Microscopic structure of spontaneously formed islands on the GaAs(001)-(2×4) reconstructed surface

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Islands are found to spontaneously form on the GaAs(001)-(2×4) reconstructed surface. The geometry and size of these islands are examined as a function of substrate temperature and island coverage. Both the islands' coverage and size increase with increasing temperature. The islands are elongated rectangles and the aspect ratio is independent of temperature. A relationship between the islands' aspect ratio and the step formation energies is presented. These results are also related to recent theoretical work on equilibrium island geometry. © *2001 American Vacuum Society.*
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I. INTRODUCTION

Numerous optoelectronic and high-speed devices found in the rapidly growing telecommunications industry are fabricated from compound semiconductors such as GaAs. Unlike silicon-based devices, these devices are typically layered heterostructures and are manufactured using epitaxial techniques where one layer of atoms is deposited onto another until the entire device structure is formed. Naturally, the growth front during epitaxy is a surface and a deeper understanding of the physics governing atoms on these surfaces would aid the understanding of the growth process.

Since the inception of molecular-beam epitaxy (MBE) over 30 years ago,[1,2] numerous studies of the GaAs(001) surface have been performed.[3,4] Our recent results have identified its atomic structure[5] and observed the effect of spontaneous island formation.[6] The latter discovery gave insight into the relationship of the arsenic flux and substrate temperature on the island coverage within the framework of the lattice-gas Ising model. In addition, the geometry of the islands was utilized to uncover the step formation energies. However, a systematic study of how the island size and shape changes with temperature has not been performed. This insight combined with recent theoretical treatments of island geometry would assist in determining the influence of strain on these islands.[7] In addition, the ability to produce a surface with a tunable island shape and coverage would be useful in studies which assess their role on transport and optical properties.

In this article, the equilibrium size, shape, and total coverage of spontaneously formed islands on the GaAs(001) surface are examined with scanning tunneling microscopy (STM) as a function of temperature. The observed behavior of the coverage of the islands with temperature is discussed in terms of fundamental thermodynamics, while the shape and size of the islands are discussed in terms of differences in step formation energies and strain.

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II. EXPERIMENTAL PROCEDURE

Experiments were carried out in an ultrahigh-vacuum (UHV) multichamber facility ($5-8\times10^{-11}$ Torr throughout) which contains a solid-source molecular-beam epitaxy growth chamber (Riber 32P) with a highly accurate (±2 °C), optical transmission thermometry system for substrate temperature determination.[8] The MBE chamber also has an all UHV connection to a surface analysis chamber, which contains a custom-integrated commercially available STM (Omicron) for surface morphology measurements.[9] Commercially available, ''epiready,'' *n*-type (Si-doped $10^{18}/cm^3$) GaAs(001) $\pm0.05°$ substrates were loaded into the MBE chamber without any chemical cleaning. The surface oxide layer was removed and a 1.5-μm-thick GaAs buffer layer was grown at 580 °C using an As_4 to Ga beam-equivalent-pressure (BEP) ratio of 15 and a growth rate of 1.0 μm/h, as determined by reflection high-energy electron diffraction (RHEED) oscillations.

Several samples were then prepared for the STM measurements by annealing them at 600 °C, with an As_4 BEP of 1.0 μTorr for 15 min, and then again at 570 °C, with the same As_4 BEP for an additional 15 min to improve the RHEED pattern. To produce the spontaneously formed islands of differing coverages, the samples were then annealed at different temperatures under a common As_4 flux of 0.03 μTorr until the surfaces were in equilibrium. To ensure equilibrium, the anneal times were successively increased until the surface morphology remained unchanged as observed in the STM images. This procedure resulted in anneal times ranging from 5 to 33 h at the highest and lowest temperatures, respectively. After the anneal, the samples were quenched to room temperature and transferred to the STM without breaking UHV, and imaged at room temperature. For each sample, 5–10, 1 μm×1 μm filled-state STM images were acquired using tips made from single-crystal ⟨111⟩-oriented tungsten wire, with a sample bias of -3.0 V and a tunneling current of 0.05–0.1 nA. To compute the fractional area of the surface covered by the islands 10–20, 200 nm×200 nm regions are cropped far from terrace edges from 5 to 10 larger images, and then thresholded to compute

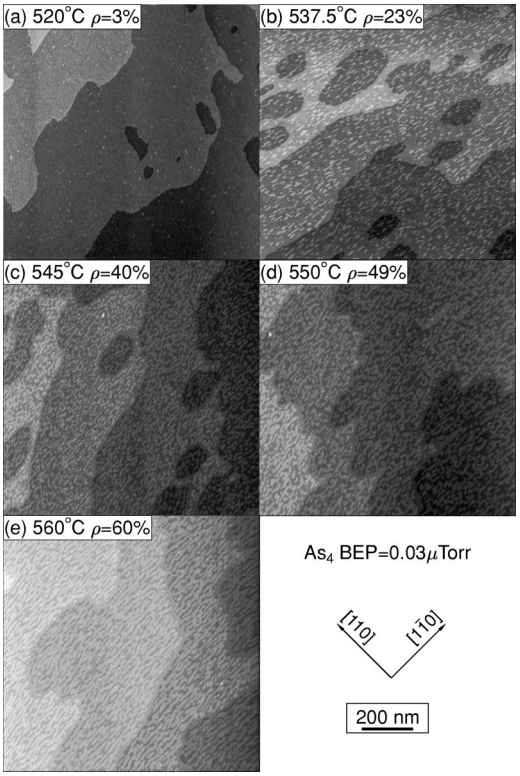

FIG. 1. Filled-state (3.0 V), 1 μm$\times$1 μm STM images showing the spontaneously formed islands created by annealing at the indicated temperatures and under a 0.03 μTorr As_4 flux. Coverage ρ increases with temperature, and all the islands are elongated in the $[1\bar{1}0]$ direction.

an average coverage, which has a uniform standard deviation of $\sim 5\%$.

III. RESULTS

The equilibrium morphology of the GaAs(001)-(2×4) surface is composed of flat terraces that are either covered or not covered with 1-ML (0.28-nm)-high islands, as displayed in the 1 μm$\times$1 μm STM images in Fig. 1. The images are unprocessed, except for having a (001) plane subtracted from them, which makes each terrace a separate color separated by a 0.28-nm-high step. Notice how the island coverage increases as the substrate temperature increases. The islands are also elongated in the $[1\bar{1}0]$ direction and tend to increase size with increasing temperature. In addition, for the higher coverages the edge of each terrace affects the formation of islands by not allowing double-height steps to form.

The microscopic structure of the elongated monolayer-high GaAs islands is more easily seen in the 200 nm $\times$200 nm STM image shown in Fig. 2(a). This sample was annealed at 560 °C, and shows a surface that is 60% covered with islands. The islands are well ordered and elongated. The thin white lines running diagonally in the $[1\bar{1}0]$ direction on both the islands and terrace are separated by 1.8 nm in the [110] direction and are a result of the ''4-by'' periodicity of the (2×4) reconstruction.[5] To quantify the island geometry a site–site correlation function is computed from the STM image and displayed in Fig. 2(b). The correlation function is

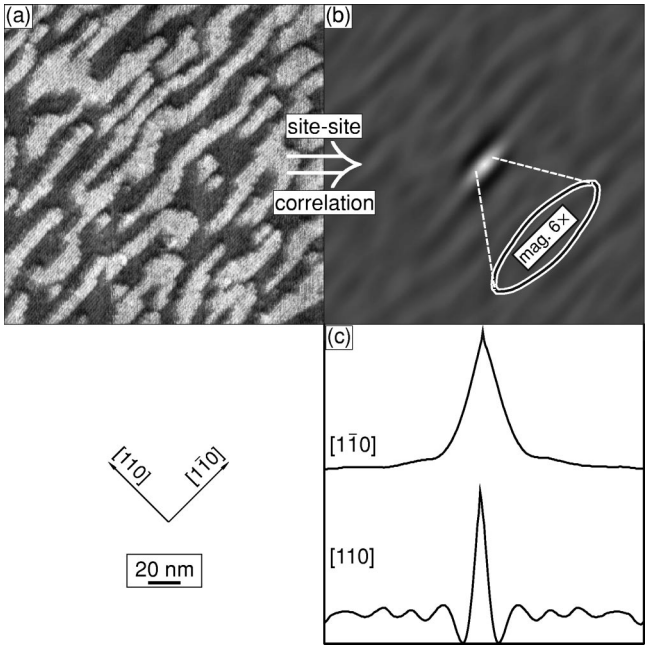

FIG. 2. (a) 200 nm$\times$200 nm STM image of the spontaneously formed GaAs islands annealed at 560 °C under an As_4 flux of 0.03 μTorr; (b) correlation function calculated from the image shown in (a); and (c) line profiles along the $[1\bar{1}0]$ and [110] directions extracted from the correlation function shown in (b).

elongated in the same direction as the islands and a cross section taken at $1/e$ of the maximum is the ellipse shown in the inset with an aspect ratio of 3.0. Line profiles of the correlation function taken along the $[1\bar{1}0]$ and the [110] directions are displayed in Fig. 2(c) and illustrate the dramatic contrast in the island correlation properties between the two directions.

The coverage is plotted versus anneal temperature for each sample and displayed in Fig. 3. This series of samples shows a Fermi-like functional relationship with temperature. To examine the island geometry a correlation function was computed for each STM image and an ellipse was extracted

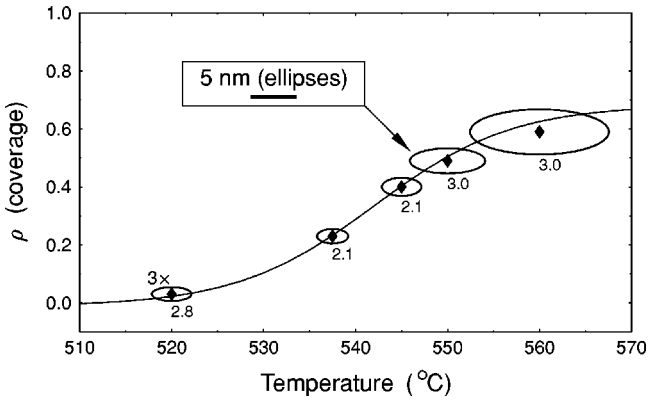

FIG. 3. Island coverage vs temperature (shown as diamonds). Ellipses are $1/e$ cuts from the correlation function, and are drawn with their centers on the corresponding diamond. The aspect ratio of each ellipse is indicated by the number underneath.

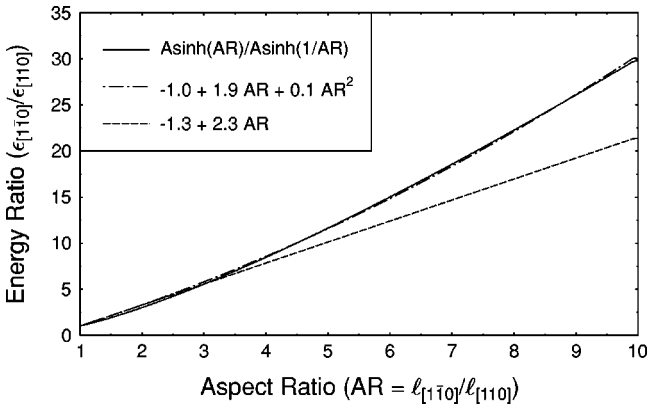

FIG. 4. Step formation energy ratio vs island aspect ratio. The exact 2D Ising model prediction is shown as a solid line. A linear approximation is shown as a dashed line. A quadratic approximation is shown as a dot-dashed line.

at the 1/*e* point. For comparison, a characteristic ellipse is plotted about the corresponding island coverage and anneal temperature point shown in Fig. 3. The ellipses get larger with increasing temperature, however, the aspect ratio stays relatively constant, between 2 and 3.

IV. DISCUSSION

Observations of islands spontaneously forming on the GaAs(001)-(2×4) reconstructed surface have been previously reported.[6,10,11] This phenomenon is surprising and counterintuitive because creating islands increases the internal energy of the system due to the additional broken bonds at the edges. However, the island formation also increases the entropy of the system. Therefore, at high temperatures and for systems where the bond breaking energy is low, island formation may be favorable. We believe this is the driving force for the equilibrium island morphology on the GaAs(001)-(2×4) surface.

The elongation of the islands is a direct consequence of the asymmetry in the energy cost to break a bond on different sides of an island. Two explicit relationships between the bond energies and the island aspect ratio were presented when the GaAs(001) surface was successfully described using the two-dimensional (2D) lattice-gas Ising model.[6] By combining these relationships, the following formula is obtained:

$$\frac{\epsilon_{[1\bar{1}0]}}{\epsilon_{[110]}} = \frac{\operatorname{arcsinh}(\mathrm{AR})}{\operatorname{arcsinh}(1/\mathrm{AR})}, \tag{1}$$

as the relationship between the aspect ratio (AR) and the energy ratio of the bond strengths in the two directions $\epsilon_{[1\bar{1}0]}/\epsilon_{[110]}$.[6,12–14] This result is plotted as a solid line in Fig. 4. For an aspect ratio of unity an energy ratio of unity is predicted, as one would expect. Using our experimentally determined aspect ratio of 3, Eq. (1) predicts an energy ratio of 5.6. This prediction means that the energy cost of forming an edge site on an island in the $[1\bar{1}0]$ direction is $\sim6\times$ greater than in the [110] direction. A linear approximation using these two data points for Eq. (1) is displayed as a dashed line in Fig. 4. A linear relationship is appealing and is commonly used as a rule of thumb, since the number of broken bonds increases linearly with the length of the island. In the literature, this is sometimes referred to as a Wulff approximation.[15–20] Notice that it deviates quickly from Eq. (1) for aspect ratios greater than 3.5. A quadratic approximation to Eq. (1) is displayed as a dot-dashed line in Fig. 4. This approximation is a good fit and simpler in form than Eq. (1).

It is interesting to compare the island shape as a function of island size with the recent theoretical work by Li, Liu, and Lagally.[7] Their study shows that the shape of the islands should change with size if there is a significant amount of strain anisotropy. Our finding that the aspect ratio stays relatively constant as the size increases indicates that the strain effects are not significant. It is known that forming the 2×4 reconstruction results in a significant amount of strain on the lattice, however, the strain must be fairly uniform in direction.

Independent of why these islands form and their size and shape, this phenomenon is remarkable and potentially useful. An ongoing debate regarding the optical and electrical properties of reduced dimensionality systems is to determine what is the explicit role of interface roughness. It is now possible to produce interfaces with systematically varying nanoscale roughness with a well-defined correlation function to test these issues.

V. CONCLUSION

It has been demonstrated that the temperature of the GaAs(001)-(2×4) reconstructed surface controls the size and coverage of the spontaneously formed islands. We find the island shape is constant with an aspect ratio of 3 and predict an energy ratio of 5.6. This energy difference explains why the islands are elongated in the $[1\bar{1}0]$ direction. These results also predict that the strain is isotropic for the 2×4 reconstructed surface. This phenomenon may prove useful in its ability to produce interfaces with a tunable amount of roughness.

ACKNOWLEDGMENTS

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