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Critical barrier thickness for the formation of InGaAs/GaAs quantum dots

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Abstract

The discovery of self-assembled quantum dots (QDs) has attracte because of its possible applications in optoelectronic devices. The low density of QDs formed makes it useful to grow several layers of dots in the form of a stacked structure. In these structures, the buried islands tend to influence the further nucleation of islands in subsequent layers. It has been experimentally found that, when the number of layers increases, island sizes and shapes become more regular with each successive layer.

Theoretical models have been proposed to elucidate the correlated vertical self-organization. The Stranski–Krastanow (SK) growth produces a tensile region, which induces the preferential nucleation of the next SK island just above the buried 3D-island.

Experimental evidence of strain below the quantum dots (QDs) has induced us to study the influence of very thin barrier GaAs layers on InGaAs/GaAs QDs structures. This In diffusion due to strain defines the Critical Barrier Thickness for the formation of quantum wells from three dimensional islands. A Critical Barrier Thickness of 6 nm was observed in the case of 1.8 nm In0.5Ga0.5As/GaAs QDs structures. Above this thickness stacked QDs show near perfect alignment, whilst below this thickness modulated QWs are observed. The structural behaviour is supported by photoluminescence (PL) characteristics.

Keywords: Quantum dot; InGaAs; TEM; MBE

1. Introduction

Under appropriate growth conditions, three-dimensional (3D) islands in strained films can be spontaneously grown with nanometer size and coherent with the substrate lattice by employing the SK growth mechanism [1]. However, a number of studies have shown that the growth mode is complex and very sensitive to the growth conditions [2–4]. The total QD volume at low temperatures is consistent with a classical Stranksi–Krastanov (SK) growth mode. By contrast, at higher substrate temperatures alloying occurs through mass transport to the dots from the wetting layer and the substrate [5].

These islands, henceforth referred to as quantum dots (QDs), differ in size, shape, chemical composition and lattice strain, parameters that strongly influence the optical properties of such complex quantum structures [6]. The optical properties of a single InGaAs QD layer, containing typically 10^{10} dots cm^{-2}, are characterized by a broad PL peak, which is related to the finite size distribution of QDs. Improvements in uniformity of size and shape are a prerequisite for their potential use in electronic or optoelectronic devices based on strongly coupled QD transitions and so a precise knowledge of the growth, especially the interaction between kinetic processes and thermodynamics is necessary.

To reach higher dot density and more pronounced size distribution promising solutions have been developed. Xie et al. [7] showed the first evidence for vertical self-organization of coherent InAs QDs separated by GaAs spacer layers. The multiple stacking of QD layers was observed to improve the island uniformity with increasing number of bilayers (the spacer layer plus the SK layer), and in this case the 3D islands become more uniform in size,

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shape, and spacing [7–12]. For sparse initial island arrays, typical of III–V systems, individual vertical columns of islands are formed with island in each column converging to a stable size and shape. The short vertical distance of such layers of several nm enables electronic coupling between adjacent island. This multiple-stacking growth concept has successfully been applied to several semiconductor systems [8,13,14,10,7].

Three mechanisms can be invoked to describe the general properties of multilayer dot systems.

1. Segregations towards the surface during the capping process that provides additional available atoms to the second and subsequent layers.
2. Roughening processes which occur during the capping process and during the deposition of the spacer layer.
3. The effect of the strain distribution on the nucleation of 3D islands at the surface of the spacer layer in the second and subsequent layers.

The relative magnitudes of the above effects are likely to be a function of mismatch and spacer layer thickness. In relatively low-misfit alloy heteroepitaxies, such as the $In_{.5}Ga_{.5}As/GaAs$ case discussed here, although the strain effect is expected to dominate, the effects of In segregation in the spacer layer and roughness enhancement cannot be entirely neglected [15]. It is clear that atomic segregation and roughening of the growth front will also contribute to the modification of the growth process, and these phenomena warrant careful studying from a theoretical point of view.

In this paper, ten repeat multilayer $In_{0.5}Ga_{0.5}As/GaAs$ QD structures, varying systematically the thickness of the GaAs spacer layer (with 12, 6 and 3 nm), were studied by Transmission Electron Microscopy (TEM) and Photoluminescence (PL). These results have shown a modification of the 2D–3D SK transition and in the degree of the vertical correlation. From these results we have determined a critical barrier thickness for the formation of uniformly stacked $In_{0.5}Ga_{0.5}As/GaAs$ QDs structures.

2. Experimental

Ten layers of $In_{0.5}Ga_{0.5}As/GaAs$ QDs were grown via Molecular Beam Epitaxy (MBE) by solid-source MBE (VG V80H system). Each InGaAs layer was 1.8 nm (six monolayers, MLs) high and it was deposited at 0.18 ML s^{-1} growth rate without growth interruption onto GaAs (001) substrates. The growth temperature was 510 °C throughout. The GaAs capping thickness was varied from 12, 6 and 3 nm for samples A, B, and C, respectively.

Photoluminescence (PL) measurements were performed at 10K using a dispersive system and Ar-ion laser excitation with a He–Ne laser emitting at 633 nm. Specimens for cross-sectional TEM (CS-TEM) observation were prepared by mechanical thinning followed by Ar^+ ion milling. The TEM studies were performed with a Philips 420 microscope operated at 120 kV.

3. Results

The growth of a nominal 6 ML of InGaAs is expected to produce the formation of QDs since the critical layer thickness for the 2D–3D transformation by SK growth mode has been observed at 4.7 ML for the $In_{0.5}Ga_{0.5}As$ alloy by several authors [16–19]. However, our study showed that the formation of QDs is not only dependent on the amount of material deposited but also on the spacer thickness.

Fig. 1 shows a cross-section image of the sample A, where the InGaAs layers from QDs with a truncated pyramid shape as a result of a complex diffusion process [20] instead of a triangular sidewall shape and rectangular base when no cap layer is deposited [21]. The dimensions of these truncated pyramid shape island, taken from a significant number of islands, were typically 7.5 nm high, 54 nm base with a 23 nm width at the apex. The micrograph also demonstrates: i) the island size increases with the number of layers, ii) there is a narrowing of the size distribution through the stack, iii) there is an effective flattening of the growth surface by the GaAs spacer. The first two effects can be explained from the strain-result is a well aligned and reproducible QDs stack in the upper layers, in marked contrast to previous observations of InAs stacks, which showed considerable variation in size and a termination of stacking within the multilayer.

Diffraction contrast images of samples B and C, with 6 and 3 nm spacer layers, respectively, are shown in Fig. 2. Contrary to our expectations, these samples do not show QDs. Instead, alternative layers of InGaAs/GaAs modulated quantum wells are observed. In both samples, the InGaAs first layer appears as a flat modulated QW. This result is surprising given that the deposition thickness is higher than the established value of critical layer thickness. However it is possible that the slow GaAs layer growth rate used (0.09

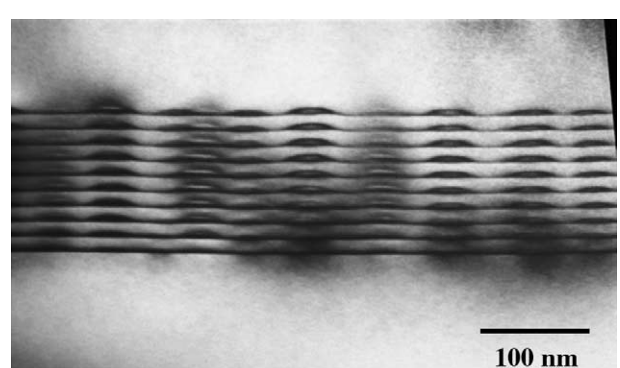

Fig. 1. Cross-sectional 200 DF image of sample A.

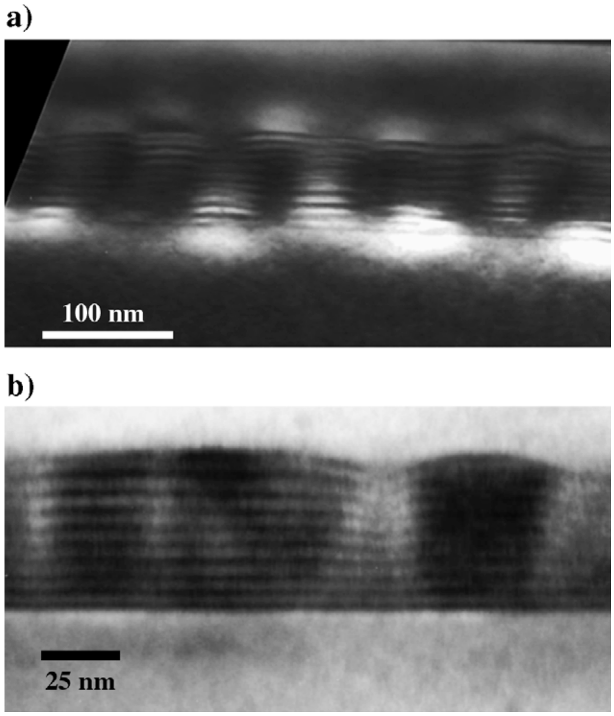

Fig. 2. Cross-sectional 200 DF image of sample (a) B and (b) C.

ML s^{-1}) has allowed re-evaporation of material from these QDs. The resulting first QD layer exhibits the behaviour of a system close to the critical layer thickness. In the case of sample C, this layer has a thickness of 1.9 nm and the 3 nm GaAs capping layer above is also flat. However, as the growth progresses, there is increasing undulation in the quantum wells, so in the upper (10th) layer the maximum InGaAs thickness is estimated as 2.9 nm and the minimum as 1.3 nm. The GaAs capping layers, although undulated, appear to have a constant thickness throughout all the structure.

Sample B, which has a barrier thickness of 6 nm, shows intermediate behaviour between A and C. The first inGaAs layer is not as flat as in sample C, but it cannot be described as a QD layer as in sample A. The structure appears as an undulated QW in a GaAs matrix. In this case, the maximum/minimum thicknesses of the InGaAs layers were estimated as 3.9/1.7 nm.

Fig. 3 shows PL measurements at low temperature (10K) of the samples for a range of excitation powers. Samples B and C (with 6 and 3 nm buffer layers) reveals a strong contribution from planar quantum well (QW) like regions in these structures. This is evidenced in the power-dependent measurements, where we find a PL peak, which shifts gradually to shorter wavelength and also broadens with increasing power levels. The magnitude of this shift, ~30 meV is consistent with the band filling (Burnstein–Moss) effect in InGaAs QWs [22]. A notably weaker contribution from QW-like regions is still found in sample A (12 nm). The general behaviour of sample A with increasing excitation power differs from that of B and C. In this case, the low excitation energy peak at 960 nm remains in all the spectra and the peak asymmetrically broadens to shorter wavelengths by up to 100 nm. The behaviour is typical of the saturation of the finite density ground state transition and population of excited states in QD systems. However, despite the excellent structural quality of this sample, PL

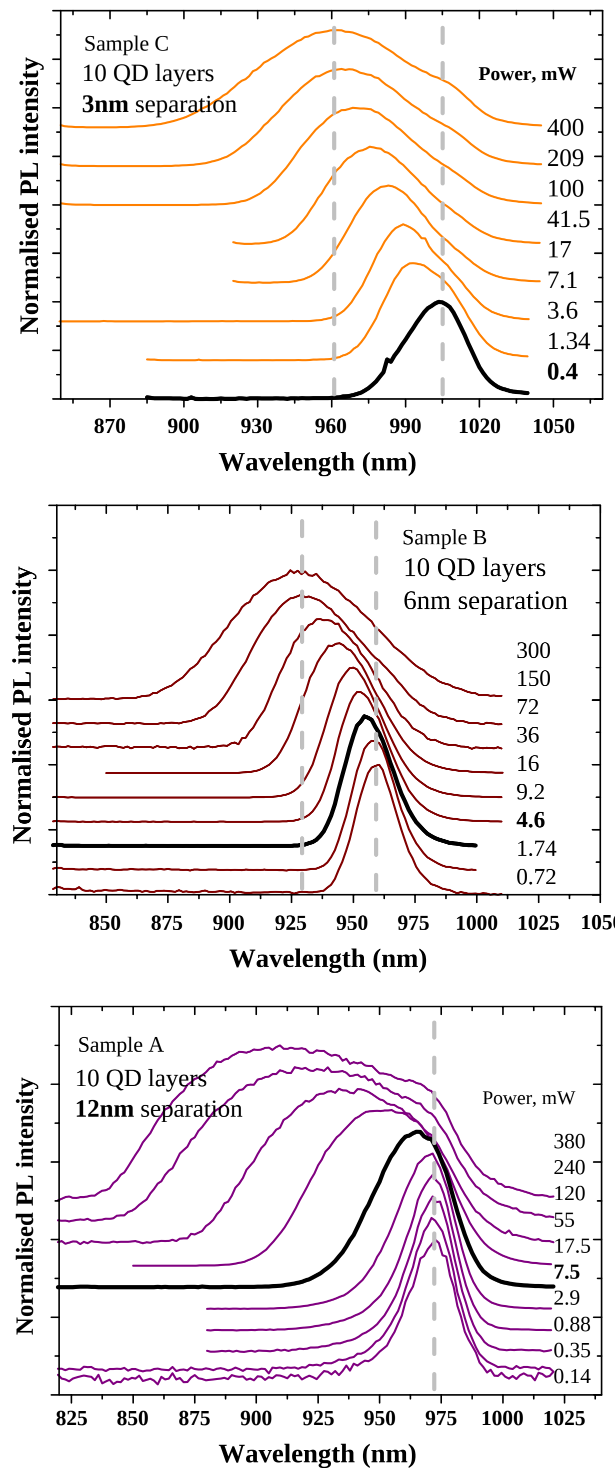

Fig. 3. Low temperature photoluminescence of the multilayer QD samples and a function of excitation power.

transitions appear relatively broad (~40 meV) and the excited state transition cannot be easily resolved.

4. Discussion

When a thin film is coherently bonded to a substrate with low lattice mismatch as in the $In_{0.5}Ga_{0.5}As$/GaAs case (ε=+3.6%), the accommodation of that mismatch is expected to be fully accounted for elastic strain. A biaxial stress is generated in a dislocation-free film of uniform thickness. However, a perfectly flat film with a constant potential along the surface is at an unstable equilibrium because the system can lower its free energy by rearrangement of atomic positions via mass transport along the surface. On this point there is a controversial behaviour discussion in the bibliography, since different systems with the same misfit show different behaviour. For example, the observed differences between the behaviour of InGaAs/GaAs and SiGe/Si systems are difficult to resolve. More specifically, the growth of a 1.8 nm layer of $In_{0.5}Ga_{0.5}As$ on GaAs, as in our studies, has been reported to develop in both a pure [23–25] or modified [26–28] SK growth mode.

From the observation of the first InGaAs layers behavior, in both Figs. 1 and 2, we cannot distinguish if these layers were grown as a rough layer, with a long-wavelength modulated composition, or if after their growth and during the capping process the dots were reduced in height and consequently diluted or depleted to form a 2D layer of none-uniform composition. In both cases, the result of having a pseudomorphic layer with different In composition zones would produce a rough surface, on top of which the GaAs spacer layer is deposited. The evolution of this roughening can be discussed in terms of a critical wavelength of modulation[29]. Periodic modulations can remain stable or unstable depending if their periods are smaller or larger than this critical wavelength, which itself is a function of the strain. In the latter case, the surface undulation creates a chemical potential which is higher at a surface peak than at a surface trough, and the process of surface diffusion smoothes out the wavy surface by transporting matter from peaks to valleys. In this case, the critical wavelength tends to infinity. As the height above the dot increases the growth of thick enough GaAs capping layers eventually smoothes the rough InGaAs providing further impetus is not provided, in the form of additional strain. This is observed in the case of sample A (12 nm barrier). The top surface of the FaAs layers is flat, but the presence of rough InGaAs layers below causes tensile regions. These regions act as a strain-modulated substrate favouring the nucleation of subsequent QDs by a strain-directed diffusion process, which results in a lowering of the overall misfit strain energy [30]. So, the formation of 3D-islands develops in the subsequent InGaAs layers and the process of preferential nucleation repeats itself creating well ordered columns of QDs.

It is well known that the interaction between the evolving strain fields induced in the substrate provides a mechanism for the preferential nucleation of QDs and for island size equalization [31]. The presence of strain fields due to InGaAs layers generates different strain regions in the GaAs cap layer (Fig. 4). Immediately after the growth of the InGaAs, a region (I) appears in which the formation of 3D islands is suppressed by the incomplete smoothing of the GaAs capping layer. At larger capping layer thicknesses, as the capping layer flattens, a second and a third region (Region II and III) appear a result of the strain field represented in Eq. (1). In these regions, the strain field is not modified by the non-planar capping layer growth and is strong enough to affect the next QD layers. Region II is defined as the region where In atoms migrate to areas where the lattice parameter is in tension and vertical-stacked islands grow on the flat capping layer. In region III, occurring at thicknesses >30–40 nm, the strain field is insufficient to nucleate QDs and a randomised distribution occurs.

Another strain region, labelled a region IV in Fig. 4, produced below a QD layer. Clearly the effect this strain field would be on layers already grown and would be through a mechanism of strain-enhanced bulk diffusion rather than the energetically more favourable surface migration effects discussed above. At present, we cannot

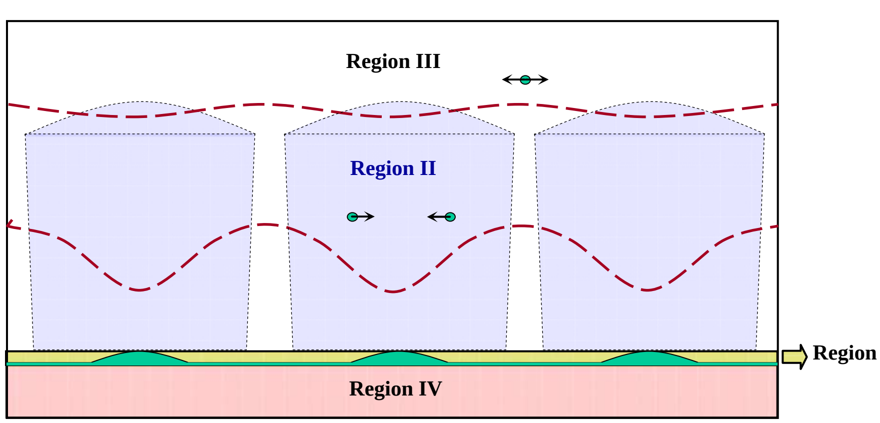

Fig. 4. Model of the different strain regions created by a strained layer.

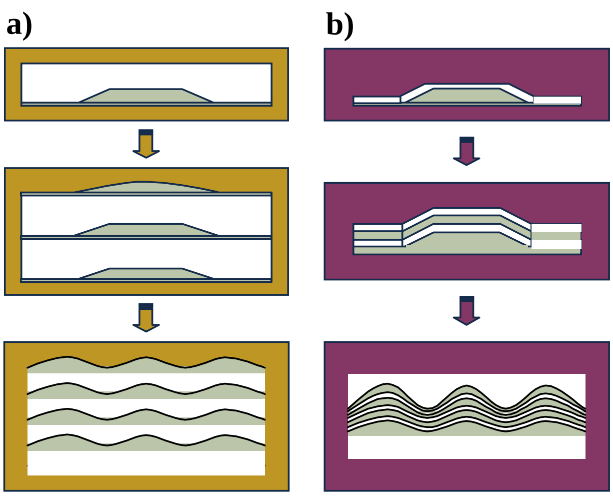

Fig. 5. Schematic of the different growth processes: (a) with flat capping layers and (b) with rough capping layers.

discount the effects of the stress on the diffusion of material already grown, although such effects appear unlikely in the structures studied in this paper.

3D islands induce strain fields in the cap layer [32] that depend on the size and shape of the buried island. Liu et al. [33] proposed that the strain at the surface of the embedding layer could, in the simplified case of a rectangular island and an isotropic elastic medium, be expressed as:

$$\epsilon(x) = -\epsilon(L)\left[\varepsilon\left(1+\varepsilon^2\right)^{-3/2}\left(2+\varepsilon^2\right) - \eta\left(1+\eta^2\right)^{-3/2}\left(2+\eta^2\right)\right] \quad (1)$$

where L is the depth which the island is placed and $\epsilon\ (L) = CH/L^2$, $\varepsilon = x + W/L$ and $\eta = x - W/L$, is C a coefficient related to misfit and elastic constants, W is one-half of the island width and H the height of the island. The resulting profiles in Fig. 4 reflect the shape of this strain distribution.

The modulated tensile stress in region II clearly explains the nucleation behaviour in sample A. For such a capping layer thickness (12 nm) subsequent QWs nucleate above the first layer and a well ordered stack is formed (Fig. 5.a). However, if the capping layer has insufficient thickness, such as in samples B and C, other factors must be sought to explain very different behaviour observed. In the case of these samples the GaAs capping layer does not fully flatten the surface and this retains a roughened profile. As result of this, an undulated capping layer is grown and the subsequent InGaAs layer is deposited on this undulated surface (Fig. 5.b). This reduces the surface tension and is clearly sufficient to avoid the formation of QDs in the second layer, as it is observed in samples B and C (Fig. 2). However, in a rough surface the lattice parameter near the valley is under a large stress, whereas the lattice parameter near the peak is relaxed. So, surface roughening of lattice-mismatched heteroepitaxial films induces a significant stress concentration at surface valleys. The stress concentration at a surface valley forces the atoms to drift farther away from the valley, which causes a deepening and sharpening of the valley into cusps. The further development of the modulated QW into cusp like features is clearly seen in Fig. 2b.

5. Conclusions

The GaAs capping layer thickness is a crucial parameter in the formation of QDs. The growth of InGaAs/GaAs multilayers, with InGaAs thickness of 1.8 nm and GaAs thickness lower that 12 nm, do not produce QDs and, in this case the growth is dominated by the incomplete smoothing of the cap layers. There exists a critical barrier thickness to form 3D islands in InGaAs/GaAs low-mismatched structures, beneath which modulated QWs are formed instead. In the $In_{0.5}Ga_{0.5}As$ case studied here this critical thickness is between 6 and 12 nm.

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