Chemical composition and strain distribution of InAs/GaAs(001) stacked quantum rings

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ABSTRACT: The strain and composition distributions of InAs/GaAs(001) stacked self-assembled quantum rings (QRs) grown by MBE have been analyzed. Transmission electron microscopy (TEM) images revealed a high degree of vertical arrangement of quantum rings for samples spaced by a GaAs spacer layer thickness $t_s \leq 6$ nm. The peak finding method was applied to high resolution transmission electron micrographs in order to plot strain maps, revealing that the higher strained areas were in the ring core and close to it. The existence of another layer with similar strain to the wetting layer was also clear from the peak finding strain analysis. The transverse compositional profiles taken from 002 DF TEM images show the existence of In-rich regions within the nano-rings and an In(Ga)As well defined layer surrounding them, which is formed during the growth process.

1. INTRODUCTION

Low-dimensional semiconductor structures have attracted much attention because they are promising for opto-electronic devices. For instance, semiconductor quantum dots confine electrons and holes in three dimensions and have therefore atomic-like properties. Furthermore, the electronic properties of low-dimensional nano-objects with annular geometries are important since these systems can exhibit a number of interesting properties: The ground state possesses a nonzero angular momentum with increasing normal magnetic field (Govorov et al 2002, Lorke et al 2000), the existence of a trapping magnetic flux and persistent current which is not affected by the presence of random scatterers (Pettersson et al 2000) and the permanent dipole moment of excitons in quantum rings can be higher (Warburton et al 2000) than for quantum dots. Likewise, Granados et al (2003) showed the possibility of tailoring PL emission by controlling the size and shape of quantum rings (QRs).

A narrow-size distribution and dislocation-free growth is crucial to yield macroscopic and tuned effects in opto-electronic devices. For thin enough spacer layers, stacking of self-assembled nanostructures produces vertical correlation and improved size homogeneity due to the strain field of the buried nanostructures at the surface of the spacer layer, where the new nanostructure layer will be formed (Xie et al 1995, Sprinholz et al 2000, Brault et al 2000).

Although conventional analyses by TEM are extremely useful and versatile tools for the characterization of material, nowadays quantitative methods applied directly to HRTEM images represent an important advance to determining the strain and composition of the epitaxial nanomaterials (Hÿtch et al 1998, Rosenauer et al 1996).

In this work we have studied the vertical arrangement of stacked InAs QRs grown at 500°C on GaAs (001) substrates, as a function of the GaAs spacer layer thickness. Compositional distribution profiles have been defined from TEM micrographs recorded with 002 reflections under dark field conditions. With the aim of extracting the strain field distribution, particularly interesting regions of the ring have been examined in depth by the peak finding method (Kret et al 2003).

2. EXPERIMENTAL

The studied samples were grown by molecular beam epitaxy (MBE). As has been explained before in detail (Granados et al 2003), the growth consisted of the deposition of InAs quantum dots (QD) on a GaAs substrate (001); after that, about 2nm of GaAs cap layer was deposited at 500°C. The competition between the change of surface energy and the Ga-In alloying led to structures which are known as quantum rings. Following the same steps, three stacks of buried InAs QRs, separated by a GaAs spacer layer were grown. We have investigated three samples with spacer layer thicknesses of 3, 4.5 and 6nm, respectively.

For TEM examination, the samples were mechanically pre-thinned down to a thickness of about 90µm. Subsequently, they were dimpled to the final thickness of approximately 20µm. Finally, ion milling (Ar+) was carried out applying a gradually decreasing acceleration voltage under cooling with liquid nitrogen to minimize damage. TEM images have been obtained with a JEOL 1200EX, while HRTEM images were taken with JEOL 2010F and JEOL 3000F microscopes operating at 120, 200 and 300kV, respectively.

3. RESULTS

Figure 1 displays a couple of 002 DF cross sectional TEM (XTEM) images that correspond to samples with $t_s = 3\text{nm}$ (a) and 6nm (b). While the sample spaced by 3nm of GaAs layers exhibits an obvious vertical ordering, a certain degree of arrangement loss is observed for the sample spaced by 6nm of GaAs layers, given that stacked rings groups, but also some non-stacked elements, can be observed. Some stacked and non-stacked QRs are pointed out by single and double arrows respectively in Fig. 1b. An intermediate case is presented for a spacer layer thickness of 4.5nm, which leads sometimes to a loss of the QR circular shape.

As reported in previous work (Granados et al 2004), if we look at the centre of each nano-object in the images of Fig. 1, several contrasts are observed. Firstly two darker contrasts are observed, one of them corresponding to the horizontal InAs wetting layer, and the another one to the InAs of the QR core with a spherical shape or a volcano-like morphology, left after the de-wetting process, whose contrast depends on the section analyzed. Moreover, two grey contrasts are shown, that are associated (i) to the compressing GaAs layer used in the generation of the QRs and, over it, (ii) an In(Ga)As layer that comes out from the original dot. This layer and the semi-empty core are the main parts which compose the annular morphology. The spacer and capped layers between the stacked rings appear with brighter contrast. These results give us a consolidation of the models proposed in previous work (Anders et al 2002, Emer et al 2000).

Fig. 1: 002 DF TEM images of stacked InAs QRs with 3 (a) and 6 (b) nm spacer layer.

Figure 2 depicts the different ring areas mentioned before using several grey levels. In this case the ring is shown without the cap layer growth as spacer. Figure 3 exemplifies a qualitative composition distribution profile for the region in between two rings extracted from the 002 DF TEM micrographs (see Pr_1 in Fig. 1a). Three peaks, P_1 , P_2 and P_3 , linked (from left to right side in the figure) to the 1st, 2nd and 3rd stacked rings layers respectively, are mainly illustrated in this profile. Each peak is deconvoluted to two gaussian curves (labelled with G_{i1} and G_{i2} on the original profile,

where $i = 1, 2$ or 3). The intensity profile due to GaAs has been subtracted from the original profile before the application of the deconvolution process. From the two deconvoluted gaussian curves, the highest intensity curve is ascribed to the InAs wetting layer and the lowest intensity curve to the above mentioned In(Ga)As layer. The In(Ga)As peak maximum is 30, 40, and 16% respectively below the InAs peak for the 1st, 2nd and 3rd stacked rings layers, respectively. This result helps to understand the Ga/In alloying process occurring during the ring generation.

Fig. 2: Schematic drawing detailing the different areas of an uncapped ring. The encircled area corresponds to profile Pr_2 of Fig. 1(a).

Fig. 3: In composition distribution profile along [001] for region in between two rings whose peaks have been adjusted to Gaussians.

We have also acquired composition distribution profiles (see Pr_2 in Fig. 1a) through regions around the edge of the ring core (corresponding to the encircled area of Fig. 2). It is worth mentioning that the In(Ga)As layer usually presents an In composition lower than that of the wetting layer, though for a few of these analysed areas that layer presents a composition similar to the wetting layer. Due to the TEM specimen preparation process, very different ring sections can be found in the electron-transparent slice. Due to this fact, we have concentrated our attention on a few noteworthy and less confusing ring areas, such as the core edge and the region in between two of rings.

Figure 4a shows one of the HRTEM images analysed by the peak finding method corresponding to a region where just the In(Ga)As upper layer is joined to the ring core. The displacement vectors between maximum intensity positions on the image and reference lattice determined in the substrate (or in another reference region of the material) are identified, and the strain field is plotted by the derivation of these vectors with respect to the spatial coordinates (Kret et al 2003). The determined strain map (measured with respect to the GaAs lattice parameter) along [001] (ϵ_{zz}) is shown in Fig. 4b.

Fig. 4: a) HRTEM image of the sample consisting of three InAs stacked rings layers with 6 nm spacer layer thickness. b) Contour plot of the experimental strain map ϵ_{zz} determined from the HRTEM image.

The strain field map of Fig. 4b reveals a more highly strained region near of the ring core for the second and third layers. That is, an increase of the strain field occurs for the upper layers (see the right side of Fig. 4b). Nevertheless, the same number of InAs monolayers has been deposited for the QD formation for the three stacked quantum ring layers, and so we can ensure that the In incorporated amount is higher than that introduced into the MBE chamber for the second and third layers. The GaAs compressing layers have a strain close to the GaAs spacer layers. Next to and on top of each ring core, a layer with a strain smaller than the wetting layer is appreciated. In a few cases this layer can have a strain similar to (or even slightly larger than) the strain of the wetting layer. From this result and the compositional profiles we conclude that the In(Ga)As layer formed during the de-wetting process has, in some few cases, similar to (or slightly larger than) the In composition of the wetting layer, decreasing as we move away from the ring core.

4. CONCLUSION

A vertical correlation of QRs is reached for spacer layer thickness $t_s \leq 6$ nm. 002 dark field TEM image profiles reveal clearly an In-rich core in the ring left after occurrence of the dot de-wetting process, and an In(Ga)As layer which begins next to this core and spreads along on the GaAs compressing layer up to the adjacent ring.

The ε_{zz} strain maps determined by the peak finding method confirm the existence of an In-rich core in the ring and an In(Ga)As layer highly strained. This method together with transversal profiles for 002 DF TEM images allow us to conclude, in some few cases, that the In(Ga)As layer composition is similar to (or slightly larger than) the wetting layer composition in the region close to ring core.

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