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Effect Of Growth Rate And Coverage On The Composition And Optical Properties Of InAs/GaAs Self-Assembled Quantum Dots

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ABSTRACT

The effect of the InAs deposition rate and coverage on the properties of InAs/GaAs quantum dots (QDs) grown on GaAs(001) substrates by molecular beam epitaxy has been studied by scanning tunnelling microscopy (STM) and photoluminescence (PL). PL spectra recorded from GaAs capped QD structures show that by changing the InAs growth rate it is possible to tune the emission properties of the QDs with low growth rates resulting in room temperature emission at 1.3 µm. STM measurements on uncapped dots reveal that the dots grown at low InAs deposition rates have a higher In content which is responsible for the shifts observed in the PL spectra. The amount of InAs deposited also provides additional control of the emission wavelength and we determine the optimum conditions for growing QD structures for applications at 1.3 µm.

INTRODUCTION

The growth of self-assembled InAs/GaAs quantum dots has been studied extensively in recent years primarily because of their potential for temperature independent ultra-low threshold lasers [1]. One of the most exciting developments is the extension of the emission wavelength, which offers the possibility of producing GaAs based devices that operate at 1.3 µm [2,3]. Although several methods have been used to control the wavelength, perhaps the most straightforward from a growth perspective involves using very low InAs deposition rates [4].

In this paper, we present a quantitative study of the effects of the InAs growth rate on the optical properties and composition of InAs/GaAs QDs grown on GaAs(001) substrates by molecular beam epitaxy (MBE). STM and PL measurements carried out on uncapped and GaAs capped QDs show that the growth rate has a strong influence on their size, composition and optical properties. Reducing the growth rate results in a decrease in the QD number density and total volume, an increase in the room temperature emission wavelength to 1.3 µm, and a systematic reduction in the emission linewidth. The volumes obtained from the STM measurements on uncapped samples indicate that the composition of the dots changes with growth rate, with the indium fraction being greatest at the lowest growth rate.

In order to maximise the emission properties of the QD structures, we also investigate the effect of InAs coverage at a fixed growth rate and show that an increase in QD size leads to a redshift in PL emission wavelength. This allows us to maximise the number density whilst still achieving 1.3 µm emission.

EXPERIMENTAL

All samples were grown in a combined MBE-STM growth system (DCA Instruments/Omicron Gmbh) that is also equipped with reflection high energy diffraction (RHEED) for in-situ monitoring of growth. The In, Ga and As cells where calibrated using RHEED oscillations on InAs(001) and GaAs(001) respectively. Epi-ready GaAs(001) substrates (n^+ Si-doped) were mounted on molybdenum plates and transferred directly into the growth chamber via a fast entry lock. After initial thermal cleaning at 300 ^{0}C the native oxide layer was removed under an As_2 flux at 620 ^{0}C. A 0.6 μm thick GaAs buffer layer was grown at 580^{0}C and the substrate temperature reduced to 510^{0}C for the deposition of 300 Å of GaAs. InAs was deposited at a range of growth rates between 0.13 and 0.0065 MLs^{-1}. A second series of samples were grown at a fixed rate of 0.016 MLs^{-1} but different InAs coverages. RHEED was used in both cases to monitor the 2D → 3D growth mode transition.

After the growth of the QDs some samples were transferred immediately into the STM chamber. The process of quenching the sample from the growth chamber is complete within a few seconds and limits additional surface effects caused by annealing. Constant current STM images were obtained with a sample bias of –3.5 V and tunnelling currents of 0.1 – 0.3 nA. A further set of samples were grown under the same conditions as those above, but they were immediately capped with GaAs. The capping process involved depositing 400 Å at the QD growth temperature of 510^{0}C followed by a further 1000 Å GaAs deposition at 580 ^{0}C. PL spectra were recorded at 10 K and 300 K. An Ar^+ laser was used for excitation and the luminescence dispersed with a SPEX 1404 monochromator and detected with a cooled Ge photodiode using standard lock-in techniques.

RESULTS AND DISCUSSION

Low temperature PL spectra recorded at a range of InAs growth rates are shown in figure 1. The emission characteristics depend strongly on the growth rate at which the QDs are grown. As the growth rate is lowered the emission wavelength shifts from 1.02 μm to 1.2 μm and there is a narrowing in the emission linewidth from 44 meV to 27 meV.

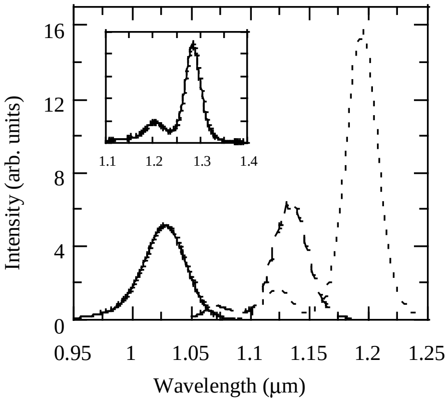

Figure 1. *PL emission spectra of GaAs capped InAs/GaAs QD samples recorded at 10 K. QD were grown at (a) 0.13 (b) 0.016 and (c) 0.0065 MLs^{-1}. The inset shows the emission from sample (c)recorded at 300K. In all cases 2.7 ML InAs was deposited at 510 ^{o}C.*

The inset in figure 1 shows the room temperature emission spectra for the QD sample grown at 0.0065 MLs^{-1}. Emission occurs at 1.3 μm, there is no increase in linewidth and the emission intensity is reduced by only a factor of eight. Although the observed redshift in wavelength with varying growth rate does correlate with the known increase in QD size [5], a simple finite square well calculation shows that this alone would not result in the measured shift and an additional explanation is required.

The two STM images in Figure 2 were taken after the deposition of 2.2 ML of InAs on GaAs(001) at 510 ^{0}C at growth rates of (a) 0.094 ML s^{-1} and (b) 0.016 ML s^{-1}. As the growth rate is lowered there is a reduction in island density and an increase in the average size of the QDs which is also accompanied by a narrowing of the size distribution . Typical statistics for the high growth rate QDs (0.094 ML s^{-1}) are; island density (N_s) ~ 1.0×10^{11} cm^{-2}, island height (h) ~ 20 Å and mean diameter (d) ~ 150 Å. Values for the low growth rate QDs (0.016 ML s^{-1}) are N_s ~ 2.0×10^{10} cm^{-2}, h ~ 35 Å and d ~ 200 Å. The respective height fluctuations $\Delta h/h$, are 22 % at 0.094 ML s^{-1} and 16 % at 0.016 ML s^{-1}. An important factor to note is that both RHEED and STM show that the growth rate has no effect on the critical thickness at which QD formation occurs , which for this growth temperature is 1.8 ML. It should also be noted that the growth temperature does have a strong influence on the critical thickness [6]. STM measurements tracing the evolution of the 3D islands show that nucleation is essentially complete within 0.3 ML of the growth mode transition. The detailed atomic structure of the underlying wetting layer appears to have no dependence on growth rate and in all cases exhibits the characteristic rather disordered (1x3) reconstruction that we have reported in detail elsewhere [7,8] .

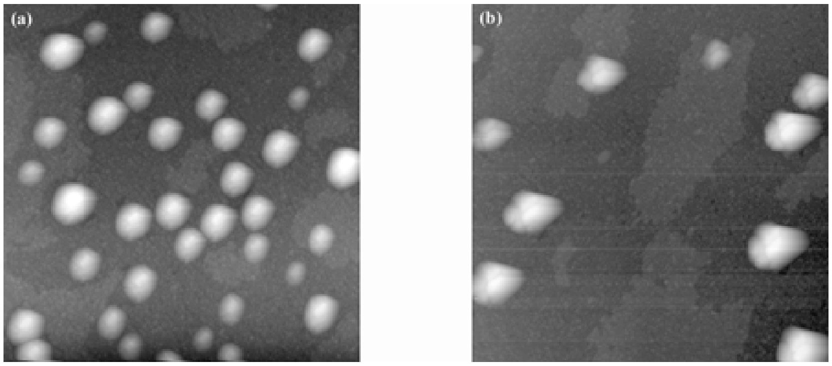

Figure 2. *STM images (0.2 μm x 0.2 μm) of 2.2 ML InAs deposited on GaAs (001) at a growth rate of (a) 0.094 (b) 0.016* MLs^{-1}.

In a previous study [6] we have shown that a considerable amount of "alloying" occurs during the formation of QDs grown at conventional growth rates of 0.13 MLs^{-1}, with the degree of Ga and In intermixing strongly dependent on the substrate temperature. The Ga content of the QDs plays an important role in determining the emission wavelength with a high Ga content tending to a lower emission wavelength. In an attempt to quantify the composition of the QDs we have used STM to measure the total amount of material (ML equivalents) that is present in the QDs. Comparison to the volume of material deposited after the growth mode transition (1.8 ML) provides a good estimate of the degree of intermixing and wetting layer erosion. Figure 3 (a) shows the total amount of material present in the QDs, as measured by STM. The solid black line represents the volume of material in the QDs that is expected if growth occurs by a classical

Stranski-Krastanov (S-K) mechanism with no alloying and a critical thickness of 1.8 ML. It is clear that there is a gradual decrease in the total QD volume as the growth rate is reduced. QDs grown at 0.13 MLs^{-1} have incorporated an additional 0.8 ML of material, whereas QDs grown with InAs growth rates below 0.02 MLs^{-1} follow the S-K growth mode.

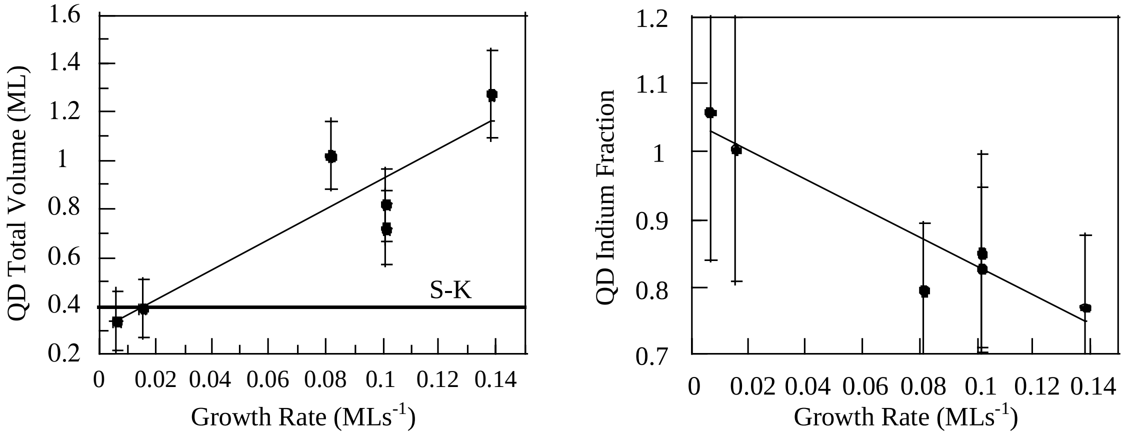

Figure 3. *(a) The average QD volume as a function of InAs growth rate volume (measured by direct integration of STM images in ML equivalents). The solid horizontal line is the volume expected assuming classic Stranski-Krastanov growth and no alloying.*

(b) The In fraction within the QDs as a function of growth rate. Each data point is calculated from the total QD volume measurements assuming that the wetting layer undergoes some erosion, but has a composition that is fixed at $In_{0.6}Ga_{0.4}As$.

The total volume measurements allow us to estimate the In composition of the uncapped QDs. We have assumed that the wetting layer undergoes some erosion but its composition is fixed at $\mathrm{In_{0.6}Ga_{0.4}As}$, as indicated by previous measurements [7,8]. We therefore estimate that QDs grown at 0.13MLs^{-1} contain ~ 20 % gallium. It is the increased Ga content that accounts for the emission wavelength that is observed in PL experiments.

We now suggest a possible mechanism for the growth rate-dependent changes in WL erosion and QD alloying. Firstly, our RHEED and STM experiments indicate that the critical thickness and WL structure are both independent of growth rate. However, lower growth rates clearly favour a smaller QD number density. A recent theoretical model for QD formation has explicitly included strain-enhanced adatom detachment from QDs [9]. The strain fields causing this effect arise from the QDs themselves, although their detailed form is not known. We believe that these strain fields may also lead to increased adatom detachment from the WL. With a high number density of QDs, there are more overlapping strain fields, favouring more detachment of atoms (both In and Ga) from the WL. These can attach to the QDs leading to Ga incorporation and a total QD volume greater than that predicted from a pure S-K mode. Hence, the change in QD alloying at fixed temperature is a consequence of the change of QD number density. This mechanism is distinct from the temperature dependent alloying of QDs at fixed growth rate [6]. The importance of the initial nucleation is also demonstrated by PL experiments investigating in-situ thermal annealing on QDs grown at high growth rates which are annealed at the growth temperature for the equivalent time as it takes to grow the low growth rate samples . The results show that the annealed samples show a further blue shift in emission which is thought to be due to additional incorporation of Ga.

The STM images in figure 2 show that due to the reduced island density at low growth rates the spacing between neighbouring QDs is much greater than at higher growth rates thus allowing more material to be deposited before coalescence is observed. Figure 4 (a) shows the effect of InAs coverage on the average volume of the QDs grown at a fixed deposition rate of 0.016 MLs^{-1}. The increase in QD size is approximately linear and calculation of the total QD volume shows no deviation from the S-K growth mode and hence no change in the composition of the uncapped QDs. Figure 4 (b) shows the emission wavelength as a function of the InAs coverage at 10 K. There is a distinct redshift of ~60 nm in the emission wavelength which we attribute to the increase in size of the QDs.

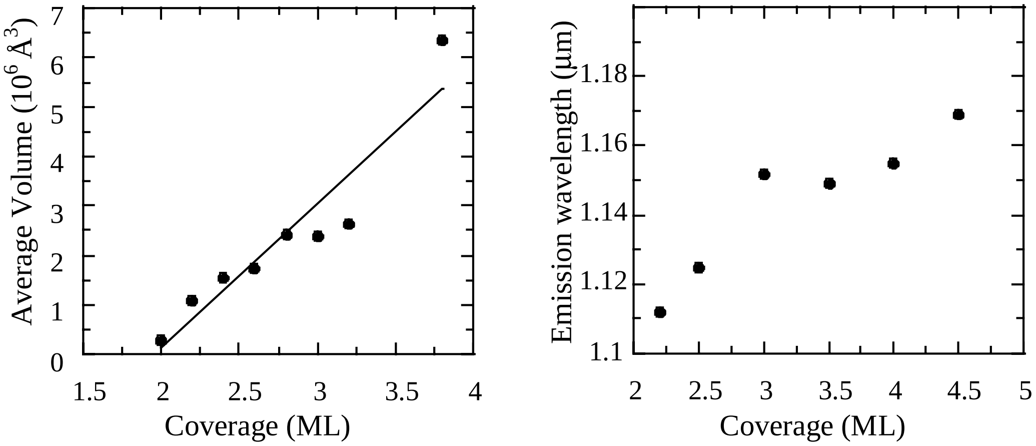

Figure 4. *(a) The average QD volume estimated by STM as a function of coverage at a fixed growth rate of 0.016 MLs^{-1}. The solid line is a linear fit.*

(b) Low temperature PL emission wavelength from capped InAs QD samples grown at 0.016 MLs^{-1}.

A consequence of lowering the growth rate in order to achieve 1.2 µm emission at 10 K is that there is a reduction in the QD number density. By depositing high InAs coverages at low growth rates it is possible to maximise the number density whilst still achieving the necessary emission wavelength. Figure 5 shows the emission wavelength obtained by varying the deposition rate at a fixed high InAs coverage (3.6 ML). The results show that deposition of 3.6 ML InAs at a rate of 0.01 MLs^{-1} is sufficient to produce QD structures which show room temperature emission at 1.3 µm. The QD number density at this growth rate is ~ 9 x 10^9 cm^{-2}, which is three times larger than the values obtained from STM analysis of QDs grown at 0.006 MLs^{-1}at the same temperature.

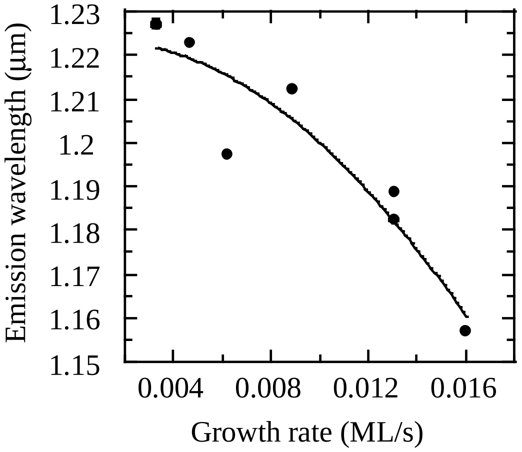

Figure 5. *The emission wavelength as a function of growth rate for a fixed InAs coverage of 3.6 ML. A growth rate of around 0.01 MLs^{-1} is required to give a low temperature emission wavelength of 1.2 μm, corresponding to 1.3 μm at room temperature.*

CONCLUSIONS

We have demonstrated a strong dependence on the InAs deposition rate on the size, composition and optical properties of InAs/GaAs QDs grown by MBE. QDs grown at the lowest rates are larger, have a narrow size distribution and a higher In content. Samples grown at the lowest growth rates exhibit strong room temperature emission at 1.3 μm with linewidths of 27 meV. The results demonstrate that the kinetics of QD formation and interplay with the WL have crucial roles in determining the QD properties. Reduction of the QD growth rate provides a straightforward means to tailor the optical properties. Increasing the InAs coverage also provides a further method to tune the optical properties and allows maximisation of the QD number density which is beneficial for device applications.

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