High quality ultra-flat BiSbTe₃ films grown by MBE

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Abstract

We report on the growth of high quality topological insulator BiSbTe₃ films (thickness of 30 nm) on sapphire (0001) substrates through the molecular beam epitaxy (MBE) technique, and discuss the possibility to improve the film quality and surface flatness through annealing. Instead of using elemental Sb, the choice of Sb₂Te₃ as well as the use of solid sources of Bi and Te assures excellent stability during thermal evaporation enabling layer-by-layer epitaxial growth of high quality films of BiSbTe₃. The crystallinity and the terrace size of the BiSbTe₃ films are found to be improved through increasing the deposition temperature and/or after annealing at 540 K - 620 K for 1 - 4 hours. The films grown at 485 K, 500 K and 515 K exhibit root-mean-square (RMS) roughness of 2.9 nm, 2.3 nm and 2.3 nm, respectively, while the RMS roughness is reduced to 0.6 nm or less when the films are annealed at 580 K for 2 - 4 hours. Annealing the film at too high temperature, such as 620 K, introduces a rougher surface due to the loss of material during annealing. A relatively low electron density of $\sim 2.2 \times 10^{18} \text{ cm}^{-3}$ (at 2 K) is achieved for the as-grown films deposited at 485 K and 500 K. Significantly enhanced electron density is found in the case of either increasing growth temperature or increasing annealing temperature. An exception is the film annealed at 620 K which became a *p*-type conductor. In addition, weak anti-localization effects are evident for the *n*-type films, but they nearly vanish for *p*-type conducting films. The significant influence of temperature on the crystallinity, the surface roughness, and the electrical transport in BiSbTe₃ films will be instructive for further investigations of the transport behavior of surface states in BiSbTe₃ films.

Keywords:

A1. Annealing, A1. Surface roughness, A3. MBE growth, B1. BiSbTe₃, B2. Topological insulator

1. Introduction

$(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ compounds have rhombohedral crystal structure (space group $R\bar{3}m$) and have been the mainstay thermoelectric materials for many decades in applications such as coolers and power generators [1-3]. Recently, these tetradymite-type structures have been predicted and confirmed to represent the simplest three-dimensional topological insulator with surface states consisting of a single Dirac cone [4-6]. Electron spins in these time-reversal-symmetry protected helical surface states are locked perpendicular to their crystal momentum due to strong spin-orbit coupling. This makes $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ one of the ideal platforms for spintronics [7], low power-consumption electronic devices [8], and other applications [9-10]. In order to better understand the surface states, studies should be carried out on high quality $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ films that have excellent crystallinity, smooth surfaces, and large terrace sizes.

Compared to bulk single-crystal growth, the molecular beam epitaxy (MBE) growth of $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ films offers a much larger parameter space for the control of intrinsic point defects and surface morphology because MBE growth is chiefly dominated by kinetic processes and also because there is some flexibility in the choice of substrates [11]. Benefiting from their layered structure and weak van der Waals (vdW) bonding between the neighboring Te-Te layers, high quality single crystal $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ films have been successfully obtained on different substrates, such as Si (111) [12-14], GaAs (111) [15], sapphire (0001) [3], and SrTiO_3 (111) [8,16]. For instance, a 20 nm thick film of $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ grown on SrTiO_3 (111) [16], showed very large terraces (possibly as large as 500 nm) but it also had large terrace steps indicating a rough surface. Using vicinal or near-vicinal GaAs (111) substrates, thick films of Sb_2Te_3 and Bi_2Te_3 were prepared with even larger terrace width of 1 μm . Unfortunately, the terrace step height was also large, reaching 35 nm [15]. It was also reported that the use of a wetting layer of Bi or Te on the Si (111) substrate promotes the growth of flat Bi_2Te_3 films with the smallest root-mean-square (RMS) roughness of about 0.4 nm [13]. However, the unsaturated bonding on the substrate surface limited the terrace size to only about 150 nm [13]. Thus, the growth of $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ films needs further optimization.

Sapphire (0001) substrates have a distinct advantage for the growth of superior $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ films due to their chemically and mechanically robust nature, the ability to heat-clean the substrate at high temperatures (1000 K), and due to the lack of unsaturated bonds on the substrate surface. In this research, we investigated the growth behavior of BiSbTe_3 films on

sapphire (0001) substrates using reflection high energy electron diffraction (RHEED), atomic force microscopy (AFM), and X-ray diffraction (XRD). Subsequently, we studied the effect of the growth temperature and annealing process by measuring the electronic transport properties of the films. Results indicate that temperature has a profound influence on the crystallinity and surface roughness as well as the transport properties of BiSbTe₃ films; this is discussed in detail in what follows.

2. Experimental Procedure

BiSbTe₃ thin films studied here were grown on sapphire substrates (0001) in an MBE chamber at a pressure of 2×10^{-8} Torr. High purity 6N Bi, Sb₂Te₃ and Te source materials were co-evaporated from three conventional Knudsen-type cells in order to provide the optimal flux ratio of 1.7 : 1 : 5.2 resulting in the deposition of stoichiometric BiSbTe₃ films. The corresponding evaporation temperatures of Bi, Sb₂Te₃, and Te were 770 K, 708 K and 591 K, respectively. The substrate temperature was calibrated prior to the deposition, which is $\sim 75\%$ of the temperature of sample holder heater up to 1000 K. This study identified substrate temperatures in the range of 485 - 515 K as suitable for the growth of high quality BiSbTe₃ ternary films. The growth conditions were monitored *in-situ* by a RHEED (KSA 400) apparatus attached to the growth chamber. The thickness of the films was controlled to 30 ± 2 nm by adjusting the deposition time based on the growth rate. The films deposited at 500 K were annealed at 540 K - 620 K for 1 - 4 hours in order to optimize their crystallinity and the films' surface roughness. X-ray diffraction was performed using a Rigaku Ultima IV X-Ray Diffractometer with Cu K_α ($\lambda = 1.5406$ Å) radiation. The AFM (Veeco Dimension Icon) was used to determine both the surface topography and the film thickness. Depth profiles of the chemical state of Bi, Sb and Te were obtained by an Auger electron spectrometer (AES, Physical Electronics Auger Nanoprobe 680) combined with Argon ion sputtering. The collection area for the Auger electron signal was $20 \mu\text{m} \times 20 \mu\text{m}$. Electrical transport properties, including Hall coefficients, electrical resistivity and magnetoresistance, were measured from 2 K to 300 K in a Quantum-Design Magnetic Property Measurement System (MPMS).

3. Results and discussion

3.1 Growth of BiSbTe₃ films

Precise calibration prior to our experiments verified that pure Sb is not an ideal source of evaporation due to its high vapor pressure and unstable flux rate during long term usage. Thus, instead of pure Sb we use Sb_2Te_3 compound to deposit binary Sb_2Te_3 and ternary $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$. Figures 1(a) and 1(b) show XRD patterns and a RHEED image of the as-deposited Sb_2Te_3 film with $T_{\text{sapphire}} = 480$ K and $T_{\text{Sb}_2\text{Te}_3} = 715$ K. Peak positions at $2\theta \approx 20.5^\circ$, 37.6° and 41.7° , shown in Fig. 1(a), correspond to the sapphire (00,*L*) peaks [17], while $2\theta \approx 40^\circ$ represents the K_β diffraction ($\lambda = 1.392$ Å) of the Sb_2Te_3 (00,15) peak. All other peaks were indexed as Sb_2Te_3 (00,*L*), indicating that the film grows parallel to the *c*-axis direction of sapphire, the usual epitaxial growth of Sb_2Te_3 . In addition, the clear RHEED streaks in Fig. 1(b) demonstrate that an excellent Sb_2Te_3 epitaxial film can be obtained by solely evaporating Sb_2Te_3 . We have also found in this study that, compared to the pure Sb, the choice of Sb_2Te_3 as the source material makes it easier to control the flux rate, resulting in stable evaporation during the growth of many BiSbTe_3 -based thin films. Based on this understanding, we studied the effect of temperature on the film crystallinity and the surface roughness of BiSbTe_3 -based thin films.

Systematical investigations reveal that high quality BiSbTe_3 films can be obtained by the above-mentioned flux ratio of Bi : Sb_2Te_3 : Te = 1.7 : 1 : 5.2 and $T_{\text{sapphire}} = 485 - 515$ K. Figure 2(a) presents the RHEED pattern of the BiSbTe_3 film grown at $T_{\text{sapphire}} = 500$ K. The observed sharp stripes indicate an atomically flat surface morphology in this as-grown film, while the weak Kikuchi lines (indicated by the dotted lines) demonstrate that the growing film possesses high quality crystalline structure which is further confirmed by the narrow rocking curve shown in Fig. 4(a) [18-19]. The reciprocal lattice parameter ($k_{\parallel}$) is marked by double arrows in Fig. 2a and is inversely proportional to the in-plane lattice parameter *a*. As shown in Fig. 2(b), *a* decreases suddenly at the very beginning of the growth with respect to the known lattice parameter of sapphire and then rapidly reaches a constant value after 90 s of growth. This reveals that after merely two quintuple layers (QLs) of BiSbTe_3 are deposited, the structure is essentially fully relaxed. We also note that the in-plane lattice parameter *a* of the BiSbTe_3 film is $\sim 11\%$ smaller than the value of sapphire, and we estimate it to be 4.25 Å, consistent with its bulk value [19]. Figure 2(c) depicts an evolution of the RHEED intensity during the growth of BiSbTe_3 films. After growing for ~ 2 min, clear sinusoidal oscillations are recorded in the RHEED intensity. The periodicity of oscillations is 63.5 s, consistent with the growth rate of ~ 0.017 nm/s determined from the AFM measurement. This result confirms advantages of the vdW epitaxial

growth of high quality films [12-16]. When the lattice parameter of a film differs considerably from that of a substrate, the lattice strain near the interface is limited to a region of 1-2 structural units and does not propagate deep into the film due to weak interactions between the substrate and the film as well as weak bonding between neighboring structural units (quintuple layers in the case of the tetradymite-type structure of Bi_2Te_3).

The chemical state of the elements is examined by AES as a function of depth down to the substrate, as shown in Fig. 2(d). The appearance of an oxygen peak in the AES spectrum after 4.5 min of argon sputtering ($E_{\text{kin}} = 1$ kV and $I = 2$ mA) represents the signal from the substrate following the total removal of the film by sputtering. No changes are observed in the chemical state of Bi, Sb, and Te as a function of depth, confirming the uniform growth of BiSbTe_3 films during the deposition.

3.2 Surface morphology of as-grown and annealed BiSbTe_3 films

Figures 3 (a-c) show AFM images ($3 \times 3 \mu\text{m}^2$ in area) of BiSbTe_3 films deposited at substrate temperatures of 485 K (a), 500 K (b), and 515 K (c). The triangular step-by-step terrace surfaces are prevalent in all BiSbTe_3 film images, tightly correlated to the trigonal (hexagonal) symmetry of the crystal structure in the basal plane. Increasing the growth temperature, the size of the terraces increases from ~ 300 nm at 485 K to ~ 500 nm at 515 K, and the terrace steps are much clearer at higher temperatures (see the inset in Fig. 3(b)), revealing the enhanced crystal quality of the surface. Height profile measurements (not shown here) indicate the height of each terrace step as ~ 1 nm which is consistent with the reported thickness of one individual quintuple layer of BiSbTe_3 . In addition, the root-mean-square (RMS) roughness is estimated to be 2.9 nm, 2.3 nm, and 2.3 nm for films grown at 485 K, 500 K, and 515 K, respectively. It is interesting that all terraces of BiSbTe_3 films are oppositely oriented as indicated by dashed triangles, representing the typical surface morphology of epitaxial A_2B_3 ($\text{A} = \text{Bi}$ and Sb , $\text{B} = \text{Se}$ and Te) films deposited on various substrates [19]. These so-called twin domains are inevitably caused by the competition of the growth directions allowed by the lattice symmetry of the film. Specifically, the A_2B_3 films can grow along any of the six equivalent crystallographic directions of sapphire's (0001) surface, while, in contrast, the crystal lattice in the basal plane of the film has a three-fold symmetry. In addition, weak vdW bonding between adjacent quintuple layers also tolerates an imperfect growth of A_2B_3 films with twin domain structure. As a result, the twin defects form in

the boundary region, as highlighted by the dashed circles in Fig. 3(c), where oppositely oriented domains align during the growth. Although in principle the density of twin defects in these as-grown films could be effectively minimized by choosing perfectly lattice matched substrates or growing on vicinal substrates [18-19], the deposition of large area and uniformly grown van der Waals epitaxial films is still a big challenge. An interesting new approach was recently demonstrated by Stoica et al. in which tuned buffer layers are used to achieve a quasi lattice match to the film being grown, with the vdW epitaxy taking care of the buffer layer/substrate interface mismatch [20].

We have also investigated the effect of annealing for the as-grown films, carried out in the presence of Te vapor to compensate for the loss of Te during the annealing process. A suitable Te flux in the annealing process is determined through evaluating the real-time RHEED images during annealing. This empirically critical value requires the observation of sharper, or brighter, RHEED streaks and Kikuchi lines but without any spotty patterns (indication of excess Te) in the RHEED image of films. This condition is used to indicate improvement of the film surface during annealing. During the annealing, the Te source temperature T_{Te} was chosen as 550 K when the annealing temperature T_{ann} was 540 K and 580 K, while $T_{\text{Te}} = 600$ K when T_{ann} was 620 K. Figure 4 plots the XRD rocking curve of the as-grown (a) and annealed (b) BiSbTe₃ films. The annealed samples are all deposited at 500 K. The rocking curve observed at $\omega \sim 22.35^\circ$ corresponds to the (00,15) diffraction peak of the BiSbTe₃ film. As shown in Fig. 4(a), with the increase of the growth temperature, the full-width-half-maximum (FWHM) of the rocking curve decreased moderately from 0.22° to 0.17° . Meanwhile, we also found a significant drop of FWHM after annealing, which reaches $\sim 0.1^\circ$ when annealing the films at 540 K - 620 K for 1 to 4 hours. By increasing the growth temperature or by annealing at elevated temperatures, the narrower rocking curve signals the enhanced crystallinity and enhanced long-range order of the structure mainly in the c-axis direction of the hexagonal structure. This indicates an improved crystallinity of the epitaxial BiSbTe₃ films. Figures 5 (a-d) display RHEED patterns of annealed BiSbTe₃ films along the azimuthal direction $[\bar{1}100]$ [21]. For any annealing temperature (540 K - 620 K) or any duration of annealing, the intensity of Kikuchi lines is remarkably increased compared to films not subjected to an annealing treatment. This indicates the improved film quality within the basal plane [19]. The temperature is also instrumental in modifying the surface morphology. As shown in Figs. 5(e-h), annealing at 540 K has a minor influence on the surface

roughness. However, there is a notable change as the annealing temperature reaches 580 K as demonstrated by a much flatter surface following annealing for 2 and 4 hours. Because of a serious loss of material upon annealing at yet higher temperature of 620 K for 1 hour, the surface then becomes less smooth as seen in Fig. 5(h). Figures 5(i-l) indicate the height profile along the line marked in Figs. 5(e-h). It is impressive that annealing the films at 580 K for 2 and 4 hours leads to a height differential between the highest and the lowest points of terraces on the surface of only about 1 nm, i.e., equal to the height of one quintuple layer. The calculated RMS roughness is about 0.6 and 0.3 nm ($5 \times 5 \mu\text{m}^2$ in area) for 2 hours and 4 hours annealing, respectively. As for the BiSbTe₃ film annealed at 620 K for 1 h, see Fig. 5(h) and 5(l), the size of the terrace is significantly increased, and the size of the largest terrace reaches up to several micrometers. The optimized annealing process (580 K for 2 - 4 hours) and the achievement of ultra-flat surfaces in BiSbTe₃ films in this study will be very useful for further improving the quality of Bi-Sb-Te-based films.

3.3 Transport properties of BiSbTe₃ films

Figure 6 plots the temperature dependent electrical resistivity (a), carrier density (b), and carrier mobility (c) of as-grown and annealed BiSbTe₃ films. All as-grown films show a slightly increasing resistivity with temperature when $T \leq 70$ K, while at higher temperatures, the resistivity decreases with temperature. The annealed films show metallic type conduction within the measuring temperature interval, except for the film annealed at 580 K for 4 hours which has a downward turn at $T \geq 140$ K. Attempts to determine the band gap of BiSbTe₃ via $\rho \propto e^{E_g/2k_B T}$, the formula relevant in the intrinsic excitation region, gave values of $E_g = 0.049$ eV, 0.038 eV and 0.017 eV for films grown at 485 K, 500 K and 515 K, respectively. We have found that with increasing growth and annealing temperatures the resistivity decreases, which is associated with an increasing carrier density, see Fig. 6(b). All as-grown films and films annealed at 580 K show *n*-type conduction, while the film annealed at 620 K exhibits *p*-type conduction. BiSbTe₃ films grown at 485 K and 500 K exhibit the lowest carrier density of $\sim 2.2 \times 10^{18} \text{ cm}^{-3}$ at 2 K. As the growth temperature is elevated to 515 K, the density of carriers increases to $5.2 \times 10^{18} \text{ cm}^{-3}$ at 2 K. Thus, for these as-grown films with relatively low carrier density, the metallic trend in the resistivity at $T \leq 70$ K could be an indication of contributions from metallic surface transport channels superposed over the bulk transport. Increasing the annealing temperature up to 580 K, it

is reasonable to expect that the extra thermal energy will lead to the formation of *n*-type antisite defects of the type Te_{Bi} and Te_{Sb} (an atom of Te sitting on a site of the Bi or Sb sublattice). As already noted, increasing the annealing temperature further to 620 K results in the loss of material (mostly Te) and this gives rise to *p*-type antisite defects Sb_{Te} and Bi_{Te} that convert the film into a *p*-type conductor [22-23]. The observed decreased carrier mobility [see Fig. 6(c)] of the annealed films thus originates mainly from the increased density of point defects. The highest carrier mobility of $408 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at 2 K is observed for the as-grown film deposited at 500 K.

Figure 7(a) displays the magneto-resistance (MR) of the BiSbTe_3 film grown at 500 K. At low magnetic fields ($B \leq 0.5 \text{ T}$), a clear cusp in the MR curve is observed which indicates the weak anti-localization (WAL) effect in the film [24-25]. The WAL effect is commonly considered a signature of surface states in topological insulators having the linear Dirac surface dispersion. Thus, the observation of WAL implies the films in this research have high quality surfaces. After annealing, as shown in Fig. 7(b), the WAL effect is still present in films annealed at 580 K, but the strength of the WAL is diminished because of an increase in its bulk carrier density. In addition, although we observe quite a large MR, the WAL effect has nearly vanished in the film annealed at 620 K for 1 hour due to the Fermi level being located deep in the valence band.

4. Conclusion

In this research we report on the MBE growth of high quality and ultra-flat BiSbTe_3 films on sapphire (0001) with a thickness of 30 nm, and we illustrate the influence of the growth temperature and annealing on the quality of the films and on their electrical resistivity. Instead of using elemental Sb, we have chosen Sb_2Te_3 compound as the source material which guarantees stable growth of high quality films. The RMS roughness of the as-grown films is estimated to be 2.9 nm, 2.3 nm, and 2.3 nm for films grown at 485 K, 500 K, and 515 K, respectively. BiSbTe_3 films annealed at 580 K for 2 to 4 hours attain an ultra-flat surface with RMS roughness of 0.6 nm or less, which is the lowest value ever reported in this system. Annealing is also important for improvement of the film crystallinity as indicated by clearer Kikuchi lines and narrower XRD rocking curves after annealing. Electrical transport measurements confirm a relatively low carrier density of $\sim 2.2 \times 10^{18} \text{ cm}^{-3}$ (at 2 K) for the as-grown films deposited at 485 K and 500 K. With the increase of the growth and annealing temperature, the free electron density increases greatly due to the increased density of point defects, believed to be mainly antisite defects. We also

observe a sign change in the dominant carriers (from *n*-type to *p*-type) when films are annealed at 620 K. This is likely due to the loss of Te and the increased density of *p*-type antisite defects such as Sb_{Te} and Bi_{Te} at elevated temperatures. Moreover, weak anti-localization is evident in *n*-type films, but nearly vanishes for the film with *p*-type conduction. This research is instructive for further improvements in the crystallinity and surface roughness of Bi-Sb-Te-based films used in investigations of surface metallic states of topological insulators.

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Figure Captions

Figure 1: (a) XRD patterns and (b) RHEED image of Sb_2Te_3 film prepared by evaporation of the Sb_2Te_3 compound.

Figure 2: (a) RHEED image of BiSbTe_3 deposited at 500 K taken along the azimuthal $[\bar{1}100]$ direction, (b) in-plane lattice parameter evolution a as a function of deposition time, (c) measured RHEED intensity oscillations, (d) Auger spectrum depth profile along the c-axis of the crystal structure. The double arrows in (a) indicate the $k_{//}$ spacing which is inversely proportional to the lattice parameter, i.e., $k_{//} \propto 1/a$.

Figure 3: AFM image ($3 \times 3 \mu\text{m}^2$ in area) of BiSbTe_3 films deposited at substrate temperatures of (a) 485 K, (b) 500 K, and (c) 515 K. Inset in (b) shows an enlarged image of Fig. 3(b). The dashed triangles mark the opposite orientation of twin domains in the MBE grown BiSbTe_3 films. The dashed circles in (c) highlight the twin defects in the boundary area among the oppositely oriented domains.

Figure 4: XRD rocking curve for (a) as-grown BiSbTe_3 films and for (b) annealed films.

Figure 5: RHEED images for BiSbTe_3 films annealed at (a) 540 K for 4 hours, (b) at 580 K for 2 hours, (c) at 580 K for 4 hours, and (d) at 620 K for 1 hour. Figures (e-h) and (i-l) show AFM images and height profile along the lines indicated, respectively.

Figure 6: Temperature dependent (a) electrical resistivity, (b) carrier density, and (c) carrier mobility of as-grown and annealed BiSbTe_3 films.

Figure 7: (a) Magneto-resistance (MR) of the BiSbTe_3 film grown at 500 K. (b) MR of the annealed BiSbTe_3 films.

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