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Strain state, film and surface morphology of epitaxial topological insulator Bi_2Se_3 films on Si(111)

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ABSTRACT

Epitaxial Bi_2Se_3 films were grown by molecular beam epitaxy on Si(111)-Bi($\sqrt{3} \times \sqrt{3}$)R30° at temperatures between 200 and 250 °C. The surface and bulk morphology was characterized by high resolution low energy electron diffraction, X-ray diffraction, and atomic force microscopy for various film thicknesses between 6 and 90 nm. The films are atomically smooth without small angle mosaics or small angle rotational domains. The precise determination of lattice parameter reveals that films grown at higher temperature exhibit a smaller value for the vertical lattice parameter. The presence of random stacking faults in the film is reflected by a parabolic increase of the width of the diffraction peaks in X-ray diffraction.

1. Introduction

Recently, topological insulators like Bi_2Se_3 have attracted much attention due to their peculiar electronic properties. While the bulk of such material is insulating or semiconducting the vacuum interface exhibits a protected surface state [1–4]. Backscattering is strongly suppressed due to spin split electronic band structure [5] which has been confirmed in many photo electron spectroscopy studies [6–10]. Such materials promise potential applications in the field of spintronics and dissipation free electronics [11]. For technologically relevant use of topological insulators it is, however, necessary to grow crystalline films of the desired material on an appropriate substrate. There are several studies on the growth of Bi_2Se_3 films on various substrates revealing the epitaxial growth mode, atomically smooth surfaces [12–18]. Here we employ in-situ high resolution low energy electron diffraction and ex-situ X-ray diffraction and atomic force microcopy to characterize the surface and film morphology and strain state of epitaxial Bi_2Se_3 films of various thicknesses grown on Si(111).

2. Experimental conditions

All experiments were performed at UHV conditions at a base pressure of less than $2 \cdot 10^{-8}$ Pa. The UHV chamber is equipped with a high resolution spot profile analysis low energy electron diffraction (SPA-LEED) instrument [19,20], a second low energy electron gun in a reflection geometry with oblique incidence [21,22], a Selenium (Se) evaporator, and a Bismuth (Bi) evaporator. The Si(111) samples were cut from a highly doped wafer with a resistivity of 4 mΩcm and a miscut of less than 0.1°. For sample heating direct current was used. All samples were degassed over-night at 600 °C in the load lock chamber at high vacuum conditions (10^{-5} Pa). Prior to material deposition and film growth a short flash annealing cycle to more than 1200 °C was used to remove the native oxide from the Si substrate. Temperature measurements were performed with an infrared pyrometer. Bi and Se were evaporated from filament heated crucibles. Both evaporators are equipped with a water cooled copper shroud to prevent contamination and heat-up of the chamber [23,24]. The Bi deposition rate was adjusted through the observation of the formation of the Bi$\left(\sqrt{3} \times \sqrt{3}\right)$ reconstruction on Si(111) at 500 °C, that occurs at a Bi coverage of 1/3 mono-layer (1 ML $= 7.83 \times 10^{14}$ atoms/cm^2, i.e., a monolayer of Si(111)) [25,26]. The Se deposition rate was estimated through the formation of the Se terminated Si(1×1) reconstruction at 570 °C that occurs at a Se coverage of 1/2 ML [27].

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As a template for Bi_2Se_3 film growth a Bi$\left(\sqrt{3}\times\sqrt{3}\right)$ [28] reconstruction was prepared through Bi adsorption at 500 °C. Subsequently epitaxial Bi_2Se_3 films were grown by co-deposition of Se and Bi at growth temperatures of 200 °C and 250 °C. To compensate for Se desorption and to assure the correct stoichiometry for the Bi_2Se_3 film the growth took place under Se rich conditions, i.e., the Se flux was adjusted 10 times higher than the Bi flux. For the thin films of 6 QL and 12 QL, the growth rate was set to 1 quintuple layer (1 QL = 0.95 nm Bi_2Se_3 [29]) in 6 min. The deposition rate was calibrated after growth through ex-situ atomic force microscope (AFM) in non-contact mode, using a Veeco Dimension 3100 AFM. To further improve the crystal quality for films grown at 250 °C post growth annealing was applied at 250 °C for 10 h. In this study we focused on the determination of the strain state and the bulk quality of the film Bi_2Se_3 and therefore did not attempt to remove residual Se precipitates from the surface after growth.

All in-situ SPA-LEED measurements were performed after growth at room temperature. A PANalytical X'Pert PRO in Bragg–Brentano (θ–2θ) geometry was used for ex-situ X-ray diffraction (XRD) with non-monochromatized X-rays (Cu-anode) of a weighted average wavelength of 1.5418 Å.

3. Results and discussion

All in-situ and ex-situ measurements shown here were performed at room temperature after cool-down of the sample. Fig. 1 shows in-situ LEED patterns and ex-situ AFM images of Bi_2Se_3 films of different film thicknesses and growth temperatures. The LEED pattern in Fig. 1a) is obtained from a 6 QL thick Bi_2Se_3 film grown at 200 °C and depicts a clear (1 × 1) surface structure. Each spot is surrounded by weak diffuse intensity. We have chosen the LEED energy of 159 eV as an out-of-phase condition to be sensitive to surface roughness of the Bi_2Se_3 film. Due to the huge step height of 0.95 nm the in- and out-of-phase conditions are very close in k-space. The electron energy of 159.4 eV corresponds to a phase condition of $S = 19.5$, i.e., almost 20 electron wavelengths for the interference between neighbored terraces. Due to this high value of S it was impossible to observe a clear variation of the spot profile with scattering phase S [30].

The line profile shown in Fig. 1a) was taken through the (00) spot and the first integer order spots. Nonlinearities in the LEED pattern were corrected using LEEDcal [31] as described in [22]. From the positions of the spots we determine a lattice parameter of 4.132 ± 0.01 Å

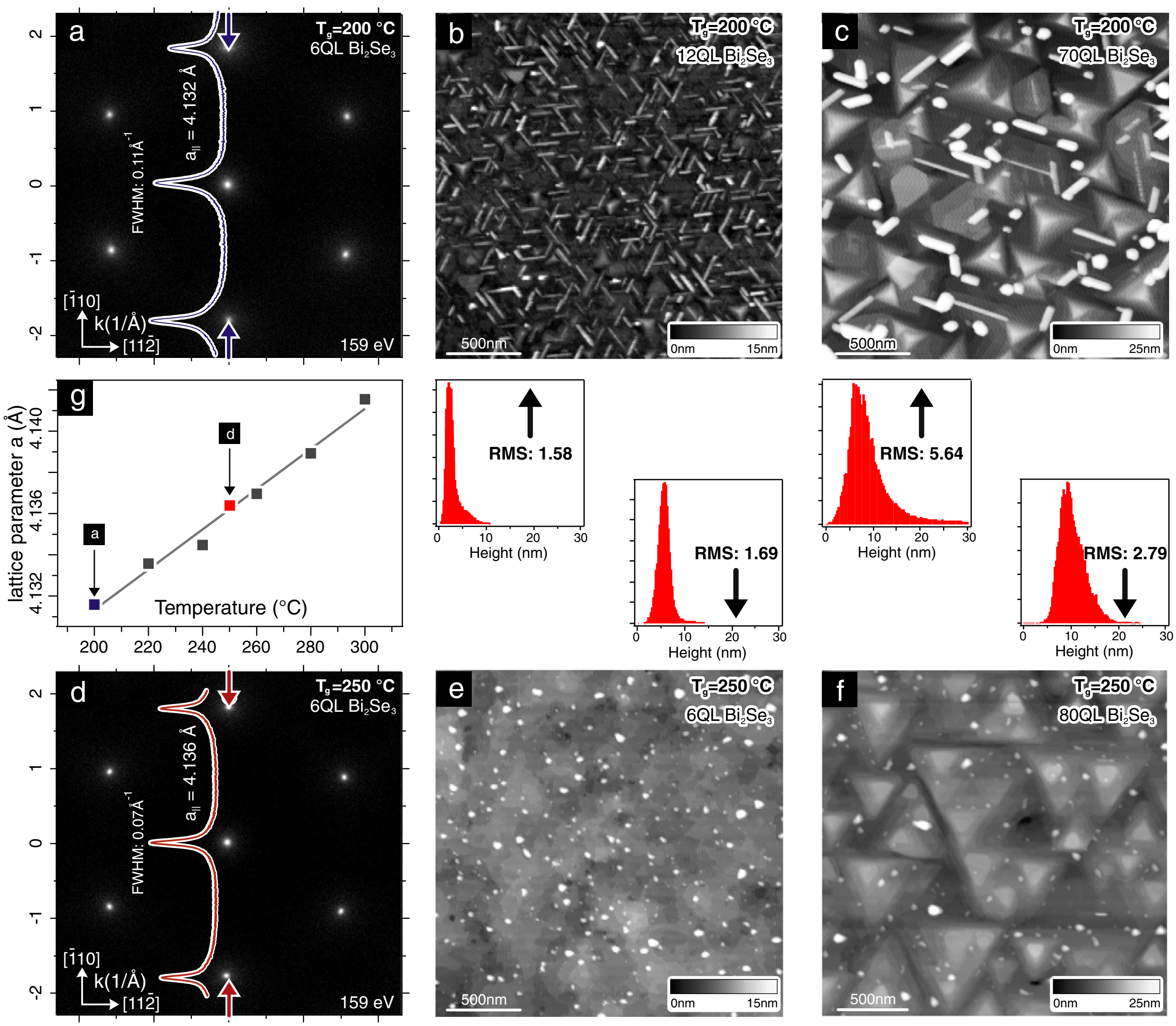

Fig. 1. a) and d) SPA-LEED pattern of 6 QL thick Bi_2Se_3 films. From the line profile through the integer order spots the lateral lattice parameter and full width of half maxima are determined. b,c) and e,f) AFM images with field of view of 2 × 2μm^2. Film thickness and gray scale look-up table are indicated. The high density of precipitates presumably arises from Se crystallites. Growth temperature is 200 °C for a–c) and 250 °C for d–f). Center plots depict histograms from the corresponding AFM images. The RMS roughness is given in nm. g) Temperature dependence of the lateral lattice parameter *a*.

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