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Strain-interactions between InAs/GaAs quantum dot layers

P. Howe[a], B. Abbey[a], E.C. Le Ru[b], R. Murray[b], T.S. Jones[a,*]

[a] Ultrafast Photonics Consortium, Centre for Electronic Materials and Devices, Department of Chemistry, Imperial College, London, SW7 2AZ, UK
[b] Ultrafast Photonics Consortium, Centre for Electronic Materials and Devices, Department of Physics, Imperial College, London, SW7 2AZ, UK

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Abstract

InAs/GaAs quantum dot (QD) bilayer and trilayer structures have been grown on GaAs(001) substrates by molecular beam epitaxy and the properties of the uncapped QDs investigated using atomic force microscopy (AFM). The emphasis is on understanding the influence a variation of the thickness of the GaAs spacer layer (*S*) between the QD layers has on the morphological properties of the QDs in the up-most layer, which is grown at a significantly reduced temperature compared to the first QD layer. The size distribution of the QDs in the second layer is shown to be exceptionally narrow for a spacer thickness of 10 nm, with a large average QD size. Larger values of *S* lead to a much broader size distribution and the appearance of significantly smaller dots. Complete strain relief is only achieved upon deposition of ∼ 50 nm GaAs in bilayer and ∼ 60 nm in trilayer structures, a result that has important implications for multiple stacking of QD layers in device applications.

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1. Introduction

Considerable effort is being devoted to the growth of InAs/GaAs quantum dots (QDs), in particular for applications at 1.3 μm and longer wavelengths. Emission at long wavelengths is difficult to achieve when using a single layer of InAs/GaAs QDs; however, we have recently shown that it is possible to fabricate bilayer QD structures that demonstrate room temperature emission at 1.4 μm using a GaAs capping layer, and 1.5 μm by including InGaAs within the capping layer [1]. A remarkably narrow (low temperature) line width of ∼ 14 meV was also reported suggesting a very narrow distribution of QD sizes.

It is well documented that vertical self-alignment of InAs/GaAs QDs occurs in multilayer structures if the thickness of the spacer layer between the dot layers (*S*) is ≤20 nm [2–5]. This stacking behaviour has been attributed to strain-interactions [4,6]. The effects of strain on the growth of multilayer QDs have also been studied [7–9]. In a previous study, the pairing probability for stacked QDs was determined using cross-sectional transmission electron microscopy (TEM) [4]. The disadvantage of cross-sectional techniques such as TEM is that, in order to obtain statistically relevant data, many images have to be recorded, since each image typically comprises only one or two stacks of dots. We have therefore employed a different method of probing the strength and the decay of the strain fields associated with the first layer of dots by growing the second layer QDs at a reduced temperature (475 °C). Without strain-interactions the low substrate temperature would lead to a significantly larger number density of QDs (N_D) and also to a reduction in the QD dimensions [10]. For small *S* however, the QD number density is fixed to approximately the same value as the first layer, which is grown at a relatively high temperature (510 °C). In this paper, we address the important issue of spacer layer thickness and investigate the size, density and uniformity of the grown (uncapped) QDs using atomic force microscopy (AFM). Measurements have been carried out on various QD bilayer and trilayer samples, grown with a range of GaAs spacer layer thicknesses (3–60 nm). The results of the trilayer structures have important implications for the growth of device structures involving multiple stacks of bilayer QDs.

* Corresponding author. Tel.: +44-20-7594-5794; fax: +44-20-7594-5801.
E-mail address: t.jones@imperial.ac.uk (T.S. Jones).

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2. Experimental details

The samples were grown in a molecular beam epitaxy (MBE) system equipped with reflection high-energy electron diffraction (RHEED) for in situ monitoring of the growth process, determination of the critical thickness for QD formation (θ_{crit}) and calibration of the incident Ga, In and As_2 fluxes. Epi-ready GaAs(001) substrates (n^+ Si-doped) were mounted on molybdenum plates and the samples introduced into the MBE chamber via a fast entry lock. After initial thermal degassing at 300 °C under an As_2 flux (As pressure = 2.6×10^{-6} mbar), the oxide layer was removed at 620 °C and a 20 nm GaAs buffer layer was then grown at 580 °C with a Ga flux of 0.5 monolayer per second (ML s^{-1}). The substrate temperature was then reduced for the growth of the InAs/GaAs QD structures.

For the bilayer structures, the two layers of QDs were separated by a GaAs spacer layer of varying thickness S_1. The InAs deposition rate in all cases was 0.016 ML s^{-1} and 2.5 ML of InAs were deposited in the first layer at 510 °C. The GaAs spacer layer was also grown at 510 °C before being annealed under an As_2 flux at 580 °C. The temperature was then reduced to 475 °C before deposition of 2.5 ML InAs (the total amount of InAs in the second QD layer was increased to 3.2 ML for the trilayer structures in order to simulate the characteristics of a structure with emission at long wavelengths) and formation of the second layer of QDs, which was again monitored by RHEED. The second layer critical coverage, $\theta_{crit,2}$, is generally (depending on the value of S) lower compared to that of the first layer [7]. For the trilayer structures, the first 15 nm of the second spacer layer (with a total thickness S_2) were deposited without growth interruption at 475 °C, with the remaining part grown at 580 °C. After annealing, 2.5 ML of InAs were deposited at 475 °C to form the third QD layer. The uncapped QD samples were removed quickly from the growth chamber for ex situ AFM imaging. This quenching is necessary to avoid any post-growth annealing processes, which are known to alter the morphological QD characteristics [11].

3. Results and discussion

The key issue for the growth of the multilayer structures is the templating effect of the seeded (first) QD layer, which dictates the second layer QD number density ($N_{D,2}$) [1,2], provided S_1 is adequately chosen. Due to the strain fields produced by the dots in the first layer, a strain energy modulation on the surface of the spacer layer is created. The lattice constant of the GaAs layer is expanded directly above a buried dot [4], and this strongly influences the migration of In adatoms [12] and therefore the growth and properties of the next layer of QDs. The low substrate temperature for the growth of the second QD layer (475 °C) is a crucial feature for two reasons. Firstly, from an application point of view, the dots in the second layer exhibit emission at long wavelengths, which can be attributed to their increased size and aspect ratio, and also to the reduced degree of In/Ga intermixing during capping [1]. Secondly, it provides an indirect tool for measuring the decay of the strain fields over large spacer layers. For large S_1, the QD characteristics should be equivalent to those obtained for single layer QD samples grown at the same low temperature (475 °C). The value for S_1 at which this happens corresponds to the thickness of the GaAs spacer at which complete strain-relief is achieved.

An AFM image for an uncapped single layer of QDs grown with 2.5 ML of InAs at 475 °C is shown in Fig. 1a. The total QD density $N_{D,1}$ is 7.5×10^{10} cm^{-2}, and the average height (h) and diameter (d) are 3.4 nm and 27.2 nm, respectively. These are typical values for single layer InAs/GaAs QDs grown under these conditions [13]. The corresponding size histograms are also shown in Fig. 1.

Fig. 1b–d show AFM images and the QD size histograms for the uncapped second QD layer of the bilayer structures, in which after the spacer layer (S_1 = 10, 20 and 50 nm), 2.5 ML of InAs were deposited at 475 °C. Significant changes in QD density ($N_{D,2}$), size and distribution are clearly visible with varying S_1. In the case of the single QD layer grown at the same low temperature (a), almost all dots have a diameter of less than 35 nm, whereas in the bilayer structure with S_1 = 10 nm (b), only dots with d>35 nm exist. In the structure with S_1 = 20 nm both classes of dots coexist and the number density of the larger dots is much greater than that of the small dots. For S_1 = 30 nm (not shown) the situation is reversed, with a majority of small dots being present. Consequently, the distributions of the second layer QD sizes for intermediate values of S_1 (20–30 nm) are relatively wide. In fact, the two distributions seen in each of the height and diameter histograms are even broader than the corresponding distributions shown in Fig. 1a and b.

A narrow size distribution characterised by σ (full width at half maximum of the distribution based on a Gaussian fit divided by the average value) is obtained for S_1 = 10 nm (diameter, σ_D = 6.4%; height, σ_H = 14.6%). This broadens for S_1 = 50 nm (σ_D = 11.9%; σ_H = 24.7%), comparable to the values obtained for the single layer (σ_D = 10.9%; σ_H = 25.1%). The distributions for the two intermediate values of S_1 (20 or 30 nm) cannot be fitted with one Gaussian, due to the existence of the two classes of dots.

Fig. 2 shows how the density of the small (d < 35 nm) and large (d >35 nm) second layer QDs and also the total $N_{D,2}$ varies with S_1. For S_1 = 10 nm the minimum in the total QD number density (1.5×10^{10} cm^{-2}; the total height of the column in Fig. 2) coincides with the maximum in the number of large dots. The total $N_{D,2}$ rises with increasing S_1 before saturating at a value of $\sim7.5\times10^{10}$ cm^{-2} for $S_1\geq40$ nm. This value is comparable to $N_{D,1}$ for the single layer QD sample grown at the same low temperature (Fig. 1a). The heights and diameters of the QDs grown for S_1>40

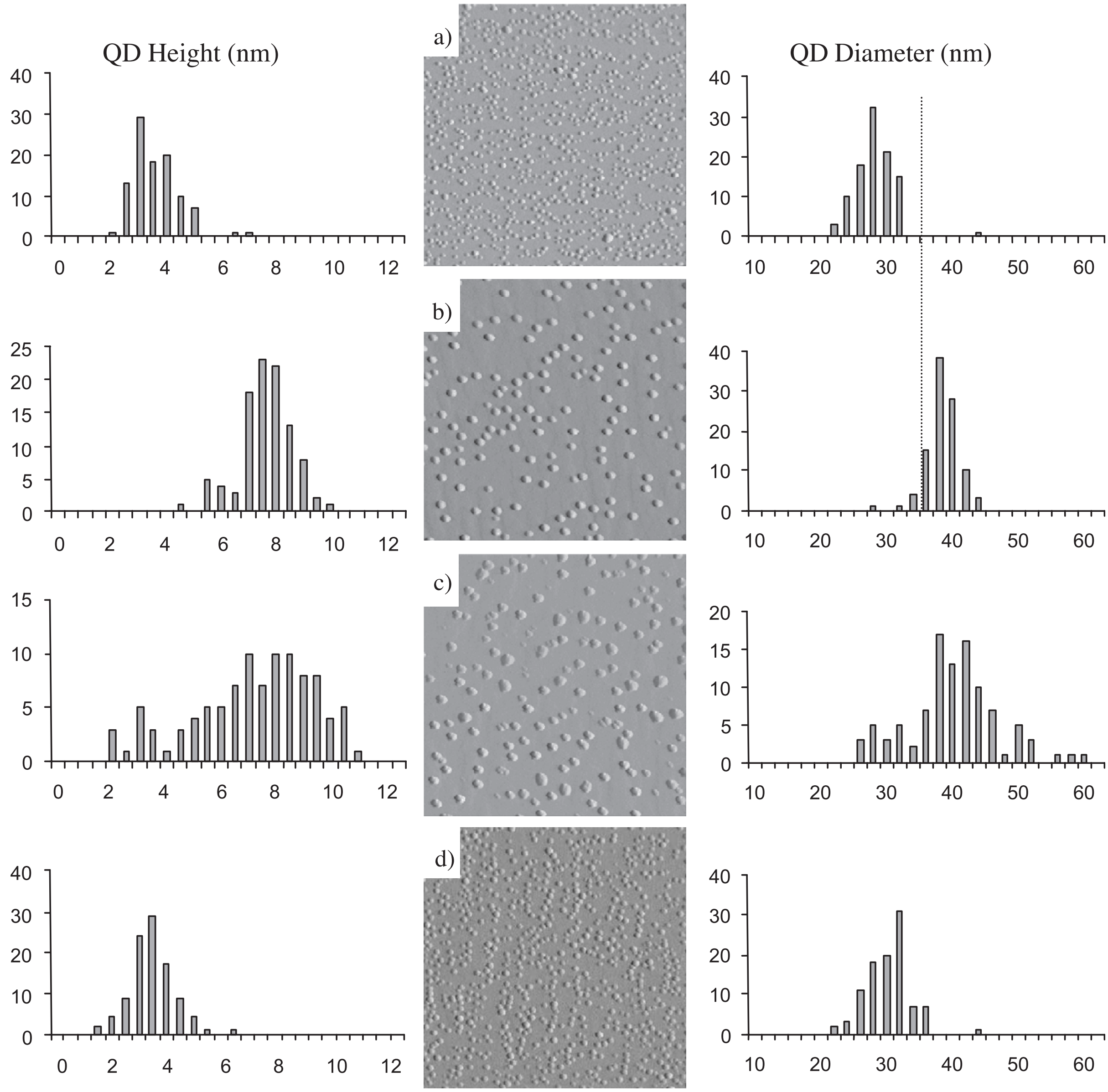

Fig. 1. Contact-mode AFM images (1×1 μm^2) of uncapped InAs/GaAs QD samples; (a) a single QD layer, (b)–(d) the second QD layer with $S_1=10$, 20 and 50 nm. Histograms of the measured QD heights and diameters are also shown. The dashed vertical line in the first two diameter histograms denotes the distinction between small and large dots (see Fig. 2). In all cases, 2.5 ML of InAs were deposited at 475 °C.

nm are also similar to those in the equivalent single layer structure. Also shown in Fig. 2 is the variation in density for the third layer of QDs ($N_{D,3}$) in a trilayer structure as a function of the thickness of the second spacer layer (S_2) in the range 40–60 nm. In this case, saturation of the total $N_{D,3}$ only occurs for $S_2>50$ nm, in which case the third layer QD dimensions are again similar to those obtained for a single QD layer grown at the same low temperature.

We propose that the large dots seen in the bilayer structure in Fig. 1b form as a direct consequence of the vertical strain fields generated by the seed layer and these are strongest for $S_1\sim10$ nm. Here, despite the lower substrate temperature used for second layer QD growth, and the resulting decrease in surface In adatom diffusion, the number density obtained is roughly equivalent to a single layer of QDs grown at 510 °C. The strong influence of the strain fields at $S_1=10$ nm also leads to the narrowest size distribution.

On the basis of QD density and size, it is also clear that the strain fields propagate up to ~40 nm in the bilayer structures, and it is only for relatively thick spacer layers ($S_1>40$ nm) that the properties of the second layer QDs become identical to those of the first layer when grown under nominally identical conditions. The strain is relieved

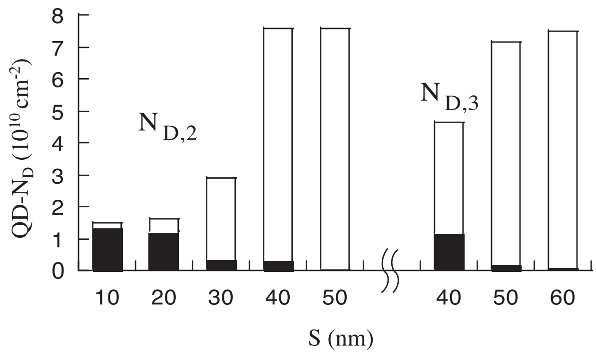

Fig. 2. Plots of the second and third layer QD density ($N_{D,2}$ and $N_{D,3}$) as a function of S. The total height of each column corresponds to the total number density. The height of the (lower) black part of the columns corresponds to large ($d > 35$ nm) dots, whereas the remainder of each column (colourless) denotes the density of dots with $d < 35$ nm.

and it is now the substrate temperature which controls the nucleation of the QDs and therefore the density and size. This results in a relatively high total $N_{D,2}$ for those structures with $S_1 \geq 40$ nm (Fig. 2), with the AFM images (Fig. 1d) showing an absence of large dots and a relatively narrow size distribution.

A different spacer layer thickness is required to relieve the strain associated with the dots in the second layer. Fig. 2 shows that for $S_2 = 50$ nm the relief of strain is not complete, since the total $N_{D,3}$ value is still lower than expected for a sample without strain-interactions. Complete strain-relief is only achieved for $S_2 = 60$ nm. The greater penetration of strain through S_2 compared with S_1 is not surprising for several reasons. Firstly, based on the QD density, the reduction of $\theta_{crit,2}$ for the second layer of QDs by approximately 0.6 ML (compared to $\theta_{crit,1}$) leads to an increase in QD height by approximately 2.6 nm. Secondly, the total coverage for the second QD layer was increased by 0.7 ML (to 3.2 ML) in order to achieve emission at long wavelengths [1]. This leads to an additional increase in QD height by 1.4 nm. Thirdly, the dots in the second layer are probably in a different strain-state than those from the first layer and the lattice constant on top of them will be more InAs-like. These differences in QD height and strain-state of the second layer QDs cause the strain-fields to propagate further through the GaAs than those associated with the first QD layer.

For intermediate spacer layers ($S_1 = 20$ or 30 nm) the influence of the strain fields is reduced and the strain energy modulation on top of the GaAs spacer layer is less pronounced. Consequently, QD formation is strain-induced, due to the preferred migration of In adatoms to regions with tensile strain [12], but in other regions it occurs as if no strain was present. In these regions the low substrate temperature dominates over the effects of strain, leading to the formation of relatively small dots due to the reduced surface diffusion of In adatoms. In the case of $S_1 = 30$ nm strain-effects are already significantly reduced and only a few large dots exist.

4. Conclusion

The strain-fields produced by the dots in the first (second) layer of a QD bilayer (trilayer) structure have been investigated by AFM. They propagate through the GaAs spacer layer and can lead to drastic changes in the properties of the QDs in subsequent layers. It can be assumed that, despite the low growth temperature, for nearly every dot in the first layer one dot is formed in the second layer for $S_1 = 10$ nm, because strain-effects dominate over the effect of reduced In adatom migration. In this case, the second layer QD density is small and the size distribution very narrow. Complete strain-relief occurs for $S_1 \sim 50$ nm, in which case the QD characteristics resemble those of a single QD layer grown at the low temperature. The process of QD nucleation and growth is then completely controlled by the temperature. For intermediate spacer thicknesses ($S_1 = 20–30$) a very large size distribution of QDs is present, which is due to the competition between strain-induced and temperature-controlled QD nucleation. Similar results are found for trilayer structures, however, due to the different size and strain-state of the second layer QDs, the propagation of strain through S_2 occurs up to ~ 50 nm and complete strain-relief is only achieved upon deposition of a spacer layer of a thickness of ~ 60 nm.

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