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Structural analysis of Si(1 1 1)-$\sqrt{21} \times \sqrt{21}$-(Ag, Au) surface by using reflection high-energy positron diffraction

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Abstract

The Au adsorption induced $\sqrt{21} \times \sqrt{21}$ super-lattice structure on the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag structure has been investigated using reflection high-energy positron diffraction. The height of the Au adatom was determined to be 0.59 Å from the underlying Ag layer from the rocking curve analysis with the dynamical diffraction theory. The adatoms were preferentially situated at the center of the large Ag triangle of the inequivalent triangle structure of the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag substrate. From the intensity distribution in the fractional-order Laue zone, the in-plane coordinate of the Au adatoms was obtained.

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

1. Introduction

The deposition of noble metal (Cu, Ag, and Au) or alkali metal (Na, K, and Cs) atoms on the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface leads to the formation of $\sqrt{21} \times \sqrt{21}$ super-lattice structures [1–4]. The $\sqrt{21} \times \sqrt{21}$ structures have a high electrical conductivity as compared to the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag surface [1,2]. The band structures have been extensively studied by photoemission spectroscopy [5–9] and optical second-harmonic generation [10]. Among the $\sqrt{21} \times \sqrt{21}$ structure family, the surface structures induced by Au and Ag atoms were expected to be the same [11]. Several structure models have been proposed of the $\sqrt{21} \times \sqrt{21}$ structures. In early studies, Nogami et al. [12] and Ichimiya et al. [13] proposed the coordinates of the Au atoms using scanning tunneling microscopy (STM), respectively, as shown in Fig. 1a and b. In the Nogami model, five Au atoms in the unit cell are situated at the center of the Ag triangle of the Si(1 1 1)-$\sqrt{3} \times \sqrt{3}$-Ag substrate. In the Ichimiya model, three Au atoms are located at the center of the Si trimer. Tong et al. also proposed the structure model using STM, as shown in Fig. 1c [14]. The model contains four Ag atoms at the center of the Ag triangle. Tajiri et al. performed the structure analysis using surface X-ray diffraction (Fig. 1d) [15]. In the Tajiri model, five Au atoms are included in the unit cell, similar to the Nogami model. Recently, we observed the Si(1 1 1)-$\sqrt{21} \times \sqrt{21}$-Ag surface using the reflection high-energy positron diffraction (RHEPD) [16]. We determined the surface structure in the case of the Ag adatoms, as shown in Fig. 1e. Three Ag atoms on the center of the large Ag triangle of the inequivalent triangle (IET) structure surround the Si trimer. We also found that the height of the Ag atoms from the underlying Ag layer is considerably low (~0.53 Å).

RHEPD is a surface-sensitive tool to determine the surface structure [17,18]. Since the positron has a positive charge, the total reflection takes place at grazing incidence.[1] In the total reflection condition, the incident

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[1] The critical angle (θ_c) at which the total reflection takes place is given in the form $\theta_c = \arcsin\left(\frac{V_0}{E}\right)^{\frac{1}{2}}$ (Snell's equation) [18], where E and V_0 are the accelerating voltage of the incident positron beam and the mean inner potential of the crystal, respectively. In the case of $E = 10$ kV and $V_0 = 12$ V for the Si(1 1 1) surface, θ_c is equal to 2.0°.

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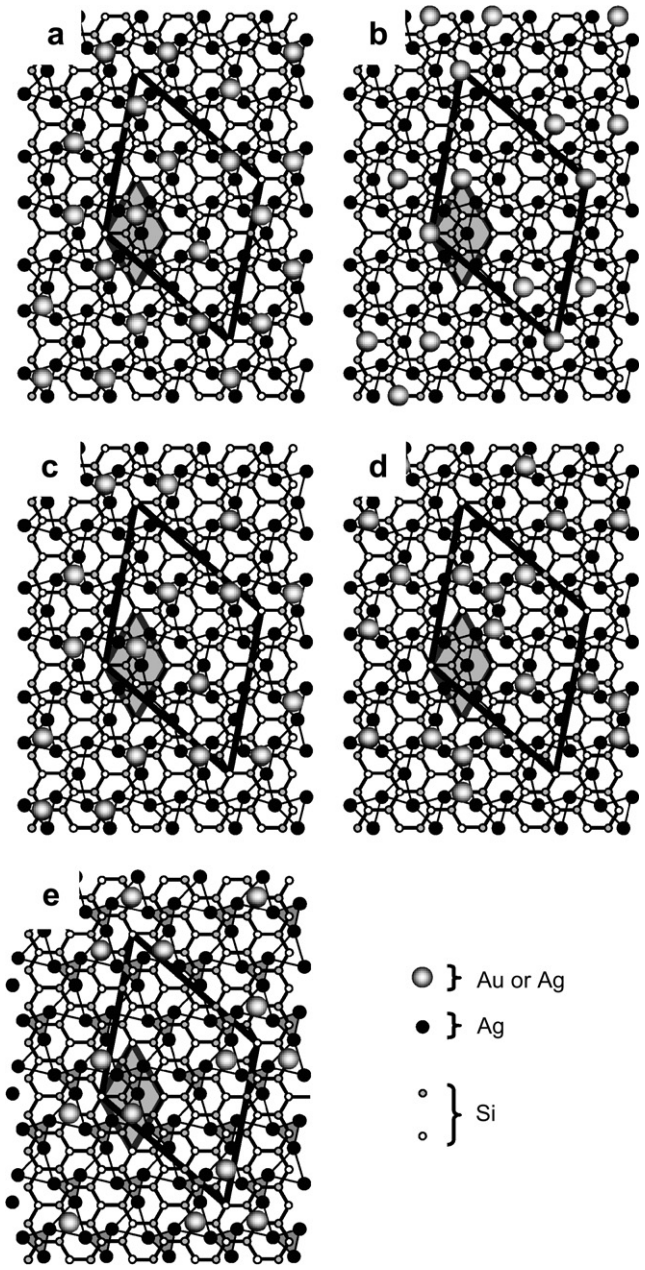

Fig. 1. Schematic drawings of the structure models for the Si(1 1 1)-$\sqrt{21}\times\sqrt{21}$-(Ag, Au) or Si(1 1 1)-$\sqrt{21}\times\sqrt{21}$-Ag surface. (a) Nogami model [12], (b) Ichmiya model [13], (c) Tong model [14], (d) Tajiri model [15], and (e) our model [16]. Large and small diamonds represent the unit cells of the $\sqrt{21}\times\sqrt{21}$ and $\sqrt{3}\times\sqrt{3}$ structures, respectively. Large and middle circles indicate the additional Ag or Au atoms and the underlying Ag atoms, respectively. The other circles show the inner Si atoms. In these figures, the Si trimers are not displayed. The underlying structures in (a)–(d) correspond to the honeycomb chained triangle (HCT) structure for the Si(1 1 1)-$\sqrt{3}\times\sqrt{3}$-Ag surface. In (e), the underlying structure is composed of the inequivalent triangle (IET) structure.

positrons are mostly reflected by the topmost surface layer without penetrating the bulk layers. Therefore, we are able to determine the adatom height and the thermal vibrational amplitude by means of the intensity analysis using the totally reflected positrons [19,20].

In this paper, we investigated the $\sqrt{21}\times\sqrt{21}$ structure induced by the deposition of Au atoms on the Si(1 1 1)-$\sqrt{3}\times\sqrt{3}$-Ag surface using the RHEPD. We measured the diffraction patterns and rocking curves at various conditions and determined the adsorption site of the Au atoms by means of the intensity analysis based on the dynamical diffraction theory. We will report the structure model of the Si(1 1 1)-$\sqrt{21}\times\sqrt{21}$-(Ag, Au) surface.

2. Experimental procedure

The substrate ($15\times5\times0.5$ mm^3) was cut from the mirror polished *n*-type Si(1 1 1) wafer with a resistivity of 1–10 Ω cm. The Si(1 1 1) substrate was rinsed in ethanol before introducing into a chamber. The sample was heated at 1470 K in 10 s a few times by passing a direct current in a ultra high-vacuum chamber. The sharp 7×7 spots were observed by using reflection high-energy electron diffraction. The Si(1 1 1)-$\sqrt{3}\times\sqrt{3}$-Ag surface was prepared by the deposition of Ag atoms on the Si(1 1 1)-7×7 surface held at 770 K. By the deposition (0.14 monolayer) of Au atoms on the Si(1 1 1)-$\sqrt{3}\times\sqrt{3}$-Ag surface at room temperature, the Si(1 1 1)-$\sqrt{21}\times\sqrt{21}$-(Ag, Au) surface was produced. The coverage of Au corresponds to three atoms per the $\sqrt{21}\times\sqrt{21}$ unit cell. The deposition rate was preliminary estimated from the formation of the Si(1 1 1)-5×2-Au surface (about 0.5 ML).

The experiment was carried out in a UHV chamber equipped with a positron source of ^{22}Na and magnetic lens system. The detail of the apparatus was described elsewhere [21]. The accelerated voltage of the incident positron beam was set at 10 kV. In the measurement of the rocking curve, the glancing angle (θ) was varied from 0.2° to 6.0° at a step of 0.1° by rotating the sample holder. The sample was cooled down to 110 K using liquid nitrogen. The temperature was measured using a thermocouple attached near the sample holder.

3. Results and discussion

Fig. 2 shows an example of the RHEPD pattern from the Si(1 1 1)-$\sqrt{21}\times\sqrt{21}$-(Ag, Au) surface. The azimuth of the incident beam corresponds to the [1 1 $\bar{2}$] direction. The glancing angle is set at 2.7°. We can clearly observe the fractional-order spots indexed by (8/21 11/21), (13/21 10/21), and (20/21 17/21). The intensity distribution in the pattern from the Si(1 1 1)-$\sqrt{21}\times\sqrt{21}$-(Ag, Au) surface is similar to that from Si(1 1 1)-$\sqrt{21}\times\sqrt{21}$-Ag surface [16]. This indicates that the atomic coordinates of these structures are similar to each other.

The determination of the atomic coordinate of the Si(1 1 1)-$\sqrt{21}\times\sqrt{21}$-(Ag, Au) surface proceeded at three steps, as described in detail in Ref. [16]. Firstly, in order to determine the height of the Au adatoms, we measured the RHEPD rocking curve under the one-beam condition. The specular spot is dominant under the one-beam condition and the intensity can be expressed by the atomic