Topological Insulators Bi_2Te_3 and Bi_2Se_3 Grown by MBE on (001) GaAs Substrates

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Abstract. Bi_2Te_3, Bi_2Se_3 and their alloy films were successfully grown by molecular beam epitaxy (MBE) on (001) GaAs substrates. The structural properties of these films were investigated by Reflection high-energy electron diffraction (RHEED), Atomic force microscopy (AFM), X-ray diffraction (XRD), High-resolution transmission electron microscopy (HRTEM) and Raman spectroscopy and mapping. The results indicate that the epitaxial films are highly uniform. High-field and low-temperature magneto-transport measurements on these films are carried out and discussed.

Keywords: Topological Insulators, MBE, XRD, TEM, Raman, and magnetotransport.
PACS: 81.05.Hd, 81.15.Hi, 61.05.-a, 68.37.-d, 78.30.-j, 73.43.Qt

Recently, photoemission measurements of the surface of topological insulators such as Bi_2Te_3 and Bi_2Se_3 confirmed that there exists a conducting surface state in these materials with a single Dirac point [1,2]. Although the growth of such topological insulators films by molecular beam epitaxy (MBE) is especially attractive, due to the pseudo-hexagonal structure of Bi_2Te_3 and Bi_2Se_3, so far most efforts to fabricate these films by MBE have been carried out using Si (111) [3,4] as substrates, with little work done on GaAs (111) substrates [5]. Because spintronic materials such as GaMnAs are usually grown on (001) GaAs substrates, in this work we discuss MBE growth of Bi_2Te_3, Bi_2Se_3 and their alloys on (001) GaAs substrates, which may open interesting opportunities for applications in spintronics.

Bi_2Te_3, Bi_2Se_3 and their alloy films were grown using a dual-chamber Riber 32 MBE system. First, a (001) GaAs substrate was heated up to 600°C for removing oxidation. The growth was initiated by the monolayer of Te-Bi-Te-Bi-Te or Se-Bi-Se-Bi-Se – a quintuple layer (QL) – in serial atomic layer epitaxy (ALE) type fashion deposited at room temperature. The substrate was then gradually heated to 300°C, and a nice streaky reflection high-energy electron diffraction (RHEED) pattern appeared (see Figs. 1a and 1b). The different RHEED patters observed in figure confirm the hexagonal surface lattice of Bi_2Te_3 and Bi_2Se_3 and indicate the *c*-axis growth of films. The MBE growth of Bi_2Te_3, Bi_2Se_3 and their alloys was then performed under Te or

15th International Conference on Narrow Gap Systems (NGS15)
AIP Conf. Proc. 1416, 105-108 (2011); doi: 10.1063/1.3671709

Se rich conditions with $T_{substrate}$ = 300°C. The film growth rate (~2nm/min) was determined by RHEED oscillations (as shown in Fig. 1c), which agrees with the thickness determined by X-ray reflectivity spectra (not shown).

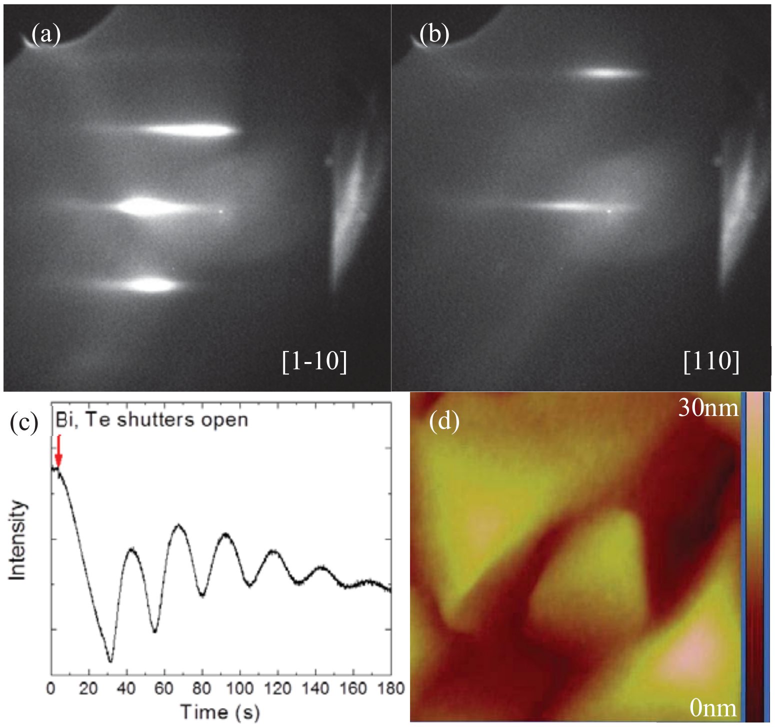

FIGURE 1. (a,b) RHEED diffraction patterns observed in two specific orientations of a (001) GaAs substrate during the growth of a Bi_2Te_3 film. (c) RHEED oscillations observed during the beginning of the growth of a Bi_2Te_3 film. (d) AFM height image of 136nm-thick Bi_2Te_3 film. The size is 1×1μm.

Atomic force microscopy (AFM) measurements (see Fig. 1d) show a surface roughness of ~ 5.27nm for a 136nm-thick Bi_2Te_3 film. The surface roughness increases as the film thickness increases, suggesting that growth conditions still need to be optimized in the future.

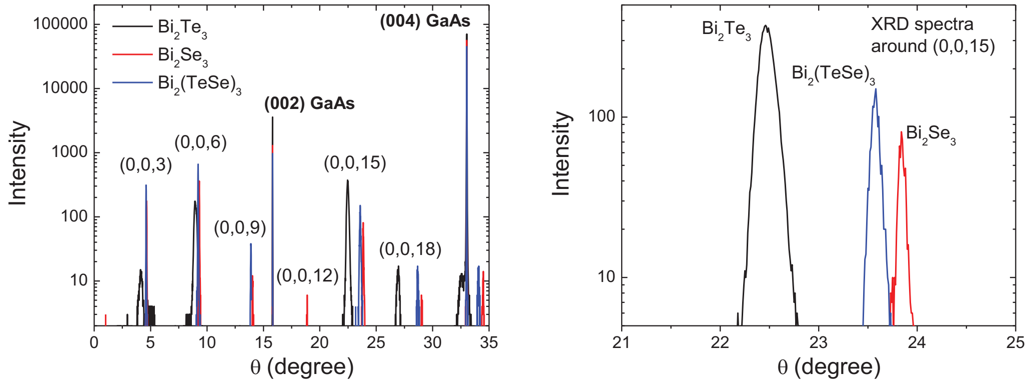

FIGURE 2. X-ray diffraction patterns obtained from 233-nm-thick Bi_2Te_3, 220-nm-thick $Bi_2(TeSe)_3$ and 180-nm-thick Bi_2Se_3 films grown by MBE on GaAs (001) substrates.

The crystalline structure of the films was confirmed by high resolution X-ray diffraction (XRD) and by transmission electron microscopy (TEM). XRD spectra shown in Fig. 2 reveal reflections only from {003}-type lattice planes, which is indicative of highly pronounced *c*-axis growth of the Bi_2Te_3 and Bi_2Se_3 films. Note that, Bi_2Te_3 and Bi_2Se_3 have a rhombohedral layered structure, which is composed of the stacking order of hexagonal Te(Se) and Bi atomic layers along the *z*-direction. The highly parallel quintuple layers – Te(Se)-Bi-Te(Se)-Bi-Te(Se) – are seen in both Bi_2Te_3 and Bi_2Se_3 films despite the slightly wavy interface of the GaAs substrate (see Fig. 3). Note that, the light and dark areas visible in the micrographs are possibly due to ion-milling damage during TEM sample preparation.

Raman spectroscopy and Raman mapping have also been performed on these materials using a 532 nm excitation laser (power ~0.8mW). The Raman spectra show three characteristic peaks for Bi_2Se_3 [at ~71cm^{-1} (A^1_{1g}), 131cm^{-1} (E^2_g) and 174cm^{-1} (A^2_{1g})] (not shown), and two peaks for Bi_2Te_3 [at ~102 cm^{-1} (E^2_g) and 134cm^{-1} (A^2_{1g})] (see Fig. 4a). The observed peaks are consistent with the lattice vibration modes reported earlier for these materials [6]. The Raman maps show that the positions of the Raman peaks measured within a scan area of 15μm×15μm differ by less than ~1 cm^{-1}, indicating a high uniformity of the films, as shown in Figs. 4b and 4c.

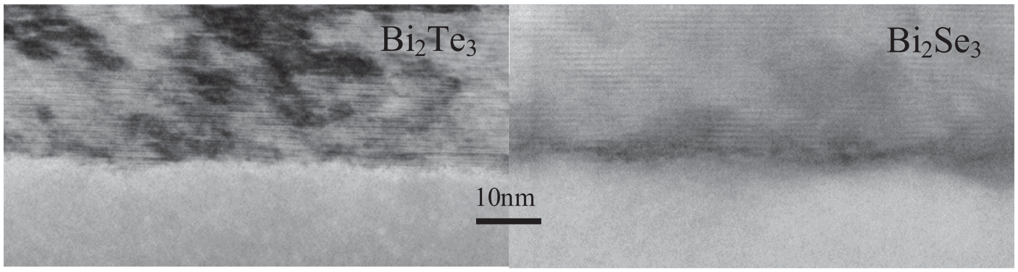

FIGURE 3. TEM images of Bi_2Te_3 and Bi_2Se_3 films.

Room temperature electron transport measurements show that the resistivity of the films strongly depends on the Group-VI/Bi flux ratio, thus suggesting a strategy for optimizing future growth conditions. Low-frequency noise measurements show 1/f-type voltage fluctuations, similar to the noise behavior occurring in conventional semiconductor films.

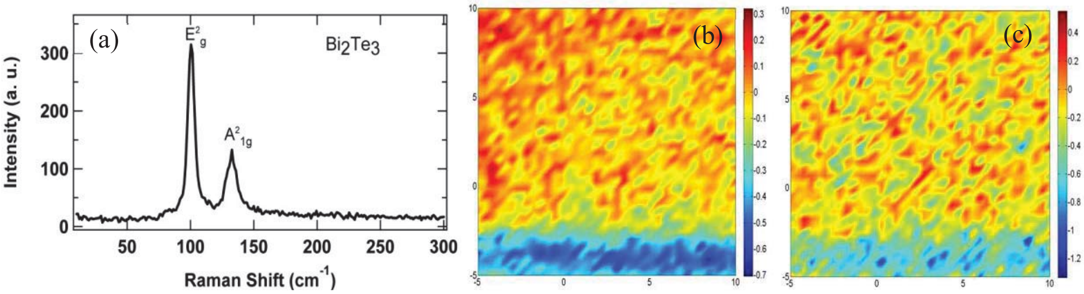

FIGURE 4. (a) Raman spectra measured in thin Bi_2Te_3 film. (b,c) Raman mappings of peak position differences compared to a bulk single crystal Bi_2Te_3 sample for the peaks E^2_g and A^2_{1g}, respectively.

High-field and low-temperature magneto-transport measurements on these films were carried out at temperatures ranging from 2K to 10K with fields up to 6T. The results confirm that the undoped topological insulator films have *n*-type conductivity, with carrier concentrations in the range of 4×10^{19} to 8×10^{19} cm^{-3} and mobilities in the range of 270 to 330 cm^2(Vs)$^{-1}$. The magnetic field dependences of the resistivity with magnetic field applied normal to the plane all show a sharp positive magnetoresistance cusp at low fields and low temperatures, which may be related to weak antilocalization corrections in a 2D system. The cusp is enhanced in thinner films, suggesting that it is a surface-related phenomenon. Our results are consistent with those reported for films grown on other substrates [5,7].

ACKNOWLEDGMENTS

This work was supported by NSF Grant 10-05851 for ND; an NSF Grant 1002114 and an AFOSR Grant FA9550-10-1-0129 for ASU; DARPA MESO program for Purdue. The authors acknowledge use of facilities in the John M. Cowley Center for HREM at ASU, and in the NIST Center for Neutron Research for XRR.

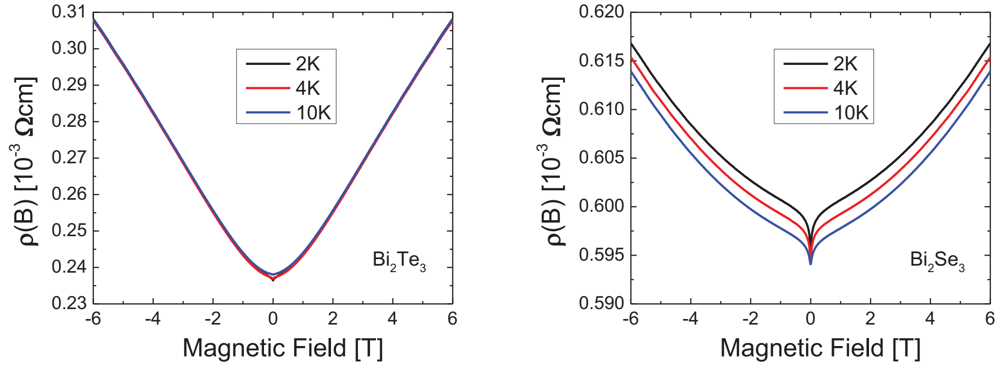

FIGURE 5. The magnetic field dependence of the resistivity for the 20-QL ultrathin Bi_2Te_3 and Bi_2Se_3 films with the field applied in the out-of-plane direction at various temperatures.

REFERENCES

1. M. Z. Hasan and C. L. Kane, *Rev. Mod. Phys.* **82**, 3045 (2010).
2. S.-C. Zhang, *Physics* **1**, 6 (2008).
3. X. Chen, X.-C. Ma, K. He, J.-F. Jia, and Q.-K. Xue, *Adv. Mater.* **23**, 1162 (2011).
4. H. D. Li et al., *Appl. Phys. Lett.* **98**, 043104 (2011).
5. A. Richardella et al., *Appl. Phys. Lett.* **97**, 262104 (2010).
6. W. Richter et al., *Phys. Stat. Sol. (b)* **84**, 619 (1977).
7. T. Hirahara et al., *Phys. Rev. B* **82**, 155309 (2010).