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Pyramids and domes in the InAs/GaAs(0 0 1) and Ge/Si(0 0 1) systems

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Abstract

A systematic study of the morphology of self-organized islands in the InAs/GaAs(0 0 1) and Ge/Si(0 0 1) systems is presented, based on high-resolution scanning tunneling microscopy measurements. We demonstrate that in both cases two main island families coexist: smaller pyramids bound by one type of shallow facets and larger multifaceted domes. Their structure and facet orientation are precisely determined, thus solving a highly debated argument in the case of InAs/GaAs(0 0 1). The comparison between the two material systems reveals the existence of striking similarities that extend even to the nature of island precursors and to the islands that form when depositing InGaAs or GeSi alloys. The implications of these observations on a possible universal description of the Stranski–Krastanow growth mode are discussed with respect to recent theoretical results.

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1. Introduction

The first stages of lattice-mismatched semiconductor heteroepitaxy are accompanied by the spontaneous formation of coherent three-dimensional (3D) nano-islands (Stranski–Krastanow growth mode). This growth technique has proved to be a viable means for the realization of quantum dots (QDs) [1], since it combines several positive aspects such as an extremely low defect density, small sizes, and thus large confinement energies, and the possibility of a direct integration into the mature semiconductor technology. Nevertheless, it is necessary to overcome the inhomogeneity

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problems bound to the intrinsic statistical nature of self-organized growth before this technique becomes truly appealing for device applications. This requires a deep understanding of the growth mechanisms, starting with a thorough characterization of the 3D islands' shape and size that, together with their composition and strain, determine the optoelectronic properties of the final QDs.

InAs/GaAs(0 0 1) is considered as the prototypical system for self-organized semiconductor QDs since most of the fundamental studies in this field have concentrated on it. Moreover it has been used as a benchmark for realizing and testing various types of novel applications: single photon sources [2,3], single electron devices [4,5] and high-performance lasers and amplifiers [6] have been already demonstrated. This system has also been considered as one of the best candidates for the practical realization of quantum gates, the building blocks of quantum computers [7,8]. Remarkably, in spite of this large interest, a commonly agreed picture of the 3D islands' morphology has not yet been established [9–17]. The situation is different for the Ge/Si(0 0 1) system, the second *drosophila* in the field of self-organized semiconductor QDs, for which a much clearer and comprehensive description has been developed over the last years [18,19].

In this work, the structure of InAs 3D islands, spontaneously formed on GaAs(0 0 1), is investigated by means of in situ scanning tunneling microscopy (STM) and, at lower resolution, by atomic force microscopy (AFM). By comparing these measurements to analogous ones on the Ge/Si(0 0 1) system, we find striking similarities that prove how a common classification of island types (domes, pyramids, hut clusters and embryos) can be applied in both cases. The implications of these observations on a possible general description of the Stranski–Krastanow growth scheme are discussed with respect to recent theoretical results.

2. Experimental procedure

The samples were prepared and analyzed in two different experimental setups. InAs islands were grown in a molecular beam epitaxy (MBE) apparatus on Si-n^+ doped GaAs wafers that had been previously thermally deoxidized (10 min at 640 °C) in ultra high vacuum (UHV) and overgrown with a Si-doped GaAs buffer (400 nm at 610 °C). 1.8 monolayers (ML) of InAs were then deposited at a growth rate of 0.01 ML/s on top of a further 10-nm-thick undoped GaAs layer with the substrate at 500 °C and under an As_4 beam equivalent pressure of 8×10^{-6} mbar. Immediately after growth, the sample heating was switched off (resulting in an initial cooling rate of ~1 °C/s) while maintaining the same As flux. The As shutter was closed when the substrate temperature reached a value of about 200 °C and the sample was moved out of the growth chamber. Some of the samples were transferred under UHV to an independent STM where they were measured in the constant current mode (current ~0.1 nA, voltage ~−3 V, filled states imaging). The others were directly removed from the MBE apparatus and analyzed under ambient condition by AFM.

Ge islands were prepared by UHV magnetron sputtering epitaxy [20]. B-p^+ doped Si(0 0 1) substrates were outgassed for about 30 min in UHV and flash cleaned by alternate current heating for removing the native oxide. Sixty nanometers thick Si buffers were subsequently grown at a rate of 0.7 ML/s and a substrate temperature ranging from 350 to 600 °C. Ge was then deposited at a rate of 0.3 ML/s with the substrate at 550 °C, until reaching a nominal thickness of 7.0 ML. Samples were cooled to room temperature and studied by UHV-STM about 20 min after their growth.

3. Results and discussion

Both material systems follow the Stranski–Krastanow growth mode as the formation of 3D islands takes place after an initial planar growth of the epilayer. The transition occurs at a critical thickness that depends on the corresponding lattice mismatch, around 1.6 ML and 7.1% for InAs/GaAs(0 0 1) and around 4.0 ML and 4.2% for Ge/Si(0 0 1), respectively. Representative morphologies of the samples' surfaces after the island formation are depicted in Figs. 1(a) and (b),

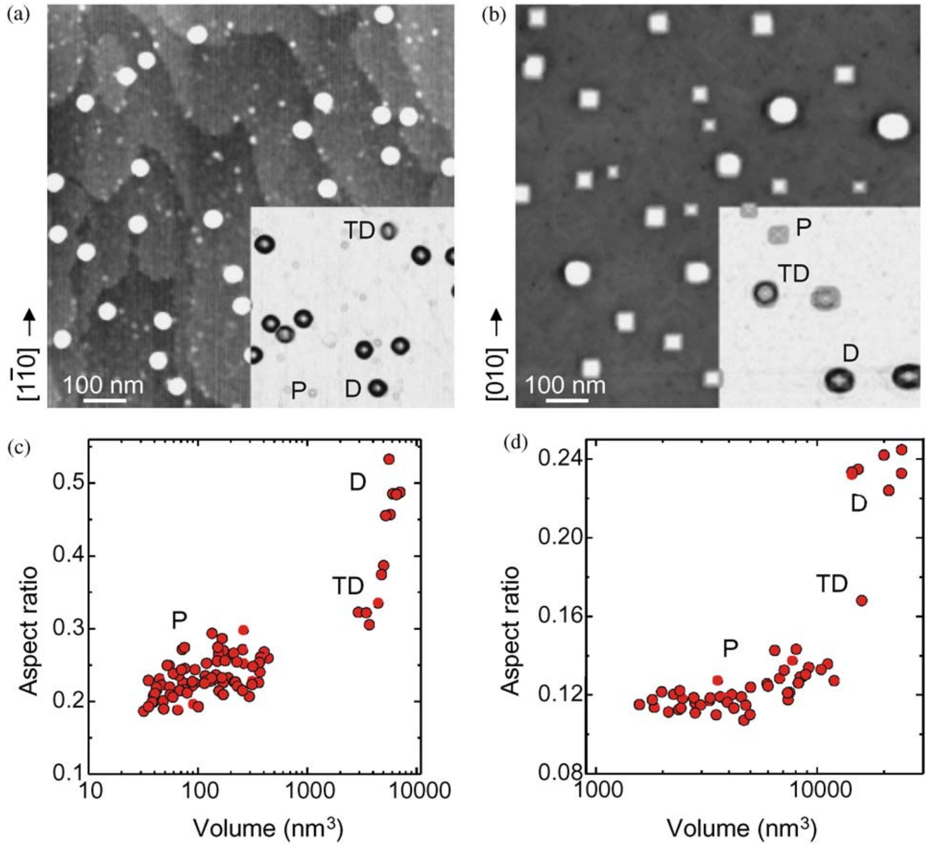

Fig. 1. Upper panel: representative AFM topographies of bimodal island distributions for (a) InAs/GaAs(0 0 1) and (b) Ge/Si(0 0 1). In the lower right corner of the images, a grayscale representing the surface slope instead of its height is used in order to enhance the visibility of islands' details. Examples of pyramids (P), domes (D) and transition domes (TD) are explicitly indicated. Lower panel: corresponding analyses of the islands' aspect ratio versus volume. Island classes are characterized by a similar aspect ratio.

showing that in both cases the chosen growth conditions produce a coexistence of small and larger islands. A quantitative analysis of these data, in which the aspect ratio of each island (defined as the height divided by the square root of the base area) is plotted versus its volume, further demonstrates that the islands divide into two main families with well-defined aspect ratios (Figs. 1(c) and (d)).

Such a bimodal distribution of small-shallow and large-steep islands is well known in the Ge/Si(0 0 1) case [18], but has also been reported for InAs/GaAs(0 0 1) [21,22]. Extending the terminology used in the former material system, we propose to generally call the two island types as *pyramids* and *domes*. A small percentage of the islands is characterized by volumes and aspect ratios that are intermediate between those of pyramids and domes, as can be verified in the scatter plots of Figs. 1(c) and (d). These islands, called *transition domes*, represent an intermediate step in the process occurring to pyramids when they increase their volume, eventually transforming into full domes. Up to now this transformation has been directly reported only in the Ge/Si(0 0 1) system [18,23,24], but the data of Figs. 1(a) and (c) constitute a first evidence that a similar phenomenon occurs also for InAs/GaAs(0 0 1).

In order to determine the actual shapes of the different islands, highly resolved in situ STM measurements become mandatory. Such images are reported in Figs. 2(a) and (c) for the pyramid

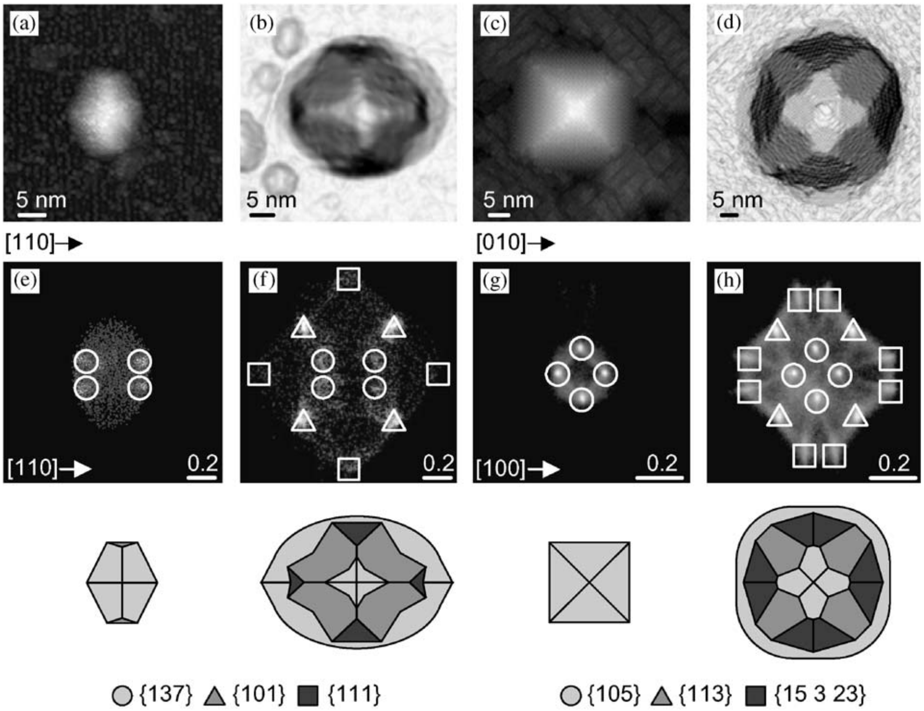

Fig. 2. Upper panel: high-resolution STM images of pyramid and domes in the InAs/GaAs(0 0 1) system—(a) and (b) and in the Ge/Si(0 0 1) system—(c) and (d), respectively. The grayscales are related to the surface height in the case of pyramids and to the surface slope in the case of domes. Central panel: corresponding histograms of the local surface gradient (facet plots, FPs). Each spot represents an island facet whose orientation is determined by the distance and angular position in respect to the (0 0 1) center of the plot. The main facets are indicated according to the corresponding symbol code. Lower panel: structural models for pyramid and dome islands in the two material systems. Different facets have different gray tones, according to the corresponding color code.

islands and in Figs. 2(b) and (d) for the domes in the case of InAs/GaAs(0 0 1) and Ge/Si(0 0 1), respectively. For pyramids, the grayscales are proportional to the actual island height while in the case of domes they are related to the local surface slope in order to display all morphological details. Already from these data, it clearly appears that the analogy between the two material systems extends far over the bimodal size distribution, since in both cases pyramids are bound by one type of shallow facet, while domes are steeper and multifaceted. This can be quantified by plotting histograms of the surface gradient averaged over a large statistics of high-resolution data [19,25]. The bright spots appearing in the resulting graphs, also called facet plots (FP), can be unambiguously assigned to facet orientations of the islands. Figs. 2(e)–(h) show the results of this analysis selectively done for pyramids and domes in the two material systems. Pyramids show four spots located at the same distance from the center of the plot, representing four equivalent and equally extended facets with {1 3 7} and {1 0 5} orientation, for InAs/GaAs(0 0 1) and Ge/Si(0 0 1), respectively. In the former case a further blurred intensity distribution around the $[1\bar{1}0]$ direction in Fig. 2(e) evidences the presence of additional steeper orientations stemming from the small upper and lower parts of the pyramid (see Fig. 2(a)). It is not possible to assign these tiny regions to definite facets [11] and the correspondent absence of clear spots in the FP seems to indicate rounded, not well-defined areas. By applying the FP analysis to the domes, several main spots are produced corresponding to {1 3 7},{1 1 1} and {1 1 1} facets in the case of InAs/GaAs(0 0 1) and {1 0 5}, {1 1 3} and

{15 3 23} facets in that of Ge/Si(0 0 1) [19,26], respectively.

The similarity between the two systems is also valid for the shallow {1 3 7} (respectively {1 0 5}) facets that can be observed at the top *and* at the base of the domes. In the case of Ge/Si(0 0 1) these have been interpreted as the remnants of the pyramids from which the domes have evolved [24]. This is a further evidence that a similar pyramid-to-dome transition can be expected in the InAs/GaAs(0 0 1) system, with a similar transformation path, essentially consisting of the bunching of incomplete {1 3 7} facets at the top of large pyramids [24].

Based on the high-resolution STM data, it becomes possible to formulate a precise morphological model for the pyramid and dome islands, shown in the lower panel of Fig. 2. While being already known for Ge/Si(0 0 1), this represents the long-sought simple and coherent description of 3D self-organized islands for the InAs/GaAs(0 0 1) system. We notice that the emerging picture is directly compatible and unifies a number of previous reports [11–13,21,27]. Moreover, slightly different assignments of the islands' facets [15–17] can also be brought back to this model, when considering the inherent imprecision of the experimental techniques by which they were obtained.

The first self-organized semiconductor 3D islands for which an accurate morphological characterization was carried out comprising an identification of their facets, were the so-called Ge *hut clusters* grown on Si(0 0 1) [28]. These have essentially the same structure as pyramids, but with two opposed {1 0 5} facets larger than the remaining ones, so that the islands become elongated in the [1 0 0] or [0 1 0] directions. Hut clusters have been reported for a wide range of growth conditions and, although having been mainly investigated in the low-temperature regime, can be observed also on our bimodal pyramids-and-domes samples, as shown in Fig. 3(b). Fig. 3(a) demonstrates that akin structures grow also in the InAs/GaAs(0 0 1) system. In this case, since the intersections of the {1 3 7} facets with the (0 0 1) substrate plane are not mutually perpendicular, the main axis of the hut clusters is along [3 $\bar{1}$ 0] or [1 $\bar{3}$ 0], thus forming an angle of about 18° with the [1 $\bar{1}$ 0] direction along which the pyramids align.

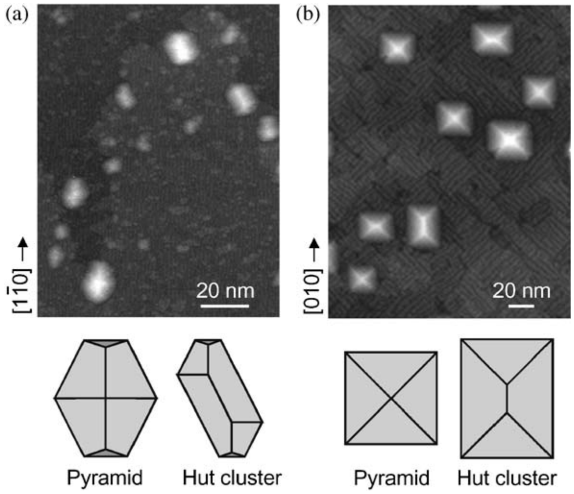

Fig. 3. Upper panel: STM images of pyramids and hut clusters for (a) InAs/GaAs(0 0 1) and (b) Ge/Si(0 0 1). Lower panel: corresponding structural models.

The aspect ratio vs. volume analysis reported in Figs. 1(c) and (d) has been performed on islands whose size was larger than a minimum threshold. This is because besides domes, pyramids and hut clusters, a certain number of smaller islands can also be found on the same samples. Some of them have an almost two-dimensional character and are constituted by few ML high platelets, but others are genuinely 3D, although not possessing any well-defined shape. Figs. 4(a) and (b) show two typical examples of this latter type of islands for InAs/GaAs(0 0 1) and Ge/Si(0 0 1), respectively. In both cases, together with indefinitely rough regions, it is easy to recognize some small faceted areas with the same {1 3 7} and {1 0 5} orientations of the respective pyramid islands. These *embryos* represent indeed the first stages of 3D island formation and are preferentially located close to step edges [29,30] or pits [31,32], since these sites allow the highest strain relaxation and might be characterized by a kinetically determined high adatom concentration [33].

In heteroepitaxial growth, the strain energy caused by lattice mismatch is the driving force for the formation of self-organized 3D islands. As a consequence, if an alloy of epilayer and substrate

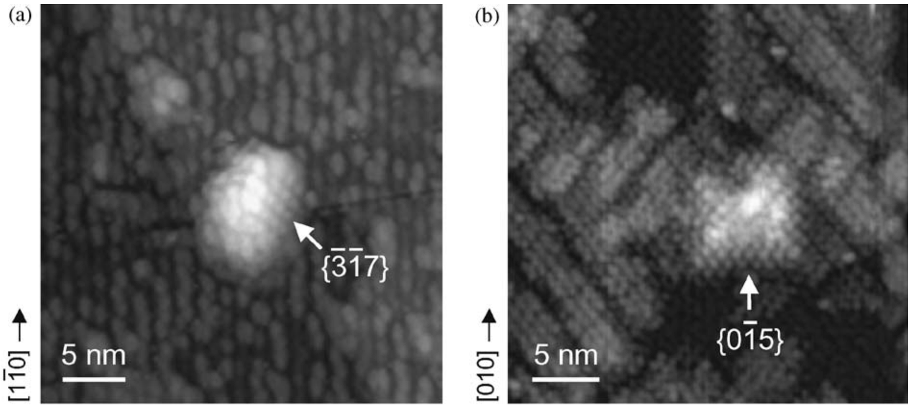

Fig. 4. Partially faceted embryo precursors of 3D pyramid islands in the case of (a) InAs/GaAs(0 0 1) and (b) Ge/Si(0 0 1). Small facets belonging to the {1 3 7} and {1 0 5} families are explicitly indicated.

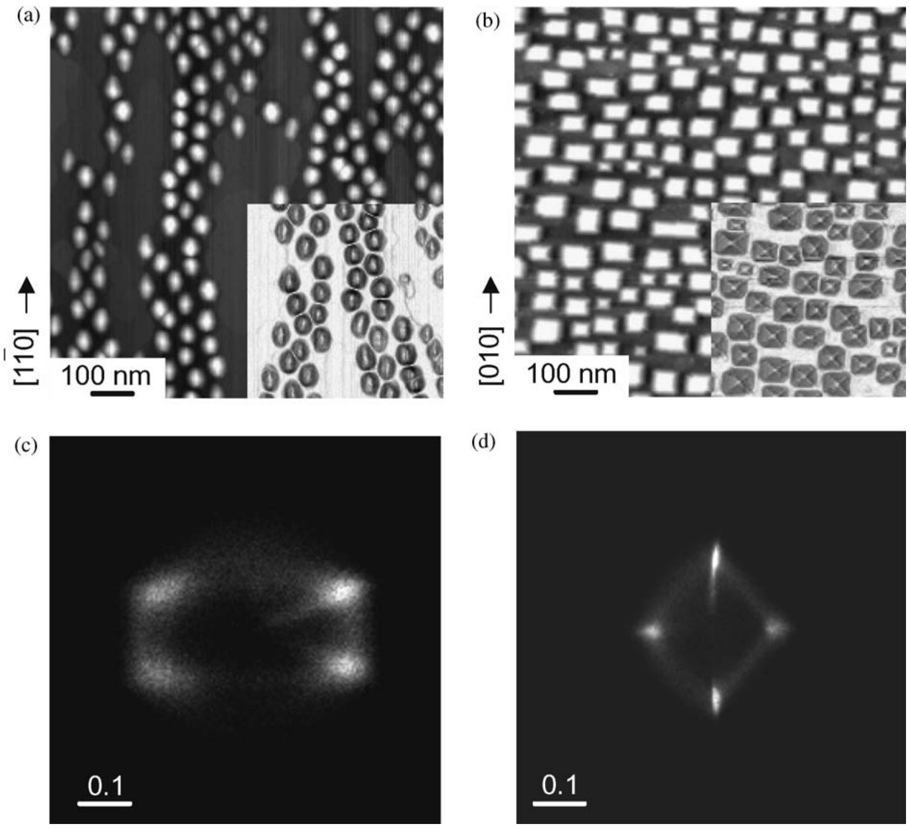

Fig. 5. Upper panel: representative AFM topographies of the surface morphology after the deposition of (a) $In_{0.5}Ga_{0.5}As$ on GaAs(0 0 1) and (b) $Ge_{0.5}Si_{0.5}$ on Si(0 0 1). Lower panel: corresponding facet plots showing that the islands are composed only of {1 3 7} and {1 0 5} facets, respectively.

material is deposited instead of the pure epilayer, one expects a sequence of morphological transitions qualitatively similar to the case of higher mismatch, but happening at higher values of total deposited material. This has been extensively proven for the Ge/Si system [34,35], but the striking analogies we have demonstrated so far, seem to predict its validity also in the InAs/GaAs case. The samples shown in Figs. 5(a) and (b) are obtained by depositing 7.0 ML of $In_{0.5}Ga_{0.5}As$ on GaAs(0 0 1) at 500 °C and 0.02 ML/s and 21 ML of $Ge_{0.5}Si_{0.5}$ on Si(0 0 1) at 600 °C and 0.7 ML/s, respectively. They clearly demonstrate the above conjecture since for both systems the observed islands, although being much larger, have exactly the same shape as the pyramids in the pure epilayer case. This is quantitatively determined by the corresponding FPs in Figs. 5(c) and (d) that show {1 3 7} and {1 0 5} facets, respectively.

4. Conclusions

All the results reported in this work point to the existence of common elementary mechanisms lying at the basis of the growth and evolution of self-organized 3D nano-islands. A theoretical model for the equilibrium shape of strained islands has been recently developed by Daruka et al. [36] and Daruka and Tersoff [37], and its predictions fit in closely with our experimental data. In fact by increasing the islands' volume, a transition from shallow to steep faceted structures is expected, as actually observed in Fig. 1. Moreover the calculated equilibrium shapes (in 2D) nicely agree with the experimentally measured pyramids and domes in Fig. 2. Finally, since the model is essentially material-independent, its predictions have a universal validity, which is demonstrated by the similarities we pointed out between the two main model systems in elemental and compound semiconductors heteroepitaxy.

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