Gate-tunable magnetoresistance in six-septuple-layer MnBi$_2$Te$_4$

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Abstract

The recently discovered antiferromagnetic topological insulator MnBi$_2$Te$_4$ hosts many exotic topological quantum phases such as the axion insulator and the Chern insulator. Here we report on systematic gate-voltage-dependent magneto-transport studies in six-septuple-layer MnBi$_2$Te$_4$. In the $p$-type carrier regime, we observe positive linear magnetoresistance (MR) when MnBi$_2$Te$_4$ is polarized in the ferromagnetic state by an out-of-plane magnetic field. Whereas in the $n$-type regime, distinct negative MR behaviors are observed. The behaviors of magnetoresistance in both regimes are highly robust against temperature up to the Néel temperature. Within the antiferromagnetic regime, the behavior of MR exhibits a transition from negative to positive under the control of gate voltage. The boundaries of the MR phase diagram can be explicitly marked by the gate-voltage-independent magnetic fields that characterize the processes of the spin-flop transition. The rich transport phenomena demonstrate the intricate interplay between topology, magnetism and dimensionality in MnBi$_2$Te$_4$.

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(Some figures may appear in color only in the online journal)

1. Introduction

Topological quantum materials such as topological insulators (TIs) [1, 2] and Weyl semimetals (WSMs) [3–5] can produce a lot of exotic topological quantum phenomena when there is interplay between the nontrivial topological band structure and other degrees of freedom. One outstanding example is the quantum anomalous Hall (QAH) effect that was first realized in magnetic doped TI films with ferromagnetic (FM) order, which is now called the Chern insulator [6–11] due to the absence of Landau levels. The axion insulator is
another topological state with nontrivial axion field $\theta$ in a bulk insulator [12–16]. Early experimental demonstrations of the axion insulator are mainly built on TI heterostructures with the top and bottom surface states gapped by opposite out-of-plane magnetization [17–19]. By applying an out-of-plane magnetic field, the axion insulator can be driven into the QAH insulator phase when the two surfaces are in the FM state.

Recently, the intrinsic magnetic TI, MnBi$_2$Te$_4$, has attracted a lot of attention in the topological matter community. It is found to be an A-type antiferromagnetic (AFM) TI below the Néel temperature $T_N$, of about 25 K [20–26]. A moderate magnetic field of about 5 T can drive it to a FM WSM [27]. When it is exfoliated into even-septuple-layer (even-SL) thin flakes, it is predicted and demonstrated to exhibit the axion insulator state in the AFM regime and the Chern insulator state in the FM regime [21, 22]. However, previous works on even-SL MnBi$_2$Te$_4$ mainly focus on the quantum phenomena with the Fermi level ($E_F$) tuned to the charge-neutral point (CNP). The gate voltage ($V_g$) dependence of magneto-transport and interplay between the band topology and magnetic order in conductive regimes have not been thoroughly investigated. Although the behavior of magnetoresistance (MR) in MnBi$_2$Te$_4$ has been reported by other recent works [28, 29], a detailed investigation of the $V_g$-dependent transport behaviors has been lacking due to the absence of quantization and insufficient gate tunability on thick samples. In addition, the WSM nature of FM MnBi$_2$Te$_4$ may further lead to exotic topological quantum phenomena, as demonstrated in other WSM systems, such as unsaturated linear MR [30–32] and chiral anomaly [30, 33, 34].

In this work, we fabricate a high-quality six-SL MnBi$_2$Te$_4$ device and measure the MR and Hall traces at different $V_g$s and temperatures ($T$s). Six-SLs is selected because it is an appropriate thickness for ensuring the nontrivial band topology and reducing the bulk carriers at the same time. In previous works, the axion insulator and Chern insulator behaviors have been demonstrated in a six-SL device [25]. Here we reproduce the axion insulator to Chern insulator transition in the insulating regime. Then the magneto-transport at different $V_g$s in the six-SL device is thoroughly investigated. In the FM state, a positive linear MR characterizing the nature of WSMs is observed until $T > T_N$ where the Hall traces indicate $p$-type transporte. In contrast, the slope of linear MR turns to negative in the $n$-type regime. While in the low-field AFM state, we observe a reversal from negative to positive MR along with the transition from the $p$-type to the $n$-type regime at $T < T_N$. The experimental phase diagram summarizes from the $V_g$-dependent MR is consistent with the origin of magnetic order in MnBi$_2$Te$_4$.

2. Methods

MnBi$_2$Te$_4$ single crystals are grown by the direct reaction of Bi$_2$Te$_3$ and MnTe with a ratio of 1:1 in a vacuum-sealed silica ampoule. The mixture is heated to 973 K and then cooled down to 864 K slowly. The quality of the MnBi$_2$Te$_4$ crystal and its precursors is examined by x-ray diffraction, energy dispersive x-ray (EDX) spectra and high-resolution x-ray photoelectron spectroscopy (XPS) spectra. The growth method and characterization are similar to those used in previous works [25, 26]. The MnBi$_2$Te$_4$ bulk crystal we use is about 2 mm in size. MnBi$_2$Te$_2$ flakes are exfoliated onto 285 nm thick SiO$_2$/Si substrates treated by air plasma and the cleaved thin flake is about 10 $\mu$m $\times$ 20 $\mu$m in size. The thickness of six-SL is determined by the optical contrast and atom force microscope measurement (see supplementary figure S1 (available online at stacks.iop.org/JPD/55/104001/mmedia)). The Hall-bar structure is fabricated by electron beam lithography followed by thermal evaporation of Cr/Au. All the device fabrication processes are carried out in an argon-filled glove box. Before transport measurement, the devices are all covered with 400 nm thick poly(methyl methacrylate). Transport measurements are performed in a commercial cryostat with the base temperature of 1.6 K and a magnetic field of up to 9 T. Standard four-probe lock-in techniques with low frequency (12.357 Hz) and an ac current of 200 nA are adopted. Both the longitudinal ($R_{xx}$) and Hall resistance ($R_{yx}$) are measured simultaneously. The $V_g$ was applied between the sample and the dielectric by a dc voltage source.

3. Results and discussion

The magnetic and crystal structures of six-SL MnBi$_2$Te$_4$ are shown in figure 1(a). At zero magnetic field, the Mn$^{2+}$ moments between neighboring SLs are antiferromagnetically aligned. As an out-of-plane magnetic field is applied, MnBi$_2$Te$_4$ is polarized to the FM state. We fabricate six-SL MnBi$_2$Te$_4$ into a standard six-probe Hall bar geometry and firstly measure the $R_{xx}$ and $R_{yx}$ at different $V_g$s at $T = 1.6$ K. Figures 1(b) and (c) show the variation of $R_{xx}$ and $R_{yx}$ with $V_g$ at $\mu_0H = 0$ and 9 T respectively. At $\mu_0H = 0$ T, a typical insulating behavior shows up at the CNP around $V_g = 30$ V as manifested by a large $R_{xx}$ over 150 k$\Omega$. In the $V_g$ range from 24 to 40 V, $E_F$ is tuned within the band gap and the system enters the axion insulator regime. When a magnetic field is applied, MnBi$_2$Te$_4$ is driven into the FM state, $R_{xx}$ exhibits a quantized plateau along with vanishing $R_{yx}$, which are hallmark of the Chern insulator state. The magnetic field driven axion-insulator-to-Chern-insulator transition is displayed in the fourth panel of figure 1(d). All these transport characters are consistent with previous reports [25, 35–37].

Then we focus on the magneto-transport in the regimes far away from the CNP. Figure 1(d) shows some representative behaviors of MR and Hall traces at different $V_g$s. For $V_g \ll 12$ V, $R_{xx}$ shows an overall $p$-type behavior in both AFM and FM states, indicating that $E_F$ lies in the valence band. In contrast, $R_{xx}$ shows the overall $n$-type behavior and $E_F$ lies in the conduction band for $V_g \geq 50$ V. Hall traces show two characteristic magnetic fields $H_{c1} \sim 2.3$ T and $H_{c2} \sim 5.2$ T, which correspond to the beginning and ending of the spin-flop process, respectively, as labeled in figure 1(d). These features can also be identified in the MR curves. Notably, the $R_{xx}$ curves show systematic and complex features with a varied
Figure 1. (a) Crystal and magnetic structure of six-SL MnBi$_2$Te$_4$ in the AFM and FM states. (b), (c) Gate-dependent $R_{xx}$ and $R_{yx}$ at $T = 1.6$ K at (b) $\mu_0 H = 0$ T and (c) $\mu_0 H = 9$ T. The CNP is about 30 V. (d) MR and Hall traces at different $V_g$s. Navy and magenta arrows mark the characteristic magnetic fields $H_{c1}$ and $H_{c2}$ respectively. Navy and magenta broken lines demote the quantized Hall and vanishing longitudinal resistance plateau, respectively.

magnetic field. In the FM state, $R_{xx}$ shows a positive linear MR in the $p$-type regime. While in the AFM state, the negative MR in the $p$-type regime evolves into a positive one in the $n$-type regime. The distinct behaviors in MR reflects the close relationship between the carrier type and magnetic structure, and the mechanism is the main focus of this work.

To further understand these MR behaviors, we perform magneto-transport on six-SL MnBi$_2$Te$_4$ at varied $T$s. In figures 2(a) and (b) we compare the $T$ dependent MR curves for $V_g = 0$ and 50 V. A field independent $R_{xx}$ is observed in the AFM state for $V_g = 0$ V, except for a weak peak at $\sim 0.2$ T (see supplementary figure S2 for details). The positive linear MR in the FM state persists up to 25 K which is above the $T_N$ of six-SL MnBi$_2$Te$_4$. As $T$ increases, $H_{c1}$ and $H_{c2}$ gradually decrease and drop to zero for $T \geq 20$ K so that the field range of linear MR increases systematically. In contrast to the positive MR at 0 V, the MR at 50 V in the FM state is negative.

To explain the opposite MR behavior in the $p$-type and $n$-type regimes, we investigate the origin of MR in the WSM phase and the asymmetry of electron and hole bands. Robust linear MR against magnetic field and temperature has been reported in many topological materials such as TIs [38–41] and WSMs [30–32, 42, 43]. And the WSM nature of the FM state of MnBi$_2$Te$_4$ is also predicted and then experimentally verified recently [21, 22, 36, 44]. In previous works the explanations of positive linear MR in WSMs are mainly based on the classical [45, 46] or quantum linear MR theory [47, 48]. Quantum theory predicts that MR is proportional to $1/n^2$, where $n$ is the carrier density. According to this model, linear MR occurs in the extreme quantum limit when only few Landau levels are occupied. Although the hole density is very low for $V_g = 0$ V due to the linear dispersion of the WSM state of MnBi$_2$Te$_4$ ($\sim 10^{12}$ cm$^{-2}$, shown in figure 2(c)), the magnetic field at which we observe linear MR is still far from the criterion of quantum limit $n \ll (eH/c\hbar)^{3/2}$ [48]. Furthermore, as plotted in figure 2(c), carrier density against $dMR/\mu_0 dH$ at different $T$s strongly deviates from the $1/n^2$ dependence predicted by the model. Here, $MR = (R_{xx}(\mu_0 H) - R_{xx}^0)/R_{xx}^0$, $R_{xx}^0$ is the intercept of the linear fitting of $R_{xx} - \mu_0 H$ in the FM state for $V_g = 0$ V and $R_{xx}$ at 0 T for other $V_g$s. Therefore, the quantum theory fails to explain the linear MR here. On the other hand, classical linear MR theory considering the mobility fluctuation predicts that the MR is proportional to the larger of average mobility ($\mu$) and the variance of mobility.
MR induced by the same mechanism can still occur. The bulk state hinders the perfect quantization but the negative scattering between the chiral edge state and the tric potential fluctuation induced by disorders, linear MR will occur \[ \mu(H) \]. When the carrier density is too low to screen the electric potential fluctuation induced by disorders, linear MR will occur \[ \mu(H) \]. As figure 2(c) shows, dMR/\mu_0 dH indeed increases with increasing mobility \( \mu \) though it slightly deviates from linear dependence. It is worth noting that the \( \mu \) we get from transport data is the average mobility and the linear relation of \( \mu \) only satisfies when \( \mu \) is larger than \( \Delta \mu \). A large fluctuation of mobility in MnBi\(_2\)Te\(_4\) may explain the deviation from linear dependence. But further experiments about the microscopic distribution of defect are needed to verify the model completely.

For \( V_g = 50 \) V, \( E_F \) is tuned out of the band gap and crosses the conduction band. However, the overall \( R_{xx} \) curve at \( V_g = 50 \) V is qualitatively the same as that at the CNP with \( V_g = 30 \) V except for the quantized transport. In the Chern insulator state of six-SL MnBi\(_2\)Te\(_4\) at the CNP, \( E_F \) is within the band gap and intersects with the dissipationless chiral edge state. Magnetic field can localize other dissipative conduction channels generated by thermal excitations and the negative MR occurs. The calculated band structure of few-layer MnBi\(_2\)Te\(_4\) in the FM state shows that the chiral edge state survives even when \( E_F \) lies beyond the band gap and coexists with the conduction band for a large energy range \[ 27, 36, 53 \]. The considerable scattering between the chiral edge state and bulk state hinders the perfect quantization but the negative MR induced by the same mechanism can still occur. The Weyl-band-induced positive MR is drowned by the chiral edge conduction, which is absent in the p-type regime. Asymmetric electron and hole bands in the FM state can explain the sign change in MR at different \( V_g \). And the metallic \( R_{xx} \) vs \( T \) relation in the FM state for \( V_g = 50 \) V is also consistent with this scenario.

Now we turn to the MR behavior in the AFM state. As figure 1(d) shows, when we set \( V_g = 12 \) V, it is still in the p-type regime. The positive linear MR in the FM state persists but a distinct negative linear MR in the AFM state emerges, which differs from that at 0 V. A MR peak at ~0.2 T is also observed. At higher \( T_s \), as shown in figure 3(a), the slope of MR decreases and the field range of negative MR shrinks. At \( T = 20 \) K and 25 K, the negative MR at low magnetic field vanishes and turns to positive in the whole field range. For clarity we only plot MR curves between \( \pm 3.5 \) T and the complete data from \(-9\) to \(9\) T is shown in supplementary figure S5. Interestingly, the MR curves for \( V_g \geq 30 \) V show the opposite behavior. At low \( T_s \), a positive MR is observed in the AFM state and the MR peak at \( V_g = 12 \) V turns into a dip. The slope of MR increases with increasing temperature up to 16 K, as shown in figure 3(b). At 20 K and 25 K, however, the slope of MR suddenly becomes negative. In figure 3(c), we display the temperature dependent dMR/\mu_0 dH. When \( T > T_N \), the AFM order vanishes and the MR shows the behavior like that in the FM state as discussed above.

At lower \( T_s \), the sign of MR and MR peak/dip are gate dependent, behaving as in Mn-doped Bi\(_2\)Te\(_{1−x}\)Se\(_x\) \[ 54 \]. The butterfly-shaped MR in the p-type regime is the conventional...
behavior associated with the suppression of spin scattering by magnetic field [24, 55]. And the linear MR seems like that in the system hosting electron–magnon scattering [56–60]. When the temperature is raised to above \( T_N \), the direction of local moments is random so that the spinwave is absent and the magnon-related negative MR vanishes. However, this behavior only emerges in a narrow gate range around \( V_g = 12 \text{V} \). The study of the excitation of the spin-wave and its gate dependence in \( \text{MnBi}_2\text{Te}_4 \) is desired to verify the scenario. When the \( E_F \) is tuned towards the CNP, the magnetic field tends to suppress the conductivity in the AFM state, turning the MR positive. We attribute this behavior to the emergence of a pair of helical hinge conducting channel in the axion insulator state, which is closely related to the AFM magnetic structure [61, 62]. When the magnetic field breaks the \( S \) symmetry in the axion insulator state, the axion field \( \theta \) deviates from the quantized value and the scattering between the helical conduction channels is enhanced. Similar MR behavior has been observed in many quantum spin Hall systems that host helical edge states protected by time-reversal symmetry [63, 64].

The competing mechanism between magnon-induced negative MR and edge-conduction-induced positive MR result in the temperature dependence of the slope of MR. The change in sign of MR in different regions can also be interpreted by this competing mechanism.

In figure 4(a), we summarise the \( \text{dMR/}\mu_0\text{dH} \) color map in the \( \mu_0\text{H–}V_g \) plane at \( 1.6 \text{K} \). There are four prominent regions \( I \sim IV \) with two clear phase boundaries \( H_{c1} \) and \( H_{c2} \). Regions I and III locate the \( p \)-type regime and II and IV cross the charge-neutral regime and the \( n \)-type regime. Below \( H_{c1} \), \( \text{dMR/}\mu_0\text{dH} \) changes from a negative value in region III to a positive one in region IV due to the competition mechanism between electron–magnon scattering and edge conduction in the axion insulator state, as discussed above. When the magnetic field is tuned above \( H_{c2} \), the Weyl-band-induced positive MR in region I changes into negative MR due to the emergence of the chiral edge state in region II. Two characteristic magnetic fields \( H_{c1} \) and \( H_{c2} \) at different \( V_g \) are both nearly constant. The carrier independent characteristic fields are consistent with the origin of magnetism in \( \text{MnBi}_2\text{Te}_4 \), that is, the superexchange interaction between Mn–Te–Mn [21, 22]. When the temperature is increased, \( H_{c1} \) and \( H_{c2} \) decrease and nearly vanish above 20 K, the \( T_N \) of six-SL \( \text{MnBi}_2\text{Te}_4 \), as shown in figure 4(b). We only plot the color map of \( \text{dMR/}\mu_0\text{dH} \) at \( V_g = 30 \text{V} \) here because the temperature evolutions of \( H_{c1} \) and \( H_{c2} \) are gate independent (see supplementary figure S4). The distinct AFM regime with a positive MR and FM regime with a weak negative MR are separated by the spin-flop regime at low \( T_s \) and merge into the paramagnetic regime above 20 K.

4. Conclusions

The gate dependent magneto-transport of six-SL \( \text{MnBi}_2\text{Te}_4 \) displays systematic and complex behaviors resulting from the interplay among the band topology, magnetic structure and scattering mechanism. Distinct MR behaviors are observed in different parameter ranges. In the AFM state, we observe negative MR in the \( p \)-type regime, while it becomes positive when \( E_F \) is tuned from the CNP to the conduction band. This behavior can be explained by the competition between the spin scattering mechanism and the edge conduction in the axion insulator state. In the FM state, due to the asymmetric band structure, we observe a reversal from positive linear MR in the \( p \)-type regime to a negative MR in the \( n \)-type regime. The characteristic magnetic fields labeling the boundary of the AFM, spin-flop progress and the FM state are independent.

Figure 4. (a) Color map of \( \text{dMR/}\mu_0\text{dH} \) at \( T = 1.6 \text{K} \) in \( \mu_0\text{H–}V_g \) plane. Regions I – IV labeled in the diagram represent the typical MR behaviors discussed in the text. \( H_{c1} \) and \( H_{c2} \) from the data in figure 1(d) plotted in navy and magenta broken lines denote the phase boundary. Black broken line marks the CNP with \( V_g = 30 \text{V} \). (b) Phase diagram of \( \text{dMR/}\mu_0\text{dH} \) at \( V_g = 30 \text{V} \) in \( T–\mu_0\text{H} \) plane. AFM, spin-flop and FM states are separated by the navy and magenta broken lines which show the temperature-dependent behaviors discussed in the text.
of $V_g$. When the temperature is increased to above $T_c$, AFM order disappears and all the critical magnetic fields are reduced to zero. With the presence of such rich MR phenomena, MnBi$_2$Te$_4$ serves as an ideal platform for exploring the interplay between band topology and intrinsic magnetism.

**Data availability statement**

All raw and derived data used to support the findings of this work are available from the authors on request.

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**Author contributions**

Y Y W and J S Z supervised the research. Y X L, C L and Y C W fabricated the devices and performed the transport measurements. H L and Y W grew the MnBi$_2$Te$_4$ crystals. J S Z, Y Y W, Y X L and C L prepared the manuscript with comments from all authors.

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