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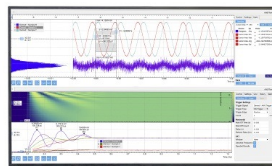

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Atomic scale analysis of self assembled GaAs/AlGaAs quantum dots grown by droplet epitaxy

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In this letter we have performed a structural analysis at the atomic scale of GaAs/AlGaAs quantum dots grown by droplet epitaxy. The shape, composition, and strain of the quantum dots and the AlGaAs matrix are investigated. We show that the GaAs quantum dots have a Gaussian shape and that minor intermixing of Al with the GaAs quantum dot takes place. A wetting layer with a thickness of less than one bilayer was observed. © *2010 American Institute of Physics*.
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Lattice-matched nanostructures such as strain free quantum dots (QDs) can be grown by using molecular beam epitaxy working in the droplet epitaxy mode (DE). First reported by Koguchi *et al.*,[1–3] the technique involves low temperature growth of unstrained group III-element droplets that are subsequently crystallized into QDs by incorporation of group V-elements. Traditionally, QDs were not grown in DE but in the Stranski–Krastanow (SK) mode. However, this latter technique is restricted to lattice-mismatched materials and therefore yields QDs that often suffer from material intermixing and strain related effects. In contrast, it has been shown that DE can be used to grow strain-free, nearly pure nanostructures with a typical size distribution of 10%–20%,[4] comparable to SK grown QDs. Recently, it has been shown that such QDs can be used in optoelectronic devices.[5,6] Furthermore, stain free QDs are highly desired in experiments devoted to the comprehension and control of single QD properties, e.g.,[7–10] In this letter we investigate GaAs QD grown by DE embedded in an AlGaAs matrix by means of cross-sectional scanning tunneling microscopy (X-STM). The shape, composition, and strain of the QDs and the AlGaAs matrix is investigated. Furthermore, the presence of a wetting layer is investigated.

All X-STM measurements were performed at room temperature under UHV ($p < 6 \times 10^{-11}$ mbar) conditions with an Omicron STM1, TS-2 Scanner. The STM was operated in constant current mode on *in situ* cleaved (110) surfaces. The preparation of the electrochemically etched tungsten STM tips is described in Ref. 11. The sample consists of three quantum ring (QR) and one QD layer. After the growth of the AlGaAs barrier layer at 580 °C, the sample is cooled down to 200 °C to form an As-stabilized $c(4 \times 4)$ surface. Subsequently, 3.75 monolayers (ML) Ga, of which the first 1.75ML changes the excess As into a two-dimensional (2D) GaAs layer,[12] is deposited at 0.5ML/s. The remainder of 2ML will form the Ga-droplets on the surface. Next, the droplets are crystallized into a QR or QD shape by supply of an As_4 flux (1×10^{-5} and 2×10^{-4} Torr beam equivalent pressure for, respectively, QR and QD). Still under As_4 flux, the sample is then annealed at 350 °C for 10 min. Subsequently the structures are capped with 50 nm AlGaAs deposited at 350 °C, followed by a second annealing step at 650 °C under As_4 flux for 5 min. This anneal step is inserted into the growth procedure to ensure that the next layer is grown on a defect free surface. Next, another capping layer of 40 nm is grown at 580°. The total structure was capped with 600 nm GaAs. A postgrowth anneal step, which is usually performed to improve the optical properties of the QDs was not performed on this sample. Although QR layers are present in the sample, this paper will only report on the QD layers.

In Fig. 1, the AlGaAs matrix is shown. All the images presented in this letter were recorded at high negative voltages (≈ -3.2 V). At these tunneling conditions and with the color scaling used, dark regions represent AlAs rich regions while bright regions represent GaAs rich regions. Although no QDs are visible in this image, two interesting features are observed. First, the position of the first annealing step is clearly visible as a dark layer, indicated by the black arrow on the top in Fig. 1. From this we conclude that a significant portion of the Ga is desorbed from the first couple of MLs during the annealing resulting in an increased AlAs concen-

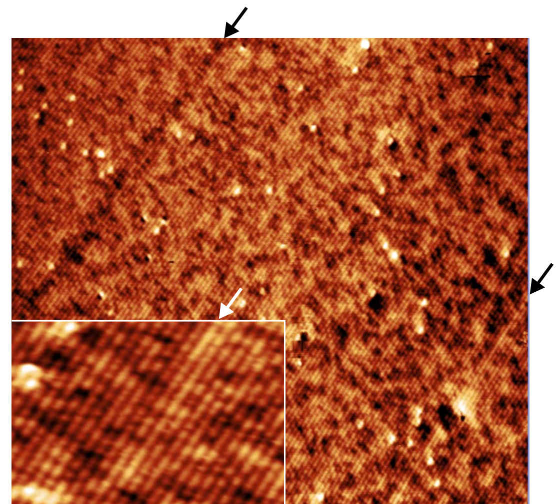

FIG. 1. (Color online) 60 nm × 54 nm topographic image of the AlGaAs matrix. The position of the first anneal step and the wetting layer is indicated by, respectively, the black arrow at the top and the black arrow on the right. The inset shows a close up of the wetting layer where the position of the wetting layer is indicated by the white arrow.

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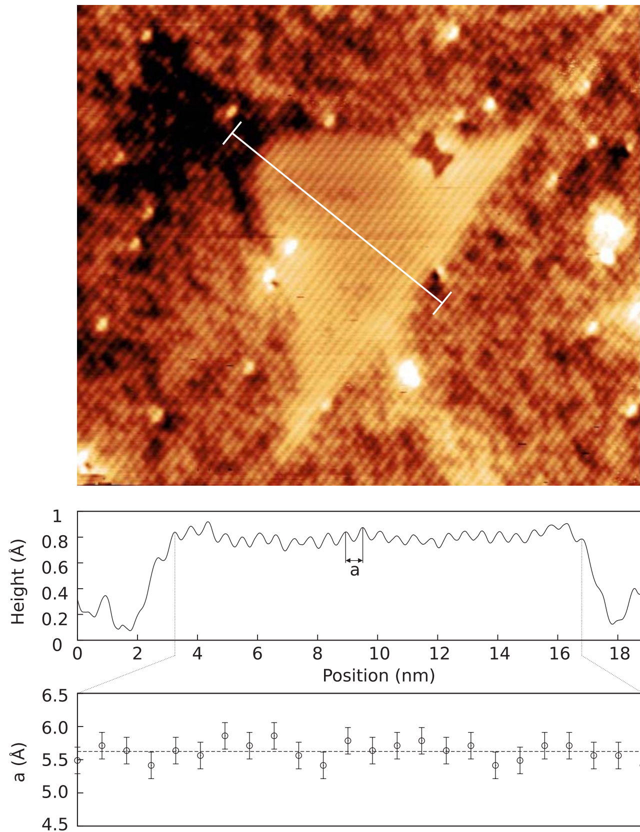

FIG. 2. (Color online) 40 nm×34 nm topographic image of a typical GaAs/AlGaAs QD (top) and an average cross-sectional profile (top graph) and separation between bilayers (bottom graph) along the line in the top figure.

tration. Second, a GaAs wetting layer, indicated by the black arrow on the right in Fig. 1, is visible as a narrow bright line. Closer inspection of the wetting layer, see the inset of Fig. 1, reveals that, as expected when depositing Ga on an As-stabilized $c(4\times4)$ surface,[2,13] the thickness of the wetting layer is less than one bilayer.

In this study a total of 11 QDs where analyzed with X-STM. A typical QD is shown in Fig. 2. As can be seen in this topographic image, the QDs are sharply defined by abrupt interfaces. Note that there is an Al rich region on top of the dot. This can be explained by the different mobility of Al and Ga atoms; the Ga atoms are more mobile and will migrate along the side of the QD during capping while the Al atoms, which are less mobile, are more likely to remain on top of the QD. The driving force behind the migration of the incoming adatoms away from the top of the QD is due to the convex curvature of the growth front at the position of the QDs.[14] Since AlAs and GaAs are lattice matched materials, the QDs are expected to be strain free. This is checked by the cross-sectional profile of the QD in Fig. 2. Three distinct regions can be observed. From left to right: an AlAs rich region, the GaAs QD, and the AlGaAs matrix. The height difference between these regions is due to electronic contrast. More importantly, all the regions are flat, there is no outward relaxation as observed in QDs grown with lattice-mismatched systems.[15] To further illustrate that the GaAs QD is strain free, the distance between adjacent bilayers along the cross-sectional profile was measured. For this analysis the STM piezoelements were calibrated by performing a 2D FFT on the AlGaAs matrix. The result is shown in

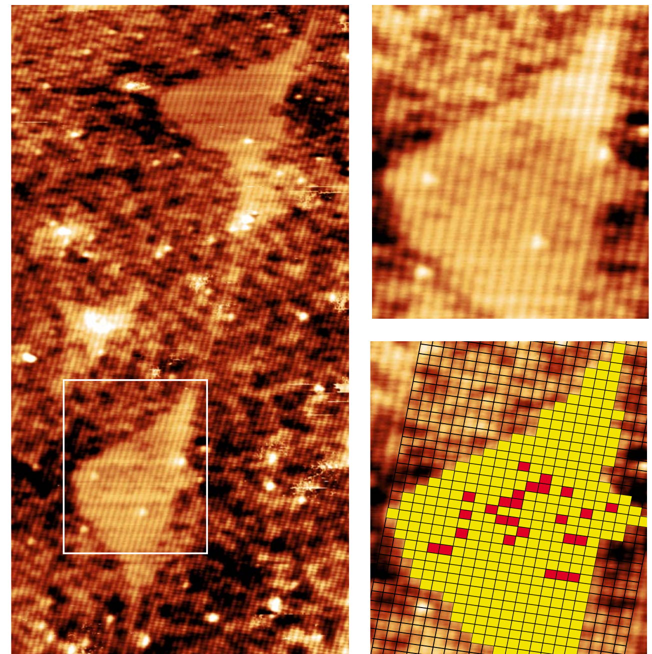

FIG. 3. (Color online) 30 nm×60 nm topographic image (left) of two QDs. An atomic grid is overlain on top of a close up of the QD dot (right). Al and Ga atoms in the QD are indicate by, respectively, red and yellow squares.

the bottom graph of Fig. 2. As can be seen, there is little deviation from the expected value of 0.565 nm (dashed line), indicating that the QD is indeed strain free. The bow tie feature in Fig. 2 is most likely a foreign atom and is of no interest in the current study.

Whether intermixing of Al is a factor of importance in the formation of GaAs/AlGaAs quantum dots grown by DE is a question frequently raised in the literature.[16–18] In all QDs imaged we have observed some degree of intermixing. In Fig. 3, two typical QDs are shown (left). Even without further analysis it is evident that some intermixing of Al has taken place. To make a more quantitative analysis we have overlaid a grid with atomic dimensions on top of a close up of one of the QDs (right). On this grid, the positions of the Al and Ga atoms are marked with, respectively, red and yellow squares. We found that the concentration of Al in this particular QD is 6%. The Al intermixing we observed varied from dot to dot, see for example Fig. 2 which shows a QD in which the intermixing is considerably lower, and thus we conclude that Al intermixing only plays a minor role in the formation of GaAs/AlGaAs QDs.

Concerning the shape of the QDs, we notice that the side facets of the measured QDs are not exactly straight. The maximum side facet angles were found to be in the range 34°–55° per QD, were the upper limit corresponds to a {111} facet (54.7°). If we assume that (1) all the QDs are approximately of equal height and (2) the observed height difference is due to the position of the cleavage plane relative to the center of the QD, this result excludes QD shapes with constant facet angles like rectangular (truncated) pyramids.[19] Since it has been reported that uncapped AlGaAs/GaAs QDs have {111} facets,[2,13,20] we conclude that the shape of the QDs is somewhat changed during capping. Figure 2 shows the highest QD we found. Since it is the highest, we assume that this QD is cleaved directly through its center. Consequently, we used the profile of this QD to generate a three-

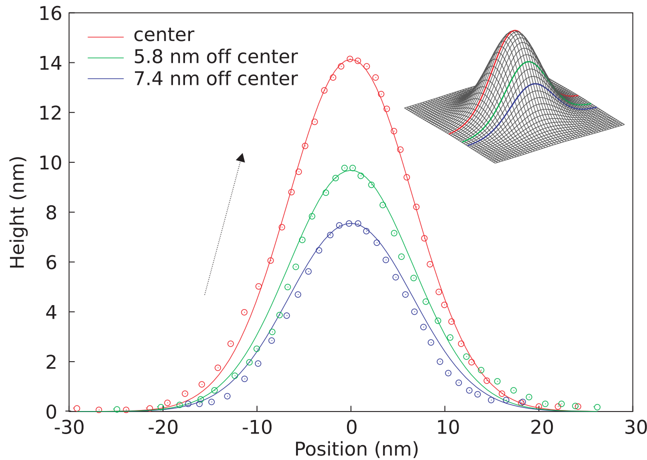

FIG. 4. (Color online) Profile of three quantum dots as measured by X-STM (open circles). A Gaussian function is fitted to the largest dot (red line). The other two dots (green and blue line) are assumed to have the same 3D-structure as the largest dot but cleaved off center. The projection of the 111-direction on the cleavage plane is given by the dashed black line.

dimensional (3D)-profile by fitting a Gaussian function, see Fig. 4 (red line), and rotating it around the symmetry axis along the growth direction. Next, we checked whether other QD profiles (green and blue lines) correspond to profiles obtained by cleaving the obtained 3D-profile at specific distances from the center. As can be seen in Fig. 4, this is the case. From this we conclude that the measured QDs are Gaussian shaped QDs of approximately the same height but cleaved at different position from their center.

To summarize, in this X-STM study we have shown that wetting layers, although not notable, form in QD layers grown with DE. The thickness of the wetting layer was found to be less than one bilayer. As expected in lattice-matched systems, we found no strain present in the QDs. Our result show that some degree of intermixing of Al in the GaAs dots is present. The shape of the QDs was found to be Gaussian. We thank STW-VICI under Grant No. 6631 for their financial support.

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