InAs/GaAs(001) quantum dots close to thermodynamic equilibrium

G. Costantini,[a] C. Manzano, R. Songmuang, O. G. Schmidt, and K. Kern
Max-Planck-Institut für Festkörperforschung, Heisenbergstr.1, D-70569 Stuttgart, Germany

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InAs/GaAs(001) quantum dots are grown at high temperature and extremely low flux and analyzed by *in situ* scanning tunneling microscopy. A bimodal distribution of dots is observed, composed of ''small'' and ''large'' islands. While the former show a broad distribution of sizes and shapes, the latter appear to be highly uniform and have a truncated pyramid shape with irregular octagonal base. (110) and (111) facets are identified and atomically resolved showing (1×1) and (2×2) surface reconstructions, respectively. The shape of the large quantum dots is in excellent agreement with recent theoretical predictions, proving that the chosen deposition conditions are close to thermodynamic equilibrium. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1572534]

Self-organized semiconductor quantum dots (QDs) produced by lattice-mismatched heteroepitaxy are considered a promising system for many developing electronic technologies and devices, such as nanoelectronics, low-threshold current lasers, memory storage, testing paradigms for quantum computers, etc.[1,2] A precise control over width, height, and shape of the QDs is of crucial importance since these morphological characteristics influence the quantum confinement of the charge carriers and therefore determine their optoelectronic properties. Nevertheless, at present, it is still very difficult to manipulate these morphological parameters in a controlled and reproducible way by acting on the experimental deposition variables. Even by restricting the research only to the most studied system, namely InAs/GaAs(001), and considering only QDs grown by molecular beam epitaxy (MBE), a number of different and sometimes contradictory experimental results have been reported in literature: widths ranging from 10 to 40 nm, heights from 2 to 10 nm, and shapes such as lenses, truncated pyramids, and a variety of multifaceted structures.[3–6] Turning to theory, very little is known, and most of the proposed models are based on thermodynamic equilibrium hypotheses. These allow one to consider only ensemble-averaged parameters, neglecting complex kinetic aspects that are difficult to treat for semiconductor systems.

Apart from the possibility of a direct comparison with theoretical predictions, the growth of semiconductor QDs under thermodynamic equilibrium would also be extremely favorable for device-oriented applications. In fact, thermodynamic equilibrium conditions are much more stable and less sensitive to small variations in the experimental parameters, and thus allow a higher degree of reproducibility and transferability of the achieved results. In order to approach this regime and to minimize the importance of kinetic effects, we grow QDs at high temperatures and at extremely low growth rates. A further positive effect of this choice is the production of ''large'' QDs, characterized by light emission wavelengths close to the technologically relevant 1.3 μm[7] and with linewidths as narrow as 16 meV.[8]

We used semi-insulating GaAs(001) wafers as substrates and removed their natural oxide by 10-min heating at 640 °C in ultra high vacuum (UHV). A 400-nm GaAs buffer was deposited thereafter by MBE at a flux of 0.6 monolayers per second (ML/s), holding the substrate at 610 °C. Flux calibration was done by means of reflection high energy electron diffraction (RHEED) intensity oscillations, while the RHEED monitoring of the $(2\times4)\rightarrow c(4\times4)$ transition in the surface reconstruction was used for calibrating the sample temperature at 500 °C. The actual QDs were grown by deposition of 1.8 ML of InAs at 0.008 ML/s with the substrate at 500 °C and an As pressure of 8×10^{-6} mbar. Immediately after closing the In shutter, the substrate heater was turned off while keeping a constant As pressure, resulting in an initial cooling rate of 1 °C/s. As soon as room temperature was reached, the samples were transferred under UHV conditions to a different chamber equipped with a homemade scanning tunneling microscope (STM). This system, similar to the one described in Ref. 9, is able to measure full-wafer samples and to analyze a large surface area. STM images were taken in the constant current mode with typical tunneling currents of 0.1 nA and voltage biases of -3.0 V (filled states).

Figure 1(a) shows a characteristic STM surface topography after the deposition of 1.8 ML InAs on GaAs(001). Two types of QDs are clearly recognizable: ''small'' and ''large'' dots, with surface densities of 7×10^{10} and 3×10^{9} cm^{-2}, respectively, that were determined by analyzing a large number of wide-area atomic force microscopy (AFM) scans similar to that shown in Fig. 1(b). The structure of the wetting layer (WL) could be determined by means of high-resolution STM images [Fig. 1(c)] and manifests the coexistence of two surface reconstructions, the (2×4) and the (1×3), in agreement with the observations in Ref. 10. A bimodal size distribution of QDs in the InAs/GaAs(001) system has already been reported by F. Patella *et al.*,[11] but in that case, the small dots had a narrower size distribution and the larger were, most probably, the result of a ripening process. Conversely, our measurements indicate that the large QDs have an extremely narrow size distribution (standard deviations of 5% for the height and 10% for the lateral dimension) while the smaller ones show large variations in height and width. Even if most of the small QDs do not have any well-defined shape and might be interpreted as precursors of fully developed

[a]Electronic mail: gio@fkf.mpg.de

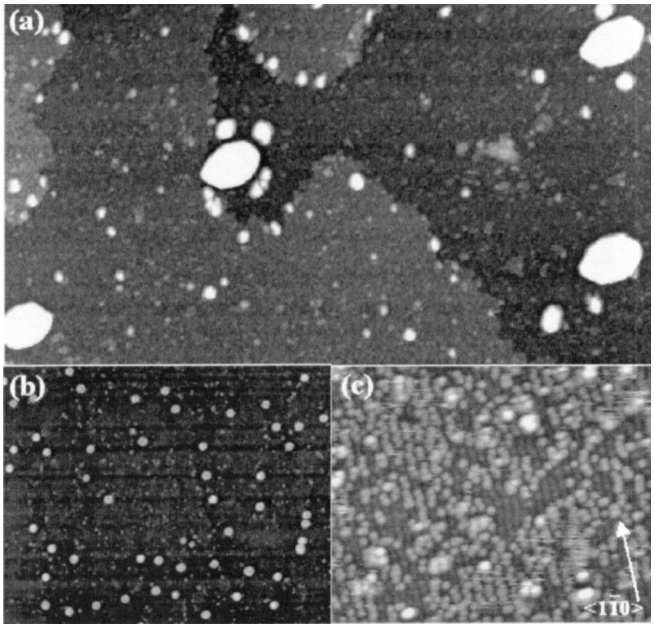

FIG. 1. (a) 500×280-nm^2 STM image of 1.8 ML InAs on GaAs(001). The image contrast has been enhanced to show surface details. (b) 2.0×1.6 μm^2 *ex situ* AFM scan of the same system. (c) High-resolution STM image of the wetting layer showing (2×4) (light gray) and (1×3) (dark gray) reconstructed domains.

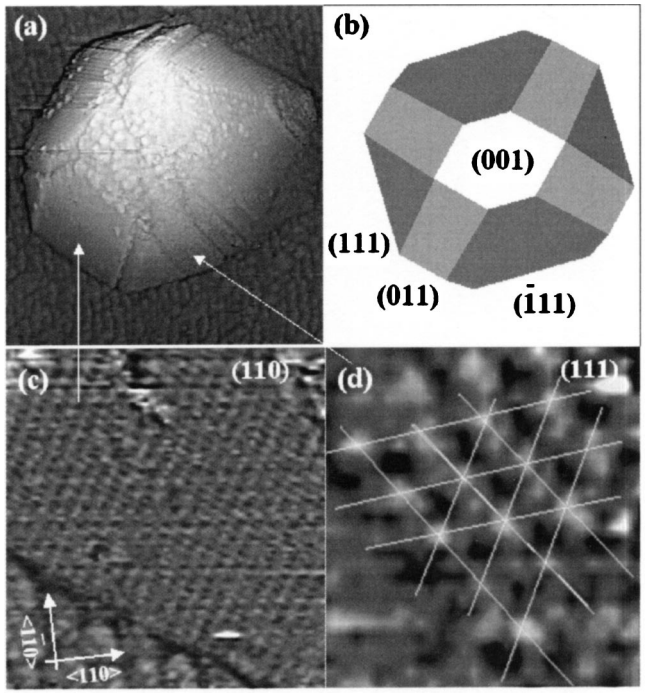

FIG. 2. (a) 50×50-nm^2 STM topography of a large island. (b) Equilibrium shape of an InAs island according to Ref. 14. High-resolution views of (c) the (110) facet (12×12 mm^2) and (d) the (111) facet (4×4 nm^2) of the same island. Lines showing the (2×2) reconstruction have been superimposed in (d).

QDs, a small fraction is composed by faceted islands with an irregular hexagonal base slightly elongated along $\langle 1\bar{1}0\rangle$. These are typically 14 ± 3 nm wide and 2.4 ± 0.5 nm high, and a measurement of the angles between their facets and the (001) plane allows an assignment to the (137) orientation, in accordance with Ref. 3. Some of us[12] have recently demonstrated that a 30-s growth interruption after the InAs deposition is sufficient for the complete disappearance of the small dots, while the larger ones remain almost unchanged. This, together with their narrow size distribution, is a first indication that the large QDs are stable structures grown close to thermodynamic equilibrium.

High-resolution STM images, such as Fig. 2(a), show that the large dots have the shape of a truncated pyramid with an octagonal base and are elongated perpendicularly to the dimer lines of the WL (i.e., along $\langle 110\rangle$). Their typical widths are 25 ± 4 nm along $\langle 1\bar{1}0\rangle$ and 44 ± 4 nm along $\langle 110\rangle$, and their height is 14.4 ± 0.7 nm. Despite these large dimensions, the dots are dislocation-free, as confirmed by good photoluminescence properties and transmission electron microscopy measurements. A closer inspection of STM topographs reveals that the sides of these QDs are composed of only two types of facets: (110) and (111). This is revealed by imaging the facets with atomic resolution [Figs. 2(c) and 2(d)]. Such measurements are much more reliable than the sole determination of facet angles, and show a rectangular lattice for the (110) planes [Fig. 2(c)] and a triangular one for the (111) planes [Fig. 2(d)]. Moreover, the evaluation of the lattice parameters (calibrated in respect to the WL reconstruction in the same image) allows even the identification of the surface reconstructions which turn out to be (1×1) for (110) and (2×2) for (111).

The accurate experimental determination of the shape of large QDs allows for a meaningful comparison with recent theoretical predictions for the equilibrium shape (ES) of coherently strained (i.e., dislocation-free) InAs islands on GaAs(001).[13–15] Scheffler and collaborators have developed an hybrid approach in which the surface reconstructions, the surface energies and their strain dependence are calculated *ab initio* by density functional theory (DFT), while the long-range strain relaxation in the QDs and in the underlying substrate are determined by continuum elasticity theory applying a finite-element approach.[15,16] The striking agreement between these theoretically predicted ES and our experimental findings is evident by the comparison between Figs. 2(a) and 2(b). This agreement extends also to the surface reconstructions of the island facets, since DFT calculations predict that the energetically preferred reconstructions are a relaxed (1×1) cleavage plane for InAs(110) and two different (2×2) reconstructions for InAs(111) and InAs$(\bar{1}\bar{1}\bar{1})$.[14] While the ES and the corresponding surface reconstructions agree nicely, the theoretically predicted volumes of the QDs are a factor of 15 smaller than the actually measured value of 5×10^6 Å^3. The reason for this is most probably the strong dependence of the ES on the lattice mismatch,[16] so that even small deviations from the theoretical value of 7.1% (due to Ga incorporation in the growing QD) can result in strong variations of the volume. Also the ratio between the extensions of (110) and (111) facets is slightly larger than predicted by theory. Apart from an experimentally smaller As partial pressure, this could also originate from residual kinetic effects, as for example the higher growing rate of (111) facets in respect to (110) ones.[17] Nevertheless, the agreement with the theory developed by Scheffler and coworkers is very good, and since the theory essentially relies on the hypothesis of thermodynamic equilibrium, this agreement represents a test on how close the chosen experimental parameters are to the thermodynamic equilibrium.

Murray *et al.*[6,10] have recently obtained quantum dot structures characterized by room-temperature emission at 1.3 μm that were grown at comparably low rates. The low resolution of their *ex situ* AFM characterization does not allow any direct comparison, but sizes and densities of the QDs are comparable with our results. The presence of (110) and (111) orientations in multifaceted islands has also been derived from x-ray scattering measurements,[18] even if the QDs were reported to be considerably smaller that what we actually measure. Apart from a difference in the deposition temperature, this could also be due to the difficulty of indirectly inferring heights and sizes of QDs from the width of broad crystal truncation rods. The major part of the literature concerned with InAs/GaAs(001) QDs concentrated on small dots (lateral dimensions <20 nm, heights <5 nm) for which a number of structural models has been proposed. Limiting only to the high-resolution measurements (both in real[3,5,19] and in reciprocal space[20]), an agreement can be found on a faceted island shape with $\langle 110 \rangle$-elongated hexagonal base and (137) main facets, according to the nice work of Márquez *et al.*[3] Our small dots strongly corroborate this model and their coexistence with the larger QDs might reconcile previous, apparently scattered and contradictory experimental results. Although both small and large dots appear to be stable structures, we would like to stress once more that only the larger ones are thermodynamic equilibrium structures, since only they show a temperature stability[12] and an agreement with theoretical equilibrium shape predictions.[13–15]

In conclusion, we have presented an accurate morphological investigation of the shape of large self-organized InAs quantum dots on GaAs(001). The very good agreement with previous theoretical previsions for the equilibrium shape of QDs indicates that the chosen deposition parameters are close to thermodynamic equilibrium, thus offering a high degree of stability and reproducibility. Moreover, the demonstration of the coexistence of stable smaller dots with larger ones reconciles previous experimental reports on different shapes and sizes of InAs/GaAs(001) QDs.

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