Scanning tunneling microscopy observation of surface superstructures during the growth of In on In/Si(111) surface

Maojie Xu a,b,e, Xiao-Ming Dou a,c, Jin-Feng Jia a, Qi-Kun Xue d, Yafei Zhang e, Arifumi Okada b, Shoji Yoshida b, Hidemi Shigekawa b

a Department of Physics and Shanghai Jiao Tong University, 200240 Shanghai, China
b Institute of Applied Physics, CREST-JST, University of Tsukuba, 305-8573 Tsukuba, Japan
b Shanghai Key Laboratory of Modern Optical System, University of Shanghai for Science and Technology, 200093 Shanghai, China
c Department of Physics, Tsinghua University, 100084 Beijing, China
d University of Shanghai for Science and Technology, 200093 Shanghai, China
f Research Institute of Micro/Nano Science and Technology, Shanghai Jiao Tong University, 200240 Shanghai, China

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A B S T R A C T

Surface superstructures are studied with scanning tunneling microscopy during the growth of In on In/Si(111). On the inhomogeneous substrate of Si(111) 4×1/√3×√3-In coexisting surface, the deposition of 1.5 monolayer (ML) In at about 0 °C leads to √7×√7 reconstruction on √3×√3 surface and the formation of one-dimensional nanowires on 4×1 surface (1.0 ML = 7.8×10^14 atoms/cm^2). Subsequent deposition of In at −100 °C gives rise to the appearance of a hexagonal superstructure with √37 × 4√3 periodicity on √7 × √7 reconstructed surface, while self-alignment of In dots along one-dimensional nanowires is observed on the initial 4×1 surface. On Si(111) √3 × √3-In surface, the growth of 2.5 ML In at about −100 °C yields 6×6 superstructure. The strain in the epitaxial In thin films provides a driving force for the formation of self-organized surface structures. © 2011 Elsevier B.V. All rights reserved.

1. Introduction

The epitaxial growth of In thin films on Si substrates has attracted considerable interests from both an experimental and theoretical point of view. Among them, In/Si(111) system is of particular interest. A variety of surface reconstructions and superstructures have been extensively studied for the growth of In on Si(111) substrate. Various phases, such as √3 × √3[1–9], √31 × √31[10–12], 4×1[2,13–22], and √7 × √7[23–27], have been reported for the growth of submonolayer or near-monolayer In on Si(111) surface. Unusual physical properties have been observed in these In thin layers. The 4×1 reconstructed surface is found to exhibit a metal-insulator phase transition at low temperature [20], and its control by optical doping has recently been explored [28]. The √7 × √7-In layer is characterized by the nearly free two-dimensional electronic gases [26,27]. The deposition of In on Si (111) √3 × √3-In reconstructed surface at room temperature results in the √3 × √3→2×2→1×1→√7 × √7→1×1 structural transformation and the variation of electronic properties of the overlayers with the increasing In coverage [29,30]. For the growth of 3.0 ML In on √3 × √3-In surface at a sample temperature of about −100 °C, 6×6 superstructure was observed using low energy electron diffraction (LEED) [31], which is associated with the lattice misfit between the In overlayer and the substrate.

In this paper, we report the results on surface superstructures observed by scanning tunneling microscopy (STM) for the growth of In thin films on In/Si(111) surface. The strain in the In overlayers induced by the lattice-mismatched substrate is found to give rise to the spontaneous formation of ordered surface structures. On Si(111) 4×1/√3×√3-In coexisting surface, the deposition of 1.5 ML In at 0 °C gives rise to √7 × √7 double-layer superstructure on √3 × √3 area and one-dimensional (1D) nanowire formation on 4×1 area. Subsequent deposition of In at about −100 °C yields a hexagonal superstructure with √37 × 4√3 periodicity on √7 × √7 reconstructed surface, while self-aligned In dots along 1D nanowires are found on the initial 4×1 area. The ordering of In dots in √37 × 4√3 hexagonal structure is attributed to the long-range indirect interaction between the dots mediated by the underlying substrate. On Si(111) √3 × √3-In surface, 6×6 superstructure of the three-layer In thin film was resolved for the growth of 2.5 ML In at −100 °C. A structural model is proposed by considering the strain relaxation through lattice incorporation between In overlayer and Si (111) substrate, and introducing both face centered cubic (fcc) and hexagonal close-packed (hcp) stacking sequences into the overlayer to further reduce the surface free energy.
2. Experimental procedure

Experiments were conducted with UNISOKU ultrahigh-vacuum STM system at a basic pressure of $1.5 \times 10^{-8}$ Pa. Si(111) samples were degassed below 600 °C overnight with a basic pressure better than $5.0 \times 10^{-8}$ Pa, and then slowly heated up to 900 °C prior to quick flashing to 1200 °C. After preparing a clean Si(111) surface, a heat-resistant evaporator was used to deposit In on the surface at a flux of 0.3–6.0 ML/min. STM images were taken at room temperature and about 80.0 K in the constant current mode.

Fig. 1. STM images of (a) $\sqrt{7} \times \sqrt{3}$ double-layer structure and 1D nanowires grown by 1.5 ML In deposition at around 0 °C on the inhomogeneous Si(111) $4 \times 1/\sqrt{3} \times \sqrt{3}$-In substrate ($V_S = -1.0$ V); (b) a hexagonal superstructure of $\sqrt{37} \times 4/\sqrt{3}$ periodicity on $\sqrt{7} \times \sqrt{3}$ double-layer structure and self-aligned In dots on 1D nanowires formed after subsequent adsorption of 0.15 ML In at about $-100$ °C ($V_S = -1.0$ V).

Fig. 2. Bias-dependent STM images of the hexagonal superstructure on $\sqrt{7} \times \sqrt{3}$ double-layer structure.
3. Results and discussion

After a Si(111) 4 × 1/√3 × √3-In coexisting surface was grown as the substrate, 1.5 ML In was deposited at around 0 °C. A √7 × √3 hexagonal double-layer structure is observed on the √3 × √3 reconstructed surface, whereas 1D nanowires are formed on 4 × 1 area. Fig. 1 (a) shows the observed surface structures. The √7 × √3 double-layer structure was previously reported in Ref. [31]. The 1D nanowire array exhibits a structure similar to that recently observed [32]. Subsequent growth of 0.15 ML In at about 0 °C leads to the appearance of a hexagonal superstructure on √7 × √3 double-layer surface and the formation of self-aligned In dots on 1D nanowires, as shown in Fig. 1(b).

In the following section, the detailed hexagonal structure is presented. Fig. 2 shows bias-dependent STM images of the hexagonal structure. At negative bias voltages of −3.0 and −2.0 V (Figs. 2(a) and (b)), the phase shows a hexagonal structure similar to that in Fig. 1 obtained at −1.0 V. Bright six protrusions form the hexagonal structure, and a dark protrusion exists at the center of the structure. On a close view, the six bright sites forming one hexagon show a slight difference in brightness. The dark site at the center of the hexagonal structure becomes bright in the STM image with a positive bias voltage. At a low bias voltage of 0.3 V (Fig. 2(c)), the central site changes but still less bright than the other sites. At 0.7 V, all sites have a similar brightness as shown in Fig. 2(d).

Fig. 3 shows the mesh analysis of the hexagonal structure relative to the underlying √7 × √3 double-layer structure. A √7 × √3 periodicity is determined for the superstructure. Fig. 3(b) shows the magnified images of the √7 × √3 and hexagonal structures with unit cells of the hexagonal structure. For comparison, a negative-bias voltage (V = −1.0 V) image is shown for the hexagonal structure (lower one) in Fig. 3(b). As the positional relation between the √7 × √3 and hexagonal structures schematically shown in Fig. 3(c), the hexagonal structure has a √7 × √3 periodicity. The atomic arrangement of the √7 × √3 double-layer and hexagonal structures are not well resolved. On the basis of STM observation, each spot of the hexagonal structure is supposed to consist of several In atoms and corresponds to an In dot. The two spot sites of the hexagonal structure, which are observed with slightly different brightness in STM image at a negative bias voltage, locate on the similar substrate sites as shown in Fig. 3. To clarify the difference in their electronic structures, further study including theoretical analysis is required.

A schematic model of the hexagonal structure is depicted in Fig. 3(c). The indirect interaction between In dots mediated by the underlying substrate is an important factor for the formation of ordered hexagonal structure [33,34]. When In atom is deposited, lattice deformation arises from the adsorbate–substrate interaction and local strain field is generated. The overlapping of the strain fields gives rise to long-range interaction between adsorbed In dots of the hexagonal structure. Thereby, long-range elastic interaction between In dots can exist and contribute to the formation of ordered hexagonal structure.

Fig. 4 shows a strain-relief pattern of 6 × 6 structure observed for the growth of 2.5 ML In at ~100 °C on Si(111) √3 × √3-In surface. The 6 × 6 superstructure of the three-layer In film was reported in LEED investigation [31]. During the initial growth stage, the √3 × √3-In surface transforms into a pseudo-morphological layer with the structure similar to the underlying Si(111) 1 × 1 substrate. As the growth proceeds, the strain generated in the In overlayer is relaxed in the third...
layer when it is larger than a critical value. Therefore, the first and second layers are pseudo-morphological, while the third layer incorporates the In atoms with a density higher than underlying layers to relieve the tensile strain. Based on STM observation and previous investigations on the formation of strain-relief patterns [33–36], a structure model is proposed by considering the lattice matching between In(111) overlayer and Si(111) substrate. The lattice constant is 3.3 Å on the average for In(111) and 3.8 Å for Si(111), yielding a lattice misfit of about 15%. To relieve the tensile strain in the In overlayer, In atoms of higher density are incorporated into the third layer of In overlayer. The incorporation of every seven In atoms into the spacing of six Si lattice constants leads to a lattice misfit less than 1.5%. Such incorporation gives rise to the 6×6 periodic corrugation at the In/Si interface, resulting in a strain-relief pattern of 6×6 structure.

An approach to more efficiently relieve the tensile strain in the In thin film is devoted to the alternative packing of triangular domains with different stacking sequences of fcc and hcp in the third In layer [33,34]. The structural model in Fig. 4(b) shows a 6×6 unit cell with the hcp and fcc half cells, in which two types of stacking sequences are introduced to realize the incorporation of seven In atoms into six Si lattice constant spacing along [211] or [121] direction in the fcc half unit cell, 7-n atoms are found to correspondingly occupy the hcp site in the hcp half unit cell. Thereby, seven In atoms are incorporated into the six Si lattice constant spacing.

The energy difference between the fcc and hcp adsorptions in the In top layer is reduced by the strain in the In thin film. In a previous study, the stacking transition from fcc to hcp is found for the adsorption of Cu atoms surrounding the Cu islands on Au(111) surface, which is attributed to the stress due to the large lattice misfit between the top island and the substrate [35]. For Si(111) 6×6-In structure, as the lattice misfit between the In layer and Si substrate is larger than 15%, a stacking transition from fcc to hcp is found to occur as well. Since the In atoms are adsorbed on the energetically favorable fcc and hcp sites in the top layer, the surface free energy would be lowered during the incorporation of every seven In atoms into the spacing of six Si lattice constants.

4. Conclusions

Surface superstructures were studied by STM during the growth of In at a high coverage (>1 ML) on In/Si(111) surfaces. The deposition of 1.5 ML In at around 0 °C on the inhomogeneous Si(111)4×1/√3 × √3-In substrate leads to √7 × √3 double-layer structure on √3 × √3-In area and 1D nanowire formation on 4×1 area. A √37 × 4√3 phase with a
hexagonal structure on $\sqrt{7} \times \sqrt{3}$ double-layer structure and self-aligned In dots on 1D nanowires were found for subsequent In deposition at $-100^\circ$C. The $6 \times 6$ superstructure was resolved for the growth of 2.5 ML In at about $-100^\circ$C on Si(111)$\sqrt{3} \times \sqrt{3}$-In surface. The strain generated in the In thin film by lattice-mismatched substrate offers a driving force for spontaneous ordering of the observed surface structures.

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References