Barrierless Formation and Faceting of SiGe Islands on Si(001)

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The initial stages of the formation of SiGe islands on Si(001) pose a long-standing puzzle. We show that the behavior can be consistently explained by one simple assumption—that for strained SiGe, (001) is a stable orientation but not a facet orientation. Calculations of energy and morphology reproduce the key features of "prepyramid" and "pyramid" islands, and explain the initial formation and subsequent shape transition. Scanning tunneling microscopy measurements confirm the key assumptions and predictions of the model.

Ge and SiGe on Si(001) have been widely used as model systems for understanding heteroepitaxy. In particular, misfit strain drives the formation of epitaxial islands, and there is wide interest in exploiting such "self-assembled quantum dots" in nanoscale technology [1]. However, this system exhibits many remarkable features—as a model system, it suggests that heteroepitaxy is surprisingly complex. Ge islands were first observed by Mo *et al.* [2] as (105)-faceted rectangular pyramids. A rich body of subsequent work showed that, in equilibrium, small islands are square pyramids, while larger islands develop a more complex multifaceted shape [3] after passing through a first-order shape transition [4,5].

Even this picture proved incomplete, as two key aspects came to light regarding the very earliest stages of island formation. First, using scanning tunneling microscopy (STM), Vailionis *et al.* [6] observed "prepyramids," tiny islands that appear to be precursors to the well-studied pyramids. This confirmed an earlier observation by Chen *et al.* [7] using atomic force microscopy. However, the nature of these islands, and their role in the growth process, has been unclear. Second, recent experiments by Sutter and Lagally [8] and by Tromp *et al.* [9] showed that, at least in some range of temperature and alloy composition, islands can evolve continuously from surface ripples. But if islands are strictly faceted, they should form by thermally activated nucleation [10].

We find that the behavior of SiGe islands on Si(001) can be consistently explained by one simple assumption—that for strained SiGe, the surface-energy anisotropy allows all orientations near (001), with the first facet being (105). With this anisotropy, we predict that tiny prepyramid islands form with no nucleation barrier. They are unfaceted, with low height/width ratio and indistinct edges. As islands increase in size, they undergo a transition in which (105) facets are introduced discontinuously at the steepest point on the island. This first-order shape transition is expected to drive anomalously abrupt coarsening [4] and the rapid disappearance of any nearby prepyramids. Our experimental observations confirm the nature of the prepyramids and of the faceting transition, as well as the key assumption of the model.

Models of heteroepitaxy and island formation have generally assumed either an unfaceted or a fully faceted morphology. (Some models invoke steps, which are discussed below.) In contrast, we propose that at typical growth temperatures, the SiGe "effective" equilibrium crystal shape (EECS) has a smoothly curved region near (001) that meets the (105) facet at a sharp edge. We say effective ECS because the surface energy may be substantially affected by misfit strain [11]. For the islands considered here, the elastic relaxation is at most a modest fraction of the misfit strain, so it is reasonable to use the surface energy $\gamma(\theta)$ evaluated at the full misfit strain and neglect variations in γ due to spatially varying strain. Since misfit strain is proportional to alloy composition, there might be an important dependence of γ on alloy composition. In addition, the anisotropy of γ may be sensitive to temperature. Here we assume a fixed temperature and misfit, and neglect possible variations in alloy composition.

For the present analysis, we use a two-dimensional (2D) model for simplicity, with the island periodically repeated [12]. The important point here is not that (105) is a true facet, but only that the surface is smooth near (001), with a range of "missing angles" between (001) and (105). We therefore replace the expected $\gamma(\theta)$ with the convenient form

$$\gamma(\theta) = \bar{\gamma}[1 - \alpha \cos(n\theta)]. \tag{1}$$

Then the EECS has no true facets in our model, but for large enough α it has smoothly curved regions meeting at finite angles. We use $n = 32$ to give regions with average orientation at $\theta = 0$ and $\theta = \pi/16 \approx 11°$. We refer to the

region around 11° as a "facet," in analogy to the (105). To avoid singular behavior at the edge, we introduce a term in the energy that rounds the corner slightly [13,14].

The chemical potential (per unit volume) is

$$\mu = S + \kappa\Gamma(\theta) + \beta C(\kappa), \qquad (2)$$

where S is the strain energy density at the surface, $\Gamma(\theta) = \gamma + \gamma''$ is the surface stiffness, and κ is the surface curvature. $C(\kappa)$ is the corner-energy function from [14], and β is the corner-energy parameter. For a prescribed island volume, as in [15] we calculate the island shape that satisfies constant μ over its surface, with a boundary condition of zero contact angle (valid for "Stranski-Krastonow" growth [15,16]). We eliminate all parameters other than α and β by quoting results in units of the strain energy density S_0 of a planar film, and a characteristic length $L = \Gamma(0)/S_0$. For the calculations here, we use $\alpha = 0.0015$ and $\beta = 5 \times 10^{-5}L^3 S_0$.

The calculated island shapes at different island sizes are shown in Fig. 1; the island energy and chemical potential are shown in Fig. 2. The smallest islands have a smooth, cosinelike shape [15]. With increasing size, the maximum angle [relative to (001)] increases. However, there is a maximum stable angle, determined entirely by the surface-energy anisotropy [13,17]. In the figures, V_3 denotes the volume at which the maximum angle reaches this stability limit. This corresponds to the largest possible unfaceted island—for $V > V_3$, the only stable shape is a faceted island with a rounded top and base. For larger islands, the rounded top and base regions continue to
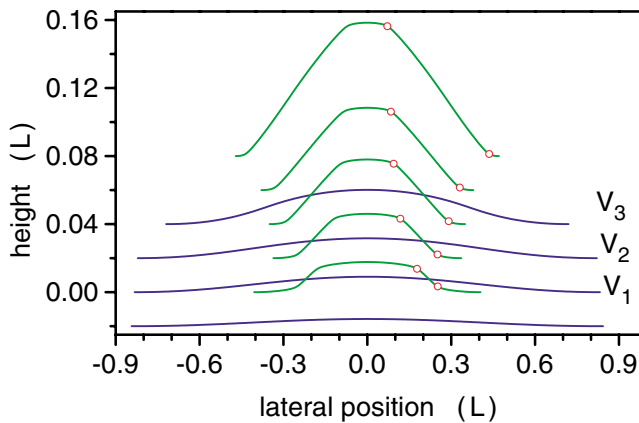
shrink, leading to a simple pyramidal appearance (not shown). At even larger sizes, there is another shape transition in which even steeper facets are introduced.

To understand the stability and the general behavior more fully, we refer to Fig. 2. In the size range of interest here, there are two distinct branches of stable (or metastable) solutions, corresponding to small smooth islands and "faceted" islands. These are connected by a branch of unstable "saddle-point" solutions. At the volume labeled V_2, the two curves cross, and the shapes are degenerate in energy. Thus, in equilibrium, there is a first-order shape transition at volume V_2. Such shape transitions have been studied previously for fully faceted islands [4,5], and they have important consequences for island growth [4]. In particular, they may play a key role in growing islands of uniform size for use as quantum dots.

From Fig. 2, there is no energy barrier to nucleate an island—the energy is a monotonically decreasing function of size, even for arbitrarily small islands. A growing island remains stable and unfaceted up to size V_2. At that point, it becomes metastable, and in equilibrium it transforms to a faceted shape. The calculated stable shapes at this size are superposed in Fig. 1. However, there is an energy barrier for this first-order transition, so the island may still grow continuously, remaining unfaceted. The energy barrier is the distance to the unstable branch in Fig. 2, and this decreases with increasing size, increasing
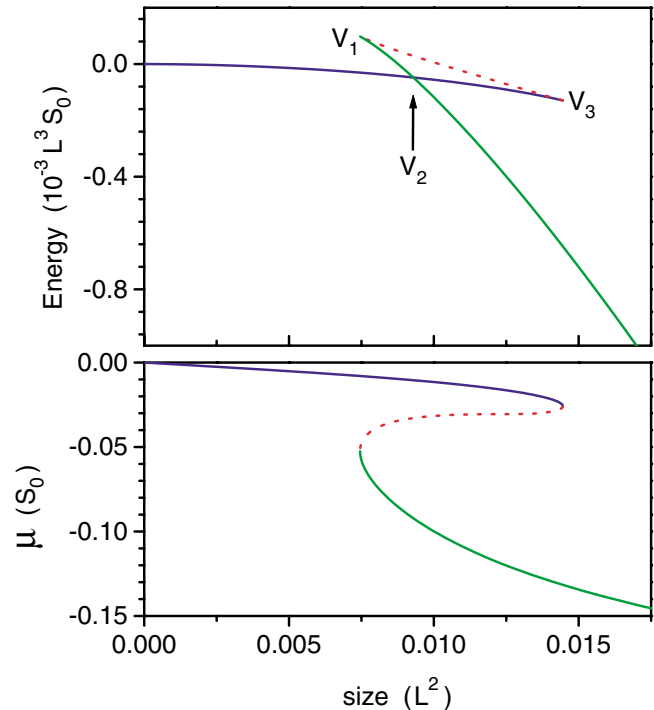

FIG. 1 (color online). Equilibrium island shapes, calculated for islands of sizes 0.0035, 0.0074 (V_1), 0.0094 (V_2), 0.0144 (V_3), 0.0200, and 0.0401 L^2, from bottom to top respectively. Shapes for different size V are offset vertically by 0.02L. When there are two (meta)stable shapes for the same V, shapes are superposed, and facet edges on the right are indicated by open circles. V_1 is the size of the smallest metastable faceted island, V_3 is the size of the largest metastable smooth island, and both shapes are stable and degenerate in energy at V_2. The substrate is not shown; island edges meet the substrate at zero contact angle.

FIG. 2 (color online). Island energy E and chemical potential (per volume) μ, relative to a planar film, vs volume V. The dotted line represents unstable solutions connecting smooth and faceted branches.

the likelihood that the island will transform via a thermally activated process of facet nucleation. The energy barrier shrinks to zero as the size reaches V_3, and the unfaceted island becomes unstable.

Thus, even if the shape transition is hindered by the nucleation barrier, it will always occur at a size between V_2 and V_3 (or, for a shrinking island, between V_2 and V_1). (We neglect the finite time over which the transition occurs, relative to the growth time or the time to initiate the transition.) Whenever the transition occurs, even at V_2, the chemical potential drops discontinuously (Fig. 2), leading to anomalous coarsening [4].

We can corroborate the key assumptions and predictions by direct comparison between theory and experiment. We have deposited $Si_{1-x}Ge_x$ layers on Si(001) at a substrate temperature of 600 °C, in a composition range x between 0.4 and 0.5, and studied them by room temperature STM. For details on experimental methodology, see Ref. [18].

First, we consider very small islands, which only have orientations near (001). According to our model, the shape should be cosinelike, meeting the substrate at zero contact angle [15,16]. Figure 3 shows an STM image of a small island, together with a line scan through the middle of the island. Aside from atomic-scale roughness, the line scan is very well fitted by a cosine, as predicted.
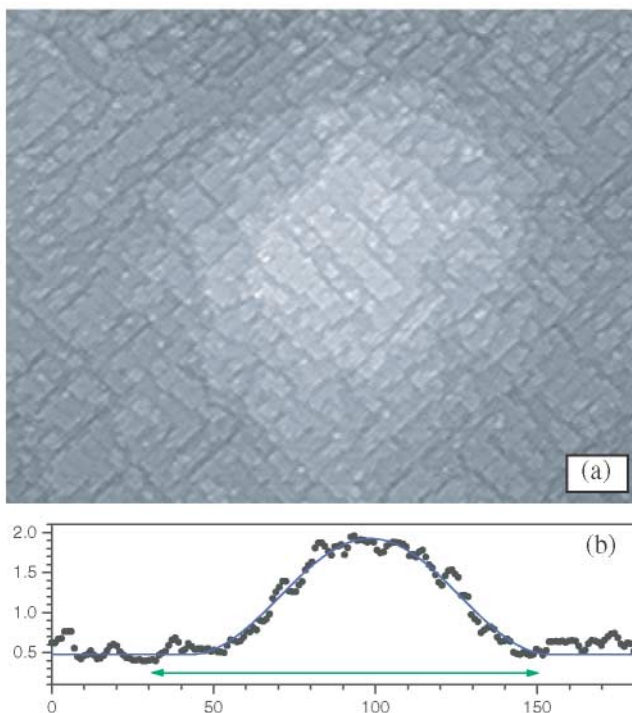

FIG. 3 (color online).   (a) STM image (114×75 nm^2) of a small island obtained by depositing $Si_{0.5}Ge_{0.5}$ on Si(001). (b) Line scan through island (dots) and cosine fit (line). Scan extends beyond the STM image, and a double arrow indicates the range of the image. Scale is in nm.

The position of the island edge is obscure in the image, precisely because it meets the substrate at zero contact angle.

A smoothly curved region on the ECS is generally associated with thermal roughness; the island in Fig. 3 clearly exhibits atomic-scale roughness. While our images are taken after the sample is cooled, the observed roughness is consistent with the hypothesis that SiGe on Si is thermally rough at the growth temperature, for orientations near (001). There is, in fact, evidence for a roughening transition of epitaxial SiGe on Si(001) in the range of typical growth temperatures [19]. This suggests that at lower temperature the (001) might be a facet orientation, in which case island formation would presumably occur by a completely different mechanism, such as nucleation [10] or step accumulation [20].

For fixed temperature, barrierless island formation is apparently suppressed at lower Ge fraction [8,9]. Ge has a lower melting point than Si, and presumably a lower roughening temperature, so the effect of lower Ge fraction might be simply to raise the roughening temperature above the temperature of the experiment. A lowering of the roughening temperature could also be tied to the compressive stress, rather than to the composition *per se*. Xie *et al.* [21] proposed that the compressive stress lowers the formation energy of steps. Thus, larger Ge fraction, by increasing the stress, would lower the step formation energy and, hence, lower the roughening temperature. Also, the surface reconstruction changes with Ge fraction as a result of the compressive stress [22]. This must change the step formation energy and the surface-energy anisotropy. The sensitivity to surface properties also suggests a likely explanation for the action of surfactants [23].

With increasing island size, we have observed that small facets first appear in the region of maximum slope on the prepyramids. Figure 4 shows a somewhat larger island, in which substantial (105) facets have formed. There is a rounded (atomically rough) region at the top, which meets the (105) facets at a sharply defined edge. For still larger islands, the rounded top region shrinks in size, leading to a pyramidal shape. All of these observations are in accord with the predictions of our model, as seen in Fig. 1.

Some models of island formation invoke a stepped morphology. However, thermodynamically well defined steps exist only on facets. From Fig. 3, it seems clear that small islands do not involve step mounds on a (001) facet, but rather a continuous deformation of an unfaceted surface, where steps are not well defined. For larger islands, the (105) is not a staircase of steps, but rather a true facet. In Fig. 4 a step is visible on the right facet, emphasizing the point that the facet itself is not composed of steps.

We can also compare with a number of published experiments. Vailionis *et al.* [6] observed prepyramids only within a narrow range of widths. This is consistent
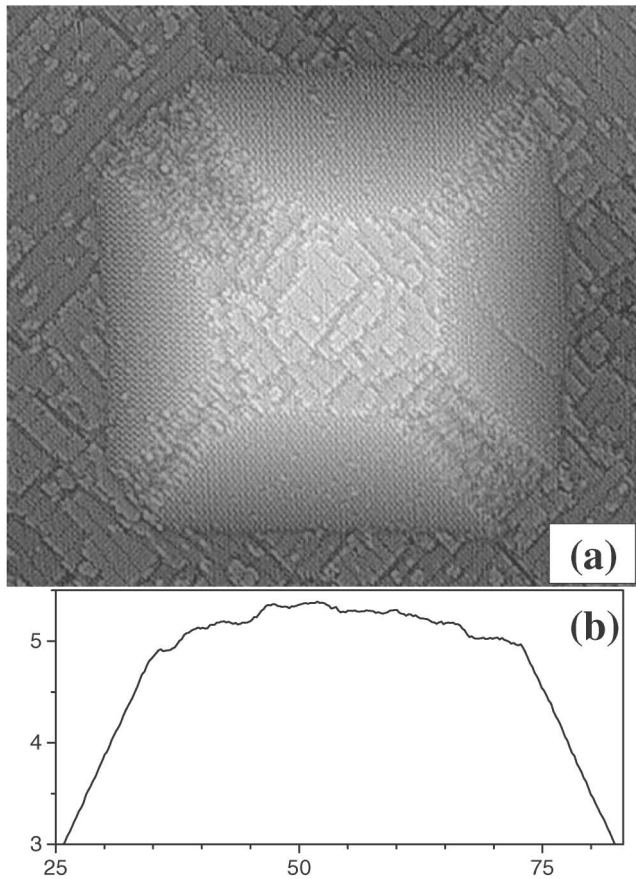
FIG. 4.   (a) STM image (108×108 nm^2) of a faceted island obtained by depositing $Si_{0.6}Ge_{0.4}$ on Si(001). (b) Cross section of a region near the top of island, from an average of 30 line scans taken from left to right close to the middle of the island. Scale is in nm.

with the predictions of our model. In fact, for the parameters used here, the width of unfaceted islands changes only 14% in going from the smallest islands (which have finite width but vanishing height [15]) to the largest (meta)stable unfaceted islands (V_3 in Fig. 2). Using low energy electron microscopy, two groups [8,9] observed formation of islands from continuous growth of ripples. This is exactly what we would predict if growth is fast enough to "outrun" the initial instability, so that a continuous layer forms initially.

In conclusion, we have presented a unified picture of the initial stages of SiGe island formation on Si(001). A broad range of experimental observations can be explained by a simple assumption about the surface-energy anisotropy. However, the anisotropy may vary significantly with temperature, alloy composition, and misfit. Thus, there may be qualitative changes in the growth as these parameters are varied.

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