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## Atomically resolved structure of InAs quantum dots

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InAs was grown by molecular-beam epitaxy onto GaAs(001) until quantum dots (QDs) formed. At this point, the growth was interrupted and the uncovered QDs were investigated *in situ* by scanning tunneling microscopy (STM). Atomically resolved STM images of the QDs revealed that four dominating bounding facets occur, whose Miller indices were identified to be {137}. The assignment of the facet orientation was based on experiments on planar high Miller index GaAs surfaces. In addition, the latter experiments indicated that {137} facets are thermodynamically stable only up to a certain size. This conclusion is assumed to explain the sharp size distribution of InAs QDs.  
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In recent years, the self-organized growth of semiconductor nanostructures that show quantum-size effects has been of considerable interest due to their enormous potential for technological applications.<sup>1</sup> Consequently, laser devices operating with self-assembled InAs quantum dots (QDs) embedded in GaAs have already been demonstrated.<sup>2</sup> So far, experimental work has focused on varying the growth conditions to control the size and improve the uniformity of the QDs.<sup>3</sup> Theoretical work has addressed the formation phenomena of quantum dots and attempted to predict their size and density.<sup>4</sup> However, the accuracy of such calculations is limited by the knowledge on the QD shape. While most theoretical calculations have assumed that only surfaces with low Miller indices occur as bounding facets of the QDs, particularly those which are based on thermodynamical descriptions,<sup>4</sup> from experiments the occurrence of facets with high Miller indices was concluded.<sup>5–9</sup> Thus, the determination of the exact shape of the QDs is needed to substantiate the theoretical models and thereby, to improve the device technology.

In this letter, we determine the exact shape of self-assembled InAs QDs from atomically resolved structures on their bounding facets. Four facets dominate the shape of the QDs, whose Miller indices were identified with the help of studies on high-index GaAs surfaces to be {137}.

The experiments were performed in a multichamber ultra-high-vacuum system.<sup>10</sup> Our samples, with a typical size of  $10 \times 10 \text{ mm}^2$ , were cut from GaAs(001) epitaxially grown wafers [Wafer Technology, Si doped,  $n = (1.1 - 4.8) \times 10^{18} \text{ cm}^{-3}$ ]. GaAs buffer layers of 20–50 nm thickness were grown by molecular-beam epitaxy at a temperature of 560 °C. After growth, the samples were cooled down to 450 °C and were kept at this temperature for 10 min until the GaAs(001) $c(4 \times 4)$ -diffraction pattern was observed by reflection high-energy electron diffraction (RHEED). Hereafter,  $1.8 \pm 0.2 \text{ ML}$  InAs were deposited at a growth rate of  $0.05 \text{ \AA s}^{-1}$ . The As:In beam equivalent pressure was 40. RHEED showed a transition from a streaky to a spotty pattern, indicating the onset of three-dimensional islanding. The samples were then

rapidly (within 15 s) transferred to the analysis chamber where they cooled down to room temperature before they were examined by scanning tunneling microscopy (STM). STM images were acquired in constant-current mode.

As a first observation, the RHEED pattern (not shown here) showed the often observed chevron-like spots along the  $[1\bar{1}0]$  azimuth, which is attributed to facets that are inclined at an angle of  $20^\circ - 25^\circ$  from the substrate. Perpendicularly, i.e., along  $[110]$ , the RHEED pattern remained spotty, indicating that no specific facet orientation occurred. Furthermore, large scale ( $0.5 \times 0.5 \text{ \mu m}^2$ ) STM images (not shown here) revealed a QD density of  $1.9 \times 10^{11} \text{ cm}^{-2}$  with a very sharp size distribution. The average diameter and height of the QDs were 125 and 22 Å, respectively. Since both RHEED and STM results were in good accord with observations of other studies (e.g., Refs. 5 and 9), we conclude that our further results should hold in general for InAs QDs. However, it should be noted that our study is based on one specific set of growth parameters, while it is well known that the size, the composition, and the density of the QDs depend on the growth conditions.<sup>3,11</sup> Therefore, our analysis does not exclude that QDs of different shapes may also occur.

In order to determine the shape of the QDs, STM images showing smaller areas ( $< 800 \times 800 \text{ \AA}^2$ ) were acquired. With the help of the atomic structures of the wetting layer, which exists between the QDs, we have been able to calibrate the scanner and to determine the principal directions ( $[1\bar{1}0]$ ,  $[110]$ ) on the substrate surface with high accuracy.

Figure 1(a) shows a STM image of one of the QDs, which is representative for all QDs considered for this letter. The QD has a pyramidal shape with a fairly sharp summit. There are four pronounced facets at the right- and left-hand sides of the image, which determine the shape of the QD. Most remarkably, atomic features are clearly visible as rows of humps on these four facets. The inclination angle between these rows and the substrate is  $26^\circ \pm 2^\circ$ . The intersections of these facets with the substrate are not parallel to the principal axes of the (001) substrate, but are inclined by  $24^\circ \pm 2^\circ$  with respect to  $[1\bar{1}0]$ . We note that the quantity of the error was derived from a total amount of 53 QDs. The ends of the QD on the lower and upper parts of the image do not show clear facets and appear fairly rounded. Atomic-scale features were

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that different shapes of the QDs would be derived if only one of these cross sections was considered: From the first one a lens-shaped QD would be derived, and a pyramidal QD in the second case. Furthermore, this anisotropy explains the difference in the RHEED patterns taken along these directions. With the electron beam directed along  $[110]$ , the RHEED pattern shows only a spotty pattern. Since the cross section of the QD along  $[1\bar{1}0]$  is rounded, the intensity of the diffracted electron beams is distributed homogeneously, and thereby no facet-related spots can be observed. In contrast, parallel to  $[1\bar{1}0]$  the electrons are diffracted from the  $\{137\}$  facets of the QDs towards specific directions, and thereby the chevrons appear in the RHEED pattern. Moreover, the agreement between RHEED and STM observations also ensures that the shape of the QDs is determined during growth and is not related to the quench procedure of the samples. This conclusion is also in accord with experiments by Guha, Madhukar, and Rajkumar,<sup>14</sup> who reported a similar cross-sectional anisotropy on buried InAs QDs by high-resolution transmission electron microscopy.

The occurrence of  $\{137\}$  facets on the QD leads to significant conclusions for the formation of InAs QDs. The study of surfaces inside the stereographic triangle<sup>12</sup> revealed that only one thermodynamically stable surface exists within the observed region, namely, the GaAs(2511) surface. This surface is composed of three As dimers wide (137) stripes, which by themselves do not fulfill the electron-counting rule (ECR).<sup>15</sup> The ECR states that the reconstructed surface remains semiconducting, and thereby it is expected to be of low energy. In contrast, the GaAs(2511) surface does fulfill the ECR, because threefold coordinated Ga atoms within the trenches between adjacent (137) stripes exist, which compensate the missing charges of the surface yielding a semiconducting surface reconstruction.<sup>13</sup> However, as demonstrated in Refs. 12 and 13, and (137) stripes with different widths also occur. These stripes extend over several 100 Å and indicate that a surface, which is composed of (137) stripes with other than three As dimers should be at least semistable. In fact, theoretical studies in Ref. 13 have shown that the surface energy of one of these surfaces, which did not fulfill the ECR either, was only a little higher than that of the GaAs(2511) surface. Thus, the experimental observations on GaAs(2511) and the theoretical calculations led us to the assumption that  $\{137\}$  areas are stable up to a certain size. Above this size, the surface energy per unit area increases rapidly. This may explain the sharp size distribution of the QDs with a modified coherent Stranski–Krastanow growth mode: When the area of the  $\{137\}$  facets on the growing QD

reaches this certain size, the energy gain due to the strain relaxation within the three-dimensional island does no longer exceed the surface energy. Hence, a certain critical QD size exists, which is determined by the size of the  $\{137\}$  facets.

To summarize, based on atomically resolved STM images of the QD facets, we proposed a model for the shape of self-assembled InAs QDs, which is determined by four  $\{137\}$  facets. The ends of the QDs in the  $[1\bar{1}0]$  direction are rounded and do not show specific facets. It is suggested that the special arrangement of As dimers within the  $\{137\}$  orientations and the resulting surface energies might be responsible for the sharp size distribution of self-assembled InAs QDs and should, therefore, be considered in further theoretical studies.

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