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Magnetotransport study of (Sb$_{1-x}$Bi$_x$)$_2$Te$_3$ thin films on mica substrate for ideal topological insulator

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We deposited high quality (Sb$_{1-x}$Bi$_x$)$_2$Te$_3$ on mica substrate by molecular beam epitaxy and investigated their magnetotransport properties. It is found that the average surface roughness of thin films is lower than 2 nm. Moreover, a local maxima on the sheet resistance is obtained with $x = 0.043$, indicating a minimization of bulk conductivity at this composition. For (Sb$_{0.957}$Bi$_{0.043}$)$_2$Te$_3$, weak antilocalization with coefficient of -0.43 is observed, confirming the existence of 2D surface states. Moreover Shubnikov-de Hass oscillation behavior occurs under high magnetic field. The 2D carrier density is then determined as $0.81 \times 10^{16}$ m$^{-2}$, which is lower than that of most TIs reported previously, indicating that (Sb$_{0.957}$Bi$_{0.043}$)$_2$Te$_3$ is close to ideal TI composition of which the Dirac point and Fermi surface cross within the bulk bandgap. Our results thus demonstrate the best estimated composition for ideal TI is close to (Sb$_{0.957}$Bi$_{0.043}$)$_2$Te$_3$ and will be helpful for designing TI-based devices. © 2016 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License. [http://dx.doi.org/10.1063/1.4943156]

I. INTRODUCTION

Topological insulators (TIs), due to their dispersionless surface state, are promising candidates for nano-electronics, spintronics, and magnetic sensors. The reason for this nontrivial surface state is the bulk band inversion which arises from the strong spin-orbit coupling. This surface state of TIs protects the electrons from back scattering which may lead to ultra-high conductivity. Experimentally, the topological surface state has been found in 3D TI materials such as Bi$_2$Te$_3$ and Sb$_2$Te$_3$. To reduce the surface crystalline defect and obtain high bulk resistivity, TIs are grown on various substrates such as sapphire and Si (111). Among these substrates, muscovite mica which enables van der Waals epitaxy shows advantages in the growth of TI with low crystalline defect and good electrical properties. Moreover, the mechanical flexibility and transparency of mica enable the application of TIs in flexible optoelectronics.

The ideal TIs require both Dirac cone and Fermi level lying inside the bulk bandgap where all the electrical conductivities are attributed by the TI surface. However, TIs such as Bi$_2$Te$_3$ and Sb$_2$Te$_3$ are reported as n-type and p-type band structure respectively. Therefore, their band structures are required to be engineered to achieve ideal TI. Due to the structural compatibility of Bi$_2$Te$_3$ and Sb$_2$Te$_3$, it is possible to tune the Fermi level and Dirac cone by doping Bi into Sb$_2$Te$_3$ to make ideal TI. Moreover, since the band structure of TIs can be affected by the epitaxial constraint from the substrate, it is still necessary to study the growth of (Sb$_{1-x}$Bi$_x$)$_2$Te$_3$ on mica substrate and experimentally locate the ideal TI composition for the practical applications.

In this work, (Sb$_{1-x}$Bi$_x$)$_2$Te$_3$ thin films were deposited on mica substrate with varying Bi concentration, aiming to study their structural characters on mica substrate and locate the ideal TI.

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composition. The magnetotransport responses of (Sb$_{1-x}$Bi$_x$)$_2$Te$_3$ near ideal TI composition will be studied in detail to understand the electrical properties of its surface state.

II. EXPERIMENTAL PROCEDURE

(Sb$_{1-x}$Bi$_x$)$_2$Te$_3$ with thickness of 12 nm were deposited on mica substrate by molecular beam epitaxy. The quality of the thin films during growth was monitored with reflection high energy electron diffraction (RHEED). The concentration of Bi in (Sb$_{1-x}$Bi$_x$)$_2$Te$_3$ was modulated by the cell temperature of Bi. X-ray diffractometer (XRD) was used to characterize structural information of thin films. The surface morphology and thickness were characterized by atomic force microscope (AFM). The composition was analyzed with the FEI Quanta FE-SEM. Room temperature sheet resistance was performed using four-point probe method. The magnetotransport measurement was performed in physical properties measurement system with magnetic field normal to the sample surface [Fig. 4(a) inset].

III. RESULTS AND DISCUSSION

A. Structure of (Sb$_{1-x}$Bi$_x$)$_2$Te$_3$ deposited on mica substrate

Surface morphology of Sb$_2$Te$_3$ grown on mica is shown in Fig. 1(a). Over a large length scale (5 × 5 µm$^2$), the thin film possesses a terrace-like structure. Along the dashed line in Fig. 1(a), the surface shows stepwise increase of its height [Fig. 1(e)] with a value of ~ 1 nm (i.e. one quintuple layer) which indicates the 2D growth of thin films on mica substrate. With doping Bi into Sb$_2$Te$_3$ [Figs. 1(b), 1(c), and 1(d)], pyramidal-shape terraces gradually appear. The highest surface roughness is ~ 1.9 nm with $x = 0.07$, which is comparable to the surface roughness (~ 1 nm) of Bi$_2$Te$_3$ with similar thickness on Si (111) substrate, indicating that mica is a suitable substrate to grow (Sb$_{1-x}$Bi$_x$)$_2$Te$_3$.

Representative XRD profile of (Sb$_{1-x}$Bi$_x$)$_2$Te$_3$ with $x = 0.043$ is shown in Fig. 1(f). Diffraction peaks from the {003} family planes of (Sb$_{0.957}$Bi$_{0.043}$)$_2$Te$_3$ are observed. Its $c$ lattice parameter is 30.42 Å, being in between that of Sb$_2$Te$_3$ (30.35 Å) and Bi$_2$Te$_3$ (30.49 Å). This is consistent with larger atomic radius of Bi than Sb in (Sb$_{1-x}$Bi$_x$)$_2$Te$_3$. Such lattice distortion introduced by Bi may

![Figure 1](image-url)
also disturb the 2D growth of \((\text{Sb}_{1-x}\text{Bi}_x)\text{Te}_3\), and then increase the surface roughness as shown in Figs. 1(a) ~ 1(d).

B. Effect of Bi on sheet resistivity of \((\text{Sb}_{1-x}\text{Bi}_x)\text{Te}_3\)

To identify the ideal TI composition in \((\text{Sb}_{1-x}\text{Bi}_x)\text{Te}_3\) grown on mica substrate, sheet resistivity of thin films were studied. Based on previous results,\(^9\) for \(\text{Sb}_2\text{Te}_3\) [Fig. 2(a1)], the Dirac point (DP) locates in the bulk bandgap while the Fermi level (FL) lies in the valence band; while for \(\text{Bi}_2\text{Te}_3\) [Fig. 2(a3)], the DP is inside the valence band with FL cutting the conduction band. The band structure of \(\text{Sb}_2\text{Te}_3\) and \(\text{Bi}_2\text{Te}_3\) thus suggests a low value of their sheet resistivity. Doping of Bi gradually shifts up the FL and pushes down the DP.\(^9\) As a result, an ideal TI with FL and DP crossing within the bulk bandgap can be formed [Fig. 2(a2)]. With extremely reducing bulk carrier density, the \((\text{Sb}_{1-x}\text{Bi}_x)\text{Te}_3\) near ideal TI should show obvious increase in sheet resistivity.\(^9\) Moreover, the energy difference between DP and FL of \(\text{Sb}_2\text{Te}_3\) \((E_{DP} - E_{FL} \sim 0.05 \text{ eV})\) is much smaller than that of \(\text{Bi}_2\text{Te}_3\) \((E_{FL} - E_{DP} \sim 0.3 \text{ eV})\).\(^9\) Thus, the ideal TI composition is expected close to \(\text{Sb}_2\text{Te}_3\) terminal.

Based on above analysis, sheet resistance measurement was performed on \((\text{Sb}_{1-x}\text{Bi}_x)\text{Te}_3\) with \(x = 0.014, 0.043, \text{ and } 0.07\). As Fig. 2(b) shows, with doping Bi, a peak in sheet resistivity is observed in \((\text{Sb}_{1-x}\text{Bi}_x)\text{Te}_3\) when \(x = 0.043\). Such experimental observation is consistent with the schematic picture in Figs. 2(a1) ~ 2(a3), indicating that \((\text{Sb}_{1-x}\text{Bi}_x)\text{Te}_3\) with \(x = 0.043\) is close to ideal TI. This doping value is consistent with previous work suggesting the ideal TI composition as \(x = 0.04 \sim 0.06\),\(^9\) indicating that epitaxial constraint generated by mica substrate is weak on determining the composition of ideal TI in \((\text{Sb}_{1-x}\text{Bi}_x)\text{Te}_3\).

C. Weak antilocalization of \((\text{Sb}_{0.957}\text{Bi}_{0.043})\text{Te}_3\) under low magnetic field

To study the topological surface state near ideal TI composition, magnetotransport responses of \((\text{Sb}_{1-x}\text{Bi}_x)\text{Te}_3\) \((x = 0.043)\) at low temperature were studied under low magnetic field. At \(T = 2.5 \text{ K}\) [Fig. 3(a)], \(\Delta G_{xx}(B)\) shows sharp upward cusps shape near zero magnetic field, demonstrating a weak antilocalization effect (WAL).\(^10,13,14\) Such behavior originated from the suppression of deconstructive interference of \(\pi\) Berry’s phase with applying magnetic field provides a clear signature on the existence of topological surface state.\(^14\) At \(T = 7.5 \text{ K}\) and \(10 \text{ K}\), the WAL becomes weak indicating the decrease of phase coherence length with increasing thermal noise. The temperature dependent longitudinal resistivity \((\rho_{xx})\) curve [Fig. 3(a) inset] shows metallic (with positive slope) behavior, which indicates the existence of surface state carriers.\(^15\) However, below 20 K, the

![FIG. 2. (a1) ~ (a3) Schematic illustrations on tuning the Fermi level and surface Dirac point of \((\text{Sb}_{1-x}\text{Bi}_x)\text{Te}_3\) with increasing Bi concentration. (b) Sheet resistivity of \((\text{Sb}_{1-x}\text{Bi}_x)\text{Te}_3\) at room temperature.](image-url)
resistivity shows a weak upturn as temperature decreases. This insulating behavior is consistent with previous results and can be explained by electron-electron interactions.16

The WAL of TI can be described with Hikami-Larkin-Nagaoka (HLN) equation as

$$\Delta G(B) = -\left(\alpha e^2/\pi h\right) \left[\Psi(0.5 + h/8\pi L_\Phi^2) - \ln(h/8\pi L_\Phi^2)\right],$$

where $\alpha$ is WAL coefficient, $e$ is electron charge, $h$ is Planck’s constant, $\Psi$ is digamma function, and $L_\Phi$ is phase coherence length.17 The best fitting at various temperature [Fig. 3(b)] yields $\alpha = -0.43$, confirming the 2D character of WAL since theoretically $\alpha$ should equal to -0.5 for 2D surface state.18 Figure 3(c) shows that the $L_\Phi$ decreases from 97 nm to 53 nm with increasing temperature. The temperature dependence of $L_\Phi$ is further fitted with a power law $L_\Phi \sim T^\beta$ with $\beta = -0.42$. Since $\beta$ is equal to -1/2 for 2D electron system,19 the present WAL results confirm the existence of 2D topological surface state in $(Sb_{1-x}Bi_x)_{2}Te_3 \ (x = 0.043)$ deposited on mica substrate.

D. Quantum oscillation of (Sb$_{0.957}$Bi$_{0.043}$)$_2$Te$_3$ under high magnetic field

To study the surface state near ideal TI composition in detail, magnetotransport responses of (Sb$_{0.957}$Bi$_{0.043}$)$_2$Te$_3$ were further studied under high magnetic field. As shown in Fig. 4(a), Hall resistance of sample demonstrates obvious Shubnikov-de Hass (SdH) oscillation at 2.5 K.6 Moreover, the oscillation can be observed at small magnetic field, indicating the minimization of bulk carrier density in the system.6 The $dR_{xy}/dB$ shows periodical oscillations with $1/B$ [Fig. 4(a) inset]. The oscillation frequency is extracted by fast Fourier transform as $F_{SdH} = 33.3$ T. The oscillation frequency is related with the area of Fermi surface $A_F$ as $F_{SdH} = [h/4\pi^2e]A_F$. Assuming a circular area, the Fermi surface can be expressed as $A_F = \pi k_F^2$, where $k_F$ is Fermi vector.6,20 The 2D
carrier density $n_{2D}$ can then be determined as $n_{2D} = k_F^2/4\pi$.\textsuperscript{20} For (Sh$_{0.957}$Bi$_{0.043}$)$_2$Te$_3$, the Fermi vector and 2D carrier density are calculated as $k_F = 3.18 \times 10^8$ m$^{-1}$ and $n_{2D} = 0.81 \times 10^{16}$ m$^{-2}$. SdH oscillation is also observed in normalized magnetoconductance [Fig. 4(b)], of which the amplitude decreases with increasing temperature [Fig. 4(c)]. The temperature dependence of oscillation amplitude can be described by Lifshitz-Kosevich (LK) model as $\Delta \sigma_{xx}(T) = \sigma_{xx}(0) \cdot |T|/\sinh(|T|)$ and $\lambda(T) = 2\pi^2 k_B m_{cy}/(he^2)$ where $m_{cy}$ is cyclotron mass.\textsuperscript{8} The effective cyclotron mass is obtained as $m_{cy} = 0.086 m_e$ where $m_e$ is mass of free electron. Notably that two dimensional electron gas (2DEG) may appear around the bottom of the bulk conduction band of n-type TI (Bi$_2$Se$_3$) due to the bending of surface band.\textsuperscript{21} However, (Sh$_{0.957}$Bi$_{0.043}$)$_2$Te$_3$ shows p-type conductivity in Fig. 4(a). Thus the contribution of 2DEG on magnetotransport is not considered here.

Compared with the surface state reported previously (for Bi$_2$Te$_3$, $n_{2D} \sim 1.2 \times 10^{16}$ m$^{-2}$, $k_F \sim 4 \times 10^8$ m$^{-1}$, and $m_{cy} \sim 0.15 m_e$),\textsuperscript{10} the 2D carrier density, cyclotron mass, and Fermi vector of (Sh$_{0.957}$Bi$_{0.043}$)$_2$Te$_3$ show lower values. It is indicated that the relative position between Dirac point and Fermi surface are close, being consistent with Fig. 2(b). Our results therefore demonstrate that (Sh$_{1-x}$Bi$_x$)$_2$Te$_3$ with $x = 0.043$ on mica substrate approaches the ideal TI [Fig. 2(a2)]. It should be noted that (Sh$_{0.957}$Bi$_{0.043}$)$_2$Te$_3$ shows p-type behavior [Fig. 4(a)], indicating that the Fermi level is slightly lower than Dirac point. Thus, more composition points and gating measurement to probe the position of Fermi level in the band gap will be studied in our future works to locate more precisely the ideal TI composition on mica substrate.

IV. CONCLUSION

In this work, high quality epitaxial (Sh$_{1-x}$Bi$_x$)$_2$Te$_3$ with $0 < x < 0.07$ were grown on mica substrate successfully. The surface roughness of the thin films increases with doping Bi, but remains lower than 2 nm. When Bi concentration is $x = 0.043$, the sheet resistance shows a local peak, indicating that it is close to ideal TI composition. Weak antilocalization effect with coefficient of -0.43 and phase coherence length of > 70 nm is observed when the temperature is lower than 10 K, confirming the existence of 2D surface state. Under high magnetic field, (Sh$_{1-x}$Bi$_x$)$_2$Te$_3$ with $x = 0.043$ demonstrates clear Shubnikov-de Hass (SdH) oscillation behaviors and shows a low value of 2D carrier density, cyclotron mass, and Fermi vector. Our results thus demonstrate that the relative position between Dirac point and Fermi surface are close in (Sh$_{1-x}$Bi$_x$)$_2$Te$_3$ with $x = 0.043$ on mica substrate. And the p-type behavior of it suggests that more Bi should be slightly doped into system to fully obtain ideal TI. Our results on the growth of (Sh$_{1-x}$Bi$_x$)$_2$Te$_3$ and the location of ideal TI composition will be helpful for the applications of TIs in flexible optoelectronics, spintronics, and magnetic sensors.

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