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Ferromagnetism of magnetically doped topological insulators in $\text{Cr}_x\text{Bi}_{2-x}\text{Te}_3$ thin films

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We investigated the effect of magnetic doping on magnetic and transport properties of $\text{Bi}_2\text{Te}_3$ thin films. $\text{Cr}_x\text{Bi}_{2-x}\text{Te}_3$ thin films with $x = 0.03$, 0.14, and 0.29 were grown epitaxially on mica substrate with low surface roughness ($\sim 0.4$ nm). It is found that Cr is an electron acceptor in $\text{Bi}_2\text{Te}_3$ and increases the magnetization of $\text{Cr}_x\text{Bi}_{2-x}\text{Te}_3$. When $x = 0.14$ and 0.29, ferromagnetism appears in $\text{Cr}_x\text{Bi}_{2-x}\text{Te}_3$ thin films, where anomalous Hall effect and weak localization of magnetoconductance were observed. The Curie temperature, coercivity, and remnant Hall resistance of thin films increase with increasing Cr concentration. The Arrott-Noakes plot demonstrates that the critical mechanism of the ferromagnetism can be described better with 3D-Heisenberg model than with mean field model. Our work may benefit for the practical applications of magnetic topological insulators in spintronics and magnetoelectric devices. © 2015 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4918560]

I. INTRODUCTION

Topological insulators (TIs) with dispersionless surface state attract attentions both for fundamental science and potential applications. The reason of this nontrivial surface state is their bulk band inversion and massless Dirac-cone-like surface state arising from the strong spin-orbit coupling.\cite{1} This surface state is protected by the time reversal symmetry which prohibits the backscattering on non-magnetic impurities and induces a weak antilocalization of Dirac fermions.\cite{2,2}

The surface state of TIs protected by time reversal symmetry can be broken by a proximity effect with interfacing an insulating ferromagnetic material or introducing magnetic impurities.\cite{3,4} Consequently, magnetic impurities in TIs can cause the opening of the surface band gap which can induce phenomena such as the quantized anomalous Hall effect and magnetoelectric effect.\cite{5,6} Due to the magnetic doping, it is shown that a weak localization (WL) behavior emerges and competes with the weak antilocalization effect (WAL).\cite{7}

Theoretically, introducing of transition metal ions can lead to ferromagnetism by either Van Vleck mechanism or Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange mechanism.\cite{4,8,9} However, the critical property of ferromagnetism in magnetic TIs is not well studied. Moreover, introducing magnetic impurities may roughen the surface of thin films and result in an increasing of carrier concentration, which prohibits the appearance of quantized anomalous Hall state.\cite{10} Thus, to realize practical applications of the nontrivial state in TIs, extensive understanding of magnetically doped TIs is still required.

In this work, we report the effect of magnetic impurity on manipulating the magnetic and transport response of $\text{Cr}_x\text{Bi}_{2-x}\text{Te}_3$ thin films with low surface roughness. We will show that Cr atoms increase the magnetization and establish ferromagnetism in $\text{Cr}_x\text{Bi}_{2-x}\text{Te}_3$ thin films. Moreover, the Arrott and Arrott-Noakes plot\cite{11,12} for $\text{Cr}_x\text{Bi}_{2-x}\text{Te}_3$ indicate that the critical mechanism of ferromagnetism in $\text{Cr}_x\text{Bi}_{2-x}\text{Te}_3$ can be described better with 3D-Heisenberg model than mean field model.

II. EXPERIMENTS

$\text{Cr}_x\text{Bi}_{2-x}\text{Te}_3$ thin films were grown on mica substrate using molecular beam epitaxy with base pressure as $\sim 5 \times 10^{-10}$ Torr. The temperature of Bi and Te source materials was adjusted to obtain a Te-rich environment. The growth rate was kept as 0.3 nm/min. The surface topography of the thin films was characterized by atomic force microscope (AFM). Root mean square (RMS) surface roughness was then quantitatively calculated. Thin films with thickness of 12 nm were deposited. The thickness was determined by measuring the height of a scratch made carefully on thin films with AFM in tapping mode.\cite{13} The composition analysis was carried out at the FEI Quanta FE-SEM. The magnetic property measurement was performed using a SQUID magnetometer. The thin film was fabricated into 1 mm wide Hall bar geometry by reactive ion etching. The transverse and longitudinal resistances under applied magnetic field were measured with physical properties measurement system. The applied AC current was 0.01 mA, with the frequency as 19 Hz.

III. RESULTS AND DISCUSSION

A. Structural characterization of $\text{Cr}_x\text{Bi}_{2-x}\text{Te}_3$ thin films

The surface morphology of $\text{Cr}_x\text{Bi}_{2-x}\text{Te}_3$ with $x = 0.00$, 0.14, and 0.29 are shown in Fig. 1(a). Without doping Cr, the $\text{Bi}_2\text{Te}_3$ shows large scale terraces. The height of a terrace is $\sim 1$ nm (i.e., one quintuple layer of $\text{Bi}_2\text{Te}_3$). When $x = 0.14$, the $\text{Bi}_2\text{Te}_3$ shows...
and 0.29, large scale terraces cannot be found. The surface morphology of thin films changes with an obvious reduction of terrace width, which may due to the competition between Bi atoms and Cr atoms at Bi site. Although the width of terraces decreases with doping Cr, layer-by-layer epitaxial growth of thin films remains. Based on the AFM images, we calculated the surface RMS roughness of thin films. As Fig. 1(b) shows, a low surface roughness for all samples (~0.4 nm) is found. The roughening of the flat surface with doping Cr reported previously is not observed in the present thin films, which may result from a low growth rate here.

The crystal quality and orientation of thin films were studied by x-ray diffractometer (XRD). Figure 1(c) shows the XRD spectrum of Cr$_x$Bi$_{2-x}$Te$_3$ with $x = 0.29$. The mica substrate is characterized as {001} family diffraction peaks, indicating its cleavage plane as the c-axis. The diffraction peaks from family of the {003} planes of Cr$_x$Bi$_{2-x}$Te$_3$ are observed, which suggests the thin films preferentially align along their c-axis. From the diffraction pattern for Cr$_x$Bi$_{2-x}$Te$_3$ with $x = 0.29$, we determine that its c-axis lattice parameter is 30.59 Å, which is larger than that of Bi$_2$Te$_3$ (30.49 Å in c-axis lattice parameter with interlayer spacing as 2.56 Å). Because the ionic radius of Cr is smaller than that of Bi, the increase of c-axis lattice parameter indicates that Cr not only substitutes Bi atoms but also locates in the Van der Waals gaps between quintuple layers, which is similar to the doping mechanism of Cu in Bi$_2$Se$_3$.

### B. Magnetization of Cr$_x$Bi$_{2-x}$Te$_3$ thin films

To investigate the magnetic properties of Cr$_x$Bi$_{2-x}$Te$_3$, the out-of-plane magnetic moment of the thin films with surface area as 0.0789 cm$^2$ was measured. As Fig. 2(a) shows, when $x = 0.03$, the magnetization of the thin film keeps as negative during cooling, indicating that the Bi$_2$Te$_3$ with slightly Cr doping is still a diamagnetic material. When $x = 0.14$, the magnetization of the thin film increases. Its value becomes positive. Moreover, the magnetization of the thin film increases with cooling below ~25 K. The saturation of magnetization with further cooling is not observed. Cr$_x$Bi$_{2-x}$Te$_3$ with $x = 0.29$ shows higher magnetization in the whole temperature window. The magnetization of the thin film starts to increase below ~50 K. The increase of the magnetization with increasing Cr concentration indicates that the magnetic moment of the samples are easier to be polarized out-of-plane, which is similar to previous report on Fe-doped Bi$_2$Se$_3$ that the c-axis is the magnetic easy axis.

The $dM/dT$-temperature curves of samples are shown in Fig. 2(b). It is found that the temperature where the $dM/dT$ curve starts to deviate from linear one increases with increasing Cr concentration. Moreover, the $dM/dT \sim T$ curve for $x = 0.29$ shows a small peak at ~20 K, indicating that there may be a magnetic transition. However, due to the low net magnetic moment of the samples and the thin film effect, this peak is not measured as clearly as it should be in bulk ferromagnetic materials; and the behavior of magnetization above the peak temperature deviates from that of a paramagnet. As a result, the existence of ferromagnetism in Cr$_x$Bi$_{2-x}$Te$_3$ is difficult to be concluded here. In Sec. III C, we investigate the magnetic properties of Cr$_x$Bi$_{2-x}$Te$_3$ by measuring the magnetic field dependence of electrical transport properties.
C. Electrical transport and anomalous Hall effect of CrxBi2–xTe3 thin films

The Hall resistance and magnetoconductance (MC) of CrxBi2–xTe3 are shown in Fig. 3. As Fig. 3(b1) shows, when \( x = 0.03 \) the WAL behavior of MC cannot be observed. Instead, a WL behavior is shown at 2.5 K. By heating up to 7.5 K, the WL behavior changes into a classical parabolic dependence of magnetic field without observing the WAL, indicating the dominance of WL behavior. Moreover, when \( x = 0.03 \), Hall resistance does not show any hysteresis with magnetic field.

With increasing the Cr concentration, features of ferromagnetism are observed. As shown in Figs. 3(a2) and 3(a3), the Hall resistance of CrxBi2–xTe3 with \( x = 0.14 \) and 0.29 shows hysteretic behavior resulting from the anomalous Hall effect, suggesting an occurrence of magnetic ordering. As in regular ferromagnetism, the coercive field and remnant Hall resistance increase with cooling. With increasing Cr, higher coercive field and remnant Hall resistances are observed; the hysteresis of Hall resistance also vanishes at a higher temperature. It is indicated that with increasing Cr concentration, ferromagnetism in CrxBi2–xTe3 is stabilized; and enhanced anomalous Hall effect can be obtained, being consistent with the results in Fig. 2.

The MC of CrxBi2–xTe3 with \( x = 0.14 \) and 0.29 consistently show the appearance of ferromagnetism in thin films. As Figs. 3(b2) and 3(b3) show, the MC of CrxBi2–xTe3 with \( x = 0.14 \) and 0.29 shows WL behavior at 2.5 K as a non-monotonic increase of MC with increasing magnetic field. Moreover, the hysteresis loop of MC shows a butterfly shape, indicating the existence of ferromagnetism. It is verified by both MC and Hall resistance that ferromagnetism is established in CrxBi2–xTe3 with \( x = 0.14 \) and 0.29. To further confirm the ferromagnetism in thin films, it is also important to directly measure the hysteresis loop of magnetization with applied magnetic field. We will address this issue in our future works.

Comparing Figs. 3(a1)–3(a3), the sign of Hall coefficient changes with incorporating Cr in Bi2Te3. For CrxBi2–xTe3 with \( x = 0.03 \), Hall coefficient shows a n-type conductivity. When \( x = 0.14 \) and 0.29, the sign of Hall coefficient becomes positive demonstrating a p-type conductivity, indicating the Cr is an electron acceptor in Bi2Te3.

We extracted the carrier concentration using Hall coefficient by examining the Hall resistivity at large field where the relation between resistivity and field is linear, where \( e \) is the electron charge and \( n \) is the carrier concentration. As Fig. 3(a1) inset shows, carrier concentration of CrxBi2–xTe3 decreases with introducing Cr, indicating that Cr reduces free carriers of n-type Bi2Te3. The increasing of carrier concentration by doping Cr in samples with high surface roughness (~1 nm)10 is not observed in this work, suggesting that the growth of low surface roughness samples helps to reduce the bulk carrier concentration.

D. Critical property of the ferromagnetism in CrxBi2–xTe3 thin films

The critical property of the ferromagnetism in CrxBi2–xTe3 was studied by Arrott plot11 and Arrott-Noakes plot12 techniques, where the Curie temperature was also determined. The Arrott plots at various temperatures for CrxBi2–xTe3 \( x = 0.14 \) and 0.29 were calculated from Figs. 3(a2) and 3(a3). As Figs. 4(a) and 4(c) show, the \( R_{xy} \) is not

![FIG. 3. (a1)–(a3) Hall resistance curves at low magnetic field for CrxBi2–xTe3 with \( x = 0.03, 0.14, \) and 0.29, respectively. (b1)–(b3) Corresponding normalized magnetoconductance curves. The temperature dependent carrier concentration of CrxBi2–xTe3 is shown in (a1) inset.](image)

![FIG. 4. (a) and (c) Arrott plot at various temperatures for CrxBi2–xTe3 with \( x = 0.14 \) and 0.29 were calculated from Figs. 3(a2) and 3(a3). As Figs. 4(a) and 4(c) show, the \( R_{xy} \) is not](image)
linear related to $\mu_0 H/R_{xy}$ at high magnetic field, indicating the ferromagnetic transition in heavily doped Cr$_{x}$Bi$_{2-x}$Te$_3$ deviates from mean field model. Thus, it is difficult to extract Curie temperature from Arrott plot.

It was predicted that the ferromagnetism in TIs may result from RKKY interaction of magnetic impurities consisting of Heisenberg-like, Ising-like, and Dzyaloshinskii-Moriya-like terms. Thus, we calculated the Arrott-Noakes plot at various temperatures for Cr$_{x}$Bi$_{2-x}$Te$_3$ $x = 0.14$ and $0.29$ from Figs. 3(a2) and 3(a3) by assuming $\beta = 0.36$ and $\gamma = 1.386$ (3D-Heisenberg model). As Fig. 4(d) shows, the $R_{xy}^{1/\beta}$ remains almost linear with respect to $(\mu_0 H/R_{xy})^{1/\gamma}$ close to 22.5 K, indicating that the critical mechanism of ferromagnetism in Cr$_{x}$Bi$_{2-x}$Te$_3$ can be described better with 3D-Heisenberg model than with mean field model. Based on Arrott-Noakes plot, we extract the Curie temperature of Cr$_{x}$Bi$_{2-x}$Te$_3$ with $x = 0.14$ and $x = 0.29$ as $\sim 12.5$ K and $\sim 23.8$ K, being consistent with the magnetic measurement shown in Fig. 2.

IV. CONCLUSION

In this work, the effect of magnetic impurities on Cr$_{x}$Bi$_{2-x}$Te$_3$ thin films with $x = 0.03$, 0.14, and 0.29 was studied. Thin films with low surface roughness ($\sim 0.4$ nm) were deposited on mica substrate. We found that Cr is an electron acceptor which reduces the carrier concentration of Bi$_2$Te$_3$. Magnetization of thin films increases with increasing Cr concentration. The ferromagnetism in Cr$_{x}$Bi$_{2-x}$Te$_3$ appears at $x = 0.14$ and 0.29, where hysteretic anomalous Hall effect and weak localization of magnetoconductance are observed. Our work is useful for achieving the gapped surface state of TIs with magnetic doping and their practical applications in magnetoelectric devices.

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