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A TEM study of the evolution of InAs/GaAs self-assembled dots on (3 1 1)B GaAs with growth interruption

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Abstract
The effect of growth interruption time on the structural properties of InAs/GaAs self-assembled quantum dots grown by molecular beam epitaxy on (3 1 1)B GaAs substrates is studied. Changes both in the density and the size of the dots in comparison with a sample without growth interruption are presented. The trends of both parameters with the interruption time are closely related to red shift and increased photoluminescence intensity observed in the set of samples. Growth interruption allows larger dots; however there is a limit after which dots exceed a critical size and become defective.

Low-dimensional semiconductor heterostructures have attracted considerable interest because of their unique micro- and optoelectronic applications [1], particularly since the formation of quantum dot (QD) structures modifies the density of states due to quantum confinement. Self-assembled QDs and the associated confinement [2] can potentially be used in new optoelectronic devices such as low threshold lasers [3], infrared detectors [3, 4] and high-density optical memories [5].

Several approaches have been used to obtain III–V semiconductors with quantum confinement, the 2D–3D growth mode transition being the most successful. In this case the dots are 'self-assembled' using the Stranski–Krastanov growth mode [6], which applies to heteroepitaxy with a lattice mismatch higher than 2%, such as InAs/GaAs and InAs/InP. The majority of studies of III–V semiconductor self-assembly have been grown on conventional (1 0 0) substrates, providing high quality materials. In order to obtain devices with the technological requirements, a precise control of the size, shape, composition, distribution and density of the QDs is essential. Nevertheless, a significant proportion of the semiconductor properties depends on the crystallographic orientation. Therefore, high Miller index surfaces and their influence on QD properties may be of interest and need further exploration. Sangster [7] suggested the (3 1 1) surface as suitable for crystal growth. This surface has different chemical potentials to the more widely used (0 0 1) and {1 1 1} surfaces, which affects the adsorption of different species. The capacity of the GaAs (3 1 1)B surface as a substrate for heterostructures makes it a viable alternative and enables an analysis of the change in QD properties as a function of the substrate orientation [8, 9].

Another parameter that can be varied in order to change QD size, shape and composition is the interruption time (τ_1) before growing the capping layer that fixes the dots at their position. The influence of this parameter on the epitaxial growth process has been studied by several groups for (0 0 1) InAs/GaAs [10–16], some studies linking optical properties with morphological and/or distribution data, obtained by atomic force microscopy of uncapped samples [12–16], or basic transmission electron microscopy (TEM) of capped samples [17]. A brief study describing the influence of annealing on size, shape and distribution of $(\bar{1}\,\bar{1}\,\bar{3})$B InAs/GaAs uncapped QDs is found in [18]. Nevertheless, there are no data on density and QD size for different growth interruption times of buried InAs QDs on (3 1 1)B GaAs.

In this paper we investigate by both conventional and high resolution transmission electron microscopy (TEM) the

effect of the growth interruption on the structural properties of InAs/GaAs self-assembled quantum dots grown on (3 1 1)B GaAs substrates and its influence on their optical properties.

The InAs QD samples were grown in a Varian Gen-II molecular beam epitaxy (MBE) system on (3 1 1)B orientated GaAs substrates. Initially a 500 nm GaAs buffer was grown, the first 200 nm at 580 °C and the remaining 300 nm at 600 °C. For dot growth, the temperature was reduced to 480 °C and a 1.8 monolayer InAs layer was deposited at 0.025 ML s^{-1}. The growth was interrupted under an As molecular flux. Finally the dots were capped with 25 nm of GaAs. Following this procedure, a series of four samples (S0, S40, S80 and S120) were grown. Interruption time, τ_1, for these samples was 40, 80 and 120s corresponding to the samples S40, S80 and S120 respectively. Sample S0 was grown under the same conditions but without any interruption time, i.e. $\tau_1 = 0$ and is used as a reference sample.

Room temperature photoluminescence (PL) measurements on all samples were performed using an Ar^+ laser tuned at 515 nm. The luminescence was dispersed by a 3/4 m monochromator and detected by a cooled Ge diode detector [19].

The microstructure of the layers was characterized by conventional transmission electron microscopy (TEM) and high resolution TEM (HRTEM) in a Jeol 2011 electron microscope working at 200 kV. Plan view TEM (PVTEM) specimens were thinned down to 100 μm by mechanical grinding and dimpled down to 10 μm followed by ion milling at 4.5 kV to electron transparency. Cross-section TEM (XTEM) specimens were prepared as described in [20].

Morphological parameters, such as the average QD diameter and dot density, were measured to determine the effect of the growth interruption time. Bright field PVTEM images taken under two-beam conditions, with $\mathbf{g} = 220$ near a $\langle 311 \rangle$ zone axis, show a clear change both in the density (ρ_{dot}) and the size of the dots (Φ_{dot}). Since the images were recorded under two-beam conditions the contrast in the images corresponds to the projection of the QD strain field and not to the real size of the quantum dot. However, all the images were recorded under identical conditions and can therefore be compared. The measured values of ρ_{dot} are summarized in figure 1(*a*). The dot density clearly decreases with $\tau_1 =$ 40 s and 80 s ($1.06 \pm 0.19 \times 10^{11}$ cm^{-2} and $1.04 \pm 0.24 \times 10^{11}$ cm^{-2}) with respect to the reference sample ($1.01 \pm 1.26 \times 10^{12}$ cm^{-2}). The size of the dots increases due to the growth interruption (figure 1(*b*)); in sample S40 the dots are clearly enlarged in comparison with the reference sample S0. Nevertheless, no significant changes in Φ_{dot} are observed in S80 in comparison with S40, even though the growth interruption was longer. Any small differences between these samples cannot be reliably measured from the images, due to intrinsic limitations of the bright field contrast of quantum dot images. When the interruption time is $\tau_1 = 120$ s, some significantly larger islands are observed. These islands could correspond to an agglomeration of smaller ones and are in the range of 4–6 times larger than the smaller coherent QDs.

The TEM results are consistent with the optical data from this set of samples. In figure 2(*a*) we compare the room temperature PL spectra of the four samples [19]. The room temperature PL from the reference sample (S0) has its

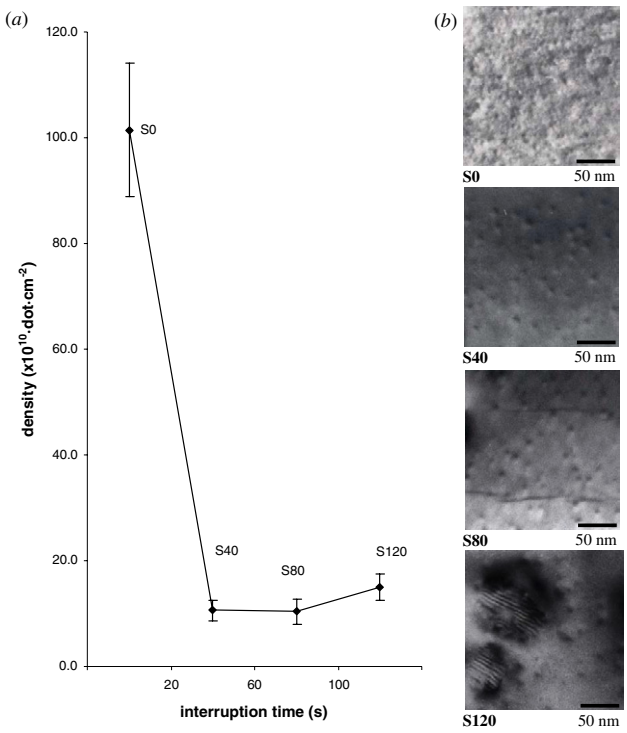

Figure 1. (*a*) Density of dots and (*b*) PVTEM images recorded under two-beam conditions with $g = 220$ close to a $\langle 1\,1\,3 \rangle$ zone axis for samples S0, S40, S80 and S120. The error bars are an estimate of uncertainty arising from the limitations of two-beam bright field images.

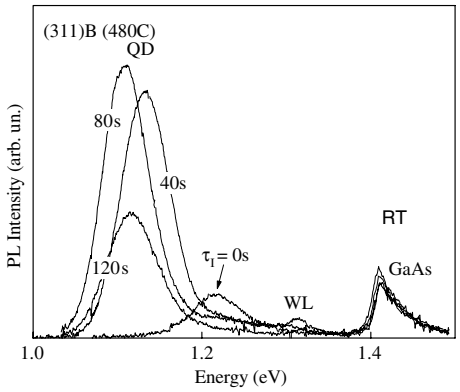

Figure 2. Room temperature PL spectra of InAs/GaAs heterostructures grown on (1 1 3)B GaAs with different growth interruption times.

maximum at 1.219 eV. The growth interruption time results in a clear red shift and increased PL intensity relative to S0. The red shift observed between S80 and S40 suggests an increment in Φ_{dot} which is not large enough to be measured by conventional TEM. When the growth interruption time increases from 80 to 120 s, a slight movement to the blue can be observed, with a broader main PL peak with reduced intensity. Thus, the optical data follow a trend correlated with the dot density and size. The PL red shift is associated with the formation of larger well-developed dots.

It is believed that the growth interruption applied after the deposition of the QDs could favour adatom transfer towards preferential sites on the growth plane, such as surface

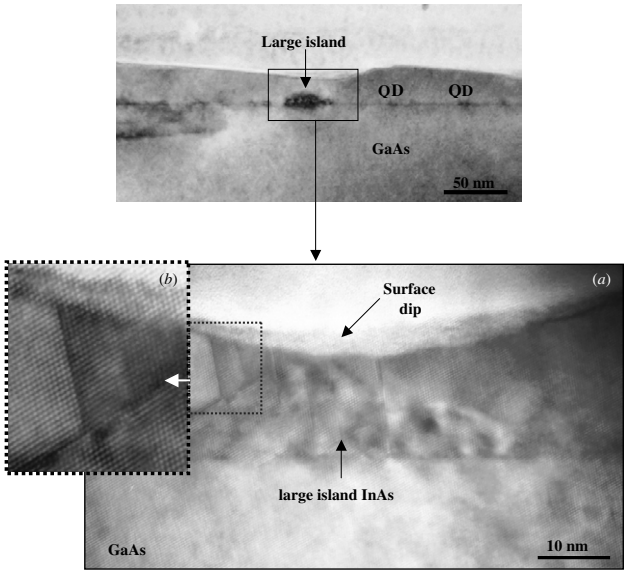

Figure 3. Conventional XTEM bright field image of sample S120, taken near the ⟨1 1 0⟩ zone axis. A bimodal distribution in the dot size is clearly observed. Image (*a*) is an enlarged part of the upper XTEM image. Image (*b*) is a ⟨1 1 0⟩ HRTEM image showing a large island in sample S120 with an associated surface dip. A higher magnification image of the area surrounded by the dotted line is inset. Planar defects can be clearly observed in the defective dot.

edge steps and previously formed dots. The dots nucleate preferentially on these sites and they have time to increase their size in order to approach an equilibrium size, although this concept is still under debate [10, 21]. Therefore, increasing τ_1 favours the formation of well-developed dots and thus a red shift of the QD PL band.

Further investigations were carried out on sample S120 in order to understand the broadening and intensity reduction observed in the PL spectra. Figure 3(*a*) is a bright field TEM recorded on a ⟨1 1 0⟩ zone axis demonstrating the occurrence of two types of microstructure, i.e. small QDs coexist with some large islands (>50 nm). These islands are often situated in areas where the surface is not flat. HRTEM was performed on various large islands along the ⟨1 1 0⟩ zone axis for atomic structure characterization. As can be observed in figure 3(*b*), these islands contain many structural defects, which is direct evidence of the proposal of Suzuki *et al* [18]. The inset in the same figure shows a more detailed image, in which planar defects are clearly observed. These defects are lying on $(1\,\bar{1}\,1)$ and $(1\,1\,\bar{1})$ planes which form angles of 80° and 30° respect to the $(3\,1\,\bar{1})$ basal plane. Therefore when $\tau_1 = 120$ s the dot size becomes larger, and during this process plastic relaxation through the generation of planar defects occurs. The dips in the overgrowth above the large incoherent islands are probably due to the fact that they are in an advanced state of relaxation, and the chemical potential for the capping atoms is higher.

Therefore, a longer growth interruption is initially beneficial since it allows more adatom diffusion and larger dots; however there is a limit after which dots exceed a critical size and become defective. This is responsible for the blue shift in the PL for the longest interruption time.

In summary, in this paper we show that the growth interruption time for InAs/GaAs self-assembled QDs grown by MBE on a (3 1 1)B GaAs substrate determines the structural and optical properties of the system. At a growth temperature of 480 °C, interruption times of 40 and 80 s are beneficial. A red shift in the PL spectra is closely related to larger dots, as well as a density reduction of one order of magnitude respect to the reference sample. Nevertheless, when the interruption time is 120 s the dots exceed the plastic relaxation critical size and planar defects clearly develop in the large islands.

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