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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.

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 × 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(4×4) 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 As4 flux (1 × 10−5 and 2 × 10−4 Torr beam equivalent pressure for, respectively, QR and QD). Still under As4 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 As4 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 °C. 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-
ing a 2D FFT on the AlGaAs matrix. The result is shown in
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. 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 par-
ticular 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 approxi-
mately 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 con-
stant facet angles like rectangular (truncated) pyramids. Since
it has been reported that uncapped AlGaAs/GaAs QDs have
\{111\} facets, 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. Conse-
quently, we used the profile of this QD to generate a three-

\begin{figure}
\centering
\includegraphics[width=0.5\textwidth]{figure2.png}
\caption{(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.}
\end{figure}

\begin{figure}
\centering
\includegraphics[width=0.5\textwidth]{figure3.png}
\caption{(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.}
\end{figure}
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.