

Effect of Strain on Surface Morphology in Highly Strained InGaAs Films

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The early stages of growth of highly strained $\text{In}_x\text{Ga}_{1-x}\text{As}$ on GaAs(100) have been investigated as a function of composition. The evolution of the film microstructure as determined by *in situ* STM and RHEED is from a two-dimensional rippled surface in the beginning stages of growth to a three-dimensional island morphology. A growth mode is proposed whereby strain relaxation is initially achieved through the kinetically limited evolution of surface morphology. In contrast to traditional critical-thickness theories, significant strain relief is accommodated by a coherent island morphology. This study represents a new view for both the growth mode and initial strain relaxation in thin films.

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The growth of strained films has become a field of intense study in recent years. In particular, $\text{In}_x\text{Ga}_{1-x}\text{As}$ on GaAs is an ideal system for studying the dependence of the growth morphology on strain because the lattice constant of the overlayer film can be varied by 7.2% as the In concentration is changed from 0 to 1. Traditionally, the evolution of the film morphology has been viewed within the context of a critical overlayer thickness. van der Merwe examined the situation of a film under uniform strain and showed that at a strain-dependent thickness (i.e., "critical thickness") it is energetically favorable for an array of misfit dislocations to partially relax the overlayer.¹ Examination of experimental results for heteroepitaxial semiconductor growth revealed that generally the transition to a relaxed state occurred at a larger thickness than predicted by this theory. This has led to the inclusion of kinetic mechanisms in the theory and the possible metastability, after critical thickness, of the uniformly strained thin film.^{2,3}

Recently, a model has been proposed to explain the critical thickness t_c and growth-mode transitions in highly strained $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}(100)$.⁴ In keeping with the traditional view, it is assumed that partial relaxation of the overlayer strain occurs through the nucleation of misfit dislocations. In particular, the experimental observation that the onset of the surface-lattice-constant relaxation (i.e., t_c) increases as either the growth temperature or In composition (i.e., strain) is reduced is explained on the basis of the kinetics of dislocation motion. In addition, it is proposed that the system undergoes a layer-to-island growth-mode transition as a result of cluster nucleation initiated by dislocations. There are several problems with this model. For low-strain films and/or low temperatures it fails to accurately predict the correct critical thickness. The data for $\text{In}_{0.25}\text{Ga}_{0.75}\text{As}/\text{GaAs}$ shown in Ref. 5 are in clear disagreement with this model.⁵ For example, at a growth temperature of 450°C a critical thickness of 340 Å is observed experimentally, whereas the model predicts a thickness which is much larger. The most pronounced disagreement with the theory is seen in some recent data by Guha, Maduhkar, and Rajkumar, where transmission-electron-microscopy (TEM) studies show that dislocations are not

present in an $\text{In}_{0.5}\text{Ga}_{0.5}\text{As}$ film grown beyond t_c , even though the surface strain has relaxed and islands have formed.⁶ These studies reveal a microstructure made up of partially relaxed, coherent islands (this microstructure is also observed in other strained systems).⁷

In light of these discrepancies we have studied the morphological evolution of $\text{In}_x\text{Ga}_{1-x}\text{As}$ on GaAs(100) using reflection high-energy electron-diffraction (RHEED) and scanning tunneling microscopy (STM). Our observations lead us to propose an alternative model for the early stages of highly strained InGaAs growth. It is based on the kinetically controlled evolution of the film toward a coherently islanded microstructure which is energetically favored over a uniform film. A link between the film morphology and strain relaxation is made. This model does not require the inclusion of dislocations for the initial relaxation of the overlayer strain or the nucleation of island growth.

Experiments are carried out in a UHV system which includes a molecular-beam-epitaxy (MBE) growth chamber equipped with RHEED and an analysis chamber containing an STM. Special care has been taken in the heating and shrouding of the sample so that the evolution of the growth surface can be quenched. Examination of RHEED patterns during and after growth allows us to conclude that there is minimal change in the growth front over the time from termination to the end of our STM measurements.⁸ Starting with a p^+ (zinc-doped) GaAs(100) substrate a 400-nm nominally undoped GaAs buffer layer was grown at 570°C under arsenic-rich conditions. Figure 1(a) shows a representative STM image of this buffer-layer surface. The dark rows running diagonally across the image in the $[\bar{1}10]$ direction are spaced 1.6 nm apart, corresponding to the spacing between missing As dimers on the (2×4) -reconstructed surface. Buffer layers onto which all of the subsequent InGaAs film were deposited were grown in this manner. In all cases, the InGaAs growth conditions were as follows: substrate temperature 520°C, deposition rate $\sim 0.15 \mu\text{m}/\text{h}$, As-rich overpressure. During growth the film evolution was monitored by RHEED. The thickness and composition were both determined from the specular intensity oscillations.

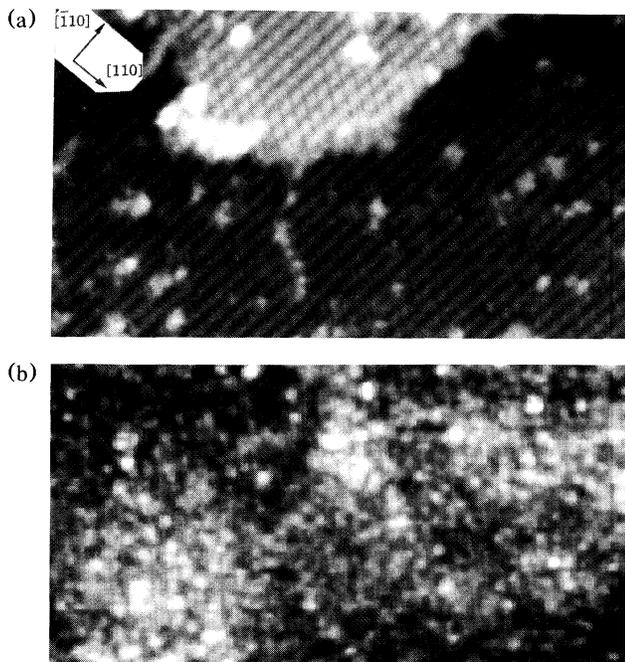


FIG. 1. (a) STM image of a GaAs(100) buffer layer. The scan range is $60 \text{ nm} \times 30 \text{ nm}$. The tunneling voltage (V_t) was $+2.8 \text{ V}$, applied to the sample, and the tunneling current (I_t) was 70 pA . (b) STM scan ($90 \text{ nm} \times 50 \text{ nm}$) of $\text{In}_{0.4}\text{Ga}_{0.6}\text{As}$ on GaAs(100), after four monolayers were deposited. $V_t = +3.0 \text{ V}$ and $I_t = 30 \text{ pA}$. The small islands covering the surface have a lateral extent of $\sim 3\text{--}5 \text{ nm}$ and a corrugation of $\sim 0.5 \text{ nm}$.

With the commencement of InGaAs growth there is a noticeable change in the observed RHEED features. The RHEED pattern transforms from a sharp 2×4 streaky pattern to a somewhat diffuse 1×1 streaky pattern, and the specular intensity drops considerably with respect to that measured during the GaAs growth. Fig-

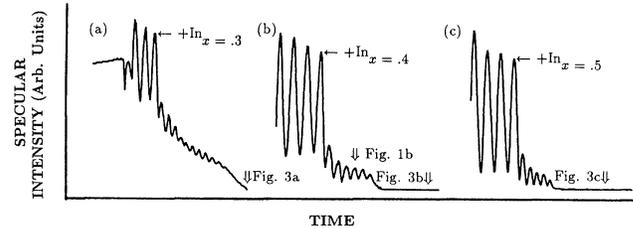


FIG. 2. RHEED specular intensity oscillations for $\text{In}_x\text{Ga}_{1-x}\text{As}$ on GaAs(100), for x equal to (a) 0.3, (b) 0.4, and (c) 0.5. $\phi = [110]$. The \leftarrow indicates where the In shutter was opened and the \Downarrow labels the point at which growth was terminated and the indicated STM image acquired.

ure 2 shows the specular intensity oscillations during the InGaAs growth for films of varying strain. The key features include an immediate reduction in the intensity of the oscillations as compared with the buffer layer, as well as a reduction in the number of discernible oscillations with increasing strain. Soon after the oscillations have damped completely, we observe that the RHEED pattern finishes its evolution to a spotty pattern and the lattice-constant relaxation saturates at the bulk value. From the standpoint of RHEED, these observations characterize the early stages of growth and they are well documented by others.^{5,9,10}

To determine the InGaAs surface morphology during growth we have terminated film growth at various thicknesses for examination by STM. Arrows (\Downarrow) in Fig. 2 indicate where these points were in the growth cycle. In the beginning stages the surface is already considerably changed from that of the buffer layer. Figure 1(b) shows a representative image of the surface morphology after 4 ML's (monolayers) of $\text{In}_{0.4}\text{Ga}_{0.6}\text{As}$ were deposited. The surface is textured with islands of height 0.5 nm and diameter $3\text{--}5 \text{ nm}$. The RHEED signature from this

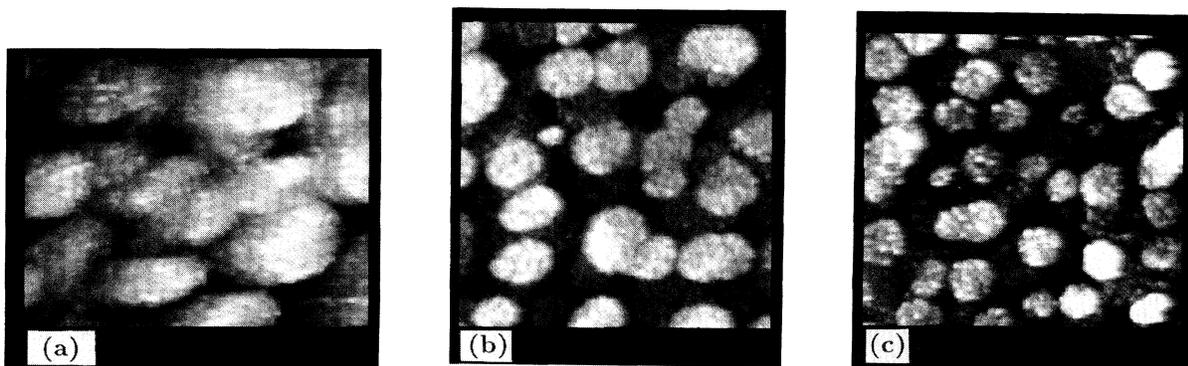


FIG. 3. STM images of $\text{In}_x\text{Ga}_{1-x}\text{As}$ on GaAs(100). (a) 14 ML's, $x = 0.3$ (2.2% mismatch), scan range = $154 \text{ nm} \times 130 \text{ nm}$, full-scale height range (from black to white) is 8.0 nm , $V_t = 2.6 \text{ V}$, $I_t = 0.4 \text{ nA}$. (b) 11 ML's, $x = 0.4$ (2.9% mismatch), scan range = $150 \text{ nm} \times 154 \text{ nm}$, full-scale height range is 8.5 nm , $V_t = 2.6 \text{ V}$, $I_t = 60 \text{ pA}$. (c) 12 ML's, $x = 0.5$ (3.6% mismatch), $150 \text{ nm} \times 147 \text{ nm}$, full-scale height range is 9.0 nm , $V_t = 2.7 \text{ V}$, $I_t = 20 \text{ pA}$. For the films represented in (b) and (c), 4–5 ML's of additional material were deposited after the RHEED pattern became spotty. During this extra growth the pattern sharpened. For the film represented in (a) there was no extra growth.

surface shows a streaky 1×1 pattern and the intensity oscillations are still occurring. We also note that the surface lattice constant has begun to relax somewhat for this film. STM images of the latter stages of growth as a function of strain are shown in Fig. 3. The RHEED signature from these surfaces shows a 3D spot pattern and the surface lattice constant has relaxed to that of the bulk. We observe in these images that each film has broken up into large, approximately circular islands. The height of the islands is approximately 2–3 times that of the overlayer thickness if it were uniformly distributed. The height and width distributions are narrow. Considering the volume of the islands and their areal density, it appears that a significant amount of the previously deposited film material has been incorporated in these clusters. Note the systematic increase in island diameter with decreasing strain.

From RHEED data there is mounting experimental evidence which suggests that in the beginning stages of growth the strained overlayer does not grow in the same layer-by-layer manner as the GaAs buffer layer. The amplitudes of the InGaAs intensity oscillations are much smaller than the GaAs oscillations. The InGaAs streaked pattern exhibits comparatively more diffuse scattering. Finally, it has been observed that the width of the specular beam increases with the start of the InGaAs growth.¹¹ The STM results confirm the diffraction data and indicate that there is both surface roughness and vestiges of layer growth in the beginning stages. These effects might be viewed as resulting from growth at too low of a substrate temperature. This seems unlikely, however, because the average cation surface migration rate for InGaAs is higher than for GaAs, so we would expect more ideal layer growth.¹² Given the strain dependence of the oscillations in Fig. 2, it is evident that the roughening is strain induced. Lending support to this view are both an atomistic energetic analysis and a continuum linear stability analysis of strained thin films.^{9,13}

In order to provide a more complete description of the InGaAs growth process throughout the early stages we have used simulations and simple analytical models.¹⁴ From the analytical models, we find that in the earliest stages the lowest-energy configuration of a strained film can be one in which partially strain relieved, coherent 3D clusters have formed. That this can be the lowest-energy configuration may be understood by considering that for an island the elastic energy reduction over its volume may outweigh the cost of additional surface energy. To reach this state, however, requires sufficient kinetics of adatom migration. For limited kinetics, it is found that the island morphology should not occur below a strain-dependent thickness. Above it, islands can form such that for a given height the diameter will increase with decreasing misfit, as in Fig. 3. This strain dependence is reasonable, since the energy difference of an island film versus a flat film is less for films with smaller misfit. If

the kinetics is very limited, but not freezing out surface diffusion, islanding may be practically suppressed and quasi layer-by-layer growth may occur.^{15,16} For this highly strained system we believe that there is no net driving force for a 2D flat film. This is in contrast to unstrained systems where 2D growth is energetically favored and kinetic limitations can produce roughening.

Utilizing these ideas, we propose a simple microscopic model for the growth of highly strained InGaAs. Excluding a wetting layer, which results from adsorbate-substrate interactions, the deposited material will nucleate small 2D islands. 3D clusters are not expected to nucleate directly because the critical cluster size, for which the reduction in strain energy dominates the additional surface energy, is too large.¹⁶ As new atoms arrive they will find it energetically favorable to nucleate at the step edges of the 2D islands rather than on top. Thus, the islands will grow laterally as the first layer nears completion. Central to our model, however, is an energy cost, which increases with strain, for these islands to merge. The strain-induced deformation of the layer beneath makes it energetically unfavorable for diffusing atoms to fill in the gaps between merging islands, making it more likely for the remaining first-layer atoms to nucleate the second layer.^{14,17} The subsequently deposited layers will follow the same scenario as they nucleate and grow on each 2D island of the preceding layer. Hence, with continued growth, the film grows in this quasi layer-by-layer fashion. This corresponds to the beginning stages of growth for which oscillations are observed and there is either a negligible or small lattice relaxation. During this roughening stage there is a strain-energy buildup with each successive layer and atoms in the top-most layers will be strain relieved compared to the bottom layers. This strain gradient manifests itself as a chemical-potential difference which is a driving force for atoms in the bottom layers to diffuse to the top. If the chemical-potential difference becomes large, a kinetically controlled coarsening process may occur in which 3D islands grow at the expense of the lower layers and other islands. Given sufficient kinetics, the coarsening will be very rapid and the microstructural change dramatic. We believe that this type of evolution, roughening followed by coarsening, could explain the high-temperature-strain morphological transformations leading to the islanded structure shown in Fig. 3. As such, it constitutes a strain-induced pathway to the 3D microstructure and a mode for strain relaxation which does not rely on dislocation generation.

Our simulations of the film-substrate strain fields associated with the islands reveal that the greatest strain occurs at the edges and the least strain at the tops. For these clusters the lattice mismatch is accommodated by elastic deformation of the substrate, not defects. Depending on the efficiency of the strain relaxation, a coherent island may have lower energy than a dislocated island because of the energy cost associated with disloca-

tions.¹⁸ It has been observed experimentally that dislocations will nucleate at the edges, if the InGaAs clusters become too large.⁶ Thus, for growth beyond the early stages we expect the islands to increase in size, edge dislocations nucleating to relieve the high strains. This additional defect-induced relaxation of the overlayer reduces the driving force for islanding, allowing for coalescence. Island merging will produce defects, perhaps threading dislocations. As the film thickens (10–100 nm), strain is relieved by dislocations and the surface will become smooth again. Eventually, the film will grow layer by layer with the bulk InGaAs lattice constant.

Using this model we believe that all of the experimental observations characterizing the early growth stages of highly strained InGaAs on GaAs(100) can be explained. The commonly observed high-temperature (520°C) characteristics (rapid transition to a spotty pattern and overlayer lattice constant) usually associated with the traditional critical thickness result from the strain-induced rapid morphological transformation discussed above. Lowering the strain (In composition) reduces the driving force for roughening and coarsening, and lowering the temperature kinetically limits it. Reduction of these parameters produces smoother films which result in larger thicknesses for the onset of relaxation, consistent with experiment. The gradual lattice-constant relaxations observed at low temperatures^{5,9,10} are explained by poor surface-diffusion kinetics. The observance of relaxation without the appearance of a spotty pattern implies that the film is constrained by kinetics and/or an insignificant driving force to remain in the roughening mode (i.e., no coarsening). Particularly troublesome for dislocation models is the distinctive observation that at low growth temperatures ($T_{\text{sub}}=450^\circ\text{C}$) surface lattice relaxation only occurs during growth.⁵ When growth is stopped, relaxation halts abruptly. In the dislocation models, the onset of lattice relaxation occurs beyond a critical thickness via thermally activated dislocation motion; thus there is no dependence on growth. In our model, surface relaxation occurs while the film grows in a quasilayered fashion. If surface diffusion is very limited, then relaxation will stop with growth termination.

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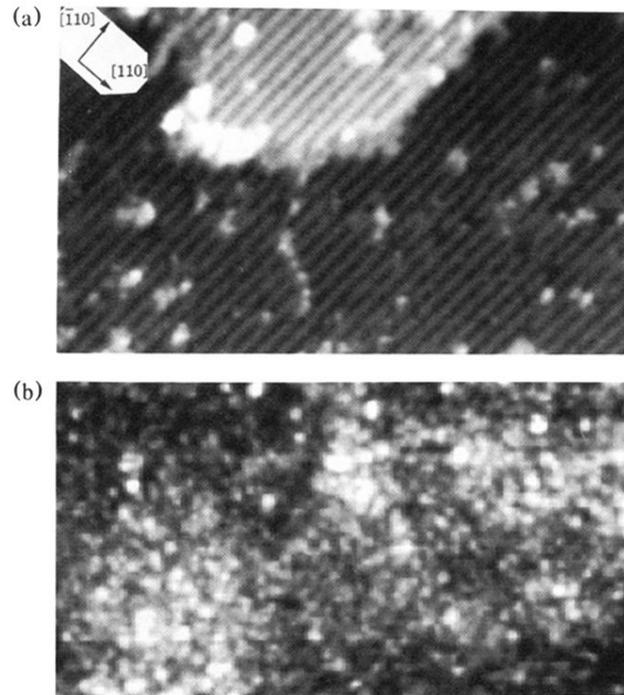


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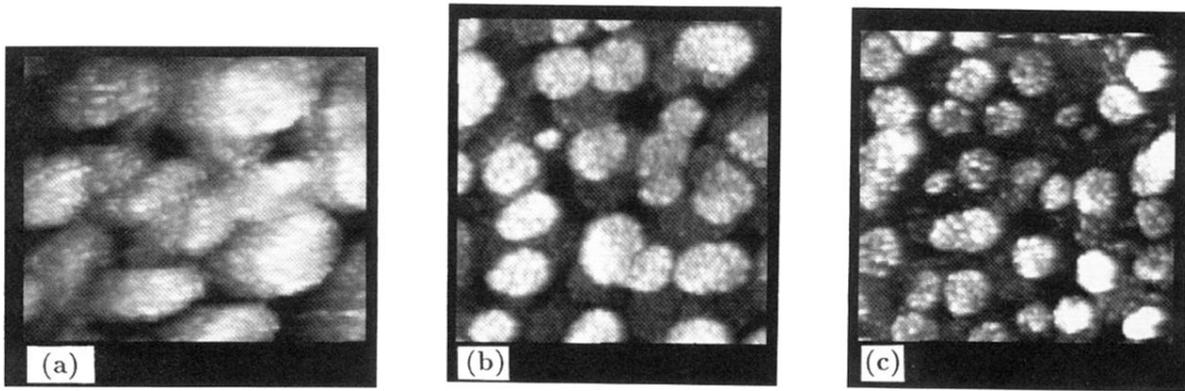


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