Evolution of island–pit surface morphologies of InAs epilayers grown on GaAs (001) substrates

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We report on a study of the morphological evolution of InAs layers grown on GaAs (001) substrates by molecular-beam epitaxy under In-rich conditions. The surface morphology of the InAs layers is characterized by a feature of island–pit combinations. We show that the vertical sizes of the islands and pits can grow simultaneously beyond the average layer thickness, up to several hundred nanometers. The composition of the islands is found to be ternary $In_xGa_{1-x}As$ rather than the expected binary InAs due to intermixing of the layer and substrate materials. We determine that this intermixing is caused by dissociation of the exposed GaAs at the pits, followed by migration of excess Ga atoms and their incorporation into the islands. The density of the island–pit combinations keeps nearly constant for different layer thicknesses. Eventually, as the layer grows beyond a certain thickness, the pits are filled up by the expanding islands, forming a nearly pure island morphology at the growth front. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1354637]

I. INTRODUCTION

The morphological instability of strained epitaxial layers has attracted great attention in the past decade due to its importance in both science and technology. Lattice misfit strain results in roughening of the growth front of a layer as a consequence of competition between strain and surface energy. Surface morphologies with characteristic features of wavy undulation,[1] island,[2–4] pit,[5,6] and their combinations[7,8] have been observed according to the amount of strain and the growth conditions of the layers. Atomic diffusion from the substrate[9] and cap layer[10] into the islands was also reported recently, indicating a rather complicated kinetics of strained epitaxial growth. Some progress has been made lately in controlling the size distribution of (InGa)As islands grown on GaAs (001) via multilayer growth,[11] tuning the composition,[12,13] or using a laterally modulated surface,[14] that in turn considerably improves the optoelectronic performances of the materials. Along different lines, studies of SiGe/Si systems have shown that the combination of islands and pits can be a very promising way to control the size, shape, and order of the self-assembled quantum dots.[7,15]

In this article we report on a study of the morphological evolution of relatively thick InAs layers grown on GaAs (001) substrates under an In-rich condition. A surface morphology characterized by island–pit combinations was observed from our InAs layers with their average thicknesses ranging from 4 to 80 nm. We show that the islands and pits first grow simultaneously as the layer deposition proceeds. Both the island height and the pit depth can be much greater than the average layer thickness. This suggests that considerable mass transport from substrate into the islands was taking place during the growth. However, when the layer becomes sufficiently thick, the pits are eventually filled up by the expanding islands, forming nearly pure island morphology at the surface.

II. EXPERIMENTAL DETAILS

The layers were grown by molecular beam epitaxy (MBE) on a V80H system. Before InAs growth, the GaAs (001) substrates were heated at 600 °C under As_2 atmosphere. The growth starts with a 200 nm thick GaAs buffer layer deposited at 550 °C under As-rich condition with a III/V flux ratio of about 15. Then, the InAs layers were grown under In-rich conditions, with a V/III flux ratio of about 6, at a temperature of 380 °C. The idea of growing an InAs layer under In-rich conditions is to try to maintain two-dimensional growth.[16–18] Arsenic flux was maintained after the layer growth was terminated. Reflection high energy electron diffraction (RHEED) showed streaky patterns at this stage. Six samples with InAs layer thicknesses of 4, 10, 20, 40, 60, and 80 nm were prepared with RHEED calibration.[19]

Surface morphologies of the InAs layers were measured in air by a NanoScope III atomic force microscope (AFM) in contact mode. X-ray diffraction measurements were performed at the Beijing Synchrotron Radiation Facility on beamline 4W1C with an x-ray wavelength of 1.537 Å. Scanning electron microscopy analyses were carried out on a S-4200 microscope operating at 20 kV.

III. RESULTS AND DISCUSSION

Figure 1 shows the AFM images of the six samples in a sequence of increasing layer thickness. The scan area is $20 \times 20\ \mu m^2$. For the 4 nm thick layer [image (a)], dense pits, mostly with a diameter of $\sim 0.1\ \mu m$ and a few larger ones

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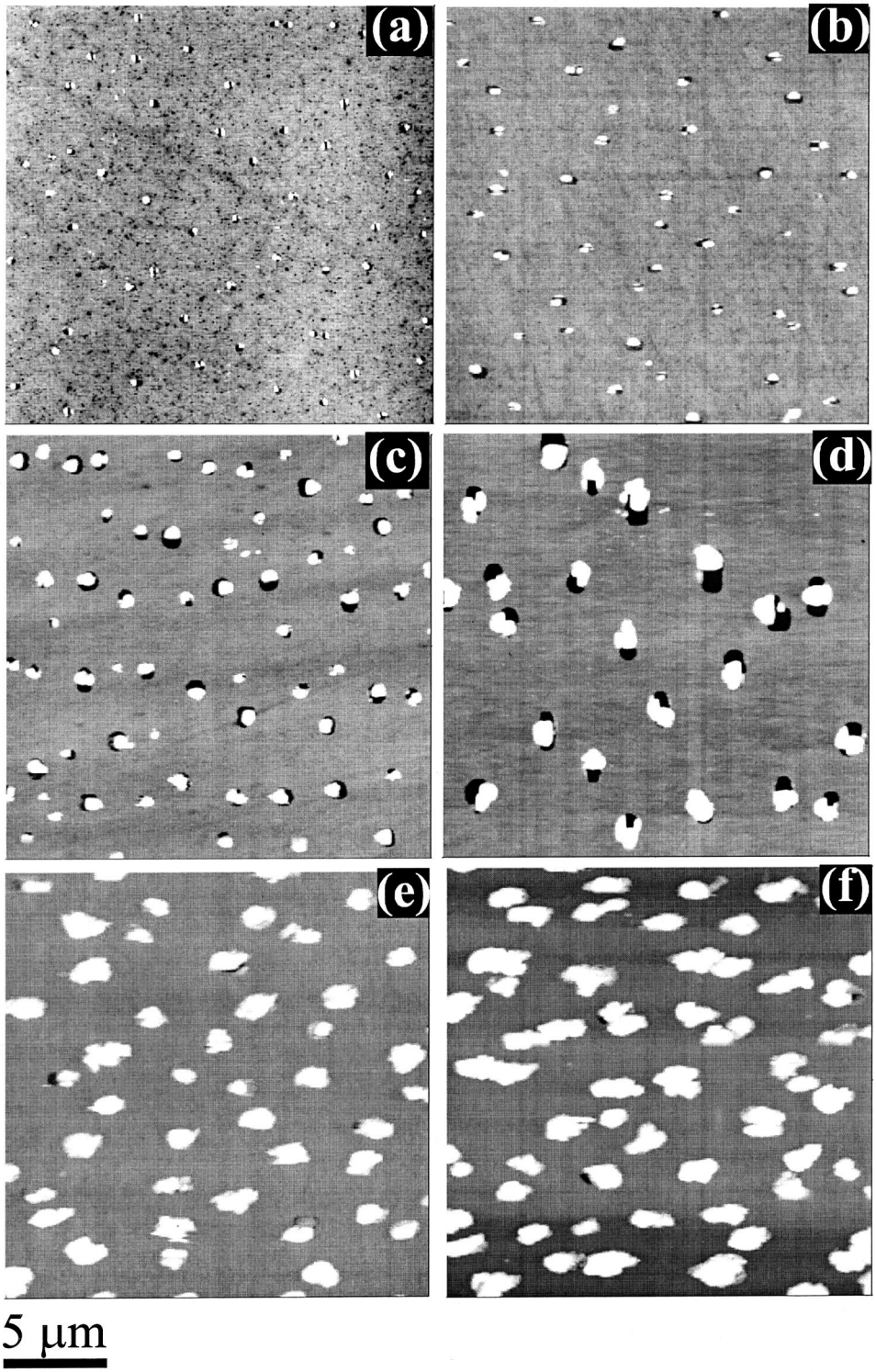

FIG. 1. AFM images of six InAs layers grown on GaAs (001) substrates. The layer thicknesses are (a) 4 nm, (b) 10 nm, (c) 20 nm, (d) 40 nm, (e) 60 nm, and (f) 80 nm, respectively.

with a diameter of ~0.25 μm, are observed. Islands are observed only beside the larger pits, forming a feature that is called an island–pit combination in this article. The islands appear either singly on one side of the pits or in pairs on opposite sides of the pits. This morphology is quite similar to that observed by Weil *et al.*[7] on Ge layers grown on Si (001). The number density of the island–pit combinations differs slightly from region to region across the wafer, but has a good statistic of about $0.13/\mu m^2$. For the 10 nm thick layer [image (b)], the size of the island–pit combinations grows, while those small pits become flattened. We also note that the density of the island–pit combinations is quite similar to that of the 4 nm thick layer. For the 20 nm thick layer, the AFM scan [image (c)] shows again a similar density of island–pit combinations. However, both the islands and the pits in this image are larger in size. As the layer thickness grows up to 40 nm [image (d)], we see that both the islands and the pits expand continuously. When the layer grows up to 60 nm thick, however, the island size increases further, but the pits have stopped expansion and have almost been filled

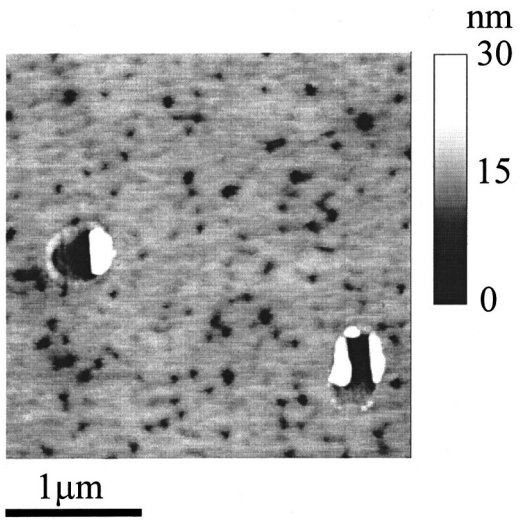

FIG. 2. A small area AFM image taking from the 4 nm thick layer.

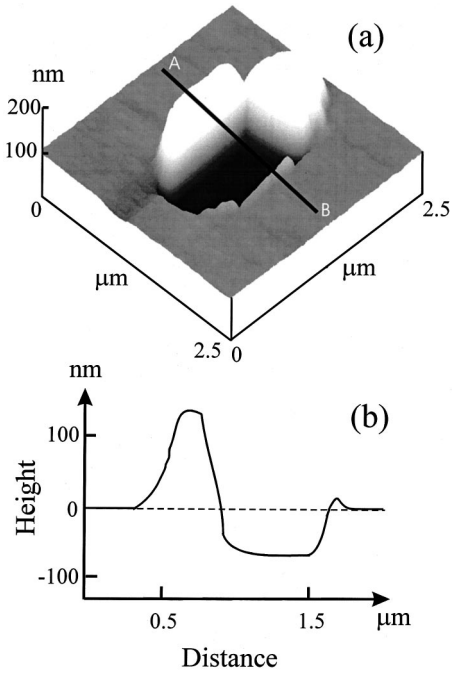

FIG. 3. (a) A small area AFM image taking from the 40 nm thick layer, and (b) the corresponding AFM cross-sectional analysis.

up by the islands [image (e)]. Nevertheless, no new nucleation of islands on the flat regions separating the island–pit combinations is observed. Further increase in the layer thickness causes further increase in the island size, but again does not lead to new nucleation of islands on the flat regions [image (f)]. Consequently, the number density of the islands in these two thickest layers is in agreement with the number density of the island–pit combinations in the thinner layers. Indeed, traces of the earlier existence of pits can be distinguished at most of the islands in images (e) and (f). Apparently some of the pits have not been completely filled up yet. It is also noticeable that the islands in images (e) and (f) are actually island clusters.

The above observations give directly the following information. (1) There is a minimum pit size requirement for combined island–pit growth. This has been clearly shown by the change from Fig. 1(a) to Fig. 1(b), where smaller pits were flattened eventually, while the larger ones became island–pit combinations. The nearly constant density of the island–pit combinations throughout the six samples further supports this conclusion. (2) The growth of the InAs layer does not lead to new island nucleation on the flat regions away from the pits, but results in continued growth of the preexisting islands and pits. The simultaneous growth of the islands and pits is an important phenomenon, which requires considerable mass transport from the pits into the islands during the growth. (3) The pits do not expand all the way as the islands do when the layer grows thicker and thicker. At a certain stage, as noted earlier, the pits stop growth and are eventually filled up completely by the expanding islands.

To have a closer view of the feature of island–pit combination, AFM scans on a smaller length scale were carried out. For example, Fig. 2 shows a $3\times3\ \mu m^2$ AFM scan for the 4 nm thick layer. Two typical island–pit combinations were captured in this image. One of the two consists of simply an island–pit couple, and the other one contains more than one island. The white ring around the peripheries of both pits is an obvious indication of mass transport from the pit center to the rim. In Fig. 3(a), a $2.5\times2.5\ \mu m^2$ scan for the 40 nm thick layer is shown. The lateral size of both the pit and island is about 1.5 μm. A cross-sectional analysis of this island–pit combination is given in Fig. 3(b), which shows that the pit bottom is quite flat. The height of the island and the depth of the pit are about 250 and 100 nm, respectively. Both are much larger than the layer thickness of 40 nm. This result is true for all samples thinner than 40 nm. In Table I the typical vertical and lateral sizes of the islands and pits measured by AFM are listed. We see that the height of the islands grows monotonically with the increase in the layer thickness. The pits, on the other hand, reach a maximum size at a layer thickness between 40 and 60 nm. Then they stop growth and begin to be filled up by the expanding islands.

The simultaneous growth of islands and pits requires significant mass transport from pits into islands. Our results suggest that the materials around the pits have been ''dug'' out and piled into the islands. The fact that the pits are deeper than the layer thickness implies that the substrate material of GaAs has been ''pumped'' out via the pits. Therefore one may expect that the islands are ternary (In, Ga)As

TABLE I. Typical sizes of the islands and pits obtained by cross-sectional AFM analyses for the InAs layers of different thicknesses. D_{iv} and D_{il} represent the height and lateral extension of the islands. D_{pd} and D_{pl} represent the depth and lateral extension of the pits.

Layer thickness (nm)	D_{iv} (nm)	D_{il} (μm)	D_{pd} (nm)	D_{pl} (μm)
4	10	0.25	6	0.25
10	40	0.70	20	0.70
20	80	1.00	45	1.00
40	150	1.50	100	1.50
60	190	1.60	50[a]	
80	240	2.10	35[a]	

[a]These data were measured from pits that were not completely filled up.

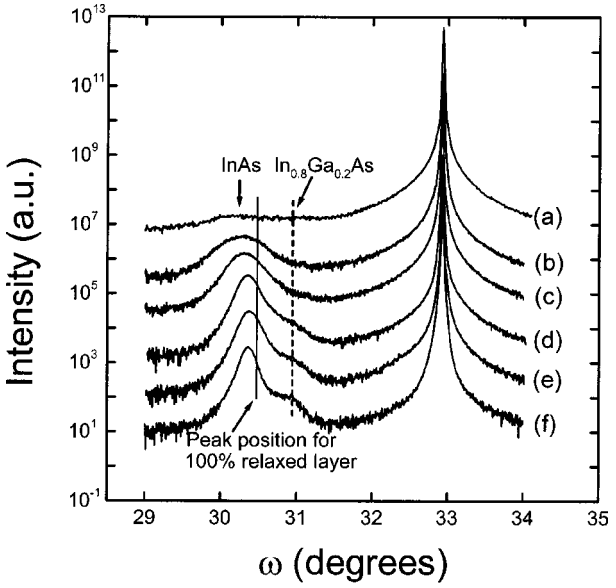

FIG. 4. X-ray double-crystal 004 rocking curves of the six samples. Curves (a)–(f) correspond to layers with thicknesses of 4, 10, 20, 40, 60, and 80 nm. The solid line indicates the position where the InAs peak should be if the layer is completely strain relaxed. The dashed line indicates the position of the shoulder peak.

rather than binary InAs. To explore this, x-ray diffraction and cross-sectional energy dispersive x-ray (EDX) analyses were made. In Fig. 4, x-ray double-crystal 004 rocking curves of the samples are displayed. In addition to the GaAs substrate reflection peak, the InAs peak is seen for all samples. Moreover, a shoulder peak (indicated by the dashed line) on the higher angle side of the InAs peak is also seen. This peak is obviously more pronounced for the thicker layers. Careful analyses of the InAs peak reveal that none of the six InAs layers is completely strain relaxed. The relaxation degrees of the InAs layers are determined to be in the range of 83% to 90%. Therefore one possible source of the shoulder peak may be the nonuniformity of strain relaxation in the layer containing three-dimensional islands, as observed by Kegel *et al.*[20] and Zhang *et al.*[21] By this we mean that a small portion of the layer is more relaxed than the rest. This will lead to a smaller lattice plane spacing along [001] for this part of the layer and consequently a diffraction peak on the higher angle side of the main layer peak, i.e., the islands are heterogeneous species. For example, the InAs islands may undergo some elastic relaxation[21] by, e.g., the bending of lattice planes at the edges, or the strain fields of the misfit dislocations may inhomogeneously distort the epilayer.[22] However, theoretical simulation of the rocking curves (not shown) indicates that even if a part of the layer were completely strain relaxed, its corresponding diffraction peak (position indicated by the vertical solid line in Fig. 4) could not reach the position of the shoulder peak on the far higher angle side (indicated by the dashed line). Thus nonuniform strain relaxation will not be able to cause the shoulder peak, though it may result in broadening and asymmetry of the InAs peak. Therefore the only plausible explanation for the shoulder peak is that a distinct part of the layer contains gallium, i.e., Ga has been mixed into the InAs deposit. In this

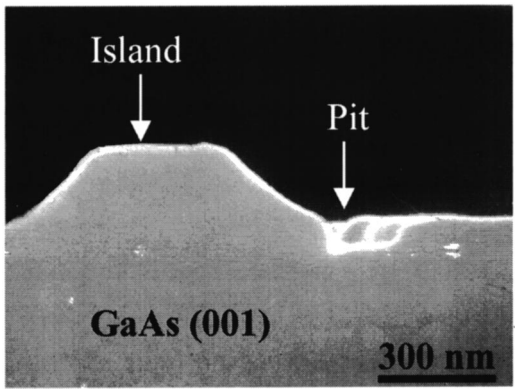

FIG. 5. A cross-sectional SEM image of an island–pit combination in the 40 nm thick layer.

manner, the lattice plane spacing of the layer will be reduced by forming (In, Ga)As alloys. This will subsequently cause a distinct x-ray peak (the InGaAs peak) on the higher angle side of the InAs peak. In our case, the substrate is the only source of gallium. The two possible places where GaAs and InAs may meet and intermix are the layer–substrate interface and the island–pit combinations. The possibility of significant intermixing of InAs and GaAs at the layer–substrate interface can be easily excluded by considering the following facts. The intensity of the InAs peak in the x-ray rocking curve of the 4 nm thick layer is overwhelming in comparison with its corresponding shoulder peak. This indicates that the intermixing at the interface, if it exists, is insignificant and has ceased well before reaching the 4 nm thickness. In this case, the intensity of the shoulder peak should not increase with the increase in the layer thickness. As a matter of fact, no significant interface mixing has ever been observed for InAs layers deposited on GaAs (001).[10,23] Thus the only possible place where the substrate material can migrate into the InAs deposit is the island–pit combinations, where the substrate is exposed during the growth. With the increase in the layer thickness, more $In_xGa_{1-x}As$ ternary alloy is formed, so that the intensity scattered by the ternary alloy increases as well. For the thin layers ($\leqslant$20 nm), the scattering from the ternary alloy is weak due to the small amount of the ternary alloy formed there, but it does result in the asymmetry of the rocking curves. For the thick layers ($\geqslant$40 nm), the amount of the ternary alloy formed can be large enough to cause a distinct diffraction peak (the shoulder peak). A cross-sectional scanning electron microscopy (SEM) image of an island–pit combination in the 40 nm thick layer is shown in Fig. 5. The picture was taken by cleaving the sample along the (110) lattice plane. EDX analysis confirms the existence of Ga in the island, although the quantitative value obtained this way is not reliable due to the limited resolution (caused by the shifting of the electron beam while making statistical counting and the possible nonuniformity of the island composition, as discussed below). From the x-ray data, we estimate the peak composition of this distinct phase in the islands is about $In_{0.8}Ga_{0.2}As$.

The exposure of GaAs substrate at pits and the diffusion of GaAs into the islands are quite interesting results. The formation of surface pits in the strained epilayers has been

investigated by Chen *et al.*[5] and Weil *et al.*[7] in their earlier works. They showed that strain induced surface diffusion associated with local perturbations at the substrate surface results in the pit formation. They suggested that at the perturbed sites, the substrate surface is strained. Thus the depositing material will prefer to not remain at these sites, but rather diffuse away, leaving behind the pits on the surface. In the extreme, if the local strain at these perturbed sites is sufficiently large, no deposited material will stay there and the substrate will eventually be exposed. In our case, because the layer was prepared under In-rich conditions, dissociation of GaAs is expected (GaAs starts to dissociate at temperature as low as 350 °C). The excess Ga may then serve as the source of the perturbation. However, only pits above a critical size will lower the energy of the strained layer.[24] Corners or edges of the large pits are the most preferred sites to attach newly deposited materials because at these sites the strain energy is most relieved. Islands are then formed there via surface diffusion. Figure 2(a) shows exactly such a result, where a dam or ridge around a pit is formed, which is clearly an effect of surface diffusion.

Continued deposition of the strained layer may lead to further relaxation at one or more of the pit corners or edges by dislocation or (In, Ga)As alloy formation. The islands at these relaxed places will grow rapidly at the expense of the material around the pits. This in turn results in the simultaneous growth of the pit size. It is possible that at this stage, the increase in the surface energy associated with the facets is sufficient to accommodate the strain energy. Figures 1(a)–1(d) show exactly such a process. However, at some critical layer thickness, the excess surface energy associated with the pit walls will become larger than that required for strain relaxation, i.e., less surface energy is required to offset the decreased strain energy. Subsequently, the pits will be gradually filled up, as shown in Figs. 1(e) and 1(f). Therefore the islands formed in the thick layers may have nonuniform composition, since less Ga is available at this stage. Eventually, we expect that the shell of the islands tends to InAs, while the core remains $In_xGa_{1-x}As$.

We now discuss the diffusion of Ga into the islands. In a recent theoretical study of the dramatic mass transport in the formation of quantum dots, Bottomley[25] calculated the Gibbs free energy of $In_{1-x}Ga_xAs$ alloy. He found that the Gibbs energy has the lowest value when x is about 0.2. Therefore there is a trend for InAs and GaAs to mix. However, for a growing layer, the surface free energy also plays an important role. For example, segregation of indium towards the surface is a well-known phenomenon when GaAs is deposited on top of the InAs,[23,26] but not vice versa.[10,23] A similar phenomenon was also observed in a Co/Cu metallic layered system, where the undermined Cu was pumped out to the surface via pinholes to form Cu–Co alloy.[27] The driving force for this pumping effect is the lower surface energy of Cu over Co. In our case, the existence of deep pits, where the GaAs substrate is exposed, provides a channel for Ga migration into InAs, which is normally not possible in the case of planar growth. The In-rich growth condition helps this process by making dissociation of GaAs much easier.

It is interesting to note that the observed Ga fraction in the islands closely agrees with the predication of 20% by Bottomley.[25] Therefore we suggest that the melting of GaAs and InAs at pits is quite plausible due to the large stress at these places. The stress-induced melt then facilitates mixing of InAs with the substrate and mass transport can occur, producing a ternary alloy of $In_xGa_{1-x}As$ in order to minimize the local free energy.

IV. CONCLUSION

We have studied the morphologies of MBE grown InAs layers on GaAs (001) under an In-rich condition. The layer surface is characterized by a feature of island–pit combinations. As the layer grows, both the islands and pits grow up simultaneously before reaching a critical layer thickness. The height of the islands and the depth of the pits at this stage are much larger than the average layer thickness. The substrate is exposed at the pits, making the continuous dissociation of GaAs and the intermixing of GaAs and InAs possible. The islands, nominally InAs, are found to be actually $In_xGa_{1-x}As$. Above the critical thickness, the pits are gradually filled up by expanding islands.

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