Island Scaling in Strained Heteroepitaxy: InAs/GaAs(001)

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Scaling properties associated with island size and separation distributions are presented for three different coverages of InAs on GaAs(001). The island size distribution is similar to that observed for homoepitaxy and obeys scaling behavior when considering the average island area. Since the island growth is anisotropic, the island size distribution was separated into [110] and [$\bar{1}$10] components, and scaling is only observed in the [$\bar{1}$10] direction. An analysis of the radial distribution of the islands shows clustering.

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There has been much recent interest in employing scaling theories to describe nonlinear systems such as the initial stages of epitaxial growth [1–4], especially in the aggregation regime which occurs when the number of free adatoms on the surface is exceeded by the number of islands. Under these circumstances film growth is completely characterized by the development of the island size, shape, and separation distributions. A scaling theory has been reported for homoepitaxy in the aggregation regime which delineates the scaling properties of the island size and separation distributions at different temperatures and coverages [1]. In this theory the island size distribution N_s can be expressed as

$$N_s \sim (\theta/\langle s\rangle^2) f(s/\langle s\rangle), \quad (1)$$

where N_s is the number of islands which contain s atoms normalized by the number of lattice sites, θ is the fractional surface coverage, $\langle s\rangle$ is the average number of atoms in an island, and $f(s/\langle s\rangle)$ is the scaling function. This scaling *ansatz* assumes that $f(s/\langle s\rangle)$ is independent of coverage and temperature in the aggregation regime [1], and it has been verified by Monte Carlo (MC) simulations assuming both irreversible and isotropic island growth and a large ratio of the surface diffusion rate to the deposition rate [1,2]. Two-dimensional (2D) island scaling, based on this theory, was observed recently in scanning tunneling microscopy (STM) images of Fe homoepitaxy [3]. Moreover, this scaling relation was also demonstrated to apply both to systems with a non-negligible probability of adatom detachment from growing islands [4] and to systems with anisotropic diffusion [1].

It would obviously be useful to determine whether this scaling ansatz characterizes the initial *heteroepitaxial growth* of semiconductors. These systems are inherently more complex due to both their covalent bonding and diffusion, reaction, and reconstruction anisotropies that dominate the growth morphologies [5]. When the heteroepitaxy is strained from lattice mismatch, the topography may also be altered significantly [6,7]. Recent theoretical work by Ratsch and Zangwill [8] has suggested that the principal effect of strain during growth is to reduce the energy barrier to adatom detachment from an island, and they have shown with MC simulations that 2D island growth in strained, isotropic, heteroepitaxial systems can also be characterized by the scaling relation of Eq. (1) [9].

We present here the scaling properties associated with the island size and separation distributions at three different coverages for InAs growth on GaAs(001)-(2 × 4) (lattice mismatch 7%). We have found that the island size distribution obeys scaling when considering the average total island size. However, considering the anisotropy of the observed island shapes [7], we have separated the average size of the island into their [110] and [$\bar{1}$10] components. *Scaling is then observed only in the* [$\bar{1}$10] *direction.* Furthermore, an analysis of the radial distribution of islands indicates that *the islands are not distributed uniformly.* Both of these important observations are a consequence of the strain which is present in this lattice-mismatched system.

The experiments were performed in an ultrahigh vacuum STM system which is coupled to a Varian Gen II molecular beam epitaxy chamber by an ion-pumped interlock shuttle [10]. Both nominally flat and 1° vicinal surfaces were investigated. The preparation of the As-rich GaAs(001)-(2 × 4) surface has been described elsewhere [7]. After the (2 × 4) surface is prepared, a reference sample is removed from the growth stage, and the remaining samples are returned to the growth chamber for InAs deposition. The samples are heated to 450 °C [11] under low As background pressure to maintain the (2 × 4) reconstruction, and various fractional coverages (θ = 0.15, 0.29, and 0.35) of InAs are deposited. The In is deposited with the As shutter closed and the As supplied by the background pressure, and the samples are then immediately transferred into the buffer chamber. This deposition and quenching procedure results in a (2 × 4) reconstruction of the GaAs on both the reference sample and the InAs-free areas of the deposited sample. Data for the island analysis were acquired from images with very few steps ($\lesssim$1 step in a 100 × 100 nm^2 area) parallel to the [110] direction (*B* type). This protocol is employed because we have

found that B-type steps significantly reduce the island density since they act as adatom sinks [7], and they alter the island shape since they limit growth in the $[\bar{1}10]$ direction. The A-type steps (parallel to the $[\bar{1}10]$ direction), on the other hand, do not influence the island density, and they do not interfere with the anisotropic growth direction [7].

We have described previously the morphology after deposition of InAs on nominally flat and vicinal ($\leq 2°$) GaAs(001)-(2 × 4) surfaces for $\theta = 0.15$ and 0.35 [7]. We observed that InAs forms 2D islands in all cases, and these islands are easily distinguished from the surrounding GaAs substrate due to their $c(4 \times 4)$ reconstruction and In termination. A typical image for $\theta = 0.29$ is shown in Fig. 1. The arsenic dimers appear in rows in the (2 × 4) reconstruction of the GaAs(001) substrate. The islands, elongated in the $[\bar{1}10]$ direction, are observed to be compact with few kinks at their edges, indicating rapid In adatom migration both along the island edge and on the terrace at the growth temperature of 450 °C. Although the islands are typically indium terminated (~1.5 Å above the substrate), some domains of arsenic termination (~3.0 Å above the substrate) can also be observed, cf. boxed area in Fig. 1. Data analysis involves a separation of coalesced islands using the method described by Bartelt and Evans [1].

The island size distribution N_s considering the total number of atoms in an island s is shown in the inset of Fig. 2(a) for the three coverages. The distributions broaden as the fractional coverage of InAs increases, and they are similar to island size distributions observed both experimentally for the homoepitaxy of Fe [3] and in MC simulations of unstrained systems [1,2,4], a surprising result considering the significant strain which is present in our system. It has been suggested that at submonolayer coverages strain energy is minimized by the formation of small, uniformly sized islands [8,9,12,13]. In other words, the dominant effect of strain is to reduce the barrier to adatom detachment so that the islands can assume an energetically favorable size. Thus, for a strained system one would expect to observe a size distribution which remains peaked at approximately the same value with an increase in the island density at higher coverages. However, these conclusions are based on an isotropic model, whereas the growth of InAs on GaAs is highly anisotropic. Anisotropic surface diffusion, different reaction probabilities at step

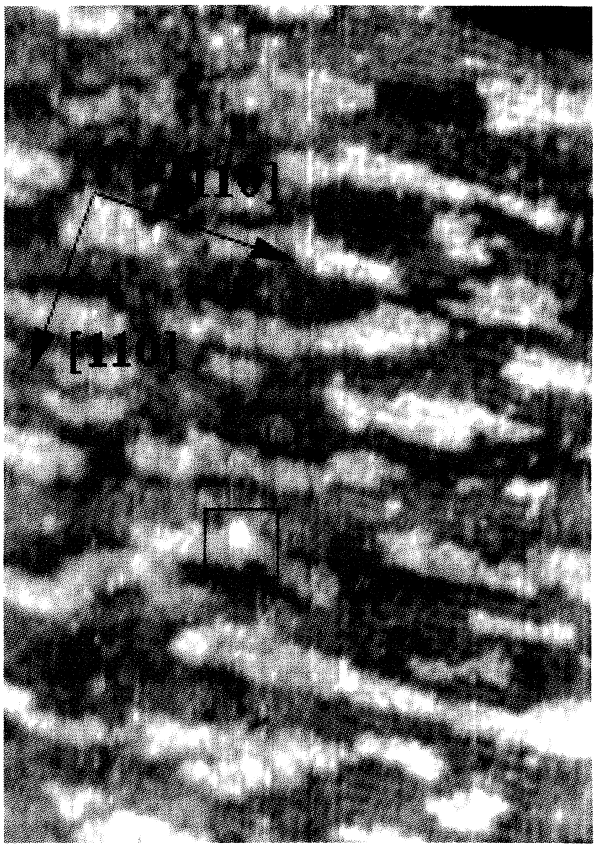

FIG. 1. A 100 × 70 nm^2 STM image of a GaAs(001)-(2 × 4) surface with an InAs fractional coverage of $\theta = 0.29$. The InAs islands are elongated in the $[\bar{1}10]$ direction, and the GaAs-related (2 × 4) reconstruction is still observed on the bare substrate. The boxed area indicates a domain of arsenic termination.

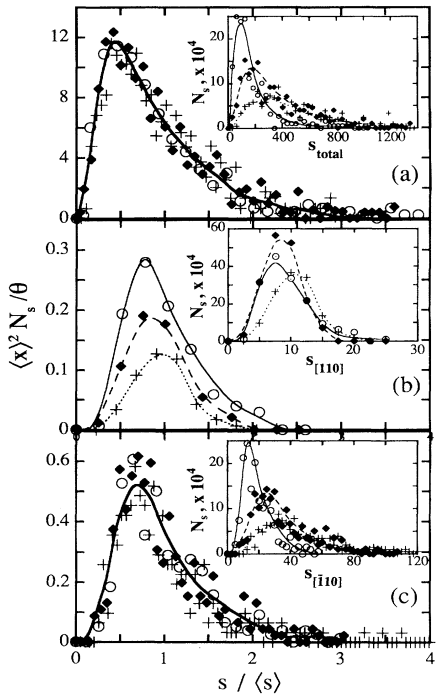

FIG. 2. The scaling properties as predicted by Eq. (1) are shown. In particular, the quantity $\langle x \rangle^2 N_s / \theta$ is plotted as a function of $s/\langle x \rangle$, with $\langle x \rangle = \langle s \rangle$, $\langle s_{[110]} \rangle$, and $\langle s_{[\bar{1}10]} \rangle$, in (a), (b), and (c), respectively. The lines are a smooth fit to the data. The insets show the corresponding size distribution for each scaling relation, as described in the text.

edges along the [110] and [$\bar{1}$10] directions, and the propensity of two surface atoms to form dimers contribute to the observed anisotropy. To clarify the effect of strain on the growth of InAs, we have evaluated the linear island size distribution along the two orthogonal step-edge directions. The insets to Fig. 2(b) and 2(c) show N_s as a function of the linear number of atoms in an island along the [110] direction $s_{[110]}$ and along the [$\bar{1}$10] direction $s_{[\bar{1}10]}$, respectively. The island size distribution in the [110] direction shows distinctly different behavior from the [$\bar{1}$10] direction. The peak in the [110] distribution shifts at most by two atoms with increasing coverage from a most probable width of 32 Å to one of 40 Å, whereas the peak in the [$\bar{1}$10] distribution shows a shift of approximately 20 atoms from a most probable length of 60 Å to one of 140 Å. Since the [$\bar{1}$10] distribution is qualitatively similar to the distribution of total atoms in an island, the growth behaves as if it is unstrained in the [$\bar{1}$10] direction; whereas growth in the [110] direction is *quenched* by strain. We can rule out the possibility that intrinsic anisotropies of the system are responsible for the essentially constant size distribution in the [110] direction. The observation of island growth on *B*-type 1° vicinal surfaces indicates that the anisotropic ratio of the diffusion barriers is not sufficiently large to account for the anisotropies observed in the islands on the *A*-type surfaces [7]. In addition, the anisotropy in the island edge reactivities required to produce the island shapes observed with increasing coverage (aspect ratio ≈5 at $\theta = 0.35$) would result in highly elongated islands at low coverage, which is clearly not observed [aspect ratio ≈2 at $\theta = 0.15$]. Furthermore, quenched growth in one dimension has *not* been observed for GaAs homoepitaxy, an unstrained system with surface anisotropies [5,14–17]. Current work with MC simulations has verified that surface anisotropies alone cannot account for the observed [110] distribution [18].

The scaling properties of the three distributions are also shown in Fig. 2. As suggested by Eq. (1), the quantity $\langle x\rangle^2 N_s/\theta$ is plotted as a function of $s/\langle x\rangle$, with the metric of the average island size taken to be $\langle x\rangle = \langle s\rangle$, $\langle s_{[110]}\rangle$, and $\langle s_{[\bar{1}10]}\rangle$ in Figs. 2(a), 2(b), and 2(c), respectively. Collapse of the data onto a single curve is observed for $\langle x\rangle = \langle s\rangle$ in Fig. 2(a), indicating that scaling is obeyed in this case and demonstrating that the scaling theory presented by Bartelt and Evans [1] to characterize homoepitaxy can be applied to strained heteroepitaxy as well, the first observation of this important phenomenon. The effect of strain on the scaling is only apparent when the distributions describing the islands in the [$\bar{1}$10] and [110] directions are plotted separately. Collapse of the data is observed for $\langle x\rangle = \langle s_{[\bar{1}10]}\rangle$, indicating that the growth behaves as if it is unstrained in this direction, cf. Fig. 2(c); when $\langle s_{[110]}\rangle^2 N_s/\theta$ is plotted as a function of $s/\langle s_{[110]}\rangle$, however, scaling is not obeyed as may be seen clearly in Fig. 2(b). The absence of scaling in the [110] direction can be understood when considering the total island density as growth proceeds. The density increase required for scaling to occur when the average island size remains constant with increasing coverage does not occur since island growth is maintained in the [$\bar{1}$10] direction. Thus, instead of many small islands with a narrow size distribution, elongated islands evolve with a size constraint in only one direction. *We expect that this is a general result for strained semiconductor epitaxy since most semiconductor surfaces are not isotropic.*

To study further the effects of strain, we have also investigated scaling in the radial distribution function of this heteroepitaxial system. For large diffusion rates the scaling relation for the island separation, $N(r)$, which is proportional to the probability of finding an island center separated by a distance, r, from the center of another island, is given by

$$N(r) \sim Ng(r/\langle R\rangle), \tag{2}$$

where N is the macroscopic number density of islands, and $\langle R\rangle \sim 1/\sqrt{N}$ is a measure of the average separation between island centers if they are uniformly separated [1]. The scaling function (the radial distribution function), $g(r/\langle R\rangle)$, has the property that $g(r/\langle R\rangle) \to 0$ for $r \to r_0$ and $g(r/\langle R\rangle) \to 1$ for $r \to \infty$ where r_0 is the average size of an individual island. Since this distribution is determined by the distribution of nucleation sites, anisotropies in a noninteracting system will not affect it.

Collapse of the distribution onto a single curve is observed for all three coverages in Fig. 3. The values of $\langle R\rangle$ ($= 1/\sqrt{N}$) for $\theta = 0.15$, 0.29, and 0.35 are 7.4 ± 0.5 nm, 12.2 ± 0.8 nm, and 12.5 ± 0.8 nm, respectively. The average separation between the island centers for these coverages, evaluated from the STM images, is 4.8 ± 0.4 nm, 7.3 ± 0.8 nm, and 7.9 ± 0.8 nm, respectively. Since these values are consistently smaller than $\langle R\rangle$, clustering of islands is occurring which results in average separations that

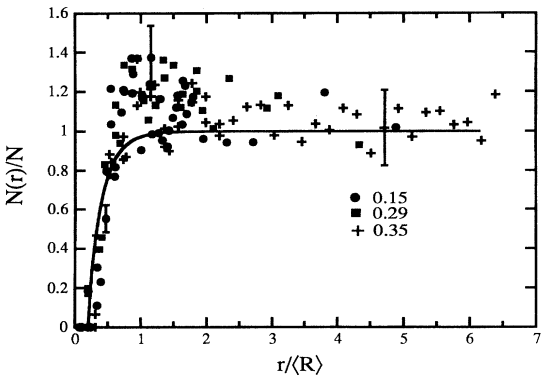

FIG. 3. Scaled radial distribution function for finding an island separated by a distance r from a given island. The solid line is given by $\{1 - K_0(r/\sqrt{D\tau})/K_0(r_0/\sqrt{D\tau})\}$, where K_0 is the modified Bessel function of order zero, and $D\tau \sim 1/4\pi N$ with $\sqrt{N} \sim 1/\langle R\rangle$ [1]. The parameter $r_0/\langle R\rangle = 0.2$. The error bars denote the standard error in the measurement.

are smaller than those expected from a uniform distribution of islands. As may be seen in Fig. 3, at $r \gtrsim \langle R \rangle$ we observe an increased probability of island formation compared to uniform nucleation. This result is unexpected from the theoretical modeling [1] and is not caused by the presence of a stepped substrate [19]. *This is the first observation, to our knowledge, of this kind of clustering of islands in epitaxial growth.* The form of the radial distribution function for a noninteracting system is obtained by solving the differential equation describing 2D diffusion [1], and the result is shown by the solid line in Fig. 3. The depletion of islands at small separations, $r_0 \lesssim r \lesssim \langle R \rangle/2$, is due to the low nucleation probability in the vicinity of an existing island, which acts as a trap for diffusing adatoms. A through-substrate strain-induced repulsion is also expected to result in a lower probability of island nucleation in this region [20].

The clustering of islands at $r \gtrsim \langle R \rangle$ suggests an increased adatom density in this region. This is a consequence of the strain which is present along the [110] direction: the $[\bar{1}10]$ edge acts as a sink for adatoms, while the [110] edge, due to the strain-limited growth, is not nearly so reactive. Adatoms undergoing detachment as a result of strain along the [110] direction are available for nucleation if they encounter another adatom. Recent modeling results also show clustering in the radial distribution thus supporting this claim [18].

In summary, we have shown that the scaling theory developed for homoepitaxy may also be applied to strained heteroepitaxy. In particular, we observed that the island size distribution is similar to that observed for homoepitaxy and obeys scaling when considering the average island size in terms of the total number of atoms in an island. The effect of strain on the initial stages of growth has been clarified by separating the size of the islands into $[\bar{1}10]$ and [110] components. Scaling is then observed in the $[\bar{1}10]$ direction but not observed in the [110] direction. Strain is also responsible for the existence of an increased density of islands at separations $r \gtrsim \langle R \rangle$ in the radial distribution function. We predict that this clustering of islands would be even greater for a heteroepitaxial system where strain affects the growth in both directions leading to an even higher adatom density which will be able to nucleate at $r \gtrsim \langle R \rangle$.

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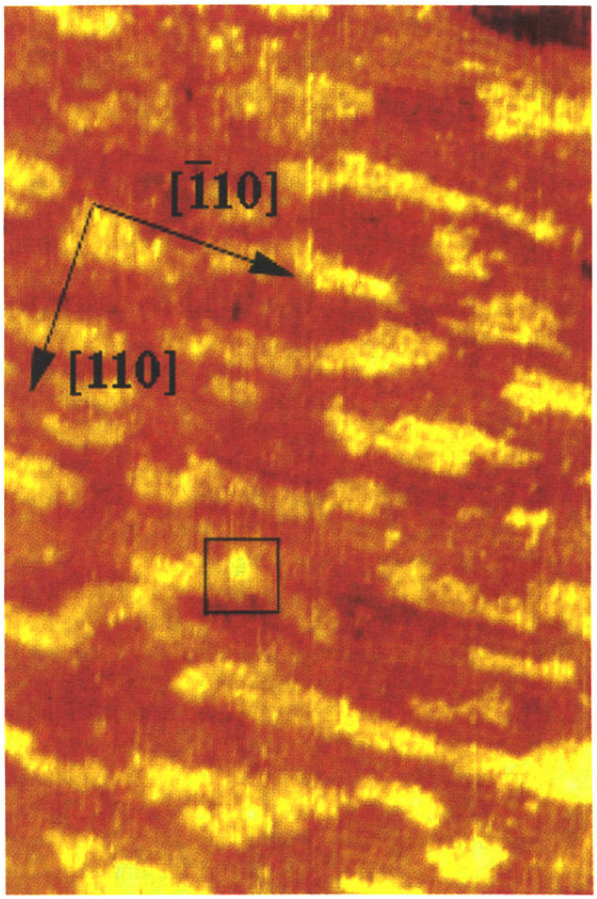

FIG. 1. A 100 × 70 nm^2 STM image of a GaAs(001)-(2 × 4) surface with an InAs fractional coverage of $\theta = 0.29$. The InAs islands are elongated in the [$\bar{1}$10] direction, and the GaAs-related (2 × 4) reconstruction is still observed on the bare substrate. The boxed area indicates a domain of arsenic termination.