Kinetic Evolution and Equilibrium Morphology of Strained Islands

A. Rastelli,[1,*] M. Stoffel,[1] J. Tersoff,[2] G. S. Kar,[1] and O. G. Schmidt[1]

[1]*Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany*
[2]*IBM Research Division, T. J. Watson Research Center, Yorktown Heights, New York 10598, USA*

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Self-assembled SiGe islands grown on Si(001) leave behind characteristic ''footprints'' that reveal that small islands shrink, losing material to nearby larger islands. The critical size, dividing shrinking from growing islands, corresponds to the pyramid-to-dome shape transition, consistent with ''anomalous coarsening.'' While shrinking, {105}-faceted pyramids transform into truncated pyramids and ultimately into unfaceted mounds. The similarity to behavior during island growth indicates that island shape and facet formation are thermodynamically determined.

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The Stranski-Krastanow (SK) growth mode has been intensely studied as an elegant and convenient method to fabricate defect-free semiconductor ''quantum dots'' [1]. This growth mode occurs during lattice-mismatched heteroepitaxy, leading to the formation of three-dimensional pseudomorphic islands (along with an atomically thin wetting layer). Ge on Si(001) serves as the prototypical system for SK growth, since it is simpler than compound semiconductors, while exhibiting strikingly similar behavior [2].

Yet even for this prototypical system, basic questions remain controversial. A long-standing issue in island growth, and more generally in crystal growth and epitaxy, is the role of thermodynamics vs kinetics in determining the behavior. For quantum-dot islands, this question applies both to the formation and growth of individual islands [3–6] and to the evolution of the ensemble, as reflected in the island size distribution [7–9].

Here we address both aspects of this question. We use a new ''footprint'' technique, analyzing traces left on the Si substrate by the evolving islands. We find compelling evidence that the shapes of individual islands reflect the equilibrium shape for a given size [3,4]. However, the *distribution* of island sizes reflects a kinetic evolution [8]. This is consistent, since equilibration among the ensemble of islands requires diffusion over much larger distances, and hence much longer times, than internal equilibration of a given island.

It is known that observed island shapes can be explained under the *assumption* that they reflect the equilibrium shape for a given island size [4]. However, recent scanning tunneling microscopy (STM) measurements have been interpreted as showing that island formation is kinetically controlled, and, in particular, that the introduction of {105} facets is kinetically limited [5,6]. We find here that islands undergo the same evolution of shape with size while *shrinking* as has been previously observed while *growing* [10,11]. In particular, the {105} facets disappear entirely at a finite island size. Thus, at least for the compositions and temperatures studied here, we find that the island shape and faceting reflect the thermodynamics, and not any kinetic limitations of growth.

Regarding the island size distribution, the pivotal question has been the origin of a bimodal distribution of island sizes and shapes [7]. Both kinetic [8] and equilibrium [7,9] models have been proposed, and both report good agreement between model calculations and experimental measurements. Our results provide a clear independent confirmation of the kinetic picture, in which the islands undergo ''anomalous coarsening'' [8]. We determine that islands smaller than a critical size are in the process of shrinking, while larger islands are growing. Moreover, this

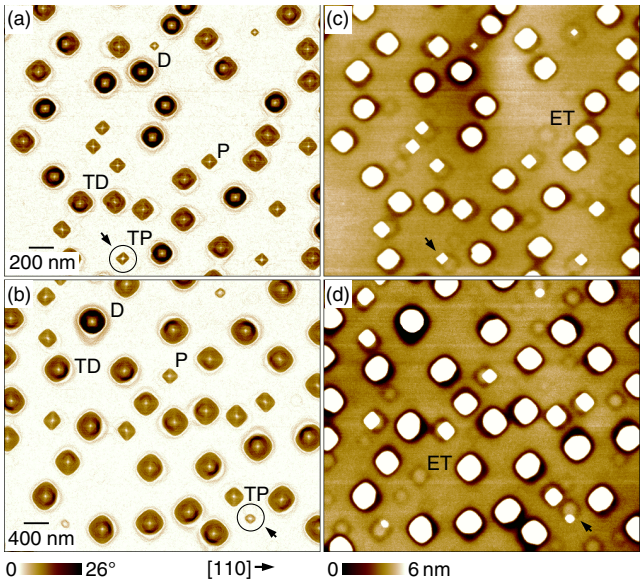

FIG. 1 (color online). AFM images of SiGe islands obtained by deposition of Ge on Si(001) at (a),(c) 740 and (b),(d) 840 °C. Note color scale reflects surface slope in (a) and (b) to highlight shape, and shallow and steep facets can be distinguished. Color reflects height in (c) and (d), with scale chosen to highlight trenches. Islands at different stages of evolution are observed: truncated pyramids (TP), pyramids (P), transitional domes (TD), domes (D). In (c) and (d), empty trenches (ET) are also seen.

critical size is precisely the transition separating pyramid islands from multifaceted "domes." A spatial correlation analysis shows that material migrates from small islands to neighboring larger islands.

The samples studied here were grown by solid-source molecular beam epitaxy. After deoxidation and Si-buffer growth, Ge was deposited at a rate of 0.04 monolayers/s at a substrate temperature T_s between 660 and 840 °C. The samples were cooled to room temperature before atomic force microscopy (AFM) characterization.

Figures 1(a) and 1(b) show AFM images of samples obtained after deposition of 5 monolayers (ML) of Ge on Si(001) at $T_s = 740$ °C and 6 ML of Ge at 840 °C, respectively. In each sample, we observe several different island morphologies: fully developed domes (D); transitional domes (TD), with shape intermediate between pyramids and domes [7,12–14]; pyramids (P); truncated pyramids (TP); and unfaceted prepyramids (PP, not shown in Fig. 1).

In Fig. 1, large islands are typically surrounded by shallow trenches. Such trenches are well known, and occur whenever the growth is performed at high enough temperature [15–18]. Remarkably, there are also "empty" trenches (ET), which do not surround any island [19,20]. These are clearly visible in Figs. 1(c) and 1(d). The most obvious interpretation is that ETs were created by islands which formed, grew, and then shrank and disappeared before the measurement was performed.

In Figs. 1(c) and 1(d) we can also identify islands, such as those indicated by arrows, whose base areas are smaller than the trench area. The trench effectively marks the maximum size of the island, so the smaller size of the island relative to the trench tells us that these islands are *shrinking*. Thus we can distinguish between shrinking and growing islands.

The main difference between samples grown at different T_s is the island size, which increases with increasing T_s, as a consequence of larger Si-Ge intermixing. Because all the samples grown at $T_s \gtrsim 680$ °C are otherwise similar [21], in the following we limit our discussion to the sample grown at $T_s = 840$ °C.

Figure 2 shows detailed statistical information about the size and shape of islands, distinguishing between growing and shrinking islands. In particular, we see that islands larger than $V_c \sim 1.3 \times 10^6$ nm^3 are generally growing, while islands smaller than V_c are shrinking. This separation is indicated by a vertical dashed line in Figs. 2(a)–2(d).

These results serve to rule out models in which pyramids and domes are in equilibrium with each other [7,9], at least for the regime studied here. Pyramids and smaller islands

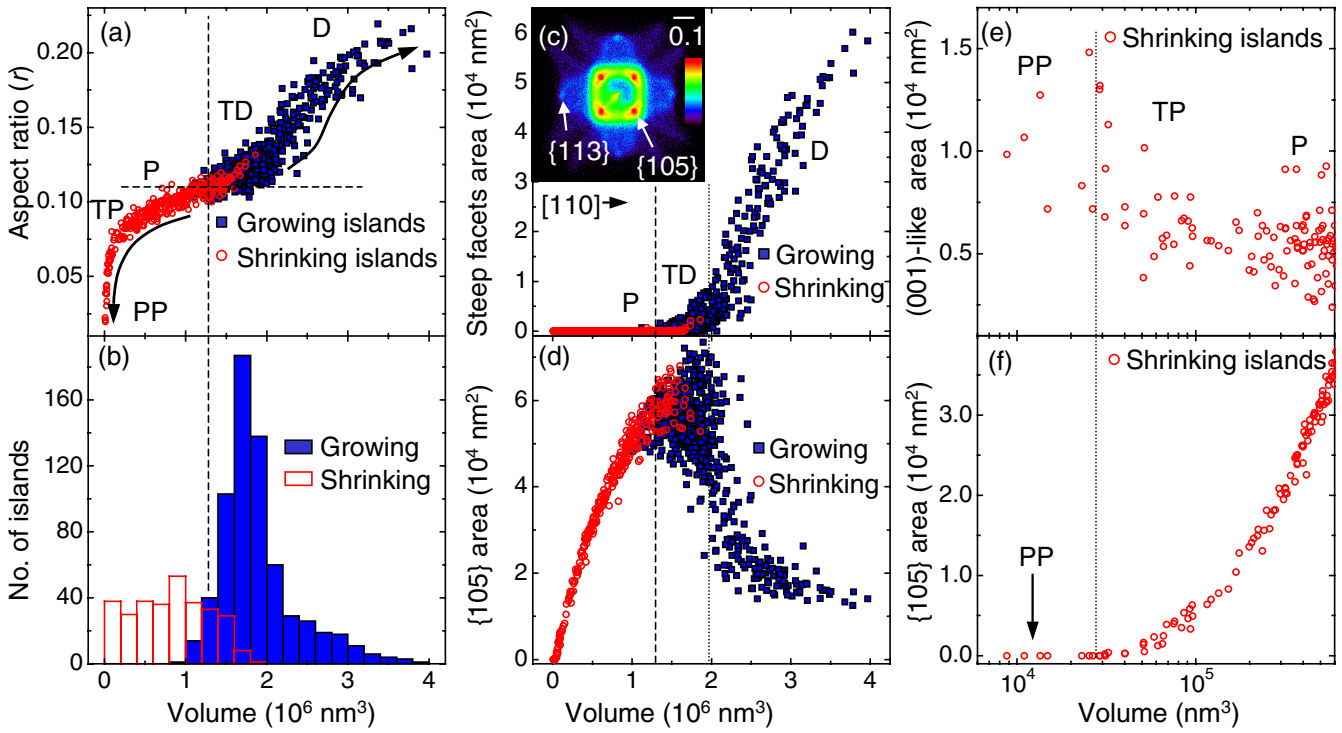

FIG. 2 (color online). Statistical analysis of islands contained in the sample grown at 840 °C: (a) Shape (aspect ratio r) vs volume and (b) histogram of volumes. The area of (c) steep, (d),(f) {105}, and (e) shallow facets vs volume were computed from the "facet plot" [22] (with logarithmic intensity) shown in the inset of (c). They show that prepyramids (PP), truncated pyramids (TP), and pyramids Ps are shrinking, while TDs and domes (D) are growing, and that the critical size distinguishing shrinking and growing islands corresponds to the P-TD shape transition.

are in the process of shrinking, and are clearly unstable with respect to domes.

To understand this, we analyze the island shapes in more detail. Pyramid islands consist primarily of {105} facets, while dome islands are composed largely of steeper facets. As indicated by the inset in Fig. 2(c), we can clearly distinguish these different facets, and in Figs. 2(c) and 2(d) we plot the island facet areas vs volume. (See Ref. [22] for details on the facet-area calculation.)

We find that V_c corresponds to the P-TD transition, where facets steeper than {105} are first introduced. This corresponds exactly to the proposed anomalous coarsening process [8], because the P-TD transition is a first-order transition marked by a discontinuous change in island chemical potential with volume [3,23]. (Normal coarsening is driven by the much weaker effect of a smoothly varying chemical potential.) The {105} facets also begin to shrink at V_c, but it is the introduction of new facets that marks the first-order transition. (In contrast, the evolution from TD to D or from TP to P is continuous and not a true transition in this sense [23].)

We now turn our attention to the smallest islands, TPs and PPs, which are in the process of shrinking but have not yet disappeared. Much work has addressed the initial stages of island formation and growth, and by studying the complementary process of shrinking and disappearing, we can shed light on the role of kinetics vs thermodynamics. (Shrinking islands have been discussed previously [11], based on the *assumption* that small islands are shrinking; but here we actually determine experimentally which islands are shrinking.)

A sequence of representative shrinking islands is shown in Fig. 3, with color scale enhancing the local slope. TPs are easily distinguished from Ps or PPs by the presence of a dark region corresponding to {105} facets, along with a lighter region at the island top, corresponding to orientations close to the (001). In contrast, Ps are composed almost entirely of dark {105} facets, and PPs show no {105}-faceted areas at all (within the resolution of our measurement). STM measurements confirm these results [24].

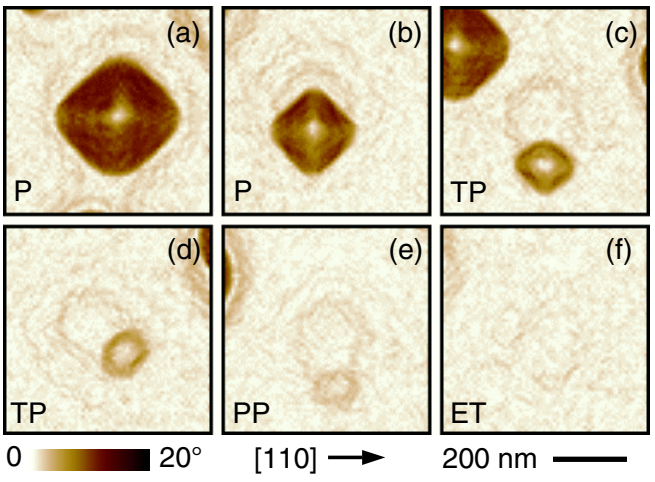

FIG. 3 (color online). Sequence of AFM magnifications illustrating shrinking pyramids and the pyramid-to-prepyramid transition occurring with decreasing volume during the ripening process. Images have color scale according to local surface slope with respect to the (001) plane.

To quantify the evolution of these shrinking islands, we plot in Fig. 2(e) the area within each island having inclination less than 5° with respect to the (001) plane, and Fig. 2(f) shows the {105} facet area for each island. As the islands shrink, the unfaceted "(001)-like" area actually increases. More importantly, the {105} facet area decreases smoothly to zero, with {105} facets disappearing at a critical volume of about 3×10^4 nm^3.

It has been proposed that the development of {105} facets is a continuous process associated with "kinetic constraints" during growth [5]. This was based on STM measurements for lower-temperature growth at a lower Ge fraction, where, indeed, kinetic constraints are more likely. Here, we can clearly rule out such an interpretation based on Fig. 2(f). The kinetic constraints for introducing {105} facets would not apply to the disappearance of these facets. Yet we see that, even for shrinking islands, the {105} facets disappear below a critical size. Evidently, based on the similar behavior seen for growing [10,11] vs shrinking islands, the presence or absence of {105} facets is determined by thermodynamics rather than kinetics.

The majority of shrinking islands sit at or near the trench. Since the trench marks the position of the island prior to shrinking, we see that shrinking does not occur symmetrically, but is consistent with the preferential removal of material from one side of the island. This is natural if the shrinking is due to coarsening, since material would flow preferentially to the closest large neighbor.

To test this, we plot in Fig. 4 the spatial correlation between the position of shrinking islands and their environment. In the inset of Fig. 4(b), we indicate the position of the island center with A and that of the trench center with B. We then denote with C the center of the island next to B, which is larger than the considered shrinking island. We measure the angle α between the segment $\overline{AB}$ and the [110] direction and compare it with the angle β between the segment $\overline{CB}$ and the [110] direction.

If material migrates from the smaller island towards this nearest large neighbor as a result of its lower chemical potential, the angles α and β should be the same. Indeed, in Fig. 4(a) the data cluster around the line $\alpha = \beta$, and the distribution of $\alpha - \beta$ in Fig. 4(b) is strongly peaked around $\alpha - \beta = 0$. This provides further direct support for the model of anomalous coarsening of the island distribution.

In conclusion, we have used a new footprint technique to study the evolution of Ge islands grown on Si(001) at a relatively high temperature. The analysis of these footprints offers a new method to study the evolution of strained islands. We expect this to be relevant to other

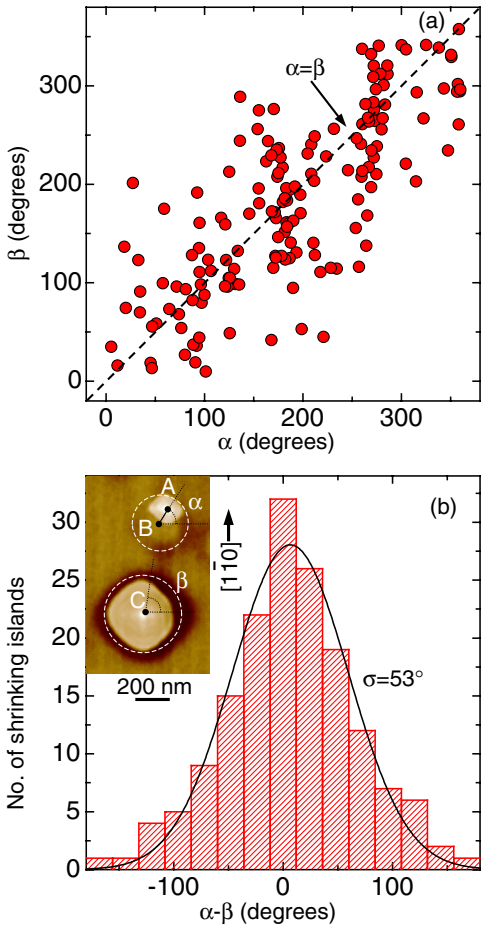

FIG. 4 (color online). Statistical analysis of the correlation between the position of shrinking islands with respect to the surrounding trenches and the environment. The inset of (b) shows the definition of the angles α and β on top of an AFM image of an island, which shrinks because of material transfer to the neighboring large island.

phenomena and material systems. Using this technique, we observe that SiGe {105}-faceted pyramids transform into partially faceted truncated pyramids and eventually to unfaceted prepyramids, as they shrink in a coarsening process. This sequence represents the inverse of that previously observed during growth and is consistent with the predictions of thermodynamic models. A spatial correlation analysis suggests that material migrates from shrinking islands to neighboring steeper islands.

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*Electronic address: A.Rastelli@fkf.mpg.de

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