Self-Limiting Growth of Strained Faceted Islands

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We consider the growth of facets associated with coherently strained semiconductor islands. Surprisingly, the island growth rate is found to rapidly self-limit, which has important consequences for island size distributions. A new explanation for the elongation of strained faceted islands is proposed as a natural consequence of facet growth kinetics. [S0031-9007(98)06262-0]

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Mechanisms of facet formation and growth are long-standing issues in surface physics and materials science [1]. In particular, faceting governs many key processes in crystal growth and etching [2]. More recently, it has been discovered that facets also play a central role in the growth of coherently strained semiconductor islands [3–7]. Here the situation is particularly intriguing from the growth physics perspective because of the spatial variation in strain across the surface of the faceted island. Despite the fact that coherently strained semiconductor islands are presently receiving considerable attention as a means of fabricating quantum dot devices [8], insight into the facet growth mechanisms has remained limited.

In this Letter we identify two surprising consequences of strained facet growth which dramatically influence island growth kinetics. First, we demonstrate that the island growth rate decreases rapidly with increasing island size. This implicates an important role of faceting as a means of inducing self-limiting growth and narrowing island size distributions, even in low misfit systems. Second, a shape instability of strained islands arises as a natural consequence of facet growth which provides a new explanation for the origin of elongated hut cluster shapes observed in strained layer epitaxy [3].

Consider the generic model of facet growth illustrated in Figs. 1(a) and 1(b). A pyramidal island of half base length s is bounded by four facets inclined at an angle θ to the surface. The three-dimensional island grows via the nucleation of two-dimensional islands of height a on the facet surfaces [Fig. 1(b)]. Such an embryo is shown originating from the bottom left hand corner of the facet in Fig. 1(a) which, as discussed later, is an energetically favorable nucleation site. We assume that the embryo shape is dominated by surface energy considerations such that the step energy γ_β is a minimum for a direction in the facet plane, inclined at an angle φ to the base of the island. This is a good approximation as long as gradients in the surface elastic energy density are small compared with the anisotropy in step energy. Details of the critical nucleus shape will, however, not influence the important qualitative features of the model.

We model the growth of strained facets as a direct transformation between the planar strained film and the embryo, which is strictly valid in the limit of zero supersaturation (i.e., annealing of the planar film in the absence of deposition). However, the analysis will also apply to deposition if the supersaturation is very small, which is true for many situations of practical significance [9]. If the adatom diffusion length is large on the scale of the spatial extent of the island strain field then the energy to form an embryo of size ℓ is

$$\Delta G = a\ell\Gamma + a\int_{\text{embryo}} \xi(\mathbf{R})\, d\mathbf{R}\,, \qquad (1)$$

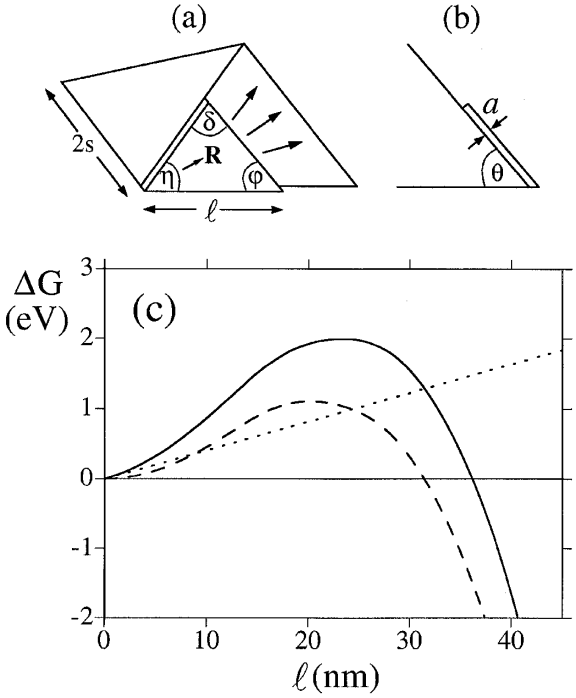

FIG. 1. (a) Schematic representation of strained facet growth. An embryo emerges from the bottom left hand corner of the facet and expands across the facet with the geometry shown. (b) The embryo of height a increments the basal dimension of the island as shown in cross section. (c) Formation energy of the embryo ΔG as a function of its size ℓ (solid line) for the geometry shown in (a). The first and second terms of Eq. (1) are represented by the dotted and dashed lines, respectively. The misfit is 2% and $s = 40$ nm. Elastic constants used in the calculations are Young's modulus $E = 102$ GJ/m^3 and Poisson's ratio $\upsilon = 0.27$. We assume a {105} facet geometry with $\theta = 11.3°$, $\varphi = 51.5°$, $a\csc\theta = 5.54$ Å, and $\Gamma = 3.8$ meV/Å^2.

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where $\Gamma = \gamma_f \sin\varphi \csc\delta + \gamma_\beta \sin\eta \csc\delta + \gamma_f \cot\theta - \gamma_s \csc\theta$ if $\ell \gg a$. The surface energies γ_f and γ_s correspond to the island facet and substrate (or wetting layer) energies, respectively, so that the first term simply reflects the additional surface energy created [10]. The second term involves the elastic relaxation of the material comprising the embryo where $\xi(\mathbf{R})$ is the elastic energy density at the surface of the embryo minus the elastic energy density of the planar strained layer. $\mathbf{R}$ is a two-dimensional vector in the facet plane [Fig. 1(a)].

To calculate the elastic energy change in creating the embryo, we evaluate the elastic energy density at the surface of a pyramidal island by finite element analysis (FEA) [11]. This is also a good approximation to the surface elastic energy density of the pyramid plus embryo, provided that the lateral dimensions of the critical nucleus are appreciably greater than its height a (i.e., we neglect self-relaxation of the embryo). Using this approximation, we then integrate $\xi(\mathbf{R})$ over the surface of the embryo and evaluate the total energy change ΔG using Eq. (1) as a function of the embryo dimension ℓ, as shown in Fig. 1(c). Clearly, the energy increases initially and reaches a maximum value ΔG^* at a critical nucleus size ℓ^*. Hence there is an energy barrier to complete the strained facet.

The physical origin of this barrier can be deduced from the finite element calculation of the elastic energy density displayed in Fig. 2(a) [12]. The elastic energy density of the planar film is 71 MJ/m^3 which corresponds to the dark pink shading in the figure. Material in the proximity of the island peak is therefore relaxed relative to the planar film but a significant stress concentration occurs at the base of the pyramid, extending from the red to the yellow regions. To grow the facet it is necessary to cover the surface with a monolayer of material which is initially strained to the dark pink level of the planar film. In order to cover the highly strained levels close to the base, this material has to be further compressed to values between the red and yellow levels. This will initially cost energy until the embryo can expand into more relaxed regions of the surface. Eventually, this reduction in elastic energy dominates the positive surface energy contribution in Eq. (1), as shown in Fig. 1(c), which is the physical origin of the energy barrier.

The energy barrier to complete the facet is therefore directly associated with the strain energy distribution on the island surface. In this regard, the apex region of the triangular facet is also a particularly favorable nucleation site since here the material is significantly relaxed compared with a corner close to the base [Fig. 2(a)]. However, Eq. (1) implies that the positive strain energy gradient away from the apex will facilitate the formation of a partially complete, metastable layer, which is not seen experimentally [3]. We attribute this to the increased value of Γ (and hence ΔG^*) associated with embryo formation at the top of the facet since, unlike basal corner nucleation (Fig. 1), it is not possible to gain energy by covering a portion of the substrate. Apex nucleation will become increasingly likely for higher aspect ratio islands associated with greater apex relaxation and basal stress concentrations.

The growth rate of strained faceted islands depends on both the frequency at which 2D nuclei form on the surface (nucleation limitation) and the rate at which they expand to cover the surface (growth limitation). In the latter case, the basal stress concentration in Fig. 2(a) can increase the energy barrier for adatom diffusion [13]. This limits the island growth rate by restricting the flow of adatoms to the 2D nuclei on the island surface. Island growth is, however, nucleation limited if the time taken for the new

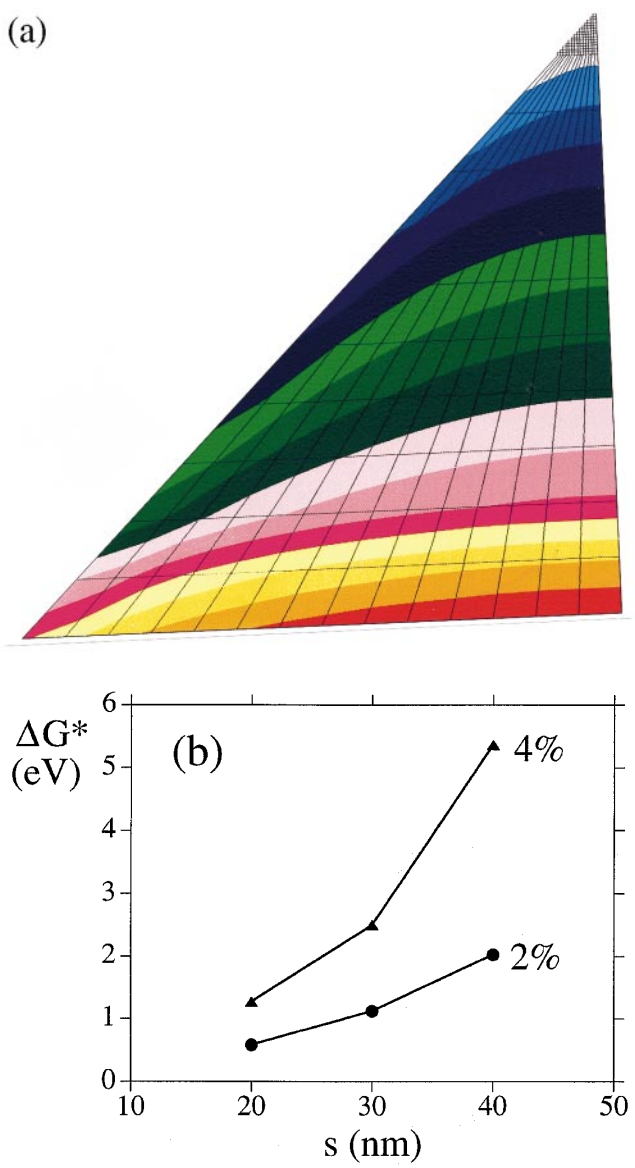

FIG. 2. (a)(color) Elastic energy density associated with half the triangular facet of a pyramidal island ($s = 40$ nm) at 2% misfit. The elastic energy density is represented by a color scale which decreases linearly from 96 MJ/m^3 (red level) at the base to 18 MJ/m^3 (white level) close to the peak. (b) Energy barrier to complete the facet ΔG^* as a function of island size s and misfit strain. Other parameters are listed in the Fig. 1 caption.

layer to cover the facet is small compared with the period between successive nucleation events.

Scanning tunneling microscopy studies of Ge/Si(001) growth do not show partially complete layers on {105} facets indicating that, after an initial nucleation event, the freshly nucleated layer completes rapidly [3]. Furthermore, the island number density was found to increase significantly with increasing Ge dose while the islands grow only slowly [3]. These observations strongly support nucleation limited growth and we now consider the important implications of this mechanism for island growth kinetics.

The magnitude of the stress concentration at the base of the island increases with increasing misfit strain. Furthermore, the spatial extent of the stress concentration increases as the islands grow larger. The energy barrier to complete the facet will therefore also increase with increasing s and misfit strain as shown in Fig. 2(b). Since the island growth rate $R \propto \exp(-\Delta G^*/kT)$, this implies a strong self-limiting growth effect in which large islands will grow more slowly than smaller islands. Smaller islands must then catch up to the larger islands in size which will narrow the island size distribution.

Unusually narrow island size distributions have now been observed for several different materials systems [14] and, in some cases, this has been attributed to specific models of self-limiting growth which do not require faceting [13,15]. Such models are particularly applicable to high misfit systems ($>4\%$) in cases where self-limiting growth sets in before well-defined facets form. The interesting feature of our model is the appreciable magnitude of the self-limiting effect which is present for faceted islands, even at relatively low misfits. This is because an embryo effectively integrates over the elastic energy of the facet surface and hence magnifies the effect of misfit strain.

To investigate this possibility experimentally we have annealed 2 nm thick $Si_{0.5}Ge_{0.5}$ alloy layers on Si(001) at 590 °C for 6 min. The layers were previously deposited at the relatively low temperature of 400 °C to ensure a nominally planar surface. The small temperature gradient of 20 ± 5 °C from the center to the edge of the wafer, as measured by NiCr-NiAl thermocouples, leads to a significant variation in the number density of square-based islands, consistent with thermally activated nucleation [16]. However, an increased size uniformity is observed in the hotter region as revealed by atomic force microscopy measurements of island size distributions in Fig. 3. The largest islands are bounded by {105} facets while the smaller islands have lower aspect ratios [17]. Interestingly, the maximum sizes in both distribution curves are almost unchanged, consistent with growth by strained facets. These observations demonstrate that, in the presence of faceting, narrow island size distributions are attainable even for low misfit systems. A bimodal island size distribution is observed on annealing the sample at a much higher temperature ($\geq$650 °C), indicating that the size uniformity attained during our annealing process is principally a kinetic effect. Now, all of the original planar film is transformed, and transitions to higher order facets [5,18] and coarsening mechanisms [19] dominate.

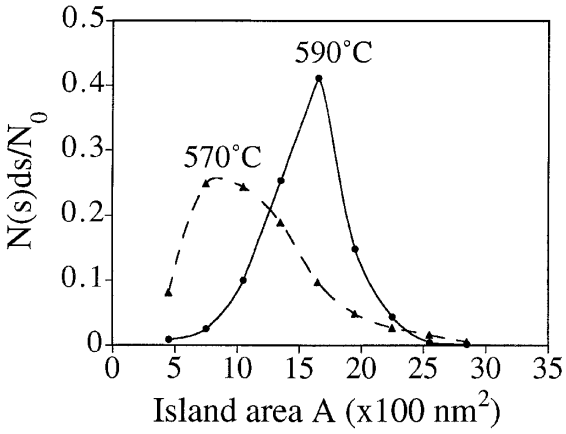

FIG. 3. Island size distributions corresponding to regions of a 2 nm thick $Si_{0.5}Ge_{0.5}$ alloy film on Si(001) annealed at 570 and 590 °C for 6 min. $N(A)dA$ represents the number of islands with projected areas between A ($= 4s^2$) and $A + dA$. N_0 is the total number of islands surveyed.

We now consider the remarkable phenomenon of island shape elongation during strained layer epitaxy, which was first reported by Mo *et al.* for Ge deposition on Si(001) [3]. Ge islands were found to elongate along elastically soft ⟨100⟩ directions, giving rise to so-called "hut cluster" shapes with rectangular rather than square bases (Fig. 4). An energetic explanation for the elongation of isolated islands of fixed height has already been proposed [20]. Here we suggest an alternative explanation which is a natural qualitative development of our facet growth model.

If the growth of a square-based pyramidal island is limited by the nucleation of new layers on the facets, elastic interactions with neighboring islands, steps [3,6,21], or pits [22] will produce a variation in energy barriers across the island. Consider, for example, the reduction in basal stress concentration due to a [100] step of height 1.36 Å located 1 nm from an island ($s = 20$ nm). This can be obtained by superimposing the strain field due to a 4% discontinuity in misfit strain at the step [23] on the island strain field calculated by FEA. Integrating the embryo over

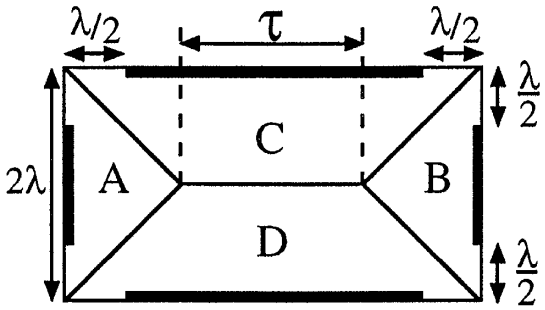

FIG. 4. Plan-view schematic of a hut cluster with a rectangular base. Shaded regions at the base of the facets represent areas of high stress concentration (see text).

the modified surface elastic energy distribution produces a 0.1 eV energy barrier difference between facets, leading to a factor of 5 in growth anisotropy at 500 °C. Although this calculation allows for no relaxation of the remainder of the facet, it provides a lower limit to the influence of the interaction.

Once an island is elongated, as in Fig. 4, the time for a freshly nucleated layer to complete extended facets such as C and D becomes increasingly significant compared with the time between nucleation events. This is because the basal stress concentration, which can act as a barrier for adatom diffusion [13], restricts the growth of extended facets (C, D) more than the short facets (A, B). For elongated islands, FEA shows that the stress concentration falls off over a characteristic length $(\sim\lambda/2)$ toward the basal corners. This is represented schematically in Fig. 4 where the shading corresponds to highly strained regions at the base of the facets [cf. the red level in Fig. 2(a)], which also extend a small distance onto the surface of the planar film. In the limit where this region acts as a perfect barrier for adatom diffusion and other regions have a negligible effect on diffusion, the ratio of the growth rates for faces A and D is $(1 + \tau/\lambda)$, assuming that a freshly nucleated monolayer acts as a perfect sink for adatoms. Although the basal stress concentration is not in general a perfect diffusion barrier [13], it is clear from these simple arguments that diffusion limited growth will eventually contribute to the further extension of elongated islands.

The link between shape elongation and elastic interactions is consistent with observations of anisotropic growth at relatively low growth temperatures where the island density is high [3,24] and there is evidence for surface defect mediated island nucleation [3,6,21]. At higher temperatures, Ge islands nucleate more homogeneously and are widely separated so that their elastic interaction is negligible. Elongated hut cluster shapes are therefore not observed [3,24].

In summary, we have investigated the growth of facets associated with coherently strained islands. A simple but generic model reveals that facets can dramatically influence island growth kinetics, inducing a self-limiting growth behavior which is useful for narrowing island size distributions at low misfits. The shape instability of strained islands can also be qualitatively explained as a natural consequence of this model.

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