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Growth of epitaxial Bi-films on vicinal Si(111)

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ABSTRACT

Vicinal semi-metallic Bi-films are expected to reveal topologically protected edge states. In this study the growth of Bi-multilayer structures on Si(557) substrates has been investigated by low energy electron diffraction. Thereby, wetting layer structures formed prior to the film deposition on Si(557) surfaces turned out to be crucial for epitaxial growth. Only in the presence of Bi-wetting layers can well-ordered films be grown. In contrast to growth on Si(111), the pseudo-cubic surface of Bi(110) dominates. In addition, Bi(221) surfaces have been obtained only on wetting layers formed by less than a monolayer. The formation of Si(335)-facets during formation of the wetting layers turns out to be essential for the growth of the these structures.

1. Introduction

The semi-metal bismuth (Bi) reveals intriguing physical properties compared to conventional metals [1–3], which partly even resemble topological insulators [4]. Surface states, e.g., are electrically much more conductive than bulk states, which become readily insulating due to quantum confinement in nano-sized structures. For very thin Bi-films even topologically protected band structures are reported [5]. Therefore, details about the surface states in epitaxially grown Bi-films have been studied intensively with transport in the recent past [6–10].

The spin-orbit coupling in Bi is evident from pronounced Rashba-splitting of the surface states, which gives rise to spin-polarized transport. It limits the allowed channels of electron scattering by defects, so that, e.g., direct back scattering without spin-flip is not possible as long as spin remains a good quantum number. This selection rule can be lifted by adsorption of magnetic impurities [11–14].

Vicinal Bi(111) surfaces reveal further attractive features, as recently shown for Bi(114) crystals. An example is the existence of a one-dimensional and spin-split electronic state similarly to edge states in spin-Hall systems [3,15]. These edge states have been characterized so far only spectroscopically. For probing these states by transport, controlled epitaxial growth of vicinal Bi-films is mandatory.

Si substrates have been shown to be ideal templates for the growth of well-defined and at the same time electronically decoupled metallic nanostructures. In this respect atomically thin films and wires represent the ultimate realization of these concepts [16,17]. For the growth of metallic films, the lattice mismatch as well as surface and interface energies are crucial parameters, which control finally the growth mode and the film quality. Sticking to the example of growth of Bi on Si(111), the film roughness is larger when grown on a $\sqrt{3} \times \sqrt{3}$-order wetting layer than for growth on Si(111)-7 × 7 [19]. In order to explain these findings as well as the film thickness dependence, also allotropy needs to be considered, which is a pronounced effect in semimetals. E.g., the growth of Bi on Si(111) starts in (110) orientation, while (111)-textures appear for larger thickness [18]. Although the preparation of vicinal semi-metallic films is highly desirable in order to study in more detail the properties just mentioned, these considerations show already that the growth of vicinal films is a challenging task.

Therefore, as first step towards this field of stepped semi-metallic films, we have systematically varied the structure of the wetting layers formed by various amounts of Bi on Si(557) and studied their influence on further film growth. As it turns out, for Bi coverages ranging from 1/3 of a monolayer (ML) to 1 ML not only the local density of Bi atoms of the wetting layer changes, but also the step morphology of the Si(557) surface is modified. For wetting layers of less than a monolayer of Bi, (335)-oriented facets are formed. Stepped Bi(110) films can be grown only on these facet structures. To the best of our knowledge this is the first study which deals with the growth of stepped Bi-films on vicinal Si substrates.

2. Experimental setup

The experiments were performed in an ultra-high vacuum system operating below a base pressure of 1×10^{-10} mbar. The system hosts a low energy electron diffraction system with the capability for spot profile analysis (SPA-LEED) in order to analyze the quality of the substrate and the films with high resolution. Clean Si(557) substrates have been obtained by several heating cycles to 1370 K for a few seconds after the sample had been degassed for hours at 870 K. Bi was evaporated from a Knudsen cell. The amount of Bi was controlled by a

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quartz microbalance and the coverage has been calibrated by characteristic reconstructions on nominally flat Si(111) surfaces, and by oscillations in conductivity due to layer-by-layer growth of Bi on preannealed Bi-films. Wetting layers with different concentrations of Bi have been realized by adsorption of 5ML at 300 K followed by annealing to various temperatures. Annealing for 1 min at 820 K and 620 K results in a remaining 1/3 ML (so-called α-phase) and 1 ML (β-phase) coverages, respectively. For intermediate annealing temperatures, e.g. 720 K, a nominal coverage of 2/3 ML remains, which we refer to as γ-phase in this work.

Bi films have been grown by deposition of Bi at 200 K followed by annealing to 350 K. The wetting layer coverage is given in monolayers with respect to Si(111) (1 ML = 7.84×10^{14} atoms/cm^2). For Bi-films we use two different calibrations: for the (111)-textured films the coverage is given in bilayers (1 BL = 1.14×10^{15} atoms/cm^2) while we refer to monolayers in the case of Bi(110) (1 ML = 9.27×10^{14} atoms/cm^2). The indexing of the Bi-films is given with respect to the rhombohedral notation. For a definition of the basic vectors $\overrightarrow{a_1}$, $\overrightarrow{a_2}$ and $\overrightarrow{a_3}$, see Fig. 5c).

3. Results and discussion

The growth of semi-metallic films depends crucially on details of the wetting layers. Therefore, the different Bi-induced facet structures depending on Bi coverage will be presented first. The growth of Bi-films of about 10 nm in thickness is studied in the second part of this paper. We want to emphasize that we restrict ourselves in this study to film thicknesses above 10 nm in order to avoid the influence of allotropic phase transitions, which have been shown to occur for thinner films [18,19].

3.1. Wetting layers of Bi on Si(557)

The Si(557) surface is a non-homogeneously stepped surface along the $[\bar{1}\bar{1}2]$-direction. The (7 × 7) reconstruction on small (111) terraces as well as dimerization at the step edges of (112)-oriented facets favor the alternating arrangement of these two facets 9 and 8 atomic units wide, respectively. They are separated by three bilayer steps so that the unit cell is $(17 \times 3.32\text{Å})^2 + (3 \times 3.13\text{Å})^2]^{1/2} = 57\text{Å}$. More details are described in refs. [21–25].

Upon adsorption of Bi and annealing, the reconstructions of the Si(557) surface changes, as illustrated by the LEED patterns after growth of Bi wetting layers with nominally 1/3, 2/3, and 1 ML shown in Fig. 1. Common to all phases is the appearance of $\sqrt{3} \times \sqrt{3}$ diffraction spots (blue dashed lines) compatible with the trimer-model for Bi adsorption on Si(111) [26].

Compared to the adsorption on Si(111), the LEED patterns shown in Fig. 1 reveal further substructures, which depend on Bi concentration, e.g. splitting of the integer spots ((1 × 1) unit cell marked by red dotted lines) along the $[\bar{1}\bar{1}2]$ direction. We will start the discussion with the situation after adsorption of 1 ML Bi, i.e. appearance of the Si(557)–γBi-phase. The line scan through the (00)-spot shown as inset in Fig. 1c) reveals a spot splitting of 5.8% SBZ (SBZ = surface Brillouin zone), equivalent to a periodicity with 17 atomic units. In other words, the initial step structure remains unchanged. Compared to diffraction patterns obtained from clean Si(557), only few spots of the step train are visible, similar to recent results obtained by adsorption of 1 ML Ag on Si(557) [27], which is indicative of modified form factors and/or reduced long-range ordering.

When wetting layers on Si(557) are formed with submonolayer coverages of Bi, the spot splitting is fundamentally different from the clean Si(557) surface. In order to quantify the strong refacetting process upon formation of these wetting layers, line scans have been taken along the $[\bar{1}\bar{1}2]$ direction through the (00)-beam at various scattering conditions S. These were plotted as a gray-scale coded picture (Fig. 2a–c). From such plots (called $(k_{||},k_\perp)$-plots in the following) the formation and inclination of faceted areas can be directly read off. The scattering phase S ($S = \frac{2\pi}{k_\perp d}$) plotted in Fig. 2 is given with respect to the step height of Si(111) of d = 3.13 Å. $k_\perp$ denotes the vertical component of the scattering vector. As deduced from the intersections of the rods with the Bragg points in Fig. 2a), (113)- and (335)-facets appear after adsorption of 1/3 ML of Bi and corroborate the Bi-induced refacetting of the Si(557) substrate. The (335)-facet has an inclination of 14.4° with respect to the (111)-planes, which is significantly larger than the average tilt angle of the (557)-surface (9.45°). A sketch of this facet orientation in real space is shown in Fig. 2d). This demands other facet orientations in order to maintain the average inclination, or, alternatively, a widening of the (111)-oriented terraces already present on the (557) surface. The high intensity along the (111)-rods of Fig. 2a indicates that this may have happened. All other orientations, are obviously not periodically arranged and are visible in LEED only as background, as already found earlier for Pb/Si(557) [28]. The (113)-facets are a minority species, as evident from the faint intensities.

Although the (335)-facets have a width of only 12.1 Å $\left(3\frac{2}{3} \times 3.32\text{Å}\right)$ the facets are covered with Bi. This becomes obvious when analyzing facet rods of the $\sqrt{3}$-reconstruction spots shown in Fig. 2b). The inclination of the rods is identical to that before for the integer-spots indicating also a strong correlation between reconstructions on adjacent terraces.

Long range-ordered arrays with (335)-facets have been found only on the Si(557)–αBi-phase, i.e. at 1/3 ML Bi coverage (Fig. 2d). This means that there is a Bi-induced minimum of (free) energy for this

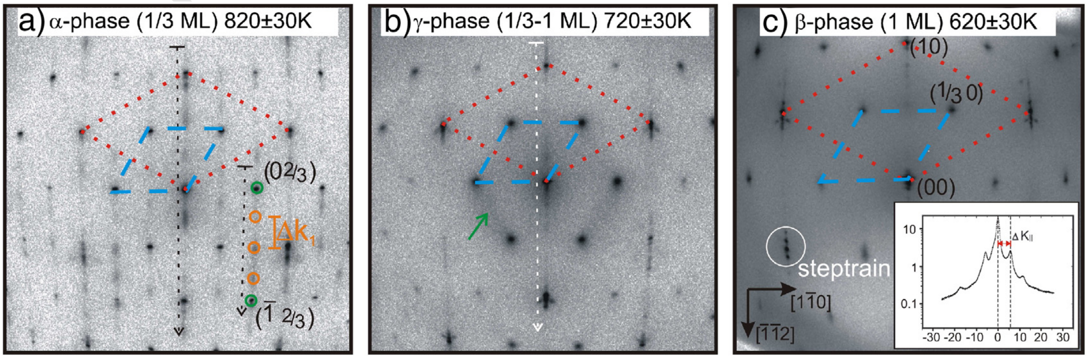

Fig. 1. Bi-wetting layers on Si(557) with coverage of (a) 1/3 ML (α-phase), (b) 2/3 ML (γ-phase) and (c) 1 ML (β-phase). The electron energies were 140 eV (a, b) and 80 eV (c). The dotted (red) and dashed (blue) rhombi denote the unit cells of the 1 × 1 and $\sqrt{3} \times \sqrt{3}$ structures, respectively. Images were taken at 80 K. The inset in c) shows a line scan taken along the $[\bar{1}\bar{1}2]$ direction through the central spot (log-intensity vs. SBZ, $\Delta K_{||} = 5.8$ % SBZ). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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