9th Int. Symp. "Nanostructures: Physics and Technology"
St Petersburg, Russia, June 18–22, 2001

Evolution of RHEED intensity in the simulated homoepitaxial growth of GaAs(001)

*M. Itoh**† and T. Ohno‡
* Department of Physics, Graduate School of Science, Osaka University,
Toyonaka, Osaka 560-0043 Japan; Electronic address
† Computer Center, Gakushuin University,
1-5-1, Mejiro, Toshima-ku, Tokyo 171-8588 Japan
‡ National Research Institute for Metals,
1-2-1, Sengen, Tsukuba, Ibaraki 305-0047, Japan

Abstract. The influence of surface reconstruction on the homoepitaxial growth of GaAs(001) has been studied by the Monte Carlo simulations. In the model, both Ga and As species are deposited onto a GaAs(001)-$\beta 2(2 \times 4)$ reconstructed surface simultaneously at $T \simeq 580^{\circ}$C as this corresponds to the ordinary growth condition of molecular beam epitaxy. The growth mechanism of the $\beta 2(2 \times 4)$ structure has been identified; also studied are the step-flow growth modes on vicinal surfaces of various misorientations.

Introduction

Molecular beam epitaxy (MBE) and reflection high-energy electron diffraction (RHEED) have been the useful tools in developing semiconductor devices by controlling growth at the accuracy of atomic layers. This is based on the fact that the oscillation period of a RHEED intensity during steady state growth mode corresponds to the growth of one atomic layer. The conventional use of these techniques relies on the claim that the evolution of the specular intensity of RHEED in MBE can be well accounted for by that of the density of atomic steps [1, 2, 3]. However, by carrying out kinetic Monte Carlo (KMC) simulations of the homoepitaxial growth on a GaAs(001)-$\beta 2(2 \times 4)$ reconstructed surface with the growth condition of the ordinary MBE, we found that the calculated density of double As dimers which characterizes the $\beta 2(2 \times 4)$ reconstruction, and not that of atomic steps, evolves synchronously with an observed RHEED intensity. We also found that the growth of an atomic step is strongly affected by the diffusion anisotropy of Ga adatoms as well as the relative phase of the $\beta 2(2 \times 4)$ structures between an upper and a lower terraces.

1. The model

Experimentally, it is well known that the $\beta 2(2 \times 4)$ reconstruction is kept stable in the homoepitaxial MBE growth of GaAs(001) when the substrate temperature is kept at $T = 580^{\circ}\text{C} \pm 20^{\circ}\text{C}$, both Ga and As species are supplied onto the substrate simultaneously, and the V/III flux ratio is sufficiently high. To simulate this, we use Ga atoms and As_2 molecules for the deposition sources of the MBE growth. The substrate is constructed on the zinc-blende structure, on which the $\beta 2(2 \times 4)$ reconstruction is realized by the Coulomb repulsion between surface As atoms located at the in-plane next nearest neighbor sites in the [110] direction [4].

For the atomistic kinetics, the diffusion of a Ga adatom is realized by the random walk. Moreover, we introduce the reservoir of As_2 species to approximate the chemisorbed as

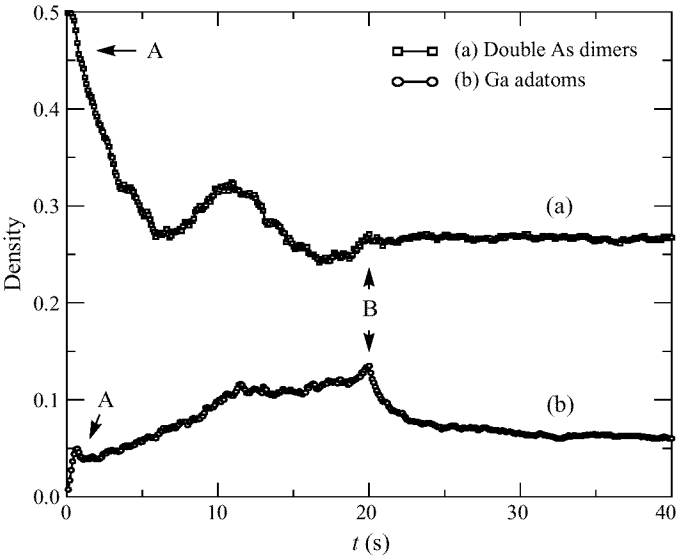

Fig. 1. (a) The density of double As dimers and (b) the density of Ga adatoms plotted as functions of growth time. The growth simulations were carried out on the $120a_s \times 120a_s$ lattice. The symbols A and B indicate the occurrence of the structural transformation and the growth interruption, respectively. The decay constant of (b) after the growth interruption is about 2.5 s.

well as physisorbed states of this species [5] in a mean-field manner. Then, when As_2 molecule is deposited from its source, it is once stored at the reservoir, from where it is either supplied onto a surface or desorbs into vacuum. It is actually the introduction of this state that makes possible the growth simulations under the realistic range of V/III flux ratio, i.e., ≥ 6 [7]. In the growth simulations which follow, the deposition fluxes are fixed to be 0.1 and 0.4 monolayers (ML)/s for Ga atoms and As_2 molecules, respectively.

Furthermore, to examine the effect of the diffusion anisotropy of a Ga adatom, we introduce the parameter G to denote the enhanced ratio of its random walk between parallel and perpendicular to the As dimer row.

2. Island growth mode

With this model, we found that the nucleation of an island occurs on top of the double As dimer row of the $\beta 2(2 \times 4)$ structure. However, the initial island thus appeared does not take the $\beta 2(2 \times 4)$ structure. Instead, an island adopts this reconstruction after it becomes wide enough in the [110] direction to split into two parts. By calculating the density of double As dimers during growth simulation and after its interruption, we found that this density, and not the density of atomic steps, evolves synchronously with an observed RHEED intensity, as seen in Figs. 1(a) and 1(b) [5]. The growth simulations were carried out on the $120a_s \times 120a_s$ lattice, where $a_s = 4.0$ Å denotes the surface lattice constant.

Moreover, we found that the structural transformation of initial growing islands can be identified *in situ* by observing RHEED. When combined with our discovery that this structural transformation is particularly prominent when the deposition coverage of Ga atoms is at around 0.1 ML, this result means that it is appropriate to interrupt growth before the oscillation of a RHEED intensity reaches its maximum by about 10% of an

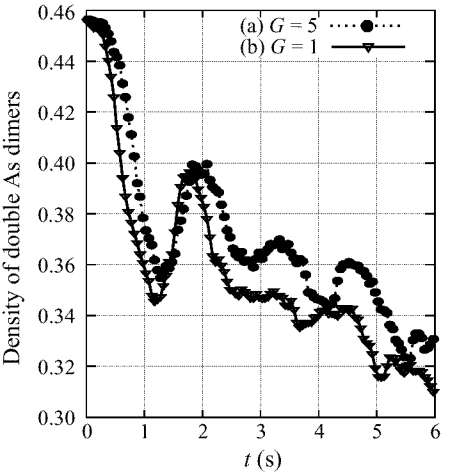

Fig. 2. Evolution of the density of double As dimers on the A surface. The simulations were carried out on a $120a_s \times 120a_s$ lattice, on which the $80a_s \times 120a_s$ strip and the $40a_s \times 120a_s$ strip are stacked consecutively. The deposition fluxes of Ga atoms and As_2 molecules are chosen to be 0.1 ML/s and 0.4 ML/s, respectively. (a) $G = 5$ and (b) $G = 1$.

oscillation period. This is contrasted with the conventional use of RHEED in which growth is interrupted when its intensity arrives exactly at its maximum of the oscillation because the evolution of a RHEED intensity has been supposed to correspond to that of surface step density.

3. Step-flow growth mode

Typical step-flow growth directions which have been studied hitherto are for the vicinal surfaces misoriented from (001) toward the [110] direction, the [$\bar{1}$10] direction, and the ⟨010⟩ directions, which are called the A, B and C surfaces, respectively [3]. For these surfaces, it was found that the RHEED intensity evolves completely differently between A and B or C surfaces. According to our simulations, this difference is caused by the diffusion anisotropy of Ga adatoms which is enhanced parallel to the As dimer row direction than to the perpendicular direction. Due to this anisotropy, once a Ga adatom is deposited onto a surface, it hardly reaches a step edge when it is running parallel to the As dimer row. Following the previous case, we calculated the density of double As dimers in the initial stage of the simulated growth with such step edges. To this end, we constructed the three-bilayer structure consisting of the stacking of the first and second strips of $80a_s \times 120a_s$ and $40a_s \times 120a_s$ in their sizes on top of the $120a_s \times 120a_s$ lattice used as the substrate. By these simulations, we found that the density of double As dimers exhibits a rapid oscillation of only one period, followed by its gradual decrease, as seen in Fig. 2 [3]. Note that, in this particular configuration, a small value of G induces the nucleation and growth of islands, whereas on a real A surface, a large value of G does this task.

In contrast, this diffusion anisotropy makes a Ga adatom to reach a step edge more easily on the B and C surfaces and, hence, it is naturally expected that the growth proceeds similarly on these surfaces. Therefore, the RHEED experiment on the A, B and C surfaces are qualitatively accounted for by these simulations.

4. Summary

By carrying out KMC simulations with the use of the realistic two-species model, we showed that the evolution of a specular intensity of RHEED is well accounted for by that of the density of double As dimers which characterize the $\beta 2(2 \times 4)$ reconstruction. On a singular surface, the structural transformation associated with the growth of the GaAs(001)-$\beta 2(2 \times 4)$ surface is found to shift the origin of the growth time by the coverage of approximately 0.1 ML. Our result suggests that one should take account of this effect in determining the time to interrupt growth in the fabrication of heterostructures. On the A surface, the similar calculation showed again that the density of double As dimers accounts for the evolution of the observed specular RHEED intensity. Thus we conclude that the qualitative difference in the evolution of a RHEED intensity between A and B or C surfaces is due to the diffusion anisotropy of Ga adatoms.

Acknowledgements

M.I. was supported by the 'Research for the Future' program of the Japan Society for the Promotion of Science (JSPS-RFTF97P00201).

References

[1] J. H. Neave, B. A. Joyce, P. J. Dobson and N. Norton, *Appl. Phys. A* **31**, 1 (1983).
[2] J. Sudijono, M. D. Johnson, C. W. Snyder, M. B. Elowitz and B. G. Orr, *Phys. Rev. Lett.* **69**, 2811 (1992).
[3] T. Shitara, D. D. Vvedensky, M. R. Wilby, J. Zhang, J. H. Neave and B. A. Joyce, *Phys. Rev. B* **46**, 6815; 6825 (1992).
[4] M. Itoh and T. Ohno, *Phys. Rev. B* **62**, 7219 (2000).
[5] J. E. Northrup and S. Froyen, *Phys. Rev. B* **50**, 2015 (1994).
[6] C. G. Morgan, P. Kratzer and M. Scheffler, *Phys. Rev. Lett.* **82**, 4886 (1999).
[7] M. Itoh, G. R. Bell, A. R. Avery, T. S. Jones, B. A. Joyce and D. D. Vvedensky, *Phys. Rev. Lett.* **81**, 633 (1998).
[8] M. Itoh and T. Ohno, *Phys. Rev. B* **63**, (2001), to appear.