Available online at www.sciencedirect.com
ScienceDirect
Applied Surface Science 254 (2008) 7733–7736
applied surface science
www.elsevier.com/locate/apsusc

Quasi-one-dimensional In atomic chains on Si(1 1 1) at low temperature studied by reflection high-energy positron diffraction and scanning tunneling microscopy

M. Hashimoto [a,*], Y. Fukaya [a], A. Kawasuso [a], A. Ichimiya [a,b]

[a] *Advanced Science Research Center, Japan Atomic Energy Agency, 1233 Watanuki, Takasaki, Gunma 370-1292, Japan*
[b] *Faculty of Science, Japan Women's University, 2-8-1 Mejirodai, Bunkyo-ku, Tokyo 112-8681, Japan*

Available online 9 February 2008

Abstract

We have investigated a quasi-one-dimensional structure of Si(1 1 1)-8 × 2-In surface using reflection high-energy positron diffraction (RHEPD) and scanning tunneling microscopy (STM). From the RHEPD rocking curve analyses, we confirmed the formation of the In hexagon structure proposed by González et al. [C. González, F. Flores, J. Ortega, Phys. Rev. Lett. 96 (2006) 136101]. Furthermore, we found that the empty-state STM image at 44 K is consistent with that calculated with the optimum hexagon structure by the RHEPD analyses.

PACS : 68.35.B-; 61.05.jh; 68.37.Ef

Keywords: Surface structure; Reflection high-energy positron diffraction (RHEPD); Scanning tunneling microscopy (STM); Silicon; Indium

1. Introduction

After depositing one monolayer of indium atoms on a Si(1 1 1)-7 × 7 surface, a Si(1 1 1)-4 × 1-In super-structure is formed. Many studies revealed that this super-structure is composed of quasi-one-dimensional metallic In chains. Each chain is further composed of two zigzag chains [1–3]. Below 130 K the phase transition from the 4 × 1 to the 8 × 2 phase occurs [4–6] and the metallic conduction vanishes [7–9]. This transition is considered to be derived by the Peierls instability with the formation of charge-density waves from angle-resolved photoelectron spectroscopy [7]. However, the atomic configuration of the 8 × 2 phase is still in debates.

From the SXRD [10] and the first-principles calculations [11,12] studies, it is proposed that each In chain of the 8 × 2 phase is composed of In trimers [12] (see Fig. 1a). It is not confirmed whether the band gap appears in this structure. Recently, González et al. proposed a hexagon structure from the first-principles calculations [13] (see Fig. 1b). In the band structure calculated with the hexagon structure, a band gap appears [13,14]. Moreover, the hexagon structure well explains the change in the electronic conductivity [5,14]. Thus, although the hexagon structure model is preferred to explain the insulating behavior of the 8 × 2 phase, no experimental evidence about the formation of this structure have been obtained.

In this study, we investigated the structure of the Si(1 1 1)-8 × 2-In surface using the reflection high-energy positron diffraction (RHEPD). The advantage of this method is the occurrence of the total reflection at low glancing angles [15,16]. In the total reflection region, the RHEPD intensity depends only on the topmost surface structure due to less penetration of positrons into the bulk. In light of the surface structure determined by the RHEPD experiment, we further examined the low-temperature scanning tunneling microscopy (STM) observations.

2. Experiment

The substrates were cut from a mirror-polished *n*-type Si(1 1 1) wafer (1–10 Ωcm). These were cleaned by direct

* Corresponding author at: Research Group for Positron Beam Materials Science, Advanced Science Research Center, Japan Atomic Energy Agency, 1233 Watanuki, Takasaki, Gunma 370-1292, Japan. Tel.: +81 27 346 9330; fax: +81 27 346 9432.
E-mail address: hashimoto.mie@jaea.go.jp (M. Hashimoto).

0169-4332/$ – see front matter
doi:10.1016/j.apsusc.2008.02.010

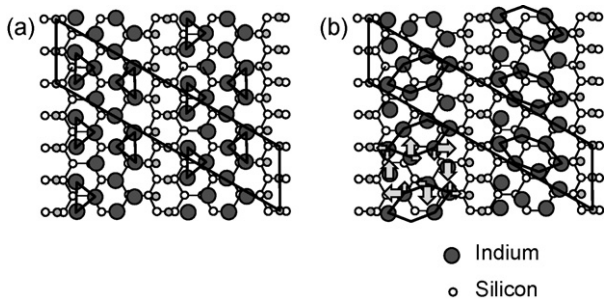

Fig. 1. Top view of the structure models for Si(1 1 1)-8 × 2-In surface; (a) trimer structure, which was determined experimentally and theoretically and (b) hexagon structure, which was proposed theoretically. The large and small arrows indicate the two different displacements of the In atoms when the surface structure is changed from the zigzag chain structure to the hexagon structure. Parallelograms indicate the unit cell of the 8 × 2 structure.

resistive heating at 1470 K for a few seconds, and then slowly cooled down to room temperature in an ultra-high vacuum (UHV) chamber evacuated to a base pressure of 5×10^{-8} Pa. The formation of clean Si(1 1 1)-7 × 7 surface was checked through the reflection high-energy electron diffraction (RHEED) and STM observations. To obtain a Si(1 1 1)-4 × 1-In surface, one monolayer of In atoms was deposited onto the Si(1 1 1)-7 × 7 surface at room temperature using alumina-coated tungsten basket and electron beam evaporator, followed by the annealing at around 670 K. The RHEPD experiments were performed with a positron beam generated by a ^{22}Na positron source and electromagnetic lenses. The details of the apparatus were described elsewhere [17]. The positron energy was set at 10 keV. The rocking curves were obtained by rotating the sample from 0.3° to 6.0° with an interval of 0.1°. The sample was cooled down to 60 K using a cryostat. The samples were also transferred to the low-temperature OMICRON VT-STM stage and the STM experiments were carried under the base pressure of less than 1×10^{-8} Pa in the temperature range of 44–104 K. All the STM images were obtained in the constant-current mode.

3. Calculation

Numerical calculation of STM image was conducted using the Advance/PHASE code [18] based on the density functional theory with a local density approximation provided by the AdvanceSoft Co. Ltd. A slab type super-cell (26.6 Å × 7.68 Å × 31.4 Å) composed of the 16 surface In atoms, 104 substrate Si atoms (three bilayers) and the 16 bottom hydrogen atoms that terminate the Si dangling bonds is constructed. The height of the vacuum layer is 21.2 Å. For the trimer structure, the theoretical In configuration was used [12]. For the hexagon structure, the In configuration determined by the above RHEPD experiment was used. For Si and In atoms, the norm conserving pseudopotential and the ultrasoft potential, respectively, are used. The cutoff energies of the plane-wave basis set are 25 Ry for the calculation of wave functions and 225 Ry for the calculation of the charge densities. The four *k*-points were sampled. The STM images were reconstructed by the charge densities.

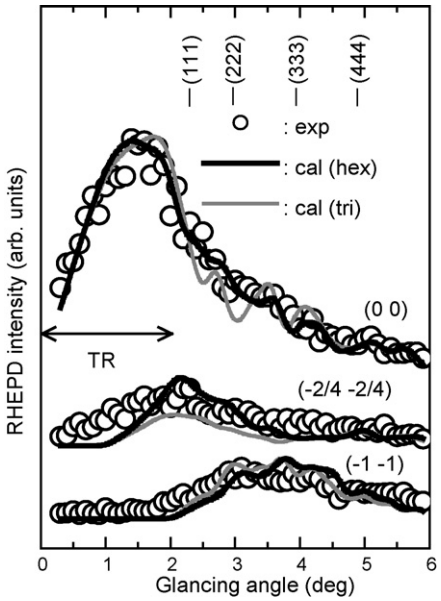

Fig. 2. RHEPD rocking curves from the Si(1 1 1)-8 × 2-In surface at the [1 $\bar{1}$ 0] direction at 60 K. The open circle and the solid line show the measured and calculated rocking curves, respectively. Gray and black lines indicate the rocking curve calculated using the theoretical trimer structure and the optimized hexagon structure, respectively. TR stands for the total reflection region.

4. Results and discussion

Fig. 2 shows the rocking curves form the Si(1 1 1)-8 × 2-In surface at 60 K and at the [1 $\bar{1}$ 0] incidence. The gray line exhibits the calculated rocking curve with the trimer structure [12] based on the dynamical diffraction theory. Using the trimer structure, the three small peaks are observed in the shoulder of the (0 0) rocking curve at glancing angles ranging from 2.5° to 4.5°. In the experimental curve, however, the small peaks observed in the shoulder at 2.5–4.5° disappear. The experimental feature at 2.5–4.5° is hardly reproduced by the trimer structure. The reliability factor defined in [19] is 2.7% [20]. Subsequently, we considered the possibility of the hexagon structure [13]. Starting from the zigzag chain structure to the hexagon structure, two particular displacement models ($\Delta\alpha$ and $\Delta\beta$) are required for individual In atoms, as indicated by the large and small arrows shown in Fig. 1b. We calculated the rocking curve by changing these two parameters so as to mimic the experimental curve. The solid line in Fig. 2 is the calculated rocking curve with the optimized hexagon structure [20]. In this case, the small peaks observed in the rocking curve calculated by assuming the trimer structure disappear. Moreover, the intensity of the (−2/4 −2/4) spot increases at around 2°. The experimental curve is reproduced in the whole angle range. We obtained $\Delta\alpha = 0.59$ Å and $\Delta\beta = 0.09$ Å that agree with those predicted from the theory ($\Delta\alpha = 0.62$ Å, $\Delta\beta = 0.21$ Å) [20]. The reliability factor is improved to be 2.1% [20], which is smaller than the experimental error (2.5%). We found that the above calculated curve is very close to the curve calculated with the theoretical hexagon structure [20]. Thus, we confirm that the hexagon structure is preferred to explain the experiment.

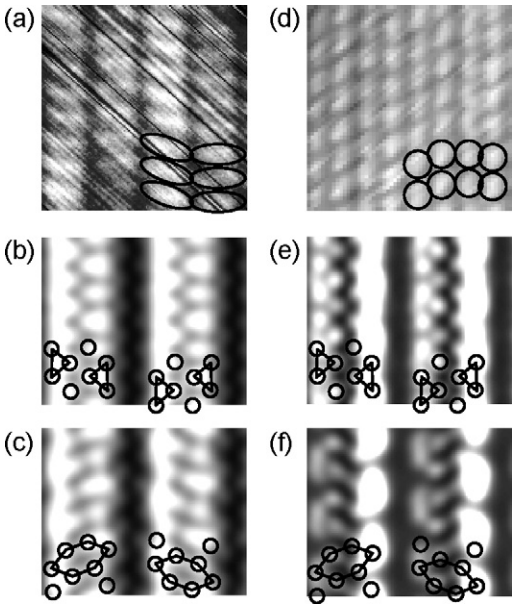

Fig. 3. Filled- (a–c) and empty-state (d–f) STM images of the Si(1 1 1)-8 × 2-In surface. The sample biases in the filled and empty states are −1.0 and +1.0 V, respectively. Experimental STM images in (a) and (d) were taken at 54 and 44 K, respectively. In (b) and (e), the STM images were calculated using the trimer structure. In (c) and (f), the images were calculated using the hexagon structure determined by the RHEPD.

Now, we confirm the validity of the structure of Si(1 1 1)-8 × 2-In determined by the RHEPD study through the STM observations and numerical calculation. Fig. 3a shows the filled-state STM images. Cocoon-like structures are observed [7,21–24]. Similar STM images at different sample biases are reported [21,22]. Thus, the experimental filled-state images are considered to be independent of sample bias. Indeed, this feature is consistent with the present numerical calculation as shown in the figure. The calculated STM image of the trimer structure (Fig. 3b) consists of bright stripes and protrusions arising from In chains and dark stripes arising from Si chains. The STM image of the trimer structure is similar to the Si(1 1 1)-4 × 1-In surface. These characteristics are in good agreement with those of the other calculations [11,12]. The calculated STM images for the hexagon structure (Fig. 3c) are very similar to those for the trimer structure. Therefore, in the filled-state case, no significant differences appear between the trimer and hexagon structures. Furthermore, the experimental STM images were different from the simulated STM images. The inconsistency may originate from the low spatial resolution for the filled-state images of the Si(1 1 1)-8 × 2-In surface [21].

Fig. 3d shows the empty-state STM images. Two different bright protrusions (indicated by circles) are clearly visible. This feature is in good agreement with the other experiments [21,22,25]. It should be noted that Park et al. and Kurata et al. took the images at much lower sample biases of +0.3 and +0.4 V, respectively. Fig. 3e shows the calculated empty-state image for the trimer structure. The dark stripes correspond to the Si chains between the In arrays. The STM image of the trimer structure is similar to the Si(1 1 1)-4 × 1-In surface and in good agreement with another simulation [11]. The calculated STM image for the trimer structure is not consistent with the experimental image. Fig. 3f shows the calculated STM image for the hexagon structure. Pair of protrusions appears on the In arrays and the trenches in the In arrays are observable. These features are in agreement with the experimental STM images. Consequently, the empty-state STM images can be explained by considering the hexagon structure.

In conclusion, the filled-state experimental STM images were different from the simulated STM images. The empty-state STM image at 44 K is consistent with that calculated with the optimum hexagon structure by the RHEPD analyses.

5. Summary

In summary, we have investigated a quasi-one-dimensional structure of Si(1 1 1)-8 × 2-In surface using the RHEPD, low-temperature STM and numerical calculation. We found that the structure of the In chain determined by the RHEPD is close to the hexagon structure obtained in the previous theoretical study [13]. The empty-state STM images are similar to the simulated images using the optimized hexagon structure. We confirmed that a quasi-one-dimensional structure of Si(1 1 1)-8 × 2-In surface is composed of the hexagon structure.

References

[1] J.J. Lander, J. Morrison, J. Appl. Phys. 36 (1965) 1706.
[2] J. Nogami, S.-I. Park, C.F. Quate, Phys. Rev. B 36 (1987) 6221.
[3] T. Abukawa, M. Sasaki, F. Hisamatsu, T. Goto, T. Kinoshita, A. Kakizaki, S. Kono, Surf. Sci. 325 (1995) 33.
[4] O. Bunk, G. Falkenberg, J.H. Zeysing, L. Lottermoser, R.L. Johnson, M. Nielsen, F. Berg-Rasmussen, J. Baker, R. Feidenhans'l, Phys. Rev. B 59 (1999) 12228.
[5] J. Nakamura, S. Watanabe, M. Aono, Phys. Rev. B 63 (2001) 193307.
[6] R.H. Miwa, G.P. Srivastava, Surf. Sci. 473 (2001) 123.
[7] H.W. Yeom, S. Takeda, E. Rotenberg, I. Matsuda, K. Horikoshi, J. Schaefer, C.M. Lee, S.D. Kevan, T. Ohta, T. Nagao, S. Hasegawa, Phys. Rev. Lett. 82 (1999) 4898.
[8] T. Kanagawa, R. Hobara, I. Matsuda, T. Tanikawa, A. Natori, S. Hasegawa, Phys. Rev. Lett. 91 (2003) 036805.
[9] T. Tanikawa, I. Matsuda, T. Kanagawa, S. Hasegawa, Phys. Rev. Lett. 93 (2004) 016801.
[10] C. Kumpf, O. Bunk, J.H. Zeysing, Y. Su, M. Nielsen, R.L. Johnson, R. Feidenhans'l, K. Bechgaared, Phys. Rev. Lett. 85 (2000) 4916.
[11] J.-H. Cho, D.-H. Oh, K.S. Kim, L. Kleinman, Phys. Rev. B 64 (2001) 235302.
[12] X. López-Lozano, A. Krivosheeva, A.A. Stekolnikov, L. Meza-Montes, C. Noguez, J. Furthmüller, F. Bechstedt, Phys. Rev. B 73 (2006) 035430.
[13] C. González, F. Flores, J. Ortega, Phys. Rev. Lett. 96 (2006) 136101.
[14] A.A. Stekolnikov, K. Seino, F. Bechstedt, S. Wippermann, W.G. Schmidt, A. Calzolari, M. Buongiorno, Phys. Rev. Lett. 98 (2007) 026105.
[15] A. Kawasuso, S. Okada, Phys. Rev. Lett. 81 (1998) 2695.
[16] A. Ichimiya, Solid State Phenom. 28–29 (1992/1993) 143.
[17] A. Kawasuso, T. Ishimoto, M. Maekawa, Y. Fukaya, K. Hayashi, A. Ichimiya, Rev. Sci. Instrum. 75 (2004) 4585.
[18] Advance/PHASE is an improved version of the *ab initio* simulation code PHASE for commercial use at AdvanceSoft Corporation. The original code PHASE has been developed at the Japanese national project Revolutionary Simulation Software 21th Century (RSS21).

[19] Y. Fukaya, A. Kawasuso, K. Hayashi, A. Ichimiya, Phys. Rev. B 70 (2004) 245422.
[20] Y. Fukaya, M. Hashimoto, A. Kawasuso, A. Ichimiya, unpublished.
[21] H. Morikawa, I. Matsude, S. Hasegawa, Phys. Rev. B 70 (2004) 085412.
[22] S.J. Park, H.W. Yeom, S.H. Min, D.H. Park, I.-W. Lyo, Phys. Rev. Lett. 93 (2004) 106402.
[23] S.V. Ryjkov, T. Nagao, V.G. Lifshits, S. Hasegawa, Surf. Sci. 488 (2001) 15.
[24] S.J. Park, H.W. Yeom, J.R. Ahn, I.-W. Lyo, Phys. Rev. Lett. 95 (2005) 126102.
[25] S. Kurata, T. Yokoyama, Phys. Rev. B 71 (2005) 121306.