Formation of InAs/GaAs quantum dots by dewetting during cooling

R. P. Mirin, A. Roshko, and M. van der Puijl[a)]
Optoelectronics Division, National Institute of Standards and Technology, 325 Broadway, Boulder, Colorado 80305-3328

A. G. Norman
National Renewable Energy Laboratory, 1617 Cole Boulevard, Golden, Colorado 80401

(Received 24 October 2001; accepted 28 May 2002)

We describe a method to form InAs quantum dots on GaAs by cooling an InAs film that is deposited at high substrate temperatures. The reflection high-energy electron diffraction pattern taken after deposition of 1.9 monolayers of InAs on (100) GaAs at 540 °C does not display the characteristic spot pattern that is seen when three-dimensional islands form on the surface. The characteristic spot pattern appears when the sample is cooled to about 330 °C, indicating that the three-dimensional islands appear at this temperature. Atomic force microscopy confirms the existence of the islands. An explanation for this behavior based on an increase in intermixing at the InAs/GaAs interface is proposed. [DOI: 10.1116/1.1495094]

I. INTRODUCTION

The formation of InAs and InGaAs self-assembled quantum dots (QDs) on GaAs by the Stranski–Krastanow transition is now a well-established procedure.[1–4] During molecular beam epitaxy (MBE), the reflection high-energy electron diffraction (RHEED) pattern is monitored in order to determine when the surface transition from two-dimensional growth to three-dimensional growth (island formation) occurs.[2] When the electron beam is directed along certain azimuths, the RHEED pattern displays a distinct spot pattern once the three-dimensional islands form on the surface. Under certain growth conditions, the RHEED pattern may display chevrons that give some information about the shape of the islands.[2,5] If structural information such as shapes of the islands is desired, the sample is often rapidly cooled to room temperature for measurement by methods such as atomic force microscopy (AFM), scanning tunneling microscopy (STM), or transmission electron microscopy (TEM). The effects, if any, of this quenching procedure are still uncertain because of the difficulty in characterizing the islands at the growth temperature.

In this article, we will describe our measurements of the evolution of the RHEED pattern during the growth of thin InAs epilayers on GaAs. These measurements show a substantial change in the pattern, from a two-dimensional pattern to a three-dimensional pattern as the substrate temperature decreases. We will also describe the reversibility of this transition by reheating and recooling the samples, where a significant hysteresis is observed.

II. EXPERIMENTAL RESULTS

The samples are grown on (100) GaAs using a solid source MBE machine equipped with a valved arsenic cracker. The substrate temperature is measured using both a noncontact thermocouple on the back of the wafer and a single wavelength pyrometer that can measure temperatures as low as 450 °C. The thermocouple reading is always higher than the pyrometer reading, and the difference increases slightly as the temperature increases. The minimum differential is 110 °C, when the pyrometer reads 450 °C and the thermocouple reads 560 °C. When reporting temperatures in this article, we use the value measured by the pyrometer for temperatures above 450 °C and the thermocouple reading minus 110 °C as an estimate below this temperature. After thermal desorption of the oxide, the following sequence of epilayers is deposited at 600 °C to obtain a smooth surface: 250 nm GaAs, 50 periods of a 2 nm AlAs/2 nm GaAs superlattice, and 250 nm GaAs. The sample is then annealed under an arsenic flux for 2 min before it is cooled to the quantum dot growth temperature. The InAs epilayers are deposited 0.1 monolayers at a time, followed by a 10 s anneal in the arsenic flux. This cycle is repeated until the total number of monolayers has been deposited.

The RHEED pattern is monitored with the electron beam incident along the $[0\bar{1}1]$ azimuth. This azimuth corresponds to the fourfold reconstruction on As-stabilized (100) GaAs when the substrate temperature is around 600 °C. We deposit about 1.9 monolayers of InAs by cycling through the above sequence 19 times. At a substrate temperature of 540 °C, we observe the RHEED pattern shown in Fig. 1(a). This pattern is very diffuse, and the only prominent feature is the specularly reflected spot. There is no indication of the diamond-like pattern consisting of two spots along the (00) streak and one spot on each of the (01) and $(0\bar{1})$ streaks that is commonly reported on this azimuth when three-dimensional islands form on the surface.

The sample is then cooled in the As flux at a rate of 25 °C/min (as measured by the thermocouple). The RHEED pattern evolves as the sample cools, acquiring first-order diffraction streaks. When the temperature is about 330 °C, we observe [Fig. 1(b)] the formation of split spots along the specular streak and individual spots on the first-order diffraction streaks. We can still observe the diffraction streaks, and

[a)]Current affiliation: Physics Dept., Fontys Hogescholen Eindhoven, Netherlands.

1490

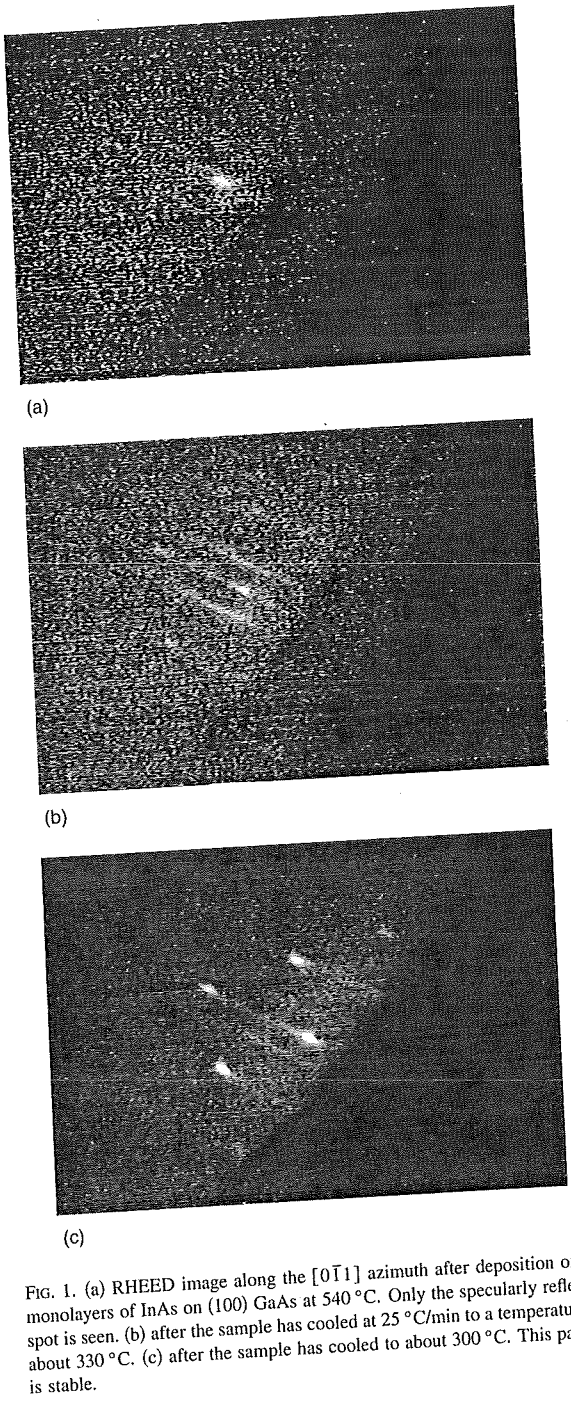

FIG. 1. (a) RHEED image along the [0$\bar{1}$1] azimuth after deposition of 1.9 monolayers of InAs on (100) GaAs at 540 °C. Only the specularly reflected spot is seen. (b) after the sample has cooled at 25 °C/min to a temperature of about 330 °C. (c) after the sample has cooled to about 300 °C. This pattern is stable.

the intensity of the spots is only slightly brighter than the intensity of the streaks. These spots continue to intensify, and at a temperature of about 300 °C the pattern shown in Fig. 1(c) becomes stable. The spots have increased in intensity so that they now have high contrast with respect to their corresponding streak. These spots are consistent with the transmission pattern seen on this azimuth during the formation of QDs at growth temperatures of 450–550 °C. We do not observe any chevrons under these growth conditions, as has sometimes been reported.[2,5] We note here that we see a similar spot pattern along the [011] azimuth, as is commonly seen when the islands form at the growth temperature.

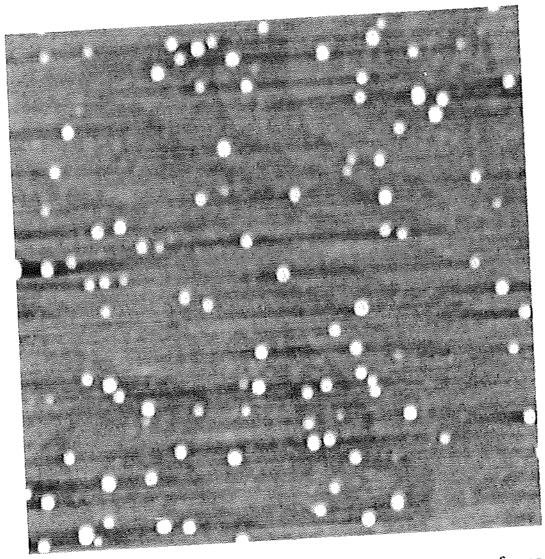

FIG. 2. 1 μm×1 μm×12 nm AFM image of the QDs that are formed by cooling the 1.9 monolayers of InAs.

The quantum dots that are formed on this sample are measured with AFM. An AFM image of these QDs is shown in Fig. 2. The areal density is about 100 μm^{-2}. The mean height is 3.37 nm with a standard deviation of 1.47 nm. This density is within the range that is often reported for InAs/GaAs quantum dot growth, but the size distribution is much larger than is typically reported. This is likely due to the low temperature at which the islands form, which reduces the adatom surface diffusion length and inhibits size equalization.

We also note that these islands are stable at temperatures that are higher than the low temperature at which the islands form. We have reheated the islands under an arsenic flux while monitoring the RHEED pattern. We continue to observe a spotty pattern until about 470 °C, at which point the diffusely scattered electron intensity is very high, and the pattern once again appears similar to what is observed in Fig. 1(a), with only the specularly reflected spot visible. When the sample is then recooled, we can once again observe the appearance of a spotty RHEED pattern.

III. DISCUSSION

This change in the RHEED pattern during cooling has not previously been reported for InAs epilayers grown on GaAs using MBE. This phenomenon has previously been observed for MBE growth of InAs on Si.[6] Hansen *et al.* deposited 1.5 monolayers of InAs on Si at 370 °C and then cooled the sample. Below 320 °C, they discovered that islands formed, and the size distribution of the islands was unusual. In the case of InAs/Si, the critical thickness required for the 2D–3D transition can vary over a wide thickness range, from about 1.5 to 5.0 monolayers, as the substrate temperature increases from 250 to 400 °C.[7] Above about 420 °C, no 3D growth mode was seen for InAs/Si. For InAs/GaAs, the thickness required for QD formation has a weaker temperature dependence, varying from 1.5 to 2.4 monolayers as the substrate temperature varies from 300 to 540 °C.[8]

The high temperature (540 °C) at which the InAs is deposited is higher than typically reported for InAs MBE growth on GaAs.[4] At temperatures above about 500 °C, theory predicts that the InAs will form a liquid-like layer on the surface of the GaAs.[9] This is consistent with the RHEED pattern shown in Fig. 1(a), where only the specularly reflected spot is observed. Such a pattern would be expected when there is little or no crystalline order in the 2 monolayers that are closest to the surface. As the sample is cooled, the InAs first forms a solid, highly strained, crystalline film in which the surface atoms have high mobilities. High mobilities allow In atoms to rearrange on the surface, thus providing a kinetic mechanism for the formation of the three-dimensional islands.

Our experiments further indicate that the critical thickness for 2D–3D transition of InAs/GaAs films is not just dependent on the substrate temperature. We have grown InAs films on GaAs at various temperatures, and we find that at a temperature of 470 °C, a 3D RHEED pattern is observed after deposition of about 1.6 monolayers of InAs. Thus we might expect that once a sample that has greater than 1.6 monolayers of InAs deposited at some higher temperature is cooled to below 470 °C, the InAs film would undergo the 2D–3D transition. This is not what we observe. As described earlier, we must cool 1.9 monolayers of InAs, deposited at 540 °C, to around 330 °C before we observe a 3D RHEED pattern. This demonstrates that the critical layer thickness for InAs quantum dot formation depends not only on the substrate temperature, but also on the substrate temperature during deposition of the InAs.

One possible explanation for this behavior is that there is some desorption of In adatoms at a substrate temperature of 540 °C, which would give an epilayer of InAs that is less than 1.9 monolayers. Previous results[8] have indicated that the required thickness for the 2D–3D transition at growth temperature is constant below about 450 °C and also that the transition is reversible. Based on these results, it is expected that an InAs epilayer that is deposited at high temperature would either be thick enough to undergo the 2D–3D transition at a temperature close to 450 °C or else would always remain below the required thickness. We observe the 2D–3D transition at around 330 °C, and the 3D–2D transition at around 470 °C. This leads us to conclude that although it is likely that some In is desorbing, that alone cannot account for the fact that the 2D–3D transition occurs at a much lower temperature than would be expected if the InAs epilayer was simply thinner due to In desorption.

Another factor that may be influencing the quantum dot formation is the reduction in the overall strain of the surface film due to increased InAs melting and intermixing with the near-surface GaAs. The intermixing of the GaAs with the InAs has been established experimentally and is consistent with a QD volume that is larger than can be attributed to the InAs alone.[10] As temperature increases, more intermixing is expected to occur, which would increase the Ga mole fraction in the InGaAs surface layer. This would lead to a reduction in the lattice mismatch and thus a reduction in the strain. Therefore, when the film begins cooling, it will contain a surface layer that has a reduced lattice mismatch compared to a film that is nominally the same composition and thickness but is deposited at a lower substrate temperature. The hysteresis in the island nucleation/denucleation that we observe during the cooling and reheating of the sample indicates some activation barrier that must be overcome for this transition to occur. This could occur if, during cooling, GaAs solidifies first, leaving the liquid layer more InAs-like. The strain would then be increasing as the sample cooled and would eventually be large enough to drive the transition to 3D islands. During reheating, the strain has been reduced by the formation of the 3D islands, and only when the temperature is sufficiently high does the 2D InAs layer again become energetically favorable, allowing remelting of the InAs and reintermixing with the GaAs. This then allows the cycle to be repeated, as we observed.

IV. SUMMARY

We report on the evolution of the RHEED pattern during the formation of InAs/GaAs QDs. We show that an initial two-dimensional epilayer of InAs evolves as the sample cools down to form three-dimensional islands. This "dewetting" phenomenon shows that the InAs films become unstable during cooling. We also show that the critical thickness for the 2D–3D transition of an InAs/GaAs film is not a simple function of substrate temperature, but instead also depends on other parameters such as deposition temperature. RHEED patterns measured during reheating and recooling of the samples demonstrate the reversibility and hysteresis of the 2D–3D transition. These results show that InAs epilayers can evolve during quenching from growth temperature and may have implications on the accuracy of measurements of the shapes and sizes of quantum dots formed at growth temperature.

ACKNOWLEDGMENT

R.P.M. would like to thank Lars Hansen for helpful discussions.

Presented at the 20th North American Conference on Molecular Beam Epitaxy, 1–3 October 2001, Providence, RI.

[1]L. Goldstein, F. Glas, J. Marzin, M. Charasse, and G. LeRoux, Appl. Phys. Lett. **47**, 1099 (1985).
[2]M. Tabuchi, S. Noda, and A. Sasaki, in *Science and Technology of Mesoscopic Structures*, edited by S. Namba, C. Hamaguchi, and T. Ando (Springer, Tokyo, 1992).
[3]D. Leonard, M. Krishnamurthy, C. R. Reaves, S. P. DenBaars, and P. M. Petroff, Appl. Phys. Lett. **63**, 3203 (1993).
[4]J. M. Moison, F. Houzay, F. Barthe, L. Leprince, E. Andre, and O. Vatel, Appl. Phys. Lett. **64**, 196 (1994).

[5]H. Lee, R. Lowe-Webb, W. Yang, and P. Sercel, Appl. Phys. Lett. **72**, 812 (1998).
[6]L. Hansen, A. Ankudinov, F. Bensing, J. Wagner, G. Ade, P. Hinze, V. Wagner, J. Geurts, and A. Waag, Mater. Res. Soc. Symp. Proc. **583**, 33 (2000).
[7]A. Yu. Egorov, A. R. Kovsh, V. M. Ustinov, A. E. Zhukov, M. V. Maksimov, G. E. Cirlin, N. N. Ledentsov, D. Bimberg, P. Werner, and Zh. I. Alferov, J. Cryst. Growth **201/202**, 1202 (1999).
[8]H. Lee, R. Lowe-Webb, W. Yang, and P. C. Sercel, Appl. Phys. Lett. **71**, 2325 (1997).
[9]D. J. Bottomley, Appl. Phys. Lett. **72**, 783 (1998).
[10]B. A. Joyce, T. S. Jones, and J. G. Belk, J. Vac. Sci. Technol. B **16**, 2373 (1998).