the interaction between topology and magnetism, as well as the potential application of topological quantum states in low-power-consumption electronics at higher temperatures.

METHODS

Crystal growth

High-quality MnBi$_2$Te$_4$ single crystals were grown by directly reacting a stoichiometric mixture of high-purity Bi$_2$Te$_3$ and MnTe, which were prepared by reacting high-purity Bi (99.99%, Adamas) and Te (99.999%, Aladdin), and Mn (99.95%, Alfa Aesar) and Te (99.999%, Aladdin), respectively. The reagents were sealed in a silica ampoule under a dynamic vacuum, which was then heated to 973 K and slowly cooled down to 864 K, followed by the prolonged annealing at the same temperature over a month. The quality of mm-sized MnBi$_2$Te$_4$ crystals was examined on a PANalytical Empyrean X-ray diffractometer with Cu Kα radiation.

Devices fabrication

The MnBi$_2$Te$_4$ nanoflakes on 300 nm-thick SiO$_2$/Si substrate were mechanically exfoliated from high quality single crystals using Scotch tape. The substrates were pre-cleaned in oxygen plasma for five minutes with $\sim$ 60 mtorr pressure. To obtain flakes with thickness down to several nanometers, we heated the substrate after covering the Scotch tape at 393 K (120°C) for one minute. Standard electron beam lithography in a FEI Helios NanoLab 600i Dual Beam System was used to define electrodes after spin-coating PMMA resist. Then, metal electrodes (Ti/Au or Cr/Au, 65/180 nm) were deposited in a LJUHV E-400 L E-Beam Evaporator after Ar plasma cleaning.

Transport measurements

Electrical transport measurements were conducted in a 16T-Physical Property Measurement System (PPMS-16T) from Quantum Design with base temperature $T = 1.9$ K and magnetic field up to 16 T. Stanford Research Systems SR830 lock-in amplifiers were used to measure longitudinal resistance and Hall signals of the device with an AC bias current of 100 nA at a frequency of 3.777 Hz. The back gate voltages were applied by a Kethiley 2912A source meter.

First-principles calculations

First-principles calculations were performed in the framework of density functional theory (DFT) by
portmeasurements. and Jiawei Luo and Jiawei Zhang for helpful discussion in trans-

WethankPuYangandZeyanYangforhelpindevicefabrication,

ACKNOWLEDGEMENTS

NSR SUPPLEMENTARY DATA available from the authors upon reasonable request.

All data analyzed to evaluate the conclusions are

DATA AVAILABILITY STATEMENT

All data analyzed to evaluate the conclusions are available from the authors upon reasonable request.

SUPPLEMENTARY DATA

Supplementary data are available at NSR online.

ACKNOWLEDGEMENTS

We thank Pu Yang and Zeyan Yang for help in device fabrication, and Jiawei Luo and Jiawei Zhang for helpful discussion in transport measurements.

FUNDING

This work was supported by the National Key Research and Development Program of China (2018YFA0305600, 2017YFA0303300 and 2018YFA0307100), the National Natural Science Foundation of China (11888101, 11774008, 51788104, 11874035, 21975140 and U1832218), the Beijing Natural Science Foundation (2180010), and the Strategic Priority Research Program of Chinese Academy of Sciences (XDB28000000).

AUTHOR CONTRIBUTIONS

J.W. conceived and supervised the experiments. J.G. and Y.L. performed transport measurements. Y.X. and J.W. carried out theoretical calculations. H.L. and Y.W. grew the MnBi2Te4 bulk crystals. J.G. fabricated devices. J.G., Y.L., T.L. and J.W. analyzed the data. J.G., Y.L., J.L., Y.X. and J.W. wrote the manuscript with input from all authors.

Conflict of interest statement. None declared.

REFERENCES

1. Klitzing K, Dorda G and Pepper M. New method for high-

accuracy determination of the fine-structure constant based on quantized Hall resistance. Phys Rev Lett 1980; 45: 494–7.
2. Laughlin RB. Quantized Hall conductivity in two dimensions. Phys Rev B 1981; 23: 5632–3.
3. Thouless DJ, Kohmoto M and Nightingale MP et al. Quantized Hall conductance in a two-dimensional periodic potential. Phys Rev Lett 1982; 49: 405–8.
4. Aarón JE, Seiler R and Simon B. Homotopy and quantization in condensed matter physics. Phys Rev Lett 1983; 51: 51–3.
5. Niu Q, Thouless DJ and Wu YS. Quantized Hall conductance as a topological invariant. Phys Rev B 1985; 31: 3372–7.
6. Haldane FDM. Model for a quantum Hall effect without Landau levels: condensed-matter realization of the ‘parity anomaly’. Phys Rev Lett 1988; 61: 2015–8.
7. Onoda M and Nagaosa N. Quantized anomalous Hall effect in two-dimensional ferromagnets: quantum Hall effect in metals. Phys Rev Lett 2003; 90: 206601.
8. Yu R, Zhang W and Zhang HJ et al. Quantized anomalous Hall effect in magnetic topological insulators. Science 2010; 329: 61–4.
9. Chang CZ, Zhang JS and Feng X et al. Experimental observation of the quantum anomalous Hall effect in a magnetic topological insulator. Science 2013; 340: 167–70.
10. Barkeshli MM and Qi XL. Topological nematic states and non-

Abelian lattice dislocations. Phys Rev X 2012; 2: 031013.
11. Li J, Li Y and Du SQ et al. Intrinsic magnetic topological insulators in van der Waals layered MnBi2Te4-family materials. Sci Adv 2019; 5: eaaw5685.
12. Zhang DQ, Shi MJ and Zhu TS et al. Topological axion states in magnetic insulator MnBi2Te4 with the quantized magnetoelectric effect. Phys Rev Lett 2019; 122: 206401.
13. Otrokov MM, Rusinov IP and Blanco RM et al. Unique thickness-
dependent properties of the van der Waals interlayer antiferromagnet MnBi2Te4 films. Phys Rev Lett 2019; 122: 107202.
14. Gong Y, Guo JW and Li JH et al. Experimental realization of an intrinsic magnetic topological insulator. Chin Phys Lett 2019; 36: 076801.
15. Otrokov MM, Klimovskikh II and Bentmann H et al. Prediction and observation of an antiferromagnetic topological insulator. Nature 2019; 576: 416–22.
16. Lee SH, Zhu YL and Wang Y et al. Spin scattering and noncollinear spin structure-induced intrinsic anomalous Hall effect in antiferromagnetic topological insulator MnBi$_2$Te$_4$. Phys Rev Res 2019; 1: 012011.
17. Chen B, Fei FC and Zhang DQ et al. Searching the Mn(Sb,Bi)$_2$Te$_4$ family of materials for the ideal intrinsic magnetic topological insulator. Nat Commun 2019; 10: 4469.
18. Deng Y, Yu YJ and Shi MZ et al. Quantum anomalous Hall effect in intrinsic magnetic topological insulator MnBi$_2$Te$_4$. Science 2020; 367: 895–900.
19. Liu C, Wang YC and Li H et al. Robust axion insulator and Chern insulator phases in a two-dimensional antiferromagnetic topological insulator. Nat Mater 2020, doi:10.1038/s41563-019-0573-3.
20. Li H, Liu SS and Liu C et al. Antiferromagnetic topological insulator MnBi$_2$Te$_4$: synthesis and magnetic properties. Phys Chem Chem Phys 2020; 22: 556–63.
21. Li J, Wang C and Zhang ZT et al. Magnetically controllable topological quantum phase transitions in antiferromagnetic topological insulator MnBi$_2$Te$_4$. Phys Rev B 2019; 100: 121103.
22. Zhang RX, Wu F and Sarma SD. Möbius insulator and higher-order topology in MnBi$_2$Te$_{3n+1}$. Phys Rev Lett 2020; 124: 136407.
23. Mong RSK, Essin AM and Moore JE. Antiferromagnetic topological insulators. Phys Rev B 2010; 81: 245209.
24. Qi XL and Zhang SC. Topological insulators and superconductors. Rev Mod Phys 2011; 83: 1057–110.
25. Xu G, Weng HM and Wang ZJ et al. Chern semimetal and the quantized anomalous Hall effect in HgCr$_2$Se$_4$. Phys Rev Lett 2011; 107: 186806.
26. Wang J, Lian B and Zhang HJ et al. Quantum anomalous Hall effect with higher plateaus. Phys Rev Lett 2013; 111: 136801.
27. Zhang H, Freimuth F and Bihlmayer G et al. Engineering quantum anomalous Hall phases with orbital and spin degrees of freedom. Phys Rev B 2013; 87: 205132.
28. Shoenberg D. Magnetic Oscillations in Metals. Cambridge: Cambridge University Press, 1984.
29. Becketta AD and Johnson ER. A simple effective potential for exchange. J Chem Phys 2006; 124: 221101.
30. Kresse G and Furthmüller J. Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set. Phys Rev B 1996; 54: 11169–86.
31. Wu Q, Zhang S and Song HF et al. WannierTools: an open-source software package for novel topological materials. Comput Phys Commun 2018; 224: 405–16.
32. Lee DS, Kim TH and Park CH et al. Crystal structure, properties and nanostructuring of a new layered chalcogenide semiconductor, Bi$_2$MnTe$_6$. CrystEngComm 2013; 15: 5532–8.