
Monograph:
Knox, C orcid.org/0000-0002-4673-5649 (2020) Growth of Bismuth Telluride: A platform for Topological Materials. Report. Materials from the Royce Deposition System (1). University of Leeds

10.48785/100/80

Growth of Bismuth Telluride: A platform for Topological Materials

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This application note describes the growth of Bismuth Telluride (Bi_2Te_3) in the Royce Institute thin films deposition facility and presents data demonstrating the material's properties.

1 Introduction

Topological insulators are materials wherein, due to their unique band structure, transport is dominated by spin-polarised, dissipationless edge states [1]. These edge states represent a fascinating opportunity for research in spintronics, superconductors and photonics. Bismuth Telluride (Bi_2Te_3) is one such topological insulator, which has been a well developed material system, due to its thermoelectric applications [2]. Additionally, modifying Bi_2Te_3 via doping and alloying with other materials has been shown to have a variety of applications, from increasing the band-gap (in order to inhibit bulk transport [2]), to adding some magnetic order to the material [3], or making the edge-states superconducting [4]. Therefore, growth of high quality Bi_2Te_3 films by MBE is essential for topological insulator research. This application note provides the growth procedure for high quality Bi_2Te_3 grown in the Royce thin film deposition system at the University of Leeds.

2 Growth

Conventionally, the epitaxial growth of material onto a clean substrate requires a good lattice match, as the length and angle of surface dangling bonds cannot be changed easily [5]. In contrast, layers of Bi_2Te_3 are not chemically bonded to each other, but are instead separated by a Van der Waals (VdW) gap. This allows Bi_2Te_3 to grow epitaxially onto a wide variety of substrates [6, 7], providing the surface dangling bonds are passivated [8]. In this application note, a high purity silicon(111) substrate was chosen in order to provide an insulating background to transport and optical spectroscopy measurements.

In preparation for the growth process, 20×20 mm Si(111) substrates were diced from a 100 mm diameter wafer and then cleaned under ultrasonic agitation in acetone and isopropyl alcohol. In order to remove any possible contaminants from the dicing process, the samples are first agitated in acetone for 5 minutes, before the acetone (which may contain residues of the tape used to hold the wafer whilst it is diced) is washed off in isopropyl alcohol and the sample is agitated in clean acetone for a further 5 minutes. That clean acetone is then washed off in more isopropyl alcohol and then dried, before immediately being loaded into the Royce thin film deposition system.

Once loaded, the substrates were out-gased at 800 ℃ for a minimum of 1.5 hours in the preparation chamber (where the temperature was measured by a thermocouple attached to the heater stage), before being allowed to cool to ambient temperature and transferred into the Royce MBE system. The sub-

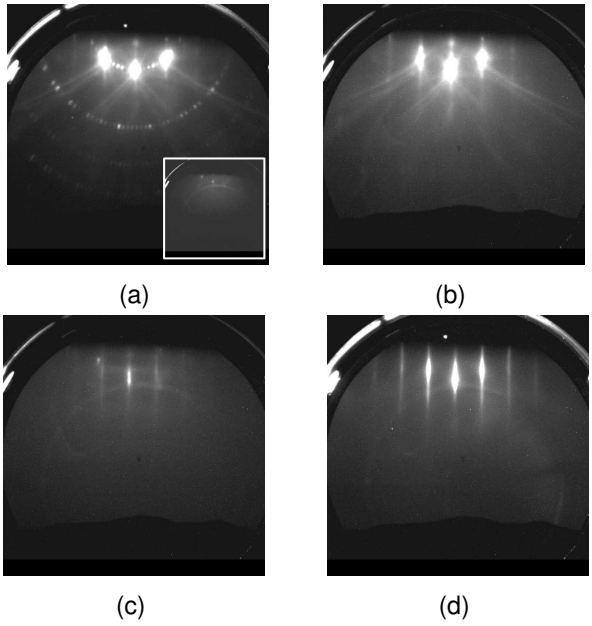

Figure 1: In situ RHEED images taken at various points in the growth process. a shows the Si (7×7) surface reconstruction. The inset shows the RHEED image after the clean surface has been exposed to a Bi flux. b shows the surface reconstruction formed after annealing the Bi treated Si surface to 360 ℃. c shows the streaky Bi_2Te_3 pattern after one minute of growth. d shows the Bi_2Te_3 RHEED pattern after 40 minutes of growth, at the end of the procedure.

strate was then heated to 800 ℃ (measured by a thermocouple attached to the sample manipulator) for further 1.5 hours for a final outgas. This additional outgas is necessary, as the base pressure of the MBE system with the heater active is lower than that of the preparation chamber. The substrate temperature was then ramped to 1250 ℃ at 10 ℃min^{-1} to evaporate the native oxide layer on the Si substrates, and to reconstruct the Si(111) surface, freeing the dangling bonds. After holding the sample temperature for 10 minutes at 1250 ℃ the temperature of the substrate, as read by a pyrometer, was approximately 900 ℃, above the 860 ℃ needed for reconstruction of the (7×7) Si(111) surface [9]. The substrate temperature was decreased to 150 ℃ at a rate of 10 ℃min^{-1}, in order to verify the (7×7) reconstruction via reflection high energy electron diffraction (RHEED), shown in Fig. 1a.

In order to improve the film quality, it is essential that the surface dangling bonds on the substrate are passivated, so that the substrate-Bi_2Te_3 interface starts with a VdW gap, and therefore no strain is applied to the initial Bi_2Te_3 layer. This is achieved by depositing a thin layer of disordered bismuth onto the Si (7×7) surface at a substrate temperature of 150 ℃ until the RHEED pattern disappears (shown in the inset of Fig 1a). The sample is then annealed to 360 ℃, where a new reconstruction is observed, as shown in Fig. 1b, and the excess bismuth is desorbed [10]. Finally, the Bi_2Te_3 film is grown on top of this passivated surface by exposing it to bismuth and tellurium fluxes simultaneously at a substrate temperature of 270 ℃. In order to obtain the correct film stoichiometry and to minimise the formation of tellurium vacancies, the film is grown in a large excess of tellurium, such that the flux of tellurium is approximately 20 times that of bismuth [7].

3 Properties

Structural characterisation was obtained by in-situ RHEED measurements during growth and X-Ray diffraction measurements. The growth of the Bi_2Te_3 film was monitored by in-situ RHEED, using a 15 kV accelerating voltage, where the beam is incident along the [100] azimuth of the Bi_2Te_3 crystal. Within 30 seconds of exposing the treated silicon surface to bismuth and tellurium fluxes, a streaky pattern develops, characteristic of the Bi_2Te_3 crystal structure [6]. This pattern is shown in Fig. 1c where 1.6 nm of Bi_2Te_3 has been deposited, after one minute of exposure to bismuth and tellurium fluxes. Additionally, the streak spacing of this pattern corresponds to an in-plane lattice parameter of 0.44 ± 0.01 nm, which is in agreement with bulk in-plane lattice parameter of Bi_2Te_3 , 0.438 nm [6]. This is indicative of good VdW epitaxy growth, where no strain is applied to the initial Bi_2Te_3 layer, as a VdW gap exists between the treated silicon surface and the first Bi_2Te_3 quintuple-layer. As the growth proceeds, this RHEED pattern becomes brighter, although it maintains the streak spacing corresponding to an in-plane lattice parameter of 0.44 ± 0.01 nm, indicative of good in-plane order throughout the crystal structure. The RHEED pattern obtained at the end of the growth procedure is shown in Fig. 1d.

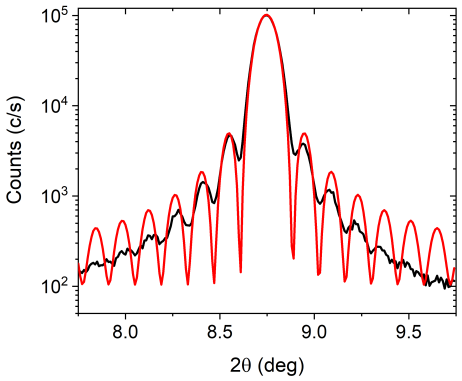

Figure 2: X-ray diffraction of the [003] peak from a Bi_2Te_3 film grown in the Royce MBE system, shown in black. The Pendellosung fringes were fitted with a c-axis lattice parameter of 3.03 ± 0.02 nm with $N_0 = 21$ lattice planes (shown in red), resulting in a total thickness of 63.6 ± 0.4 nm.

The out-of-plane lattice characteristics were subsequently characterised by X-ray diffraction measurements, using copper Kα radiation, the results of which are shown in Fig. 2. The presence of Pendellosung satellite fringes around the [003] diffraction peak indicate that the material is well ordered along its c-axis throughout the sample thickness. The profile of the [003] diffraction peak and the associated Pendellosung fringes is fitted with a modified Scherrer equation, following the approach of Sürgers et al [11]. Here, the effect of disorder is accounted for by taking a sum of different oscillators, each with a different number of lattice planes (N), weighted around a mean

(N_0) by a Gaussian distribution of width σ;

$$I = \sum_N \frac{1}{\sigma} e^{-\left[\frac{(N-N_0)^2}{\sigma^2}\right]} \left(\frac{\sin(\frac{Nqc}{2})}{\sin(\frac{qc}{2})}\right)^2 \quad (1)$$

where I is the observed X-ray intensity, c is the c-axis lattice parameter and $q = 4\pi\sin(\theta)/\lambda$, the X-ray scattering vector. This fit yields a c-axis lattice parameter of 3.03 ± 0.02 nm (in line with the bulk c-axis lattice parameter of Bi_2Te_3, 3.045 nm [6]), a variance of 0.004 ± 0.001 lattice planes and a mean thickness of 63.6 ± 0.4 nm. Therefore, as the film was deposited over the course of 40 minutes, the growth rate is 0.265 ± 0.002 Ås^{-1}. It is important to note that, while the lattice constant of Bi_2Te_3 is 3.03 ± 0.02 nm, this includes a total of 15 atomic layers and three instances of the inter-layer VdW gap, and as such the height of one covalently bonded quintuple layer of Bi_2Te_3 (approximately 1 nm high) is much smaller than the lattice constant [12].

4 Further Information

The Royce Deposition System is a multichamber, multitechnique thin film deposition tool based at the University of Leeds as part of the Henry Royce Institute. Materials from the Royce Deposition System are available as a facility service and for collaborations.

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