Charge carrier mediation and ferromagnetism induced in MnBi$_6$Te$_{10}$ magnetic topological insulators by antimony doping

Hangkai Xie$^{1,2}$, Fucong Fei$^{1,2,}$*, Fenzhen Fang$^{1,}$*, Bo Chen$^{1,2}$, Jingwen Guo$^{1,2}$, Yu Du$^{1,2}$, Wuyi Qi$^{1,2}$, Yufan Pei$^{1,2}$, Tianqi Wang$^{1,2}$, Muhammad Naveed$^{1,2}$, Shuai Zhang$^{1,2}$, Minhao Zhang$^{1,2}$, Xuefeng Wang$^{2,3}$, and Fengqi Song$^{1,2}$

$^1$National Laboratory of Solid State Microstructures, Collaborative Innovation Center of Advanced Microstructures, and College of Physics, Nanjing University, Nanjing 210093, People’s Republic of China
$^2$Atomic Manufacture Institute (AMI), Nanjing 211805, People’s Republic of China
$^3$National Laboratory of Solid State Microstructures, Collaborative Innovation Center of Advanced Microstructures, and School of Electronic Science and Engineering, Nanjing University, Nanjing 210093, People’s Republic of China

E-mail: feifucong@nju.edu.cn and fenzhen@nju.edu.cn

Received 21 May 2021, revised 1 November 2021
Accepted for publication 8 November 2021
Published 6 December 2021

Abstract
A new kind of intrinsic magnetic topological insulator (MTI) MnBi$_4$Te$_7$ family has shed light on the observation of novel topological quantum effects such as the quantum anomalous Hall effect (QAHE). However, strong anti-ferromagnetic (AFM) coupling and high carrier concentration in the bulk hinder practical applications. In closely related materials MnBi$_4$Te$_7$ and MnBi$_6$Te$_{10}$, the interlayer magnetic coupling is greatly suppressed by Bi$_2$Te$_3$ layer intercalation. However, AFM is still the ground state in these compounds. Here, by magnetic and transport measurements, we demonstrate that a Sb substitutional dopant plays a dual role in MnBi$_6$Te$_{10}$, which can not only adjust the charge carrier type and concentration, but also induces the solid into a ferromagnetic (FM) ground state. The AFM ground state region, which is also close to the charge neutral point, can be found in the phase diagram of Mn(Sb$_x$Bi$_{1-x}$)$_6$Te$_{10}$ when $x \sim 0.25$. An intrinsic FM-MTI candidate is thus demonstrated, which may take us a step closer to realizing a high-quality and high-temperature QAHE and related topological quantum effects in the future.

Supplementary material for this article is available online

Keywords: anomalous Hall effect, ferromagnetism, transport measurement, topological materials, topological insulator

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

1. Introduction

Novel topological quantum effects such as the quantum anomalous Hall effect (QAHE), the topological axion state, and the related topological electromagnetic effect can be achieved when combining magnetic property and topological insulators (TIs) [1–4]. However, from the first realization of the QAHE in Cr-doped (Bi,Sb)$_2$Te$_3$ in 2013 [5], the observation temperature is still extremely low, which is limited by extrinsic magnetic elemental dopants or the magnetic proximity heterostructures in magnetic topological insulators (MTIs) [6–9]. Recently, a
new class of MTI-MnBi$_2$Te$_4$(Bi$_x$Te$_{3-x}$)$_h$ family ($n = 0, 1, 2$) materials has been discovered that is proposed to hold intrinsic magnetic property instead of external magnetic impurities [10–16]. Because of the strong anti-ferromagnetic (AFM) coupling between MnBi$_2$Te$_4$ septuple layers (SLs), an experimentally observed QAHE in MnBi$_2$Te$_4$ needs the assistance of a large external magnetic field ($\sim 6$–$8$ T) to form a ferromagnetic (FM) state in MnBi$_2$Te$_4$ [17–20], or with an extremely high bias voltage up to 200 V in a five SLs MnBi$_2$Te$_4$ device under zero field [21]. Fortunately, the interlayer AFM exchange interaction can be significantly reduced by intercalating Bi$_x$Te$_{3-x}$ quintuple layers (QLs) into the MnBi$_2$Te$_4$ matrix [22–28]. The MnBi$_2$Te$_{7}$ ($n = 1$) can significantly reduce the AFM coupling [25, 29–31] and then the MnBi$_6$Te$_{10}$ ($n = 2$) can almost fully hold a FM state at zero field [22, 23, 28, 32], but the AFM state still possesses lower energy than the FM state as the ground state so that a small field is still essential to overcome the AFM coupling. Furthermore, in all of these intrinsic MTI candidates, MnBi$_2$Te$_{2n}$($\text{Bi}_2\text{Te}_3$)$_n$ ($n = 0, 1, 2$), the heavy n-type trivial bulk carriers will be induced because of the ineluctable antisite defects between the manganese and bismuth atoms, hindering the novel transport signatures contributed by the chiral edge state located in the exchange gap opening at the Dirac point of the surface states. Further applications of MnBi$_2$Te$_4$ and related intercalated materials in spintronics and dissipationless quantum devices are also obstructed.

In MnBi$_2$Te$_4$, Sb substitution at the Bi sites can neutralize the excess bulk carrier efficiently [33–35], while the AFM coupling is still robust. On the other hand, Sb substitution in MnBi$_4$Te$_7$ can tune the excess bulk carrier and adjust the magnetic interaction between MnBi$_2$Te$_4$ SLs at the same time [36, 37], but only a subtle FM state can be achieved when the Fermi level aligns with a van Hove singularity in the bulk conduction band [36], and no truly FM-MTI can be obtained. Fortunately, in the case of the MnBi$_6$Te$_{10}$ discussed in this work, the AFM coupling can be completely overcome and the bulk carrier concentration can be restrained at the same time by Sb substitution at the Bi sites, with no-magnetic elements doping, paving the way to demonstrating a real intrinsic FM-TI candidate for the QAHE.

Single-crystal serials of Mn(Sb$_{1-x}$Bi$_x$)$_{10}$Te$_{10}$ from $x = 0$ to $x = 0.32$ were grown. Then, magnetic and electrical transport measurements were conducted to explore the influence of the Sb substitution in MnBi$_6$Te$_{10}$. From the magnetic measurements, we find that the FM state takes the place of the AFM state as the ground state in Mn(Sb$_{1-x}$Bi$_x$)$_{10}$Te$_{10}$, even in a low level of Sb substitution when $x = 0.11$, implying that the AFM coupling between the SLs has been completely overcome, which is also consistent with a previous report about magnetic measurements in Sb-doped MnBi$_6$Te$_{10}$ [38]. Furthermore, according to the electrical transport, similar to the case of MnBi$_2$Te$_4$ and MnBi$_4$Te$_7$, one can realize that the Sb doping is also an effective approach to modulating the bulk carrier densities. Specifically, the heavy n-type bulk carriers in pure MnBi$_6$Te$_{10}$ can be significantly suppressed by Sb doping, and the charge neutral point (CNP) in the n–p transition process appears near $x \approx 0.25$, which is also in the FM state region with full net magnetization at zero field. Our work demonstrates the engineering of both the magnetic property and the charge carriers in MnBi$_6$Te$_{10}$ by Sb doping, and Mn(Sb$_{1-x}$Bi$_x$)$_{10}$Te$_{10}$ is a promising candidate for FM-TIs and low bulk carrier concentrations under certain doping ratios, which provides a prospective avenue to accomplish the QAHE at the zero field as well as other exotic topological effects.

2. Results

Mn(Sb$_{1-x}$Bi$_x$)$_{10}$Te$_{10}$ is a layered rhombohedral material with the space group $R 3 m$ [22, 28, 38, 39] stacked with one SL of Mn(Sb$_{1-x}$Bi$_x$)$_2$Te$_4$ and two QLSs of (Sb$_{1-x}$Bi$_x$)$_2$Te$_4$ alternately through the weak van der Waals forces along the $c$ axis, as seen in figure 1(a). The high-quality crystals of Mn(Sb$_{1-x}$Bi$_x$)$_{10}$Te$_{10}$ were obtained by the self-flux method [40]. The molar ratio of the reactants was around MnTe:(Sb$_2$Te$_3$ + Bi$_2$Te$_3$) $= 1:8$, then they were put in an alumina crucible and sealed by a quartz tube before being put into a muffle furnace. The temperature rose up to 950 $^\circ$C in 24 h and was maintained at this temperature for 6 h, then slowly cooled down to around 580 $^\circ$C, followed by centrifugation to separate the lustrous flakes from the excess flux. The detailed molar ratio of the reactants and centrifugation temperature for every different Sb-doped sample can be seen in table S1 in the supplementary material (available online at stacks.iop.org/JPD/55/104002/mmedia). The inset of figure 1(b) is the morphology image of the as-grown Mn(Sb$_{1-x}$Bi$_x$)$_{10}$Te$_{10}$ crystals. Figure 1(c) displays the energy dispersive spectra (EDS), which show that the stoichiometric ratio of Mn:Bi+Sb:Te is close to 1:6:10. The concrete composition ratio measured by EDS for every sample can be seen in table S1 in the supplementary material. The relative intensity of Bi at 3.8 keV and Sb at 3.6 keV evolve as expected after normalizing the counts with the Te peaks, elucidating the valid substitution of the two elements. The real ratio between Mn and Bi is a little deviated from the stoichiometric ratio, and this is believed to be the antisite defects of Mn and Bi(Sb) [41–43]. We then checked the single crystal x-ray diffraction (XRD) patterns for every Sb-doped sample, which show prominent peaks labeling (00n) Miller indexes in figure 1(b), and one can see that all the XRD patterns in figure 1(b) only show a pure MnBi$_6$Te$_{10}$ phase pattern without intermixing the other phase pattern from MnBi$_2$Te$_7$ or MnBi$_2$Te$_4$ or some other phases [22, 28, 38], which means the Sb element has entered into the MnBi$_6$Te$_{10}$ structure, introducing no other composition. The extracted value of the $c$ axis from the XRD pattern is nearly at $\sim 102$ Å with just a tiny change at a different Sb-doped ratio, as listed in table S1 in the supplementary material for every sample, indicating the similar structure constants and identical layered crystal structure for different Sb-doped Mn(Sb$_{1-x}$Bi$_x$)$_{10}$Te$_{10}$ samples [38]. Both the XRD and EDS results demonstrate the favorable crystallinity and the precise molar ratio during the procedure of the substitution in Mn(Sb$_{1-x}$Bi$_x$)$_{10}$Te$_{10}$.

To determine the magnetic properties in the Mn(Sb$_{1-x}$Bi$_x$)$_{10}$Te$_{10}$, magnetic measurements using a vibrating sample magnetometer equipped on a physical property
measurement system (Quantum Design PPMS-14T) were carried out. First we measured the field cool (FC) and zero-field cool (ZFC) processes from 300 K to 2 K for each different Sb-doped sample, as shown in figure 2(a). The magnetic field is along the c axis of the sample cleave surfaces with a magnitude at 500 Oe. In the pure MnBi$_2$Te$_4$, the FC-ZFC curves show clear, typical AFM peaks at 10.1 K and the overlapped ones diverge at a lower temperature (≈5 K), which corresponds with previous reports [22, 32]. The magnetic field dependence of the magnetization curves in figure 2(b) exhibit the zigzag shape and the hysteresis loop divides into two jump-points deviating from the zero field at around ±0.07 T obviously from 2 K to 8 K, which also indicates the existence of AFM coupling. Although the saturation field to approach the FM region in the MnBi$_6$Te$_{10}$ is just around 0.2 T, which is much smaller than the one in MnBi$_2$Te$_4$ (6–8 T), the magnetic ground state in MnBi$_6$Te$_{10}$ is still an AFM state and the external magnetic field is still essential to form a FM state.

The Bi$_2$Te$_3$ QLs intercalation enlarges the distance between the neighboring MnBi$_2$Te$_4$ SLs, and the energy difference between the AFM and FM becomes much smaller from MnBi$_2$Te$_4$ to MnBi$_4$Te$_7$ then to MnBi$_6$Te$_{10}$ [32]. Weak inter-layer AFM coupling paves the way for magnetic property modulation by various approaches. In addition, when substituting Bi with the no-magnetic Sb element in MnBi$_6$Te$_{10}$, the AFM ground state can be completely eliminated without introducing undesired outer magnetic impurities, and is then replaced by the FM ground state. As seen in figure 2(a), the FC-ZFC curves of all the Sb-doped samples show the λ shaped typical of FM properties. Then we can get the transition temperature by differentiating the magnetization versus temperature curves. It shows clear kinks around 12 K in all the Sb-doped samples, indicating the Curie temperature ($T_c$) [25], which is also slightly higher than the Néel temperature ($T_N$) in pure MnBi$_6$Te$_{10}$, as shown in the figure 2(a). Furthermore, the magnitudes of divergence between the FC-ZFC curves in the

Figure 1. Crystal growth and characterization of the Mn(Sb$_x$Bi$_{1-x}$)$_2$Te$_{10}$. (a) The crystal structures of the Mn(Sb$_x$Bi$_{1-x}$)$_2$Te$_{10}$/Sb$_x$Bi$_{1-x}$Te$_3$ superlattice. (b) Single crystal x-ray diffraction of the five samples. The curves are offset for clarity. Inset: optical images of the as-grown sample, the scale bar is 2 mm. (c) Energy dispersive spectra after normalization with the intensity of the Te element.
Figure 2. Magnetic measurement of the Mn(Sb\(_{x}\)Bi\(_{1-x}\))\(_6\)Te\(_{10}\). (a) FC-ZFC curves of the different Sb-doped ratio samples when the field is 500 Oe along the \(c\) axis, and the Néel temperature (\(T_N\)) or Curie temperature (\(T_C\)) for each sample. The curves are offset for clarity. (b)–(g) The magnetic field dependence of the magnetization from 2 K to 15 K when the field is along the \(c\) axis.

Sb-doped samples at lower temperatures (~5 K) rise much larger than the divergence value in the pure sample with an AFM transition point at 10.1 K, proving the observable enhancement of the FM ingredient compared with pure MnBi\(_6\)Te\(_{10}\). This implies that Sb substitution at the Bi site in the MnBi\(_6\)Te\(_{10}\) is an effective way to modulate the magnetic interaction between SLs from the AFM to the FM state.

To further explore the evolution of magnetic property of the various Sb doping ratios, magnetization versus magnetic field (\(M-H\)) measurements were conducted, shown in figures 2(b)–(g). In pure MnBi\(_6\)Te\(_{10}\), one can see the feature of zigzag-shaped hysteresis loops at different temperatures, which evolve to the inclined rectangle-shaped loops at around ±0.07 T in figure 2(b). Of course, one can notice at 2 K the zigzag-shaped hysteresis loops merge to be one hysteresis loop to a large extent, confirming that the interlayer AFM exchange coupling between MnBi\(_2\)Te\(_4\) SLs is significantly weakened by the intercalating Bi\(_2\)Te\(_3\) QLs, but there is still the main domination of the AFM feature in MnBi\(_6\)Te\(_{10}\), especially at higher temperatures above 2 K. On the other hand, even at a low level of Sb doping (\(x = 0.11\)), the zigzag shape and the hysteresis loop separation in the \(M-H\) curves are completely gone, as is shown in the figure 2(c). The hysteresis loop demonstrates one pure large inclined rectangle-shaped loop with a fully polarized FM state stabilized at zero fields at 2 K, which is beneficial to realizing the QAHE under zero field. In addition, when increasing the temperature, the loop does not split into two separated parts with a zigzag-shaped curve but simply shrinks and is always pinned at the zero field. Along with the Sb doping ratio increasing continuously, the FM state at Mn(Sb\(_{1-x}\)Bi\(_x\))\(_6\)Te\(_{10}\) still holds, and all of them are capable of holding a fully polarized FM state at zero fields at 2 K from \(x = 0.11\) to \(x = 0.25\), as shown in figures 2(c)–(f). When \(x\) rises up to around 0.32, the FM state cannot be fully maintained at zero fields even at 2 K, as is shown in figure 2(g).

Although the magnetic transition temperature increases with Sb doping, the coercive field drops down to around 300 Oe or even less for the Sb-doped samples. This may be explained by the effect of magnetic anisotropy caused by the introduction of Sb element impurities, or some other different type of magnetic interaction appears, which can compete with the single FM interaction between the SLs [38]. Anyway, the results suggest that the good FM state region can be held at a Sb-doping level range from \(x = 0.11\) to \(x = 0.25\) in Mn(Sb\(_{1-x}\)Bi\(_x\))\(_6\)Te\(_{10}\). This kind of hysteresis behavior demonstrates a typical FM ordering after Sb doping in MnBi\(_6\)Te\(_{10}\), consistent with the FC-ZFC behavior discussed above. It can be believed that, after Sb doping, the Mn-Sb site mixing where Mn occupies the Sb site probably mediates an FM coupling between Mn layers in the MnBi\(_2\)Te\(_4\) family as shown in some recent researches, which leads to the changing of energy between the AFM and FM states [35, 44–47].

Accompanied by engineering of the magnetic property, Sb doping can also modify the level of carrier density in MnBi\(_6\)Te\(_{10}\). Due to the internal defects of the Mn and Bi antisites, the as-grown MnBi\(_2\)Te\(_4\)(Bi\(_2\)Te\(_3\))\(_n\) (\(n = 0, 1, 2\) family
are all serious n-type doped regardless of the growth methods [12, 42], which is also consistent with previous angle resolved photoemission spectroscopy measurement reports [15, 16, 23, 25, 28, 48, 49, 50]. Heavy carriers in bulk will be a big obstacle for the capacity of regulating gate voltage in ultra-thin films in the device-running process. Fortunately, no-magnetic element doping is an efficient way to adjust the Fermi level of the samples without destroying the topological property and the carrier mobility according to several previous reports of these related materials [33–35, 51, 52].

We performed electrical transport measurements in MnBi$_6$Te$_{10}$ after Sb doping including magneto and Hall resistivity measurements. As shown in figure 3(a), the resistivity versus temperature curves display characteristic kinks marked by arrows at around 12 K in Mn(Sb$_x$Bi$_{1-x}$)$_6$Te$_{10}$ for all different Sb-doping concentrations, and the kink of the $x = 0$ sample is around 10 K, which is consistent with the magnetic transition temperature obtained from the magnetic measurement in figure 2(a). It is also noticeable that the resistivity firstly rises when the Sb doping ratio is increased, and reaches a maximum when $x = 0.25$. When further increasing the value of $x$ above 0.25, the resistivity drops. Distinct to the metallic behavior in other samples, the sample with $x = 0.25$ also seems to show a semiconductor behavior with a slight increase of resistivity when the temperature drops to around 200 K. Thus, it suggests that the carrier concentration is suppressed and exhibits properties similar to semiconductors, which also indicates that the Fermi level is close to the n–p transition point in the MnBi$_6$Te$_{10}$ sample.

One can then pay attention to the longitudinal and Hall resistivity, as displayed in figures 3(b)–(e). The anomalous Hall effect (AHE) can be clearly seen in Sb-doped MnBi$_6$Te$_{10}$ samples. For pure MnBi$_6$Te$_{10}$, one can see the AHE hysteresis curves display zigzag-shaped loops and show two parts of the inclined rectangle-shaped loops at around $\pm 0.07$ T at 2 K in figure 3(b), as well as the butterfly-shaped hysteresis behavior in longitudinal resistivity as shown in figure 3(c). When the temperature rises up to 5 K, the inclined rectangle-shaped loops start to separate into two isolated small loops, again confirming the AFM ground state in pure MnBi$_6$Te$_{10}$. 

Figure 3. Electrical transport of the Mn(Sb$_x$Bi$_{1-x}$)$_6$Te$_{10}$. (a) Resistivity versus temperature from 2 K to 300 K with the obvious kinks marked by arrows. (b) Hall and anomalous Hall (after subtracting the background) signal at selected ratios: $x = 0.00, 0.11, 0.20,$ and 0.32. (c) Magneto resistivity signal at selected ratios: $x = 0.00, 0.11, 0.20,$ and 0.32. (d) Hall signal in the $x = 0.25$ Sb-doped sample. (e) Magneto resistivity signal in the $x = 0.25$ Sb-doped sample.
Then let us shift the sight to the AHE signal in the Sb-doped samples. Consistent with the magnetic measurements, the AHE signal shows typical FM-type shaped loops with a fully polarized FM state stabilized at zero fields at 2 K. The loop simply shrinks and still performs as a form of FM state as the temperature goes up until TC is from x = 0.11 to x = 0.20, as shown in figure 3(b). In the meantime, the longitudinal resistivity shows clear butterfly curves at 2 K in figure 3(c), which further confirms the net magnetization emerging near zero fields. It is clear that the AHE signal hysteresis is always maintained when x = 0.11 and x = 0.2. When the Sb doping concentration is up to 0.25, the hysteresis loop cannot be clearly seen and the Hall signal shows round, S-shaped curves, which implies the Fermi level in the Sb-doped sample with x = 0.25 is close to the Dirac point, so the Hall measurement demonstrates a two-band feature, as is shown in figure 3(d). When x rises up to 0.32, the FM-type AHE emerges again, with the hysteresis loops shrinking compared with previous lower Sb-doped samples, and the fully AHE signal at zero field cannot be held at 2 K, the same as the magnetic measurement in figure 2(g).

It is also worth noticing that the Hall signal completely changes its sign from negative to positive, which is similar to the MnBi2Te4 series reported in previous studies, and can be attributed to the competition of intrinsic Berry curvature and extrinsic skew scattering [53, 54], as is shown in figure 3(b). This also explains the disappearing AHE signal hysteresis in the Mn(Sb0.25Bi0.75)6Te10 sample: the two opposite AHE signals from two distinct origins coincidently cancel each other out.

One can then focus on the carrier densities by extracting the Hall coefficient according to the slope of the Hall measurements up to 9 T at 2 K for every Sb-doped sample, as seen in figure S1 in the supplementary material. In pure MnBi6Te10, there is an n-type carrier with a high concentration at around 2.68 × 10^20 cm^-3 at 2 K, similar to MnBi2Te7 and MnBi2Te4 with a high n-type carrier concentration, which can also show the metallic properties [22, 25, 33, 55–57]. After Sb doping, the carrier concentration is about 2.58 × 10^20 cm^-3 when x = 0.11 for Sb doping at 2 K, almost the same with the pure MnBi6Te10. Then, when x goes up to 0.17, the concentration is a little lower at around 2.07 × 10^20 cm^-3, but still has a high bulk carrier density. When x = 0.2 for Sb doping, one can notice that the carrier concentration has dropped to around 8.07 × 10^19 cm^-3, much smaller than the pure sample, and it is still an n-type carrier in bulk. As for the sample with x = 0.32, the carrier concentration is about 5.3 × 10^19 cm^-3. More importantly, the symbol of the Hall coefficient changes from negative to positive and the bulk carrier is a p-type now, which means that the n–p transition occurs in this region, corresponding to the behavior in the Hall signal in the x = 0.25 Mn(Sb0.1 Bi0.9)6Te10 sample, as shown in figure 3(d). Here we use the classical two-band magneto transport model [58] to fit the Hall data at 2 K in the x = 0.25 Sb-doped sample to extract two kinds of carrier concentration, as seen in figure S2 in the supplementary material. The Hall resistivity can be described as

\[
\rho_{xy} = \frac{(p_0 \mu_n^2 - n_0 \mu_p^2)B + \mu_n^2(B - n_0 \mu_p^2)(p - n)B^2}{\mu_n^2 + (p_0 \mu_n + \mu_n^2)(p - n)^2 \mu_n^2 B^2}
\]

where n (p) and \( \mu_n (\mu_p) \) are the carrier density and mobility for electrons (holes), respectively, and B is the magnetic field. Then we get 6.1 × 10^19 cm^-3 and 2.0 × 10^19 cm^-3 for n-type and p-type carrier density, respectively, and both of them are much lower than the carrier density in the pure MnBi6Te10 sample and other Sb-doped samples, proving again that the Fermi level in the x = 0.25 sample is around the Dirac point and Sb element substitution at the Bi site in the MnBi6Te10 is an effective way to modulate the chemical potential.

Furthermore, let us pay more attention to the magneto transport behavior of the x = 0.25 Sb-doped sample. The longitudinal resistivity also shows clear butterfly curves, displayed in figure 3(e), the same as the other Sb-doped MnBi6Te10, indicating that the spontaneous magnetization with the FM ground state is also maintained in Mn(Sb0.25Bi0.75)6Te10, which also corresponds to the magnetic property in Mn(Sb0.25Bi0.75)6Te10 that we measured above in figure 2(f). We also measured the transport signals for some other Sb-doped MnBi6Te10 samples around x = 0.25 for further step verification. The semiconductor-like behaviors are repeatable, and the carrier densities are also in a very low level compared to other different Sb ratio samples, as seen in figures S3–S5 in the supplementary material. Contrary to the stubborn AFM state in the Sb-doped MnBi2Te4 [33] and the complex competition between the AFM and FM states in the Sb-doped MnBi4Te7, it is apparent that the FM ground state can be approached accompanied with the successful modulation of carrier densities in Sb-doped MnBi6Te10 samples.

To clearly elucidate the properties of the magnetism and the electrical transport during the procedure of Sb doping in MnBi6Te10, we summarize the magnetic transition temperature and the charge carrier concentrations of the Mn(Sb, Bi1−x)6Te10 samples with various Sb doping ratios in the diagram displayed in figure 4. In pure MnBi6Te10, the n-type carrier concentration is high and totally in an AFM state region with the Néel temperature at 10.1 K. As the Sb doping ratio increases, Mn(Sb, Bi1−x)6Te10 quickly takes a step into the FM ground state region and there is a slight rise to around 12 K for the magnetic transition point. In the meantime, the bulk carrier density keeps falling continuously. More importantly, the n-type carrier concentration is significantly reduced at the Sb doping ratio of 20%, and the bulk carrier changes its sign, being a p-type at 32% ratio, implying that the CNP is in this Sb ratio region. This can be explained by the fact that the Sb vacancies or Sb-on-Te antisites (SbTe) are incrementally introduced into Mn(Sb, Bi1−x)6Te10 [59–61], then the n-type carrier is suppressed by Sb element substitution at the Bi sites until x ≈ 0.25. After the Sb doping ratio increases to around 0.32, the Sb vacancies or SbTe antisites take a remarkable effect for the p-type background. Furthermore, from the phase diagram, one can clearly see that the CNP takes place deeply in the FM state region, colored by the cyan background, ensuring strong spontaneous magnetization.
with parallel magnetic moment arrangement in the bulk carrier suppressed Mn(Sb,Bi$_{1-x}$)$_6$Te$_{10}$ with x around 0.25. This is also confirmed by our measurement combined with the magnetization and electrical measurement of the x = 0.25 Sb-doped sample, which combine both n-type and p-type carriers near the Dirac point, and two types of carrier densities are both restrained at a relatively low level. Then we can demonstrate the real intrinsic FM-TI in Mn(Sb,Bi$_{1-x}$)$_6$Te$_{10}$, especially when x = 0.25, with the Fermi level close to the Dirac point, which provides a promising platform to explore and realize the QAHE and other topological quantum effects.

3. Conclusion

In summary, the magnetic property and carrier density have been modulated simultaneously in Mn(Sb,Bi$_{1-x}$)$_6$Te$_{10}$. The AFM ground state has evolved into an FM state after Sb doping, and is always kept in the FM state region at different doping ratios. The AFM coupling between the SLs has been completely suppressed. Besides, the dominated charge carriers can be modified from n-type to p-type effectively, and the CNP with greatly suppressed bulk carrier concentrations can be achieved in the sample with a Sb ratio around x = 0.25. Meanwhile, these samples close to the CNP also hold the FM ground state at zero external field, demonstrating a promising candidate for MTIs with a FM ground state and lower bulk carrier concentrations. The intriguing magnetic and charge carrier transitions induced by Sb dopants also provide an excellent avenue to study the interaction coupling between the magnetism and carriers in MTIs. And the most important thing is that Mn(Sb,Bi$_{1-x}$)$_6$Te$_{10}$ displays improved performance on both magnetic property and electrical transport than the pure MnBi$_2$Te$_6$/Bi$_2$Te$_3$$_n$ series, shedding light on the exploration and realization of the long-expected QAHE and related quantum topological effects.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

Acknowledgments

The authors gratefully acknowledge the financial support of the National Key R&D Program of China (2017YFA0303203); the National Natural Science Foundation of China (12025404, 91622115, 11522432, 11574217, U1732273, U1732159, 61822403, 11874203, 11904165, and 11904166); the Natural Science Foundation of Jiangsu Province (BK20190286); the Fundamental Research Funds for the Central Universities (020414380150, 020414380151,
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