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Observation of Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface at room temperature by reflection high-energy positron diffraction

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Abstract

We have observed the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface using reflection high-energy positron diffraction (RHEPD). The RHEPD pattern clearly displays the intense ($\pm 2/3$ $\pm 2/3$) spots, which results from the large magnitude of the scattering factor for the Ag atoms. The rocking curves of the RHPED at room temperature have been measured and analyzed by means of the dynamical diffraction theory. We found that the atomic height of the topmost Ag triangle is 0.77 Å and is well in accordance with those determined by the other methods.

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Keywords: Surface structure; Reflection high-energy positron diffraction (RHEPD); Total reflection; Silicon; Silver

1. Introduction

The Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface has been widely studied as a typical example of metal/semiconductor system. The surface structure of the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag was revealed as the honeycomb chained triangle (HCT) model by X-ray diffraction analysis [1], which is the Ag atoms of one monolayer adsorbed on the missing top layer of the Si crystal (Fig. 1). In 1999, the first-principles calculations and the low-temperature scanning tunneling microscopy (STM) observation showed that the ground-state structure is the inequivalent triangle (IET) model [2], in which the topmost Ag triangles are slightly rotated from those in the HCT model. Then, the question arose as to whether the room temperature phase consists of the IET or HCT structure [3,4]. The phase transition of the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface between low and room temperatures attracts much attention.

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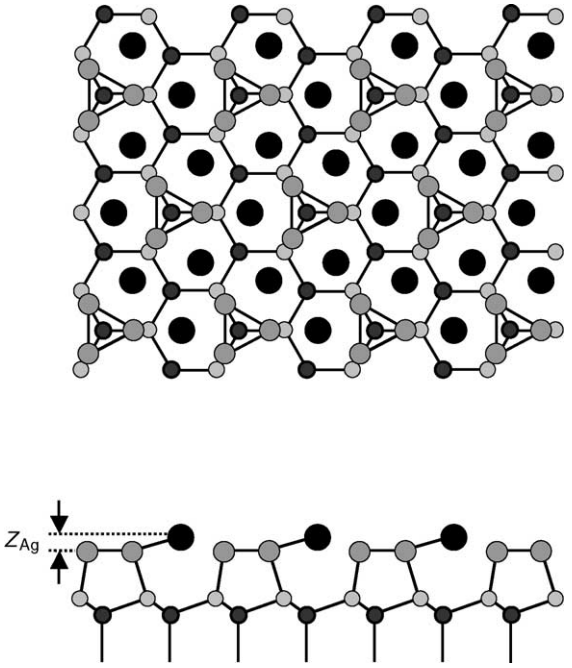

Fig. 1. Schematic drawing of the honeycomb chained triangle (HCT) model for the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface. The large black circle indicates the topmost Ag atom and the others are the inner Si atoms.

Reflection high-energy positron diffraction (RHEPD) is a new tool for the surface structure analysis and powerful to study the topmost surface structure and vibrational state [5,6]. It is a great advantage that the total reflection takes place because of the positive charge of positrons. The RHEPD intensity is very sensitive to the topmost surface because the incident positrons hardly reach the bulk region. Therefore, we are able to obtain selectively the information about the topmost atoms by using the RHEPD [7–9].

In this paper, we demonstrate the first observation of the RHEPD pattern from the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface. For the first step toward the solution of the phase transition, we also measured the rocking curves of the RHEPD from the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface at room temperature and determined the vertical atomic positions of the topmost Ag triangle by means of the intensity analysis based on the dynamical diffraction theory.

2. Experimental procedure

The substrate cut from an n-type Si(1 1 1) wafer was used (size: 15 mm × 5 mm × 0.5 mm, resistivity: 1–10 Ω cm). This was heated at 700 K for 6 h and flashed at 1500 K for 10 s a few times. After the observation of clear 7 × 7 pattern by reflection high-energy electron diffraction (RHEED), the Ag atoms of one monolayer was deposited on the Si(1 1 1)-(7 × 7) surface at 793 K using a W basket. The formation of the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag structure was confirmed by the RHEED.

The experiments were carried out in a UHV chamber equipped with electrostatic lens system and positron source of ^{22}Na. The details of the RHEPD apparatus were described elsewhere [10,11]. The incident positron energy was set at 20 keV. In the measurement of the rocking curve, the glancing angle (θ) was changed from 0.5° to 4.5° with a step of 0.1° by rotating the sample mounted on the manipulator. The RHEPD rocking curve was measured under the one-beam condition [12]. The RHEPD intensity under this condition is very sensitive to the change in the atomic positions and the number density perpendicular to the surface because the simultaneous reflections parallel to the surface are suppressed.

3. Results and discussion

Fig. 2 shows the RHEPD pattern from the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface at room temperature along the [1 1 $\bar{2}$] direction. In addition to the (0 0) spot, the intense ($\pm$2/3 $\pm$2/3) spots are observable in the relatively wide range of θ. The intensity of the integer-order spots such as (1 1) spot is very weak in the RHEPD, although in the RHEED the intense integer-order spots generally appear. The appearance of the bright ($\pm$2/3 $\pm$2/3) spots is explained by the kinematical diffraction theory taking into account only the Ag layer. Therefore, the appearance of the intense ($\pm$2/3 $\pm$2/3) spots is considered to be due to the large magnitude of the scattering factor for the Ag atoms.

Fig. 3 shows the RHEPD rocking curve from the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface at room temperature under the one-beam condition. In this case, the critical angle of the total reflection can be evaluated to be 1.4° using the mean inner potential of the Si crystal

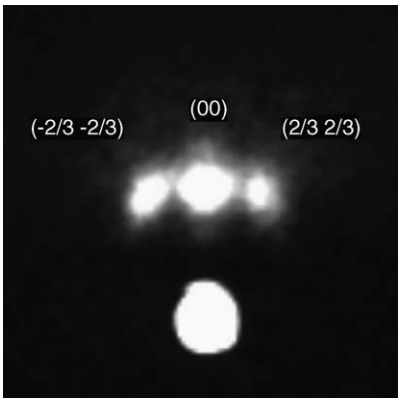

Fig. 2. RHEPD pattern from the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface at $\theta = 2^\circ$. The accelerating voltage is 20 kV and the incident azimuth is the [1 1 $\bar{2}$] direction.

(12 eV). Thus, the RHEPD intensity in the total reflection region ($\theta < 1.4^\circ$) is high. In addition to the intense (1 1 1) Bragg peak, (4 4 4) and (5 5 5) Bragg peaks are clearly seen, while (2 2 2) and (3 3 3) are not so clear. The incident positrons are diffracted at the topmost Ag layer under the total reflection condition. When the glancing angle is over the critical angle ($\theta = 1.4^\circ$) of the total reflection region, the incident positron penetrates the bulk Si layers. Thus, we are able to observe the Bragg peaks in the rocking curve at $\theta > 1.4^\circ$. These Bragg peak positions correspond to those expected from the bulk Si. However, since the scattering factor for Ag atoms much lager than that for Si atoms, it is considered that these peak positions depend on the interlayer distance between the topmost Ag layer and the underlying Si layers.

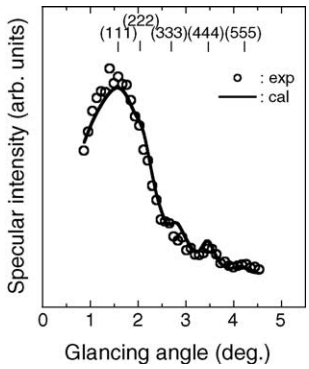

Fig. 3. RHEPD rocking curves from the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface under the one-beam condition. The open circle denotes the measured rocking curve. The solid line indicates the rocking curve calculated based on the dynamical diffraction theory using the optimum values of the Z_{Ag} and V'_{el}.

Now, we determine the atomic heights of the topmost Ag triangle comparing the measured rocking curve and the calculated one. The rocking curve is calculated from the dynamical diffraction theory taking into account the thermal diffuse scattering and the electronic excitation [8,13,14]. In the calculation, the atomic coordinates for the IET model, which were evaluated by the first-principles calculations [2], were used with respect to the Si layers below the topmost Ag layer. The thermal vibrational amplitudes of the topmost Ag atoms and the underlying Si atoms were 0.14 and 0.07 Å, respectively. The absorption potential due to the electronic excitation for the Si atoms was taken to be 1.22 V [15]. The RHEPD rocking curve under the total reflection condition is very sensitive to the height of the topmost atoms and the electronic excitation [7]. Thus, the height of the Ag atoms (Z_{Ag}) (see Fig. 1) and the absorption potential (V'_{el}) due to the electronic excitation were changed so as to reproduce the measured rocking curve. In order to compare the experiment and calculation qualitatively, we adopted the reliability factor (R) defined as

$$R = \sqrt{\sum_{\theta} (I_{\theta}^{exp} - I_{\theta}^{cal})^2},$$

where $\sum_\theta I_\theta^{exp} = \sum_\theta I_\theta^{cal} = 100\%$ [8]. Smaller R indicates better agreement between measured and calculated rocking curves.

From the minimum R ($R = 1.70\%$), the Z_{Ag} is determined to be 0.77 Å. On the contrary, the V'_{el} is nearly 0 V. Therefore, the electronic excitation does not play a significant role of the absorption effect, which is consistent with the result previously reported for the Si(1 1 1)-(7 × 7) surface [7]. It should be noted that even if the atomic coordinates below the Ag layer for the HCT model was used, the result does not almost changed. The solid line in Fig. 3 represents the rocking curve calculated using the optimum values. The calculated rocking curve is in good agreement with the measured one.

The Z_{Ag} obtained in this study is consistent with the values estimated by X-ray diffraction [16], RHEED [17], and first-principles calculation [2]. Our result is very close to the interlayer spacing (0.78 Å) of the double layer in the bulk Si. As mentioned above, the

each Bragg peak position in the measured rocking curve corresponds to that evaluated by the kinematical calculation using the ideal bulk Si positions. In the dynamical calculation, the peak positions shift when the Z_{Ag} is varied. Thus, the agreement of the Bragg peak positions suggests that each layer is situated at the ideal bulk position.

Finally, owing to the first-principles calculations, the atomic coordinates normal to the surface for the HCT and IET models have no sufficient difference [2]. Therefore, we hardly identify whether the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface at room temperature is made from the HCT or IET structures from the analysis of the rocking curve under the one-beam condition. In order to distinguish them, analyses of the rocking curves along the symmetric azimuth are now under the progress.

4. Summary

In summary, we demonstrated the first observation of the RHEPD patterns from the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface. Moreover, we determined the atomic height of the topmost Ag triangle using the RHEPD rocking curve analysis under the one-beam condition. The topmost interlayer distance of 0.77 Å obtained in this study is in good agreement with those determined by the other methods.

References

[1] T. Takahashi, S. Nakatani, N. Okamoto, T. Ishikawa, S. Kikuta, Jpn. J. Appl. Phys. 27 (1988) L753.
[2] H. Aizawa, M. Tsukada, N. Sato, S. Hasegawa, Surf. Sci. 429 (1999) L509.
[3] H. Tajiri, K. Sumitani, S. Nakatani, A. Nojima, T. Takahashi, K. Akimoto, H. Sugiyama, X. Zhang, H. Kawata, Phys. Rev. B 68 (2003) 035330.
[4] I. Matsuda, H. Morikawa, C. Liu, S. Ohuchi, S. Hasegawa, T. Okuda, T. Kinoshita, C. Ottaviani, A. Cricenti, M. D'angelo, P. Soukiassian, G.L. Lay, Phys. Rev. B 68 (2003) 085407.
[5] A. Kawasuso, S. Okada, Phys. Rev. Lett. 81 (1998) 2695.
[6] A. Ichimiya, Solid State Phenom. 28/29 (1992/1993) 143.
[7] A. Kawasuso, Y. Fukaya, K. Hayashi, M. Maekawa, S. Okada, A. Ichimiya, Phys. Rev. B 68 (2003) 241313.
[8] Y. Fukaya, A. Kawasuso, K. Hayashi, A. Ichimiya, Phys. Rev. B 70, in press.
[9] A. Kawasuso, T. Ishimoto, Y. Fukaya, K. Hayashi, A. Ichimiya, J. Surf. Sci. Nanotechnol. 1 (2003) 152.
[10] A. Kawasuso, S. Okada, A. Ichimiya, Nucl. Inst. Meth. Phys. Res. B 171 (2000) 219.
[11] T. Ishimoto, A. Kawasuso, H. Itoh, Appl. Surf. Sci. 194 (2002) 43.
[12] A. Ichimiya, Surf. Sci. 192 (1987) 893.
[13] A. Ichimiya, Jpn. J. Appl. Phys., Part 1 22 (1983) 176.
[14] S.L. Dudarev, L.-M. Peng, M.J. Whelan, Surf. Sci. 330 (1995) 86.
[15] G. Radi, Acta Crystallogr. 26 (1970) 41.
[16] T. Takahashi, S. Nakatani, Surf. Sci. 282 (1993) 17.
[17] A. Ichimiya, S. Kohmoto, T. Fujii, Y. Horio, Appl. Surf. Sci. 41/42 (1989) 82.