Available online at www.sciencedirect.com
ScienceDirect
Thin Solid Films 516 (2008) 8227 – 8231
www.elsevier.com/locate/tsf

Epitaxial Bi(111) films on Si(001): Strain state, surface morphology, and defect structure

H. Hattab *, E. Zubkov, A. Bernhart, G. Jnawali, C. Bobisch, B. Krenzer, M. Acet, R. Möller, M. Horn-von Hoegen

Department of Physics and Center for Nanointegration Duisburg-Essen (CeNIDE), University of Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Germany

Received 21 September 2007; received in revised form 15 January 2008; accepted 26 February 2008
Available online 2 March 2008

Abstract

Smooth and epitaxial thin bismuth (Bi) films with low defect density were grown on Si(001) by molecular beam epitaxy. The film quality is characterized by *in situ* spot profile analysis low-energy electron diffraction and scanning tunneling microscopy, and *ex situ* atomic force microscopy and X-ray diffraction. The complete process is accomplished in three steps. Firstly, a template of a strained 6 nm Bi(111) film is grown at 150 K. Secondly, during annealing to 450 K the strain is relieved by the formation of an ordered array of misfit dislocations at the interface. Finally, additional Bi is deposited at 450 K up to the desired thicknesses of the Bi film. The film consists of 90° rotated and twinned μm size crystallites with a terrace size larger than 100 nm and an overall roughness of only 0.6 nm. A 25 nm thick Bi film is relaxed to Bi bulk lattice constant which is confirmed by X-ray diffraction.

Keywords: Bismuth; Low-energy electron diffraction; Surface roughness; Defects; Scanning tunneling microscopy; X-ray diffraction

1. Introduction

The fabrication of high quality nanofilms on semiconductors has always been of high interest for applications in the fields of technology and basic research. The goal is the preparation of relaxed (or homogeneously strained) atomically flat single crystalline thin films with very low defect density. Such films are indispensable for the fabrication of microdevices or for the study of physical phenomena.

In particular, the preparation of bismuth (Bi) films has received much attention due to the remarkable electronic properties of Bi which result from its low carrier concentration, the small effective masses $m*$ and a highly anisotropic Fermi surface [1–3]. Due to its large Fermi wavelength $\lambda \sim 40$ nm, and large carrier mean free path l, Bi is a good candidate to study the quantum transport and finite-size effect in thin films [4,5]. The small value of $m*$ and the large value of l lead to a large magnetoresistance effect observed in bulk Bi [6], Bi nanowires [7,8], and Bi thin films [9]. Additionally, the semimetal Bi film transforms to a semiconductor at a critical thickness on order of 30 nm [10].

Bi was grown on various substrates by molecular beam epitaxy [10,11], electrodeposition [9], and RF magnetron sputtering [12]. On Si(111) epitaxial thin films were grown at room temperature (RT) by Nagao *et al.* [13,14] and Kammler *et al.* [15]. Those films are used to observe quantum well states [2]. Concerning the deposition on Si(001) polycrystalline Bi films of various thicknesses were grown at different temperatures [16]. In addition, there are few studies reporting on the monolayer regime of Bi adsorption and related reconstructions [17–19].

In this study we present the morphological and crystallographic proprieties of high quality epitaxial Bi films on Si(001) adopting a kinetic pathway of deposition. A 6 nm Bi template is deposited at low temperature, followed by an annealing step at a higher temperature, and additional deposition of Bi up to the desired thickness at 450 K. Here we report on a 25 nm Bi(111) film consisting of homogeneous, twinned and 90° rotated μm sized Bi crystallites with an overall roughness of only 0.6 nm. The film was characterized by *in situ* spot profile analysis

* Corresponding author.
E-mail address: hichem.hattab@uni-due.de (H. Hattab).

0040-6090/$ - see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.tsf.2008.02.038

low-energy electron diffraction (SPA-LEED) and scanning tunneling microscopy (STM), and *ex situ* atomic force microscopy (AFM) and X-ray diffraction (XRD).

2. Experiment

The experiments were carried out under ultra-high-vacuum (UHV) conditions at a base pressure $<2\times10^{-8}$ Pa. The chamber was equipped with SPA-LEED for *in situ* analysis of the surface morphology. As substrate, highly oriented Si(001) (Boron doped, miscut less than 0.2°) was used. The sample was cleaned with ethanol before inserting to the chamber and outgassed at 600 °C in UHV for several hours. After a flash annealing up to 1150 °C heated by direct current the native oxide layer was removed, and the LEED pattern shows a (2×1) reconstruction at 300 K and *c*(4×2) at 80 K indicating a clean Si(001) surface [20]. The sample was cooled by a liquid nitrogen cryostat attached to the sample holder. Bi (Mateck GmbH, Purity 99.9999%) was evaporated from a thermally heated ceramic crucible [21] at a pressure of 3×10^{-8} Pa. The evaporation rate (0.28 nm/min) was monitored *in situ* by a quartz microbalance. The film thickness was calibrated using the intensity oscillations of the (00)-spot during the deposition at 150 K [22] which occur in a bilayer-by-bilayer mode [14]. Additionally the total coverage of the film was also measured by *ex situ* AFM. The bulk quality of the film was characterized using *ex situ* XRD (Cu-K_α radiation). The local structure of the Bi(111) surface was studied in a separate UHV chamber equipped with STM. Those films were prepared at a residual pressure of 1×10^{-7} Pa.

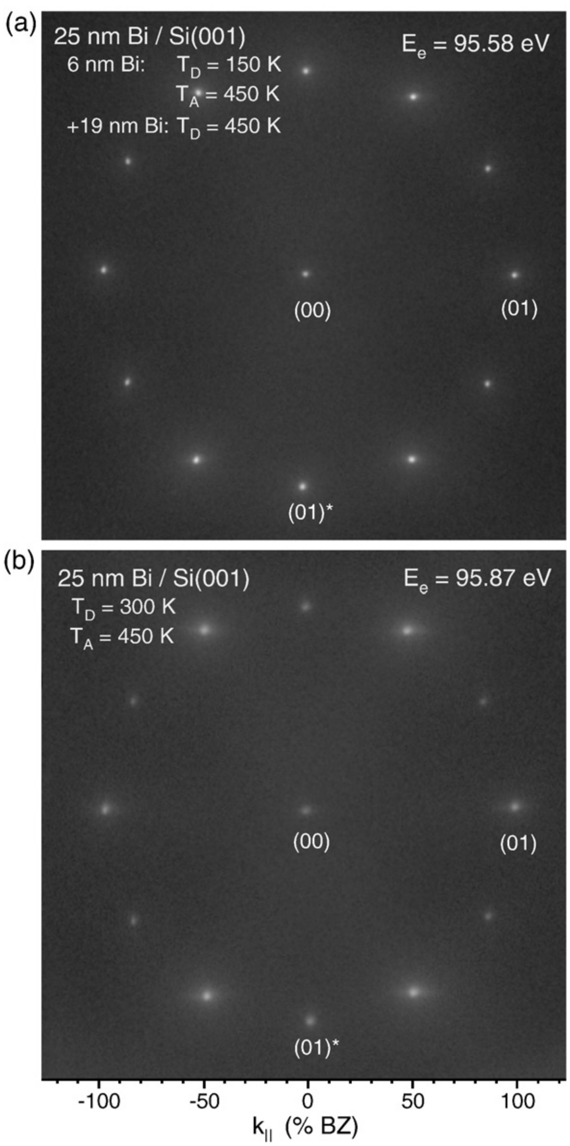

Fig. 1. LEED patterns of 25 nm Bi film on Si(001): (a) prepared with the method described in the text, (b) deposited at RT and annealed to $T_A=450$ K.

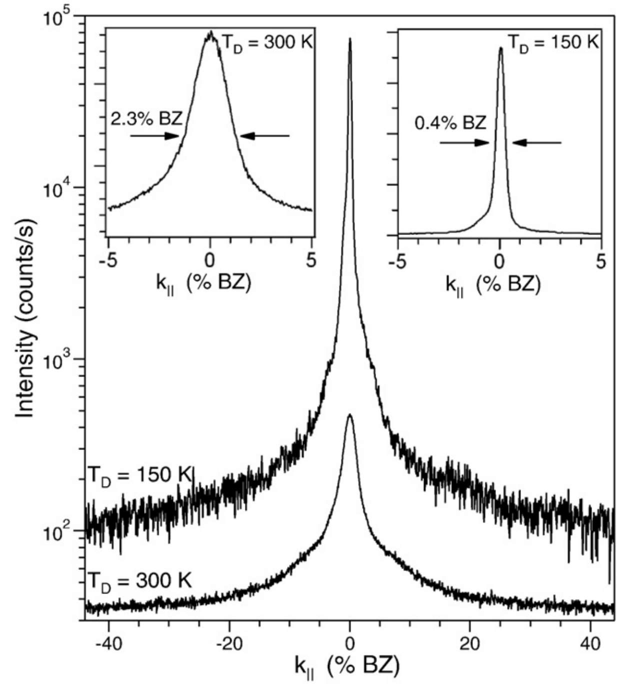

Fig. 2. LEED (00)-spot profiles in a logarithmic scale of a 25 nm Bi film on Si(001) taken at in-phase condition: upper curve prepared with the method described in the text, bottom curve deposited at RT and annealed to $T_A=450$ K. Insets show the profiles in a linear intensity scale close-up of the (00)-spot taken at the in-phase condition, which is sensitive to bulk defects only (small angle mosaics, screw dislocations, stacking faults *etc.*) [26,27].

3. Results and discussion

The deposition of a 6 nm Bi film at 150 K onto Si(001)*c*(4×2) results in a LEED pattern which has a quasi-12-fold symmetry of the integer order spots. This pattern can be explained by a superposition of two hexagonal Bi(111) LEED patterns, which are rotated by 90° with respect to each other (see also Fig. 1).

From the separation between (00)-spot and the first integer order spots we derived a row distance of 3.88±0.02 Å of the Bi atoms. This corresponds to a lattice parameter of 4.48 Å. Comparing with the Bulk lattice constant of Bi ($a_{Bi}=4.54$ Å [3,23]) the film is compressively strained by 1.3%.

Annealing up to 450 K (annealing rate = 10 K/min) results in the generation of an ordered array of misfit dislocations at the interface accommodating the lattice mismatch of 2.3% between