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Island size scaling for submonolayer growth of InAs on GaAs(001)-(2×4): Strain and surface reconstruction effects

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The submonolayer growth by molecular beam epitaxy of InAs on the GaAs(001)-(2×4) surface has been studied using rapid-quench scanning tunneling microscopy. InAs islands exhibiting the (2×4) reconstruction are formed and show remarkably similar characteristics to GaAs submonolayer homoepitaxy on this surface. Detailed analysis of the islands indicates that strain plays a negligible role in their nucleation, and the (2×4) reconstruction dominates both island growth and island anisotropy.

The growth behavior on the (001) surface of GaAs is unique when compared with the other low index surface orientations in both GaAs homoepitaxy and InAs/GaAs heteroepitaxy.[1,2] In submonolayer GaAs *homoepitaxy*, the island size distributions obtained on the As-terminated (001) surface differ markedly from those on (110) and (111)A,[3] due to the complexities of island growth on the (2×4) reconstructed surface, which are now understood in considerable detail at the atomistic level.[4–6] For InAs/GaAs *heteroepitaxy* (~7%) lattice mismatch), the (001) surface is again unique for growth under As-rich conditions.[7] InAs grows in a two-dimensional (2D) layer-by-layer mode on (110) (Refs. 8,9) and (111)A (Ref. 10) surfaces, with strain relief involving the formation of misfit dislocations. By contrast, a growth mode transition occurs on (001) surfaces and coherent three-dimensional (3D) islands (quantum dots) are formed by a modified Stranski-Krastanov growth process.[11]

The growth of InAs/GaAs(001) quantum dots is a complex process and one that is still not fully understood. Under conventional conditions growth takes place on the $c(4\times4)$ reconstructed surface of GaAs, and initial deposition of InAs leads to a 2D wetting layer that exhibits a (1×3) surface reconstruction and is an $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ alloy.[12,13] Furthermore, the 3D islands that form as a consequence of the growth mode transition also contain significant amounts of Ga.[14] It is clear that significant intermixing of the group III species occurs both in the formation of the wetting layer and the quantum dots. A detailed study of the initial stages of InAs/GaAs heteroepitaxy is therefore crucial for developing a more detailed understanding of quantum dot formation in this complex material system.

In this paper we present the results and analysis of the submonolayer growth of InAs on the (2×4) reconstructed surface of GaAs(001). This surface reconstruction is much better understood[15–17] than the more As-rich $c(4\times4)$ structure, and its smoother surface morphology permits a more straightforward study of island formation.[18] Studies of heteroepitaxy on the (2×4) surface also allow direct comparison with recent detailed studies of GaAs homoepitaxy on the same surface.[4,5] In an earlier paper, Bressler-Hill *et al.*[19] reported a similar study, but in their work the (2×4) surface was maintained by shutting off the arsenic flux (relying on the background arsenic pressure to stabilize the reconstruction), before reduction of the substrate temperature and subsequent deposition of indium; in effect they were not studying the actual growth of InAs on GaAs. The distributions found for both the island area and length along $[\bar{1}10]$ collapsed on to a single universal curve for different indium coverages, while the island widths along [110] fell on to a different curve for each coverage.[19] The departure from universal scaling in this quantity was attributed to the anisotropic effects of strain, although it should be stressed that the effects on the *highly anisotropic* (2×4) surface reconstruction were not considered. The key difference in our experiments is the availability of a valved arsenic cracker cell that allows easy adjustment of the incident arsenic flux so that true codeposition of In and As can be performed under controlled conditions. This procedure results in the formation of true InAs islands (height ~3 Å) on the GaAs surface, the (2×4) reconstruction being present in both the substrate and the islands at sub-ML coverages. Island size distributions are presented and compared to our previous studies of GaAs(001) homoepitaxy.

Our experimental approach has been described previously for both GaAs(001) (Ref. 4) and InAs(001) (Ref. 20) homoepitaxy. The combined MBE-STM facility and efficient sample transfer allows the (small) substrate to be removed very quickly (seconds) from the growth environment so that quenching to room temperature occurs in an As-free, ultrahigh vacuum STM system. This effectively ''freezes'' the surface structure and allows the observation of surface reconstructions and island morphologies.

Epiready GaAs(001) substrates were prepared by standard means, with buffer layers exhibiting a (2×4) reconstruction grown using an As_2 flux of $\sim2.5\times10^{14}$ molecules $\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$. The As_2 flux was then reduced to $(9.0\pm2.0)\times10^{13}\,\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$ using a needle valve mounted on the cracker cell. These values were measured by a standard ion gauge, calibrated using reflection high energy electron diffraction (RHEED) intensity oscillation measurements on a Ga-rich GaAs(001) surface.[21] The (2×4) reconstruction was maintained as the substrate temperature was reduced from the growth temperature of 580 °C to 470 °C. InAs was then deposited at a rate of $(0.016\pm0.002)\,\mathrm{ML\,s}^{-1}$, corresponding to an As_2:In flux ratio of approximately 9:1. This ratio is somewhat larger than that used in previous GaAs/GaAs (Ref. 4) and InAs/InAs homoepitaxy experiments,[20] and well

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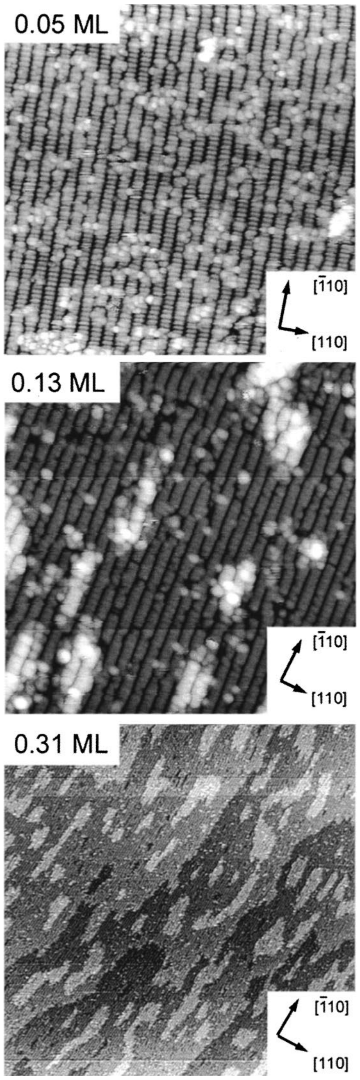

FIG. 1. Filled states STM images after deposition of different InAs coverages on GaAs(001)-(2×4); 0.05 ML (400 Å ×400 Å), (b) 0.13 ML (400 Å ×400 Å), and (c) 0.31 ML (2000 Å ×2000 Å).

above the value at which the effects of flux ratio can be seen in island statistics in GaAs homoepitaxy.[22] InAs coverages of between 0.05 and 0.32 ML were deposited and the samples were then quenched immediately, or left to anneal for five min at the growth temperature (and still under the As_2 flux) before subsequent quenching. STM measurements were made using a constant current of 0.2 nA and a sample bias of −3.5 V.

Three examples of STM images at different InAs coverages (0.05, 0.13, and 0.31 ML) are shown in Fig. 1. The image size for (a) and (b) is 400 Å ×400 Å, whilst for (c) it is 2000 Å ×2000 Å. The (2×4) reconstruction of the underlying GaAs surface is clearly observed as dark and light stripes running along the $[\bar{1}10]$ direction and these represent the As dimer-pair rows and trenches respectively.[15–17] The dimer rows tend to remain very straight in the $[\bar{1}10]$ direction, showing kinks only in the vicinity of islands. The islands are the brightest features in the images and are observed frequently at 0.13 and 0.31 ML (for example, ten islands are visible in the 0.13 ML image, but only two very small islands can be seen in the 0.05 ML image). The height of the islands is ∼3.0 Å and each one exhibits a (2×4) reconstruction, indicating they are most likely to be InAs.

TABLE I. The island density, mean island size and island anisotropy measured from STM images for InAs islands grown on GaAs(001)-(2×4) as a function of coverage and after a rapid quench from the growth temperature. For the two lowest coverages, the total number of islands observed was rather small and statistics are not given. Also shown for comparison are statistics for GaAs homoepitaxy on the GaAs(001)-(2×4) surface (a coverage of 0.13 ML and an As_2:Ga flux ratio of 3:2).

InAs coverage (ML)	Island density (10^{11} cm^{-2})	Mean island size $\langle s \rangle$ (2×4 mesh areas)	Island anisotropy
0.05	∼0.3		
0.08	∼0.5		
0.13	3.0	30	2.5
0.18	2.6	42	2.9
0.24	1.5	129	3.0
0.31	1.3	142	3.1
GaAs/GaAs 0.13	5.8	16	1.6

The number density of islands at different InAs coverages is summarized in Table I, along with their mean size and anisotropy, the latter quantity being defined as the maximal length along [110] divided by the maximal width along [110]. The number densities are derived from measuring several images (size 2000 Å ×2000 Å) for each InAs deposition. The island density rises sharply between 0.08 and 0.13 ML, suggesting that a critical surface concentration of In atoms is required for the nucleation of (2×4) reconstructed InAs islands. We have observed a similar ''induction period'' in island formation for GaAs homoepitaxy on the (2 ×4) surface, although the delay appears to be rather longer for InAs/GaAs (∼0.08 ML) than for GaAs/GaAs (∼0.04 ML), suggesting that nucleation events are somewhat less probable. Direct comparison of the absolute island densities to our previous GaAs homoepitaxy experiments must be approached with caution since the growth rates and As_2 fluxes are rather different in the two cases. However, a result for GaAs homoepitaxy is also shown in Table I, for a coverage of 0.13 ML, a growth temperature of 550 °C, a growth rate of 0.05 ML s^{-1}, and an As_2:Ga flux ratio of 3:2. The island density in this case is considerably larger than for InAs/GaAs at the equivalent coverage suggesting that nucleation is less probable than island growth in the heteroepitaxial case. It should also be noted that the island densities are significantly larger at the ∼0.1 ML stage for InAs/InAs homoepitaxy on the (001)-(2×4) surface.[20]

It is clear from Table I that the islands become more anisotropic as the InAs coverage increases. The precoalescence values of 2.5 and 2.9 are typical for InAs homoepitaxy[20] and significantly greater than the value of 1.6 observed for GaAs homoepitaxy at a coverage of 0.13 ML. In terms of anisotropy, the islands observed in the present heteroepitaxial case are much more ''InAs-like.'' The reduction of island density at the two highest coverages (0.24 and 0.31 ML) is due to the onset of coalescence. The large scale STM image at 0.31 ML (Fig. 1) clearly shows large, irregularly shaped islands, many of which will have developed by coalescence. This also corresponds to a sharp increase in the mean island size at the highest two coverages (Table I).

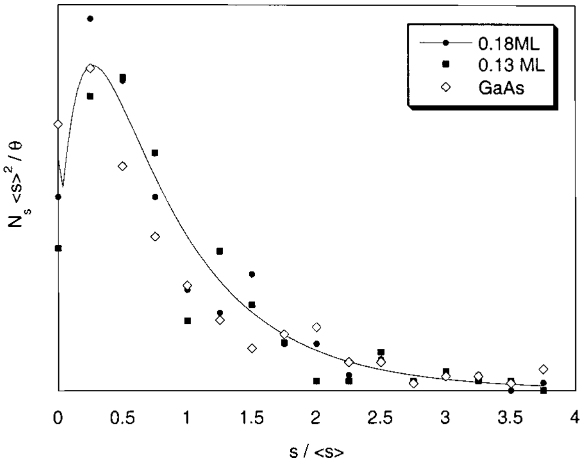

FIG. 2. Island size distributions for 0.13 and 0.18 ML InAs grown on GaAs(001)-(2×4). Also shown for comparison is the distribution for 0.13 ML GaAs grown on GaAs(001)-(2×4) using a low As_2:Ga ratio (3:2). The solid line is a smooth fit to guide the eye.

Some insight into island growth can be gained by measuring island size distributions.[3,19] Distributions at two precoalescence InAs coverages (0.13 and 0.18 ML) are shown in Fig. 2, together with the distribution for 0.13 ML GaAs/GaAs growth using the low As_2:Ga flux ratio of 3:2. The data are normalized conventionally and based on counts of 150–200 islands. The variable s is the island area, $\langle s\rangle$ is the mean island area for each coverage, N_s is the island number density, and θ the coverage. The solid line is a smooth fit to guide the eye, and it is clear that all three sets of points fall roughly on to a single curve. Two features of the distribution should be noted; it peaks well below the point $s=\langle s\rangle$ and it remains nonzero at the lowest sizes. We have observed similar distributions for both GaAs and InAs homoepitaxy on the (001)-(2×4) surface,[20] and they are very different from the more conventional island size distributions measured in GaAs homoepitaxy on (111)*A* and (110) surfaces.[3] The distributions obtained on the (001) surface for InAs/GaAs, GaAs/GaAs, and InAs/InAs are due to the strong influence the (2×4) surface reconstruction has on island nucleation and growth.[4,20] In effect, InAs island growth on GaAs(001) occurs as though the system were a homoepitaxial one, with the island size distribution characteristic of growth on a (2×4) reconstructed surface. It should be noted that one discrepancy between the InAs/GaAs and GaAs/GaAs size distributions occurs at the lowest size bin (island sizes less than 5 unit mesh areas) and there are significantly more of these islands in the heteroepitaxial case. This again points to the different balance between nucleation and growth probability in the two systems.

Following the work of Bressler-Hill *et al.*,[19] we have also measured the lengths and widths of the islands in both InAs/GaAs and GaAs/GaAs. The corresponding distributions are shown in Figs. 3(a) and 3(b); the length l is measured along $[\bar{1}10]$, the width w is measured along [110]. All the length scaling data collapses on to the same curve and is rather similar in shape to that shown in Ref. 19. By contrast, the width data does not fall on to a single curve and there is a large discrepancy between InAs/GaAs and GaAs/GaAs, but a much smaller one between the two coverages for InAs/GaAs. Both are essentially due to the different island anisotropies and absolute densities between the two material systems, and also between InAs islands at different coverages. These strongly influence the scaled frequencies plotted in Figs. 2 and 3. There is no reason to suspect that strain is the cause of the effect since island anisotropies vary significantly in unstrained homoepitaxy on both InAs and GaAs(001) (2×4).[20,22] The mechanism by which the (2×4) surface reconstruction itself influences the island anisotropy in island nucleation and reconstruction is one of Coulomb repulsion between As dimers and does not depend on strain.[4] Essentially, islands must pass through an energetically unfavorable configuration in order to grow into an adjacent dimer-pair row, limiting growth along [110]. This has been investigated in detail at lower coverages (0.07–0.13 ML) in GaAs homoepitaxy, where it dramatically affects the island size distributions. Since it also occurs in InAs homoepitaxy (and indeed is somewhat stronger, as expected from the larger charge transfer to As dimers[20]) this mechanism is suggested as the source of the anisotropic properties of the island size distributions observed in InAs/GaAs(001)-(2×4).

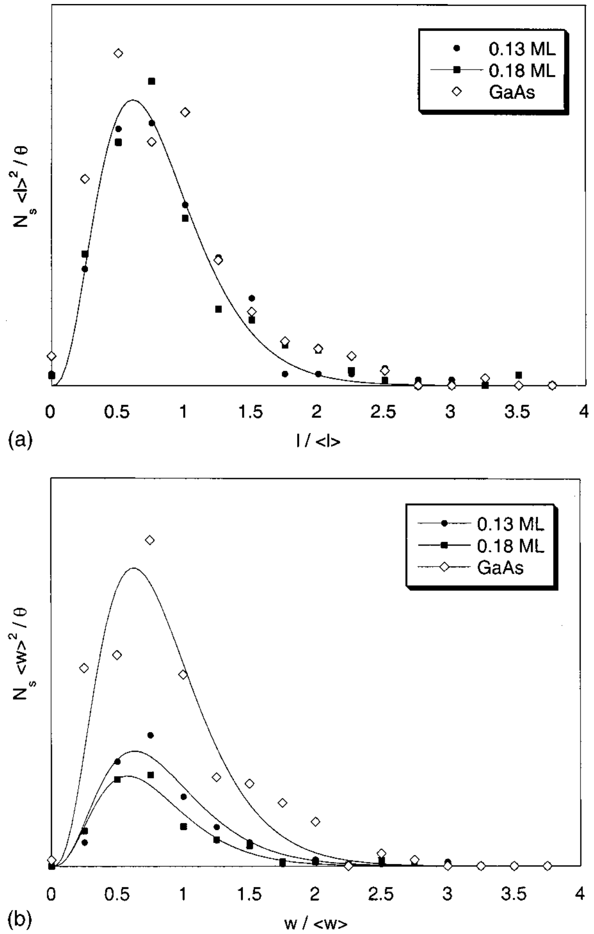

FIG. 3. Island distributions for (a) the length and (b) the width of islands in InAs/GaAs heteroepitaxy and GaAs/GaAs homoepitaxy on (001)-(2×4). The length l was measured along $[\bar{1}10]$ and the width w measured along [110]. The three sets of points in each case represent InAs coverages of 0.13 and 0.18 ML, and a GaAs coverage of 0.13 ML.

Finally, it should be noted that the growth conditions used for the heteroepitaxial studies presented in this paper meant that the structure of the starting GaAs(001) surface was very

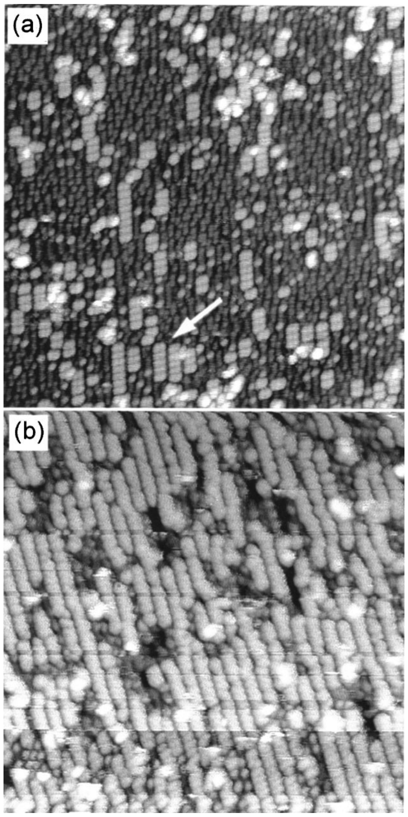

FIG. 4. STM images (400 Å ×400 Å) of 0.08 ML InAs deposited on GaAs(001)-(2×4) followed by a 5 min postgrowth anneal at the growth temperature. In (a) the surface has undergone a partial transition to the $c(4\times4)$ phase during the postgrowth anneal leaving (2×4) domains (white arrow) in the layer above the $c(4\times4)$. In (b) the surface remains (2×4), but is rather disordered with many kinks and patches of missing As dimers.

close to the onset of the phase transition between the (2×4) and $c(4\times4)$ reconstructions.[18,23] If the temperature is too low, or the As flux too high, the surface can become partly $c(4\times4)$ reconstructed during island growth or a postgrowth anneal. An example is given in Fig. 4(a), which shows an STM image for 0.08 ML InAs on GaAs(001)-(2×4) after annealing for 5 min at the growth temperature. The underlying surface structure has $c(4\times4)$ symmetry, but the characteristic blocks of six As atoms that form the main component of the ideal structure are rarely complete,[24] and the residual (2×4) domains lie in the layer *above* the $c(4\times4)$ surface (see white arrow in the figure). Both of these observations have been made for the two-phase GaAs surface in the absence of In.[18] The small white features dotted about the surface, with an apparent height of ~1 Å , could well be due to In atoms (their density is around 0.02 ML) and are not observed for the In-free two-phase surface. Through careful control of the conditions, the surface can remain (2×4) after equilibration. The RHEED patterns change in the [110] direction and show a sharply reduced 2/4 order streak, characteristic of a (2×4) surface with many kink defects in the dimer-pair rows.[16,24] A typical STM image of an equilibrated InAs/GaAs-(001) (2×4) surface is shown in Fig. 4(b). There are many kink defects and quite substantial patches of missing As dimers, some of which show short-range ordering. In GaAs homoepitaxy the surfaces return to very good (2×4) ordering, but the disorder observed in the heteroepitaxial case is likely to arise from strain accomodation due to the significant alloying that occurs on the $c(4\times4)$ surface.[12,13]

In conclusion, island size statistics have been measured for 2D InAs islands grown by MBE on GaAs(001)-(2×4). The islands appear to grow in a very similar way to the cases of GaAs/GaAs and InAs/InAs homoepitaxy on the (001)-(2×4) surface, and exhibit the same basic scaling properties. The key difference between heteroepitaxy and homoepitaxy appears to be a slight favoring of island growth compared with nucleation in the former case. Strain is not necessarily the cause of the anisotropic scaling properties involved; instead, we believe the same reconstruction-based mechanism that operates in homoepitaxy leads to anisotropies in the island size distributions obtained for InAs/GaAs heteroepitaxy.

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