TRANSMISSION ELECTRON MICROSCOPY STUDY OF InGaAs/GaAs STRUCTURAL EVOLUTION NEAR THE STRANSKI-KRASTANOW TRANSFORMATION

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ABSTRACT

An experimental study of the microstructure during formation and evolution of MOCVD-grown $In_{0.6}Ga_{0.4}As/GaAs$ quantum dots (QDs) was undertaken to provide a more thorough understanding of the underlying growth principles. Transmission Electron Microscopy (TEM) was used to examine the evolution of the $In_{0.6}Ga_{0.4}As/GaAs$ system in order to correlate photoluminescence (PL) spectra with structural data. In particular, we have examined the QD size evolution, capped and uncapped, and its possible contribution to the slight QD PL blueshift observed before QD saturation. TEM studies in the QD coalescence regime clarify the microstructural origins of the sharp decrease in QD PL due to large, incoherent islands observed in AFM and TEM images.

INTRODUCTION

Recent experiments[1] in the study of morphological and photoluminescence evolution in $In_xGa_{1-x}As/GaAs$ quantum dots have shown various features in their optical properties that can be related to microstructural development during the Stranski-Krastanow (SK) growth process. For example, during deposition of the wetting layer, the luminescence peak continuously shifts to lower energies as the wetting layer thickens. At the onset of dot formation, the sharp wetting layer peak no longer redshifts, but remains at a specific energy value. This is strong evidence that the thickness of the wetting layer remains constant once islands begin to form and that all subsequent deposition goes into quantum dot formation and growth. With further deposition the quantum dot emission blueshifts slightly as the island concentration increases to saturation. Force microscopy images of uncapped InGaAs/GaAs quantum dots show a decrease in island size as island density increases. However, it has been suggested that the size of buried dots does not change significantly with concentration[2] and that island size evolution is not a satisfactory explanation of the quantum dot blueshift. Once the islands begin to coalesce, there is a sudden decrease in the QD luminescence intensity, which is accompanied by an increase in the number of large, possibly incoherent, islands observed in force microscopy images.

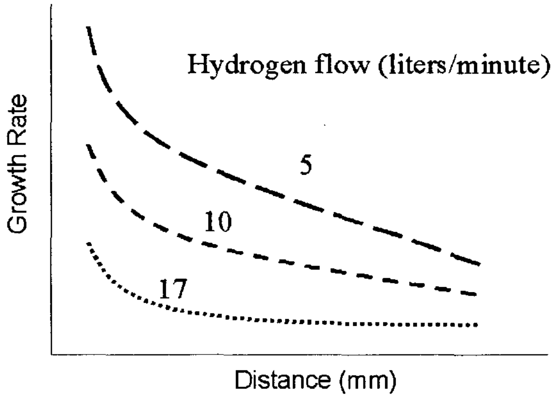

Figure 1. Growth rate profiles for different H_2 flow values as a function of distance from the MOCVD susceptor edge. As the H_2 flow is decreased, the growth becomes more graded.

Motivated by these recent findings, we carried out a more detailed TEM study of capped and uncapped quantum dot morphology. We observed that, despite the trend of decreasing island size with in increasing concentration in surface dots, there is no significant size evolution in buried InGaAs/GaAs islands. In addition, we confirmed that the large islands present at quantum dot coalescence are incoherent, as shown by a high density of defects in the TEM images.

EXPERIMENT

In_xGa_{1-x} As /GaAs structures were grown by metal-organic chemical vapor deposition (MOCVD)[1]. A horizontal reactor operating at 76 torr and $(CH_3)_3Ga$, $(CH_3)_3In$, and AsH_3 were used as precursors ($(CH_3)_3Ga$ - 5.36 x10^{-6}, $(CH_3)_3In$ - 5.18 x10^{-6}, AsH_3 - 2.5 x 10^{-4}). The H_2 carrier flow rate used was 5 standard liters/min (slm), which gave spatially graded deposition (flows of 17.5 slm and greater give large areas of uniform growth, while lower flow rates result in graded growth, see Figure l). The flow of $(CH_3)_3In$ was monitored and controlled by an EPISON ultrasonic sensor. After growth of GaAs buffer layers at 650°C, the temperature was lowered to 550°C and nanometer sized $In_xGa_{1-x}As$ islands were grown by depositing ~5ML of $In_{0.6}Ga_{0.4}As$. These nominal compositions were determined from PL measurements in thick relaxed films and PL emission from thin In_xGa_{1-x} As /GaAs quantum wells (QWs). Growth rates ranged from 0.5 to 0.75 ML/s. GaAs capping layer thickness was 30 nm (not graded). Substrates were (100) semi-insulating GaAs.

Force microscopy (FM) with standard etched silicon nitride tips was used to obtain statistical information on island surface densities. Similar experiments and prior work comparing FM and transmission electron microscope images indicate that concentrations in capped and uncapped samples are equivalent.

With Transmission Electron Microscopy (TEM) images we obtained structural information on both uncapped quantum dots and buried islands. The plan-view specimens were first mechanically thinned to ~100µm and locally thinned to ~50µm with a dimpler/grinder. The final thinning was accomplished with a bromine/methanol etch. A Philips 430 (300keV) and CM12 (120keV) TEM was used to obtain on-zone bright field images of both capped and uncapped dots.

RESULTS

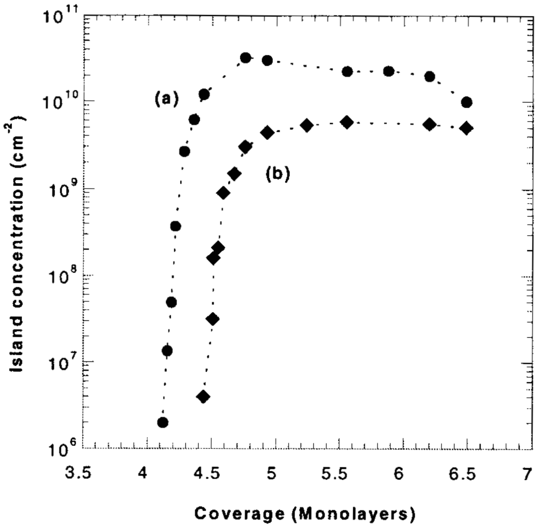

Figure 2. Evolution in island concentration[3] over depositions between 4.0 to 6.5 ML. Island growth in (a) and (b) differ only in AsH_3 pp, with lower pp in (a) than (b).

Figure 2 shows the island concentration as a function of coverage in units of monolayers (MLs) for two values of AsH_3 partial pressure (low in a and higher in b)[3]. Most of the experimental results presented here were obtained from samples grown using the parameters of curve (a). The coverage has been determined from calculated curves for different hydrogen carrier flows using the dimensions and conditions in our cell[4]. These describe growth rates in laminar flow systems on the basis of concentration profiles under diffusion controlled conditions. The deposition scale and gradients have been calibrated in this work by measurements of the PL emission and the corresponding shifts as a function of distance from graded In_xGa_{1-x} As /GaAs capped quantum wells. The latter structures were grown under the same conditions as the graded QD samples but a shorter deposition time was used so the SK transformation did not occur in the wafer strip. This allowed establishing a growth rate in MLs as a function of distance from the edge of the MOCVD susceptor. From Figure 2a, we determined the critical thickness for the 2D to 3D transition in the MOCVD growth of $In_{0.6}Ga_{0.4}As$/GaAs to occur after 4.0 ML deposition. The onset of island coalescence becomes apparent with the decrease in island concentration with further deposition.

Figure 3 shows a series of atomic force microscopy images of $In_{0.6}Ga_{0.4}As$/GaAs quantum dot morphology from island formation to near the saturation regime (4.20 ML - 4.50 ML)[1]. A representative dot (scaled by 200%) from each 250 nm × 250 nm image is shown above the corresponding frame. From the visual aid of the parallel lines, we see that the average island diameter of these uncapped quantum dots is decreasing as the concentration approaches saturation.

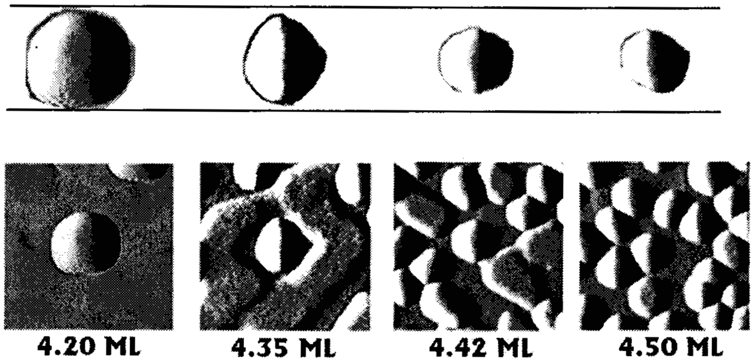

Figure 3. Deflection force microscopy images of InGaAs/GaAs islands showing island size evolution from low island concentration to saturation. Above each 250 nm × 250 nm frame is a magnified (200%) image of a typical island.

Plan-view TEM images of uncapped islands on the same graded growth sample are shown in Figure 4a - b. Recent cross-section and plan-view TEM studies[5,6] suggest that $In_{0.6}Ga_{0.4}As/GaAs$ islands are lens-shaped and have a circular base, which can be measured from bright field on-zone images of plan-view specimens. Comparing a low (4a) and high (4b) concentration of uncapped islands, we see a significant increase in average island diameter for the lower concentration, which is in agreement with the trend observed in the force microscopy images. For buried islands, the low (4c) and high (4d) concentration islands do not show a similar change in average island size.

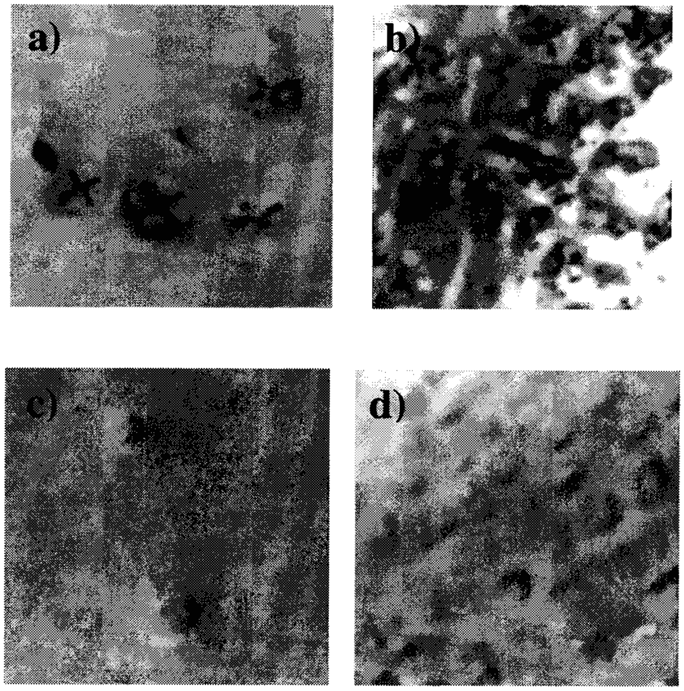

Figure 4. Plan-view on-zone bright field Transmission Electron Microscopy images of uncapped (a - b) and capped (c – d) $In_{0.6}Ga_{0.4}As/GaAs$ islands. Each image is approximately 280 nm × 280 nm. The details of the strain contrast in the surface dots is discussed elsewhere[6].

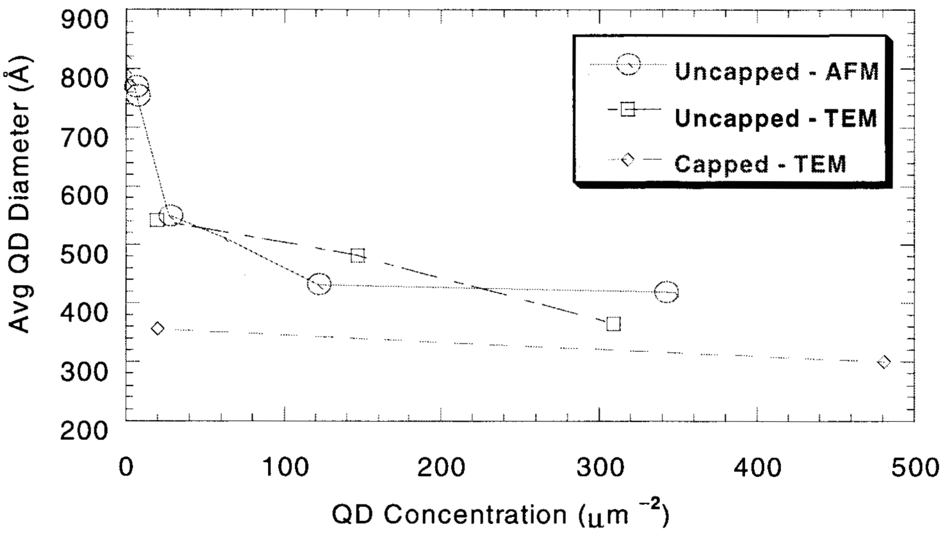

Figure 5. Size evolution of $In_{0.6}GaAs_{0.4}$/GaAs quantum dots. The uncapped islands (circles and squares) clearly show a trend of decreasing size with increasing quantum dot concentration. The capped islands (diamonds) do not exhibit this size evolution.

A plot of average island diameter as a function of concentration is shown in Figure 5. For the uncapped quantum dots, there is a clear trend of decreasing island size with increasing island concentration, while the capped islands do not show such a significant change in island diameter. Because photoluminescence experiments are performed on GaAs-capped quantum dots, island size evolution is not sufficient to explain the blueshift of the buried quantum dot peak from island formation to saturation. A recent study shows that the strain field of an isolated InGaAs/GaAs quantum dot is modified by the presence of other dots in close proximity[2], which leads to an increase in the luminescence peak position. This picture of the QD blueshift does not depend on large changes in the average island size and is therefore consistent with our microstructural observations.

Plan view TEM images of large dislocated islands are shown in Figure 6. These samples were grown using a higher arsine partial pressure (pp) than in the above experimental results. As shown in Figure 2b, $In_{0.6}Ga_{0.4}As$/GaAs quantum dots grown under these conditions form at a higher critical wetting layer thickness and saturate at a much lower island density value. There is also evidence that during high arsine pp growth, the large, incoherent islands ripen at the expense of the smaller, coherent quantum dots[3]. The resulting lower concentration and larger incoherent island size makes the higher arsine pp samples ideal for a structural study. As shown in Figure 6, the islands exhibit a large defect density. This is in agreement with the observation of the sharp drop in photoluminescence intensity, giving structural confirmation that these large islands are optically inactive.

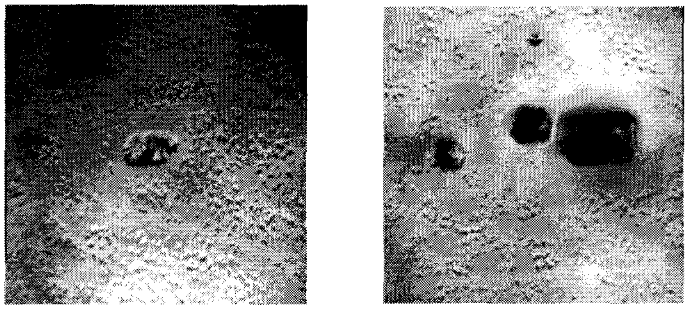

Figure 6. Plan-view bright field on-zone TEM images of large uncapped islands. Images are approximately 1µm × 1 µm.

CONCLUSIONS

In this structural study of the microstructural evolution of MOCVD-grown $In_{0.6}Ga_{0.4}As/GaAs$ quantum dots we report a discrepancy in the size evolution of surface dots and buried dots from low concentration to saturation. We have also shown that the large islands, which begin to dominate the surface morphology after saturation, are incoherent and contain dislocations. The sudden drop in quantum dot photoluminescence intensity after island saturation indicates that these large incoherent islands are optically inactive.

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