APPLIED PHYSICS LETTERS **86**, 071918 (2005)

Vertical order in stacked layers of self-assembled In(Ga)As quantum rings on GaAs (001)

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(Received 3 September 2004; accepted 21 December 2004; published online 10 February 2005)

Stacked layers of self-assembled In(Ga)As quantum rings on GaAs grown by solid source molecular beam epitaxy are studied by *ex situ* atomic force microscopy (AFM), low temperature photoluminescence (PL) and cross-sectional transmission electron microscopy (XTEM). The influence of the strain field and InAs segregation on the surface morphology, optical properties and vertical ordering of three quantum ring layers is analyzed for GaAs spacers between layers from 1.5 to 14 nm. AFM and PL results show that samples with spacers >6 nm have surface morphology and optical properties similar to single layers samples. XTEM results on samples with 3 and 6 nm GaAs spacers show that the rings are preserved after capping with GaAs, and evidence the existence of vertically ordered quantum rings. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1866228]

Molecular beam epitaxy (MBE) is a powerful technique for the fabrication of lattice mismatched semiconductor nanostructures. In particular, InAs on GaAs (001) self-assembled quantum dots (QDs) are one of the most studied systems. QDs are particularly interesting for their potential use in detectors, memories, quantum computing and photonic devices applications.[1–4] The development and design of QDs based devices requires a precise control of the morphology and composition of the QDs. As an example of the achieved capabilities to control QDs size and shape it has been proved that it is possible to self-assemble In(Ga)As quantum rings (QRs).[5] QRs are obtained by covering a layer of QDs with a thin cap (~20% of the dot height) followed by a growth pause. In this way, the original islands reshape into ring-like structures.[6]

Several authors have reported interesting differences in the optical and confining properties between QDs and QRs. Pettersson *et al.*[7] found an oscillator strength for the fundamental transition three times higher for QRs than for QDs. Warburton *et al.*[8] showed that the permanent dipole moment of exciton in QRs is three times higher and with opposite sign than those found for lens shaped QDs. Lorke *et al.*[9] proved that the ground state of QRs with zero angular momentum transits into a chiral state under the influence of an external magnetic field, concluding that ring shaped morphology translates into a ring-like electronic structure. Their nontrivial geometry, together with the possibility of controlling the energy levels, oscillator strength, polarizability and magnetic properties, make QRs good candidates for the development of new devices. The use of stacked layers increases quantum efficiency and avoids saturation gain effects in laser devices.[10] In the case of QRs, stacking the nanostructures is even more important, as the large in-plane mean dimensions of the rings (100 nm × 90 nm by AFM) require low density ring ensembles ($1–9\times10^9$ cm^{-2}) to avoid overlap problems.

In spite of all the work focused on optical characterization, little is known about the structural properties of embedded rings. This work mainly presents structural characterization on stacked layers of In(Ga)As QRs.

The samples are grown in a MBE reactor on GaAs(001) semi-insulating substrates. Reflection high energy electron diffraction is used to monitor dot formation and to calibrate temperatures and growth rates.[6] The surface morphological properties of the samples are characterized by *ex situ* contact mode atomic force microscopy (AFM). The setup details for low temperature photoluminescence (PL) are described elsewhere.[6] Cross-sectional transmission electron microscopy (XTEM) is used to characterize morphology and composition of buried nanostructures using (002) dark field images registered near two beam conditions.

AFM samples are grown using three stacked layers of QRs with a GaAs spacer thickness of 1.5, 3, 4.5, 6, 10, and 14 nm, leaving the top layer uncovered. These samples are cooled down immediately after growth and removed from the MBE chamber. In all the samples, QRs are obtained by capping the QDs with a thin GaAs layer of 2 nm. The spacer thickness is measured from this thin GaAs capping layer to the InAs deposition of the subsequent layer.

The samples for PL and XTEM characterization are similar to the AFM ones but capping the top layer with 60 nm of GaAs. Meanwhile the GaAs spacer is grown at 500 °C under As_2 flux, the rest of the GaAs is grown at 615 °C under As_4. Additional samples of single layer capped and uncapped QDs and QRs are also grown for comparison with stacked layer samples.

The QRs are obtained in each layer from an ensemble of QDs using the minimum amount of InAs coverage (ϕ_c) required for the onset of the three-dimensional transition. While ϕ_c is 1.65 ML for the first layer, in the samples with 1.5 and 3 nm GaAs spacers the nucleation of the second and the third layers takes place for a coverage significantly below

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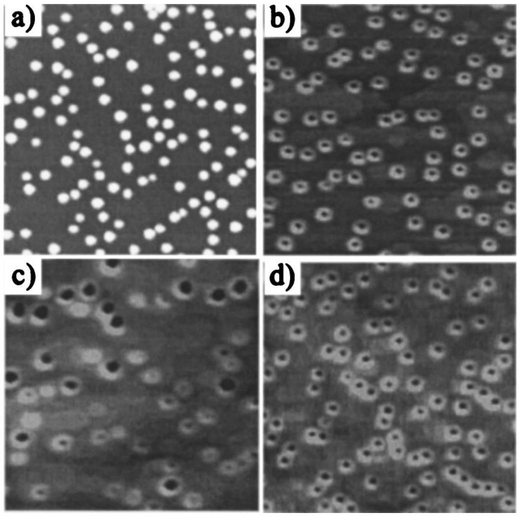

FIG. 1. AFM images 1×1 micron^2 of samples with a single layer of (a) QDs, (b) QRs, and the top layer of three stacked QRs layers separated by (c) 3 nm of GaAs and (d) by 6 nm.

($\sim$1.35 ML).[11] This effect is also observed in stacked QDs due to strong In segregation and strain propagation effect.[12–14] For spacers of 4.5 nm or thicker, ϕ_c is 1.65 ML for all QD layers.

Figure 1(a) shows the AFM characterization of a single layer of QDs (height 11 ± 2 nm and diameter 40 ± 7 nm), with a density of $\sim1\times10^{10}$ cm^{-2}. The QR density [Fig. 2(b)] is 9.7×10^{9} cm^{-2}, showing a QR formation efficiency close to one. The QRs are 1 ± 0.1 nm high and 100 ± 7 nm in external diameter along the [1-10] direction, with an internal diameter of 20 ± 5 nm. AFM images of stacked QR samples are presented in Figs. 2(c) and 2(d) for spacers of 3 and 6 nm, respectively. The sample with 3 nm spacer presents several families of ring-like and dot-like nanostructures. To elucidate whether the origin of this effect was a broad size distribution of the dots employed to form the rings, two more samples were grown with two layers of QRs and a third layer of uncovered QDs with GaAs spacers of 3 and 6 nm between stacked layers. In both samples narrow size distributions of the top QD layer are observed by AFM, similar to Fig. 1(a). These results suggest that the InAs segregation and the strain field propagation between stacked layers of QRs affects the ring formation dynamics significantly more than the dot formation. For GaAs spacers >6 nm [Fig. 1(d)] the QR aspect ratio and the density are similar to the one in the single layer.

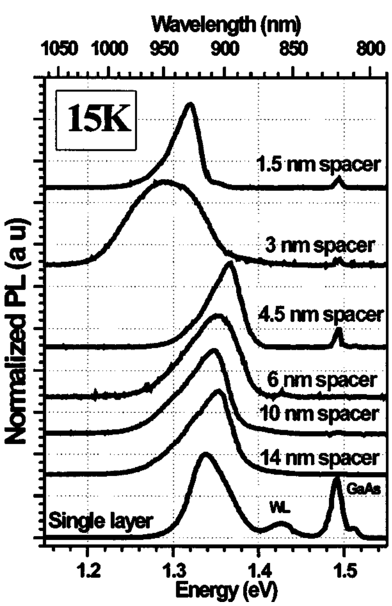

FIG. 2. PL at low temperature of a single layer of embedded QRs in a GaAs matrix (lower plot) and with various GaAs spacers between three stacked layers of QRs.

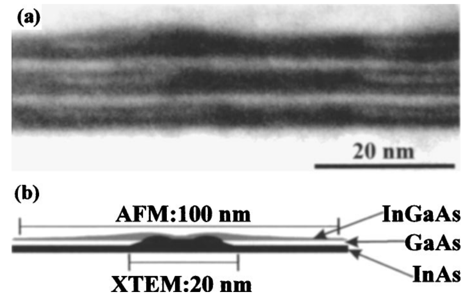

FIG. 3. (a) The (002) dark field XTEM of a sample with three stacks of QRs with 3 nm GaAs spacer between them, showing clear vertical ordering of the QRs. (b) Scheme of the proposed material distribution of a single QR depicted from XTEM measurements.

The PL emission of buried QRs is shown in Fig. 2. The emission at 15 K of a single layer is centered at 926 nm. The samples with spacers <4.5 nm show a strong inhomogeneous broadening (corresponding with the broad size distribution observed by AFM) and redshifted spectra (probably associated with a stronger electronic coupling between layers). Stacked rings with spacers 4.5, 6, 10 and 14 nm emit between 900 and 920 nm, close to the emission of the single layer one.

Figure 3(a) shows a (002) dark field XTEM image of a sample with a spacer of 3 nm of GaAs. Clear vertical ordering of the QRs can be observed in the image. Vertical ordering of the QRs has also been observed in samples with 6 nm spacers (not shown). The analysis of ensembles of ring geometry in cross-section images is not simple. It is even more difficult for XTEM technique in which slabs of material are explored. The QRs appear mostly as dark-gray disks (InAs rich) and a few times it is possible to observe two lobes corresponding to a cut through the middle of a ring. Surprisingly the sizes measured by XTEM and AFM are not the same. Meanwhile, the XTEM separation between lobes is $\sim$20 nm, the AFM dimension of the outer diameter of the ring is $\sim$100 nm. This inconsistency will be discussed next.

In Fig. 3(a) the InAs wetting layer (WL) is observed as a dark-gray region at the bottom of each layer. Moreover, on top of the WL a light-gray GaAs region surrounds each QR and corresponds to the 2 nm GaAs cap employed to form the QRs. On top of this layer there is a second gray region which corresponds to an InGaAs alloy coming from the InAs ejected out of the center of each QD during QR formation.[6,15] Most likely it is this InGaAs top layer that we measure by AFM in uncapped samples. The location of each QR can be determined by looking for dark-gray (InAs rich) regions surrounded by these trilayers.

It is possible to draw a picture of the structure of the rings and of their formation process, which agrees with previously proposed ring formation.[6,15] When the dots are partially capped with a 2 nm GaAs layer, the core of the dot is ejected outward forming a rich InAs ring-shaped core (20 nm wide) and an InGaAs ring-shaped region (100 nm wide) on top of the 2 nm GaAs layer [see Fig. 3(b)]. When the rings are capped afterward, the hole is filled with GaAs and the shape is preserved. The XTEM measured size of the InAs rich region (~20 nm) is in accordance with the estimated size for similar rings from magnetic measurements (~14 nm).[9] Generally these observations agree with experiments performed by cross-sectional scanning tunneling microscopy.[16]

The observed vertical ordering of the QRs is only present when the corresponding QR of the first (bottom) layer is isolated. Whenever several QRs are bonded together on the first layer, the vertical ordering is tilted and even some QRs are missing on the third (top) layer, maybe due to the fact that they are out of the explored slab of material. QRs are never observed on the second or third layers without a QR below on the underneath layer. The apparent size of the rings increases with the layer number for the sample with 3 nm spacer [see Fig. 3(a)]. For the 6 nm spacer the vertical ordering is observed (not shown) in the same cases as for the 3 nm spacer, though there is not an increase of the apparent size of the QR. This is reasonable, since even though the strain field propagation is strong enough to vertically order the QRs, the In segregation influence and electronic coupling are weaker and they do not affect the morphological or optical properties.

In conclusion, we have shown that the formation of rings in stacked layers is possible. The strain and segregation mainly affect the ring (more than the dot) formation. For spacers >6 nm the PL and surface morphology are similar to those of a single layer. The ejected InAs from the center of the dot forms an InGaAs ring-like structure much wider than a central InAs ring-like region. These ring structures are preserved after capping. Both 3 and 6 nm spacer samples show regions where the QRs are vertically ordered.

This work was supported by Spanish MCyT under NANOSELF project (TIC2002-04096), by NANOMAT project of the EC Growth Program (Contract No. G5RD-CT-2001-00545) and by the SANDiE Network of excellence (Contract No. NMP4-CT-2004-500101). The authors also thank the Junta de Andalucía (PAI research group TEP-0120). TEM measurements were carried out in the DME-SCCYT. The authors acknowledge P. Offermans and P. Koenraad for fruitful discussions.

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[16] In a collaboration with Peter Offermans and Paul Koenraad cross-sectional scanning tunneling microscopy measurements have been performed on our ring structures. The results will be published separately.