Structure of Ga-stabilized GaAs(0 0 1) surfaces at high temperatures
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Abstract
We have studied the atomic structure of the Ga-stabilized GaAs(0 0 1)-c(8/2√2) surface using rocking-curve analysis of reflection high-energy electron diffraction (RHEED). The c(8 × 2) surface is stable only at temperatures higher than 600 °C, but changes to the (2 × 6)/(3 × 6) structure at lower temperatures. The atomic structure of the c(8 × 2) surface at high temperatures is basically the same as that determined by the analysis at room temperature. We propose that the surface atomic configurations are locally fluctuating at high temperatures.

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1. Introduction

It is generally believed that the atomic structures of the GaAs(0 0 1) surface are commonly characterized by the formation of dimers at the outermost layer. However, the dimerization at the subsurface layer is prominent in the structure models recently proposed for the GaAs(0 0 1)-c(8 × 2) surface [1–3]. Lee et al. proposed the ζ model (Fig. 1(a)) using density-functional theory [1]. This model explains quite well previous low-energy electron diffraction [4] and scanning tunneling microscopy (STM) data [5,6], and is also supported by X-ray diffraction (XRD) analysis [7]. A similar structure model shown in Fig. 1(b) has been proposed by an independent study on the basis of the XRD analysis using direct methods [2,3]. The two models shown in Fig. 1(a) and (b) have basically the same atomic structures, but differ in the presence of Ga adatoms and in the partial absence of surface Ga-dimers in the latter model (Fig. 1(b)).

Both models in Fig. 1(a) and (b) explains rather well experimental data obtained at room temperature [1–3,7]. However, as we will show later, the c(8 × 2) structure is stable only at high temperatures, but changes to the (2 × 6)/(3 × 6) structures as the temperature is decreased [8–10]. Thus, in order to study the actual atomic structure of the c(8 × 2) surface without considering the possible coexistence of other phases, a structure analysis at high temperatures is particularly important.

This paper reports the surface structure analysis of the GaAs(0 0 1)-c(8 × 2) at temperatures higher than 600 °C. Rocking-curve analysis of reflection high-energy electron diffraction (RHEED) based on dynamical diffraction theory, which is suited for the
structure determination at high temperatures [11], has been used for this purpose. The results show that the atomic structure of the GaAs(0 0 1)-c(8 √2) surface is basically the same as that at room temperature [2,3]. We found that the dimerization at the outermost layer is not necessarily favorable at high temperatures.

2. Experiment

The experiments were performed in a dual-chamber molecular-beam epitaxy (MBE) system which is equipped with an X-ray photoelectron spectroscopy and a STM for on-line surface characterization. Surfaces of GaAs(0 0 1)-c(8 × 2) were obtained by heating the GaAs(0 0 1)-(2 × 4) surfaces above 600 °C without As fluxes (at a base pressure of 5 × 10⁻¹¹ Torr). RHEED rocking-curves were measured using the extended beam-rocking facility (Staib, EK-35-R and k-Space, kSA400). The energy of the incident-electron beam was set at 15 keV. Integrated intensities of 17 spots, usually, (0 0), (+1 0), (+2 0), (+3 0), (±1 0), (+± 0), (++ 0), (+− 0), and (±2 0) for the [1 1 0] direction, and five spots, (0 0), (0 ±1), and (0 ±2) for the [1 1 0] direction were used in the structural analysis. Averaging of the intensities of symmetry-equivalent spots led to nine and three independent rocking-curves for the [1 1 0] and [1 1 0] directions, respectively. RHEED intensities were calculated by the multislice method [12], using 24 fractional-order and 9 integer-order reflections for the [1 1 0] electron-incidence azimuth. On the other hand, for the [1 1 0] azimuth, only integer-order reflections were used in the calculation because no fractional-order reflections were observed on the zeroth-order Laue zone. The Fourier coefficients of the elastic scattering potential were obtained from the atomic scattering factors for free atoms calculated by Doyle and Turner [13]. A correction due to condensation was made to fit the positions of bulk Bragg peaks at large glancing angles. For instance, the resulting mean inner potential of bulk GaAs was 13.6 eV. The Debye–Waller parameters were taken to be 1.70 and 1.47 Å² for Ga and As atoms in bulk layers [14], while those for atoms in the surface bilayer were treated as fitting parameters. In order to quantify the agreement between the calculated rocking-curves and the experimental ones, the R factor defined in [15] was used.

3. Results and discussion

When the GaAs(0 0 1)-(2 × 4) surface was heated above 450 °C, the (2 × 4) RHEED patterns begins to give way to the (2 × 6)/(3 × 6) patterns. As the temperature is increased above ~580 °C the reflections associated with a c(8 × 2) reconstruction emerged and the (2 × 6)/(3 × 6) reflections disappeared at ~600 °C. The c(8 × 2) structure is stable in the range of 600–640 °C, beyond which the surface roughened. On the other hand, as the substrate temperature is decreased below 600 °C, the (2 × 6)/(3 × 6) structure begins to coexist with the c(8 × 2) structure. While the c(8 × 2) reconstruction was partially preserved on the surface when the samples were quenched from 600 °C to room temperature, our STM observation at room temperature could not detect the c(8 × 2) structure when the substrate temperature is gradually decreased below ~550 °C. These results prompted us to study the atomic structure of the c(8 × 2) surface at high temperatures.

Fig. 2 shows RHEED rocking-curves measured from the GaAs (0 0 1)-c(8 × 2) surface at 610 °C.
(solid curves), together with the calculated ones (dashed curves) from the optimized structure model (Fig. 3). The present RHEED calculations assumed that the surface relaxation extends no further than the third atomic layer. Most of features in the measured rocking-curve were reproduced in the calculated curves (Fig. 2) after the structure optimization for the model in Fig. 1(b). The $R$ factor for the optimized model is 0.089, showing a good agreement between the experiment and calculation. On the other hand, the $\zeta$ model (Fig. 1(a)) gives a larger $R$ factor of 0.142. These results support the presence of the Ga adatoms and the partial absence of surface Ga-dimers in the $c(8 \times 2)$ structure. We have also tested other structure models [4–6,16,17]. As shown in Fig. 4, most of structure models give $R$ factors larger than 0.25, even after the structure optimization.

The atomic coordinates obtained by the present analysis are in good agreement with those from XRD analysis. In addition, the site occupancies ($\theta$) for Ga adatoms (Ga(1)) and surface Ga-dimer (Ga(2)) are $0.33 \pm 0.13$ and $0.45 \pm 0.13$, respectively, which are close to the corresponding values in [2,3]. The reduced site occupancy of Ga(2) suggests that an atomic arrangement without Ga(2) atoms locally exists on the $c(8 \times 2)$ surface. However, further studies are needed to examine the stability of such a local structure.

The bond lengths between the Ga(1) atom and its As nearest neighbors (As(2) and As(3)) fall in the range of 2.92–3.46 Å, being compatible with the values (3.40–3.45 Å) obtained by the XRD analysis [3]. These values are significantly larger than the Ga–As bond length in bulk GaAs (2.45 Å), suggesting that the Ga(1) atoms are weakly bonded to the surface. This seems to be consistent with the recent first-principles calculations showing that the existence of Ga(1) adatom is energetically unstable [7].

We have also performed RHEED analysis for the model in which Ga(2) atoms do not form dimers.
We have confirmed that the model revealed clear minimum in an $R$ factor of 0.089–0.093, irrespective of their atomic coordinate in the [1 1 0] direction. This result is consistent with the fact that the atomic coordinate of Ga(2) has a significantly larger error in the [1 1 0] direction (∼1 Å) than other atoms [10]. Thus, we conclude that the formation of Ga(2) dimers is not necessarily favorable at a high temperature of 610 °C. Although the XRD analysis at room temperature support the formation of Ga(2) dimers, less dimerization has been found for the $c(8 \times 2)$ structures of InAs(0 0 1) and InSb(0 0 1) [2,3]. The present analysis suggests that thermally-activated Ga(2) atoms at a high temperature of 610 °C migrate along the [1 1 0] direction, changing their local atomic configurations.

4. Conclusion

We have studied the surface structure of GaAs(0 0 1)-$c(8 \times 2)$ at high temperatures. The $c(8 \times 2)$ structure

1 While such a random positioning of Ga(2) atoms results in the (4 × 1) periodicity in the outermost layer, the subsurface dimerization of Ga(6) produces the $c(8 \times 2)$ periodicity.
is stable at 600–640 °C, but changes to the (2 × 6)/
(3 × 6) surface below 600 °C. We performed RHEED
rocking-curve analysis above 600 °C and confirmed
that the c(8 × 2) surface has the structure model
proposed by Kumpf et al. We proposed that surface
Ga atoms are thermally activated at high temperatures
and dynamically change their local atomic configu-
rations without disturbing the c(8 × 2) symmetry.

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