Applied Surface Science 254 (2008) 7827–7830

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Surface structure and phase transition of Ge(1 1 1)-3 × 3-Pb studied by reflection high-energy positron diffraction

Y. Fukaya [a,*], M. Hashimoto [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

ARTICLE INFO

Article history:
Available online 29 February 2008

PACS:
68.35.Rh
68.35.Bs
61.14.Hg

Keywords:
Phase transition
Surface structure
Reflection high-energy positron diffraction (RHEPD)
Total reflection
Germanium
Lead

ABSTRACT

We studied the structures and the phase transition of Pb/Ge(1 1 1) surface by using the reflection high-energy positron diffraction. The surface structures at 60 K and 293 K have the 3×3 and $\sqrt{3} \times \sqrt{3}$ periodicities, respectively. The rocking curves measured at both temperatures are nearly the same. This indicates that the equilibrium positions of the surface atoms do not change according to the phase transition. From the analysis of the rocking curve based on the dynamical diffraction theory, we found that at both temperatures the surface structures are composed of the so-called one-up and two-down model. The 3×3–$\sqrt{3} \times \sqrt{3}$ phase transition for the Pb/Ge(1 1 1) surface is interpreted in terms of order–disorder transition.

© 2008 Published by Elsevier B.V.

1. Introduction

It is known that Pb and Sn atoms occupy the T_4 site of Ge(1 1 1)-c2 × 8 surface and the super-structures with $\sqrt{3} \times \sqrt{3}$ periodicity appear at room temperature when the coverage is 1/3 monolayer (see Fig. 1). The Pb/Ge(1 1 1) and Sn/Ge(1 1 1) surfaces have been extensively studied experimentally and theoretically as a prototypical two-dimensional metal system [1–4]. These surfaces undergo the 3×3–$\sqrt{3} \times \sqrt{3}$ phase transition at around 200 K. Recently, it has been reported that on the Sn/Ge(1 1 1) surface new $\sqrt{3} \times \sqrt{3}$ structure, which is a Mott insulator, appears below 30 K [5]. Due to various phases on this surface, the Pb or Sn adsorbed Ge(1 1 1) surface system attracts much attentions [6–19].

It has been reported that in the 3×3 phase, one adatom is located at the higher position compared to the other two adatoms [4,9–11]. This model is called one-up and two-down (1U2D) model (Fig. 1(b)). The 1U2D model is energetically favored from the density functional calculations [11]. In actual, the structure model has been supported by the experimental study [4]. However, the two-up and one-down, 2U1D, model has been reported as a 3×3 phase of the Sn/Ge(1 1 1) surface [8,18]. In the 2U1D model, the two adatoms are located at the higher position compared to another adatom (Fig. 1(c)). Thus, the atomic configurations of the 3×3 phase are still in debates [20]. Similarly, the mechanism of the phase transition is also still unknown [20]. In this study, we focus on the atomic configurations of the Pb-adsorbed Ge(1 1 1) surface at around 200 K.

Reflection high-energy positron diffraction (RHEPD) is a powerful tool to study the surface structures and properties. When the positron beam is incident on the surface at low glancing angle, the total reflection takes place [21,22]. The critical angle of the total reflection is estimated to be 2.2° using the Snell's law when the accelerating voltage and the inner potential of the crystal are 10 kV and 14.3 V, respectively [21]. In the total reflection region, the incident positron beam is not able to penetrate the bulk. Therefore, the diffraction beam is very sensitive to the topmost surface. By means of the analyses of the total reflection intensity, we are able to investigate the adatom heights and the thermal vibrational amplitudes [23,24]. In this paper, we investigated the

* 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: fukaya.yuki99@jaea.go.jp (Y. Fukaya).

0169-4332/$ – see front matter © 2008 Published by Elsevier B.V.
doi:10.1016/j.apsusc.2008.02.093

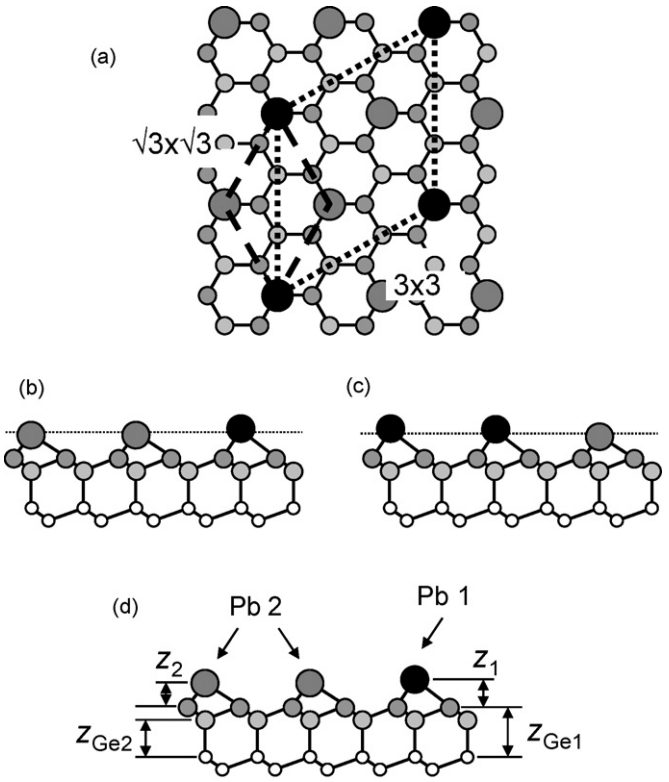

Fig. 1. (a) Schematic drawing of the surface structure of Pb/Ge(1 1 1) (plane view). Large circles denote Pb atoms. Middle and small circles indicate Ge atoms. Dotted and broken lines show the unit cells of the 3×3 and $\sqrt{3} \times \sqrt{3}$ structures, respectively. Two different possible models for the 3×3 structure (cross-sectional view); (b) one Pb atom is located at the higher position compared to the others (one-up and two-down, 1U2D model) and (c) two Pb atoms are located at the higher position compared to the other (two-up and one-down, 2U1D model). In (d), the fitting parameters used in the calculations are denoted (see text).

phase transitions of the Pb/Ge(1 1 1) surfaces using a reflection high-energy positron diffraction.

2. Experimental procedure

Samples were cut from a mirror-polished n-type Ge(1 1 1) wafer with a resistivity of ~35 Ω cm. These were cleaned by repeated cycles of Ar^+ sputtering and annealing at 910 K in 10 min until sharp c2 × 8 patterns appeared in reflection high-energy electron diffraction observation. To obtain the Ge(1 1 1)–$\sqrt{3} \times \sqrt{3}$-Pb surface, 1/3 monolayers (ML) of Pb atoms were deposited on the Ge(1 1 1)-c2 × 8 surface at room temperature using an electron beam evaporator, followed by the annealing at 600 K. Here, the amount of 1 ML corresponds to 7.2×10^{14} cm^{-2}.

Experiments were carried out in a ultra-high vacuum chamber equipped with a positron source of ^{22}Na and magnetic lenses. The detail of the apparatus was described elsewhere [25]. The accelerated voltage of the incident positron beam was 10 kV. The diffraction pattern was enhanced with a micro-channel plate with a phosphor plane and recorded using a charge coupled device camera. In the measurement of the rocking curves, the glancing angle (θ) was changed from 0.6° to 6° with a step of 0.1° by rotating the sample. The sample was cooled down with a cryostat.

3. Results and discussion

Fig. 2(a) and (b) show the RHEPD patterns observed from the Pb/Ge(1 1 1) surface at 60 K and 293 K, respectively. The incident azimuth corresponds to the $[1\bar{1}0]$ direction. At both temperatures, intense (0 0) spots were clearly observed together with the integer-order spots of $(\bar{1}\,0)$ and (1 0). Fig. 2(c) and (d) represent the corresponding line profiles between $(\bar{1}\,0)$ and (0 0) spots at 60 K and 293 K, respectively (denoted as white lines in Fig. 2(a) and (b)). At 60 K, the fractional-order spots between the integer-order spots apparently appear. These additional spots correspond to the 3×3 periodicity. On the other hand, at 293 K, there are no additional peaks other than the integer-order spots. Thus, the phase transition takes place in the temperature range of 60–293 K and the 3×3 phase is transformed into the $\sqrt{3} \times \sqrt{3}$ phase.

Fig. 3 shows the RHEPD rocking curves of the specular spots under the one-beam condition at 60 K and 293 K. The one-beam condition corresponds to 7.5° away from the $[11\bar{2}]$ direction [26]. Under the one-beam condition, the diffraction intensity is very sensitive to the perpendicular components of the atomic position to the surface because the simultaneous reflections parallel to the surface are sufficiently suppressed [26]. In the rocking curve at 60 K, intense peak in the total reflection region ($\theta < 2.2°$) is observed. In the total reflection region, a dip structure is also clearly observed. The position of the dip reflects the height of the adatoms via the Bragg equations, $2d\sin\theta = \lambda$, where d and λ are the height of the adatoms from the substrate and the wavelength of the incident beam, respectively [17]. In this case, the averaged height of the adatoms from the first Ge layer (see Fig. 1(d)) is estimated to be 2.06 Å because the glancing angle at which the dip appears is 1.7°. The height of 2.06 Å is relatively high as compared with that (1.84 Å) of the Sn/Ge(1 1 1) surface [17]. This is a rough estimation of the adatom height and hence the detailed atomic positions were determined on the bases of the dynamical diffraction theory, as shown below.

The rocking curves measured from the both phases show no significant differences, as shown in Fig. 3. In the rocking curve at 293 K, the intense peak and the dip in the total reflection region are observed, similar to the curve at 60 K. This indicates that at both temperatures the equilibrium positions of the surface atoms are almost identical. To determine the atomic positions in the 3×3 phase, we calculated the rocking curves based on the dynamical diffraction theory [27]. Two independent heights of Pb atoms (denoted as z_1 (1/9 ML) and z_2 (2/9 ML)) and two heights of the first and second Ge layers (denoted as z_{Ge1} and z_{Ge2}) shown in Fig. 1(d) were varied so as to minimize the reliability factor (R) between the measured and calculated curves [24]. In the calculation, the thermal vibrational amplitudes of Pb and Ge atoms at 60 K were assumed to be 0.064 Å and 0.040 Å, respectively. The absorption potentials for the Pb and Ge layers were taken to be 0 V and 1.1 V, respectively [23,28].

The solid line in Fig. 3 shows the calculated rocking curve using the optimized atomic positions. In the lower glancing angle range including the total reflection, the calculated curve is in good agreement with the measured curve (R = 4.5%). However, in the higher glancing angle region, the calculated curve slightly deviates from the measured one. This deviation indicates the relaxations with respects to the heights of deeper Ge layers. In the optimization, we obtained z_1 = 2.32 Å and z_2 = 1.81 Å at 60 K. Since z_1 is greater than z_2, the 1U2D structure is responsible for the Ge(1 1 1)-3 × 3-Pb surface. These values are compatible with the surface X-ray diffraction study [4], as shown in Table 1. The height

Table 1
Atomic heights of the Ge(1 1 1)-3 × 3-Pb surface

	z_1 (Å)	z_2 (Å)	Δz (Å)	z_{Ge1} (Å)	z_{Ge2} (Å)
This study	2.32 ± 0.26	1.81 ± 0.09	0.51	3.06 ± 0.07	2.63 ± 0.09
SXRD [4]	2.23	1.81	0.42	3.24	2.54

These parameters were defined in Fig. 1. For the comparison, the atomic heights determined by the surface X-ray diffraction (SXRD) are also listed.

Download English Version:

https://daneshyari.com/en/article/5360778

Download Persian Version:

https://daneshyari.com/article/5360778

Daneshyari.com