PHYSICAL REVIEW B VOLUME 46, NUMBER 3 15 JULY 1992-I

Direct observation of the growth-interruption effect for molecular-beam-epitaxy growth on GaAs(001) by scanning tunneling microscopy

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(Received 2 March 1992)

Using a scanning tunneling microscope, we examine the evolution of the surface configuration of GaAs(001) upon annealing after molecular-beam-epitaxy growth interruption. Soon after growth interruption, the surface exhibits many two-dimensional islands elongated along the $[1\bar{1}0]$ direction and a ragged step configuration. Continuous annealing after growth interruption causes changes in the surface topography. Annealing for 2 min makes the surface very smooth by decreasing the number of islands and smoothing the step shapes. Further annealing for about 20 min causes step bunching. The decrease in the number of islands and smoothing of step shapes are explained by shortening of the total step length.

Control of the step structure of GaAs(001) surfaces grown by molecular-beam epitaxy (MBE) is of technological importance for developing superlattice structures. Step configurations on the surface or at interfaces have been intensively studied by reflection high-energy electron diffraction (RHEED) and cross-sectional transmission electron microscopy (TEM) observations. Both methods provide one-dimensional information about the surface or interface structures. However, MBE growth on GaAs surfaces oriented exactly to (001) is generally in a two-dimensional island (2D island) growth mode, because of the large terrace width compared to the diffusion length of the growth-determining species on the surface. Photoluminescence (PL) studies of quantum wells[1-3] suggest that after growth interruption these 2D islands disappear by atomic diffusion on the surface. Cross-sectional high-resolution TEM observations show a very smooth heterointerface for AlAs films on GaAs(001) grown by MBE with growth interruption.[4] To study this growth-interruption effect by direct two-dimensional observations, we examined a GaAs(001) surface grown by MBE using a scanning tunneling microscope (STM). STM observations of various reconstructed GaAs(001) structures have been reported by Pashley *et al.*[5] and Biegelsen *et al.*[6] In this report, we observe by STM evolution of the GaAs(001)2×4 surface configurations due to growth interruption, and discuss the anisotropy of island and step shapes.

Our STM is the same type as the field ion-scanning tunneling microscope apparatus developed by Sakurai *et al.*,[7] but equipped with an MBE chamber to observe MBE grown surfaces without exposing them to air. Substrates were exactly oriented N^+-type GaAs(001) wafers, prepared by chemical etching using H_2SO_4 and H_2O_2 solutions. The surface oxide layers on the substrates were removed by heating above 600 °C in an As flux in the MBE chamber. The GaAs epitaxial layer growth was started by opening a Ga cell shutter and was stopped by closing it. This was the growth-interruption step. (In MBE growth interruption, the Ga shutter is closed but the As shutter remains open.) During the MBE growth and after growth interruption the substrate was kept at 590 °C while being exposed to the As flux. The Ga and As flux were 3×10^{-8} and 1×10^{-6} Torr, respectively. The fluxes were measured by a flux gauge at the same position as the sample during growth. The thicknesses of the epitaxial layers were more than 2000 Å, in order to eliminate completely the surface roughness caused by the chemical etching. The STM tips were made of tungsten wire by electrochemical etching. Oxide layers on the tips were removed by field evaporation during field ion microscopy (FIM) observation, and before every STM observation we checked the tungsten FIM image with an applied voltage of several kV in the same UHV chamber. The STM was able to observe silicon (111)7×7 and (100)2×1 structures and also GaAs(001)2×4 with atomic resolution.

Figures 1(a)–1(d) show GaAs(001) STM images prepared by MBE growth. Figure 1(a) shows a sample which was rapidly cooled soon after growth interruption. Figures 1(b), 1(c), and 1(d) show samples annealed for 10 sec, 2 min, and 20 min after growth interruption, respectively. The temperature of the rapidly cooled sample was decreased at a rate of about 20 °C/sec immediately after growth interruption, while the other samples were cooled at a rate of 2 °C/sec after annealing. All STM observations were carried out at room temperature. Each sample shown in Figs. 1(a)–(d) was prepared independently by MBE growth. Every image covers approximately a 4000×4000 Å^2 area. Atomic steps and 2D islands can be clearly seen in the images. Directions in the (001) surface plane are shown below Fig. 1. Some of these images are somewhat inclined by drift during acquisitions of the STM images. Figure 2 shows an atomic-resolution image of the GaAs(001)2×4 surface annealed for 2 min after growth interruption. All surfaces annealed after growth interruption exhibit the same 2×4 reconstruction together with 1 monolayer height (2.8 Å) steps built up from complete 2×4 unit cells. On the GaAs(001)2×4 surface there are two different types of steps. The steps running parallel to the $[1\bar{1}0]$ direction (i.e., 2× or dimer direction) and parallel to the [110] direction (i.e., ×4 direction) are referred to as *A*-type steps and *B*-type steps, respectively.

Figures 1(a)–1(d) apparently show the gradual disap-

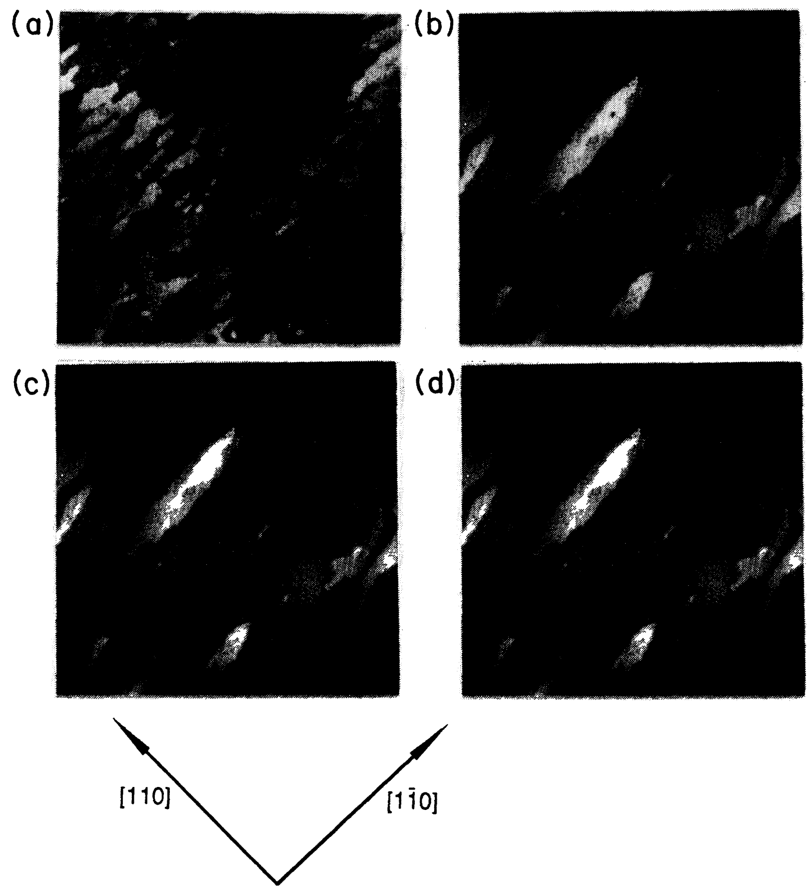

FIG. 1. STM images of GaAs(001) surfaces grown by MBE taken after growth interruption followed by (a) rapid cooling, or (b) 10 sec, (c) 2 min, or (d) 20 min anneals. These images were obtained with a tunnel current of 2 nA and sample bias of −2.5 V.

pearance of 2D islands and smoothing of the step shapes by annealing after growth interruption. According to the RHEED study by Däweritz *et al.*,[8] RHEED specular-beam intensity increases in three stages during annealing after growth interruption. The first rise occurs within 1 sec, the second rise happens after about 2 min, and the third after about 20 min after closing the Ga cell shutter. In our experiments, the change of the surface in the first stage could not be observed due to the cooling time from growth temperature to room temperature. Comparison of Figs. 1(a)–1(c) indicates that the 2D islands disappear and step smoothing occurs during the second stage. As a result of these configurational changes, the area of the 2D islands and peninsular shapes of steps are gradually enlarged by annealing, and thus the total step edge length is shortened. We ascribe this growth-interruption effect, by which high-quality heterointerface structures can be built,[1–3] to a lowering of the total step energy by shortening the total step edge length. A comparison of Fig. 1(c) with Fig. 1(d) indicates that the third stage is step bunching. This contrasts with the case of Si(100)2×1 where such step bunching was not observed even after repeated annealing above 1200 °C.

The shapes of the 2D islands in Figs. 1(a)–1(d) are elongated in the [1$\bar{1}$0] direction. During MBE growth, the surface configuration is affected by kinetic effects of atomic diffusion. By annealing after growth interruption, the surface gradually approaches thermodynamical equilibrium by a redistribution of atoms. The shape anisotropy of the 2D islands on the sample annealed for 2 min [e.g., the island at the left-hand side of Fig. 1(c) indicated by an arrow] does not change even on the sample annealed

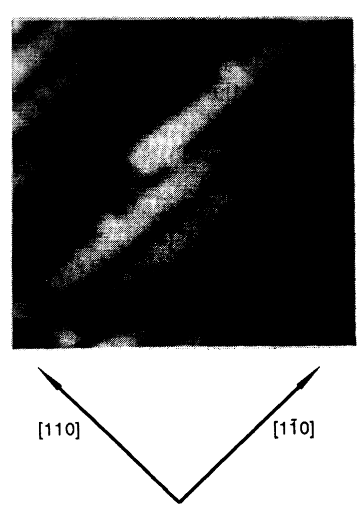

FIG. 2. High-resolution STM image of the GaAs(001)2×4 surface shown in Fig. 1(c). Wide dark stripes running parallel to the [1$\bar{1}$0] direction correspond to missing dimer rows.

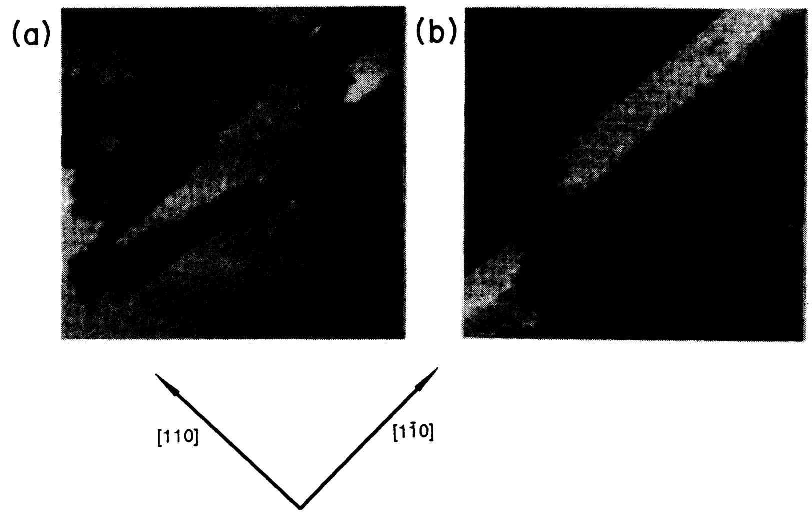

FIG. 3. STM images of the GaAs(001) surfaces grown by MBE with growth interruption showing step shapes after (a) rapid cooling and (b) 20 min annealing.

for 20 min [e.g., the island at the top of Fig. 1(d) indicated by an arrow]. This invariability suggests that the anisotropies of these islands are equilibrium shapes. The shapes of such islands at thermodynamical equilibrium are determined by the difference in step formation energies for A- and B-type steps. From this anisotropy the formation energy of B-type steps can be roughly estimated to be 5–10 times higher than that of A-type steps.[9]

Large area STM observations [Figs. 1(a)–1(d)] show the gradual smoothing of step shapes by continuous annealing after growth interruption. However, it should be noted that thermal fluctuations affect the step shapes on an atomic scale, even after annealing for 20 min, as shown below. Figures 3(a) and 3(b) are STM images showing step shapes with a 2×4 unit-cell arrangement. Figure 3(a) is observed on the rapidly cooled sample and Fig. 3(b) is from the sample annealed for 20 min after growth interruption. In these images stripes running parallel to the [1$\bar{1}$0] direction correspond to missing dimer rows which appear in Fig. 2 as wide dark lines along the [1$\bar{1}$0] direction. Both Figs. 3(a) and 3(b) show that steps along the [1$\bar{1}$0] direction mainly consist of A-type steps which line up straight, while steps along the [110] direction are ragged. These observations are in accordance with experiments on vicinal surfaces.[10] In order to lower the total step energy the step shapes become smooth, as observed in the large area images (Fig. 1). However, the atomic scale observation shows that the step shapes running along the [110] direction are still ragged, having many kinks, even after annealing for 20 min [e.g., lower right-hand side of Fig. 3(b)]. These kinks, which are energetically unfavorable are thought to appear due to thermal fluctuations. On the other hand, A-type steps have a tendency to line up straight even on the rapidly cooled sample. This straightness may be affected by kinetic effects but the details are not yet known.

In summary, the evolution of the surface configuration of GaAs(001) upon annealing after MBE growth interruption was examined using STM. Soon after growth interruption the surface exhibits many 2D islands elongated along the [1$\bar{1}$0] direction and ragged step configurations. Continuous annealing for 2 min makes the surface very smooth by decreasing the number of islands and smoothing the step shapes. These phenomena correspond to the growth-interruption effect that has been reported by PL studies of heterointerface structures. Further annealing for about 20 min causes step bunching. The decreasing number of islands and the smoothing of step shapes are due to shortening of the total step length to lower the total step energy.

The authors would like to thank K. Ishida and H. Tsuda for useful discussions, and Y. Matsumoto, K. Ishida, and K. Hirose for their encouragement.

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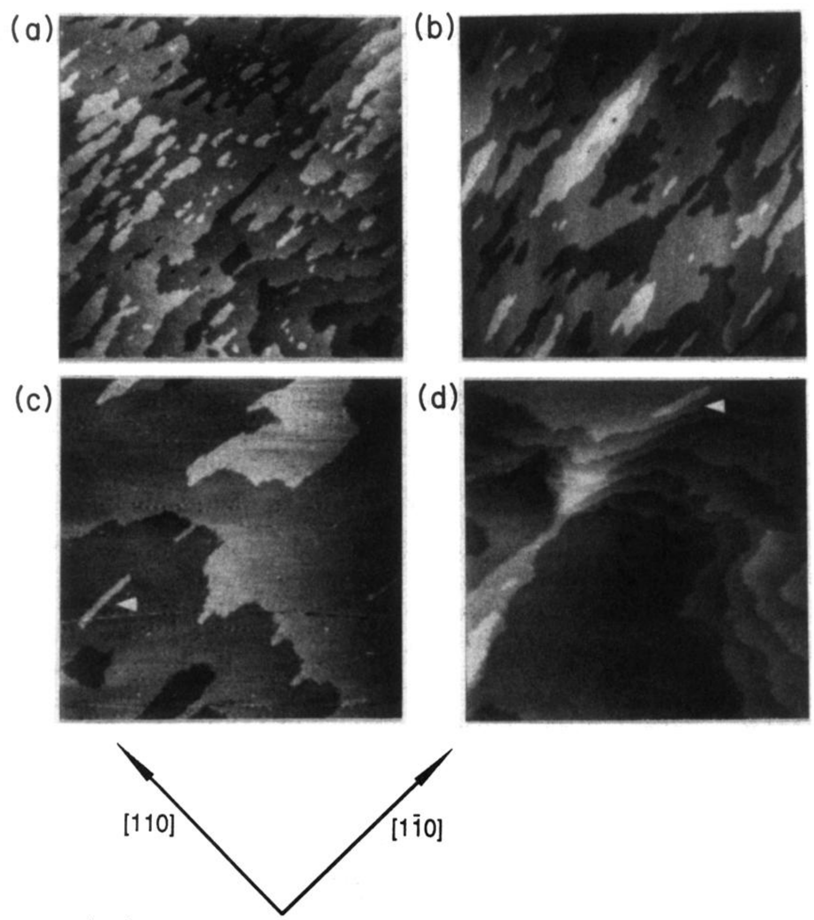

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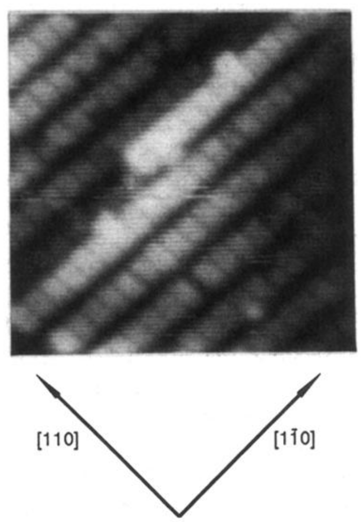

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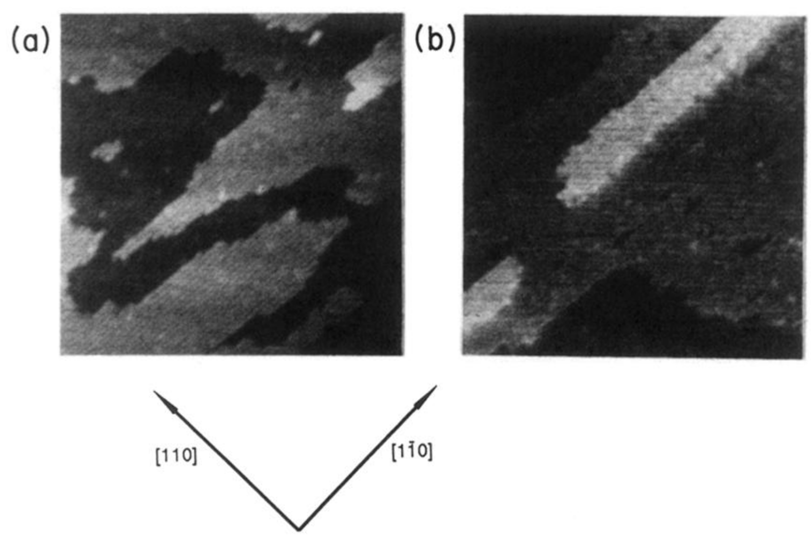

FIG. 3. STM images of the GaAs(001) surfaces grown by MBE with growth interruption showing step shapes after (a) rapid cooling and (b) 20 min annealing.