Equilibrium Shape of Two-Dimensional Islands under Stress

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We show that the equilibrium shape anisotropy of two-dimensional islands in heteroepitaxial growth depends on island size, a consequence of the presence of strain. Even in homoepitaxy, in which the island shape has conventionally been equated with the ratio of step energies, a substrate surface stress anisotropy can influence island shape.

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Much of our understanding of the fundamental mechanisms of film growth originates from investigations of two-dimensional (2D) islands at the very early stage of epitaxial growth. Both kinetic and thermodynamic mechanisms can be determined. Examples of kinetic parameters include the surface diffusion coefficient, extracted from the number density of 2D islands as a function of growth temperature [1]; the anisotropy in surface diffusion [2] and in adatom sticking to island edges [3], inferred from the shape anisotropy of islands; and the kinetics of island-edge diffusion [4] and corner crossing [5], derived from the shape and compactness of the islands. Examples of thermodynamic parameters include step energies, determined from equilibrium island shapes and their thermal fluctuations [6–9]. In particular, the ratio of step free energies on a surface is commonly believed to define the aspect ratio of equilibrium islands [6–9].

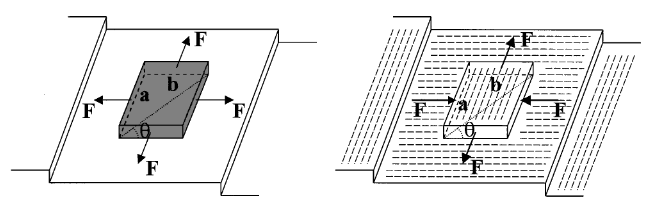
Most quantitative studies of 2D island morphology have been limited to homoepitaxial systems. In heteroepitaxy, where the growing material has a different lattice constant from that of the substrate, such studies become much more complicated because misfit strain can change both the thermodynamics and kinetics of 2D island formation. For example, strain causes spontaneous formation of long-range domain structures [10], whose properties (such as domain size and topology) are well understood.

In this Letter, we describe the effect of strain (lattice mismatch as well as intrinsic anisotropic surface stress) on the equilibrium shape of a 2D island. We demonstrate that the conventional wisdom that the equilibrium shape of a 2D island is determined by the ratio of step free energies is in general incorrect, even for homoepitaxial systems if a surface stress anisotropy is present. We show that strain drives islands to a great anisotropy as island size increases.

We use continuum elastic theory to investigate the stability of a single 2D island under biaxial isotropic stress on the surface of a semi-infinite substrate. We focus on a single island isolated from other islands and steps, to eliminate possible complications of elastic island-island or island-step interactions on island shape. Minimization of strain energy for different island sizes leads to a complex evolution of island shape with increasing island size that depends on the relative strengths of step and strain energies and on the anisotropy of step energies. Biaxial isotropic stress induces a spontaneous shape instability: for isotropic step energies, an island adopts an isotropic shape at small sizes and transforms into an anisotropic shape beyond a critical size; for anisotropic step energies, the island always has an anisotropic shape, but its aspect ratio increases continuously with increasing island size as the strain energy becomes a more significant contribution to the total free energy. The same behavior also occurs for a homoepitaxial 2D island growing under stress induced by substrate surface stress anisotropy.

Consider a biaxially strained epitaxial 2D island on a surface with twofold symmetry [e.g., the (001) surface of a material with the diamond structure]. For simplicity, we assume it has a rectangular shape [11] as shown in Fig. 1. The lattice mismatch between the island and the substrate introduces an elastic-force monopole along the island periphery [10] proportional to the misfit strain and the height of the step that forms the edges of the island. The strain energy of the whole island can then be expressed as

$$E_{\mathrm{strain}} = \frac{1}{2} \int \int \boldsymbol{u}(\boldsymbol{r}_1, \boldsymbol{F}(\boldsymbol{r}_2)) \cdot \boldsymbol{F}(\boldsymbol{r}_1)\, d\boldsymbol{r}_1\, d\boldsymbol{r}_2 \,, \quad (1)$$



(a) (b)

FIG. 1. Schematic views of 2D islands grown on a surface of twofold symmetry, with a rectangular shape of length a and width b. $\theta = \arctan(a/b)$ defines the aspect ratio of the island. (a) Heteroepitaxial growth. F represents the elastic force monopole along the island periphery induced by the lattice mismatch between the island and substrate. (b) Homoepitaxial growth on a surface with anisotropic surface stress. Dashed lines indicate alternating stress domains arising from surface stress anisotropy. F represents the elastic force monopole induced by the surface stress anisotropy. Note that the force monopole on the two a sides points in a direction opposite to the force monopole in heteroepitaxy, shown in (a).

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where $\boldsymbol{u}[\boldsymbol{r}_1, \boldsymbol{F}(\boldsymbol{r}_2)]$ is the displacement at point $\boldsymbol{r}_1$ induced by the force $\boldsymbol{F}$ at point $\boldsymbol{r}_2$.

The integration of Eq. (1) for a rectangular island of length a and width b gives

$$E_{\text{strain}} = E_s\left[2(1-\nu)\left(a\ln\frac{\sqrt{a^2+b^2}+a}{\sqrt{a^2+b^2}-a} + b\ln\frac{\sqrt{a^2+b^2}+b}{\sqrt{a^2+b^2}-b} - 2a\ln\frac{a}{ea_0} - 2b\ln\frac{b}{ea_0}\right) + 4[a+b-2(1-\nu)\sqrt{a^2+b^2}]\right], \tag{2}$$

where $E_s = \frac{1+\nu}{2\pi\mu}F^2$ is the unit strain energy, representing the interaction energy of two parallel force monopoles at unit separation, $F = |\boldsymbol{F}|$ is the force density along the periphery of the island, μ and ν are the Young's modulus and Poisson's ratio of the substrate, respectively, and a_0 is a cutoff length in the range of the surface lattice constant.

Equation (2) can be rearranged into a generic compact form as

$$\frac{E_{\text{strain}}}{E_s} = PG(c) - P \times 2(1-\nu)\ln\frac{D}{a_0}, \tag{3}$$

where $P = 2(a+b)$ is the perimeter, $c^2 = a/b$ is the aspect ratio, and $D = \sqrt{ab}$ is the diameter of the island. $G(c)$ is a dimensionless geometric factor which depends on the island aspect ratio c^2 as follows:

$$G(c) = \frac{1}{2(c+\frac{1}{c})}\left[2(1-\nu)\left(c\ln\frac{\sqrt{c^2+\frac{1}{c^2}}+c}{\sqrt{c^2+\frac{1}{c^2}}-c} + \frac{1}{c}\ln\frac{\sqrt{c^2+\frac{1}{c^2}}+\frac{1}{c}}{\sqrt{c^2+\frac{1}{c^2}}-\frac{1}{c}} - 2c\ln\frac{c}{e} - 2\frac{1}{c}\ln\frac{1}{ce}\right) + 4\left(c+\frac{1}{c}-2(1-\nu)\sqrt{c^2+\frac{1}{c^2}}\right)\right]. \tag{4}$$

For an isotropic island (square), $G(c)$ reduces to a constant.

The island's strain energy, E_{strain}, has two contributions [Eq. (3)]: both are proportional to the perimeter (P) of the island but with opposite signs. The first term, which is positive, arises primarily from the elastic interactions between force monopoles along the same island edge (either a or b); the second term, which is negative, arises from the interactions between force monopoles on opposite island edges (a and a, or b and b) separated by the average island dimension $D = \sqrt{ab}$. The balance of these two terms defines the optimal island shape at a given size D [12] (neglecting step energy contributions). For small island sizes, the first term dominates [i.e., $G(c) \gg \ln(D/a_0)$], and the energy minimization requires minimizing P, favoring an isotropic island shape; for large island sizes, the second term begins to dominate, and the energy minimization requires maximizing P, favoring an anisotropic shape.

The free energies of steps bounding the island of course also contribute in defining the island shape. If E_a and E_b are, respectively, the free energies of unit length for island edges a and b, the total energy of the island is

$$\frac{E_{\text{total}}}{E_s} = \frac{2aE_a+2bE_b}{E_s} + \left[PG_1(c) - P\times 2(1-\nu)\ln\frac{D}{a_0}\right] = P\times\left[\alpha\frac{\beta^2c^2+1}{\beta c^2+\beta} + G(c)\right] - P\times 2(1-\nu)\ln\frac{D}{a_0}, \tag{5}$$

where $\alpha = \sqrt{E_aE_b}/E_s$ defines the ratio of the average step energy to the unit strain energy and $\beta^2 = E_a/E_b$ denotes the ratio of the step free energy of edge a and b.

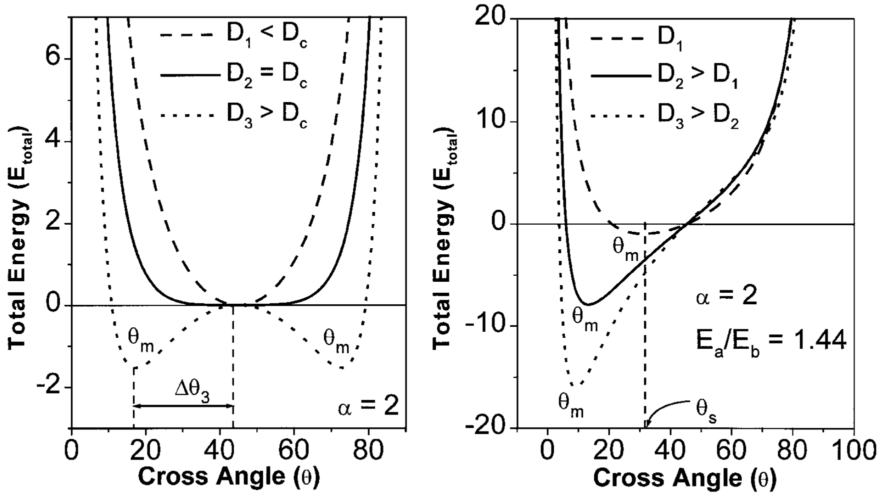
Figure 2 illustrates the strain-induced shape instability. In Fig. 2a, the calculated total energy of an island is shown as a function of $\theta = \arctan(a/b)$ (see Fig. 1) for different island sizes with isotropic step free energies ($E_a = E_b$). We use θ instead of the aspect ratio a/b as the variable for island shape, because the energy is symmetric about $\theta = 45°$. The islands originally adopt an isotropic (square) shape, with an energy minimum at $\theta_m = 45°$. As the islands grow beyond a critical size D_c, strain induces a spontaneous shape instability: the islands adopt an elongated rectangular shape in either of the two orthogonal directions with two degenerate energy minima at $\theta_m = 45° \pm \Delta\theta$. $\Delta\theta$, and hence the aspect ratio of the elongated islands, increases with increasing island size (D). The critical size D_c is defined by the condition

$$\frac{d^2}{d\theta^2}E_{\text{total}}|_{\theta=45°} = 0,$$

which gives rise to

$$D_c = a_0\exp\left[\frac{\alpha+2}{2(1-\nu)} + 1.30\right]. \tag{6}$$

The existence of the spontaneous shape instability originates from the strain relaxation energy. It is especially obvious when the step free energy is zero ($\alpha = 0$). An isotropic step energy shifts the critical size D_c to a larger value, because it would act to drive the island toward an isotropic shape for all sizes. The step energy becomes the dominant factor in defining the critical size when the step



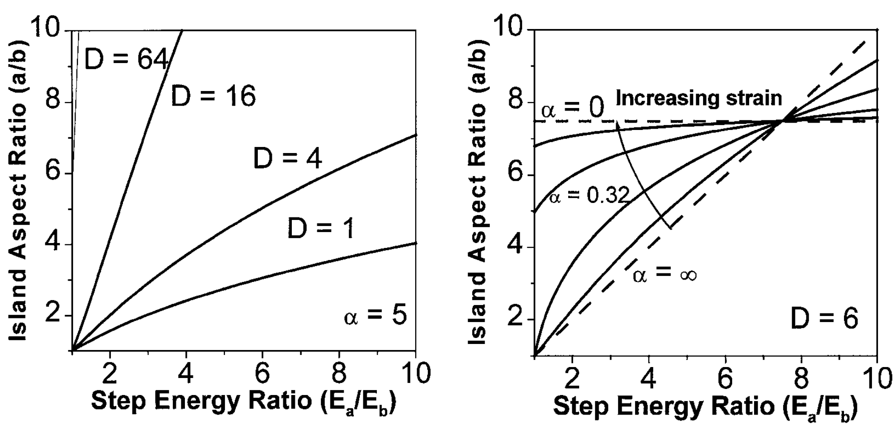
(a) (b)

FIG. 2. Total energy of strained 2D islands vs angle $\theta = \arctan(a/b)$ demonstrating the strain-induced shape instability. The ratio of average step free energy to unit strain energy α is chosen to be 2. (a) 2D islands with isotropic step free energy ($E_a = E_b$). (b) 2D islands with anisotropic step free energies with $\beta^2 = E_a/E_b = 1.44$. The vertical dashed line marks $\theta_s = \arctan(E_b/E_a)$, defined by the step free-energy ratio.

energy is much stronger than the strain energy ($\alpha \gg 1$). In principle, such an instability exists only for 2D islands, because a 3D coherently strained island can always lower its strain energy by increasing its height [13]. However, if the increase in height of a 3D island is *kinetically* limited, the island may grow only laterally. It then can exhibit a shape instability [13–15] similar to the one we describe here for 2D island, driven now by the competition between strain energy and island surface (facet) energy [13].

The strain-induced shape instability redefines the traditionally assumed relationship between the equilibrium island shape and the step free-energy ratio [6–9], namely that the shape reflects the step free-energy anisotropy. When the step free energy is anisotropic, the symmetry between the two orthogonal directions of strain-induced island anisotropy is broken. The anisotropic step free energies (Fig. 2b) cause islands to elongate along the low-step-free-energy direction, in which both the step free energy and strain energy are minimized. (The other direction becomes energetically unfavorable because the step free energy would not be optimized.) For any given island size, in general, the strain favors an optimal island aspect ratio different from what could have been defined solely by the step free-energy ratio; the total energy of the island has one deep minimum at $\theta_m = 45° - \Delta\theta$ for $E_a > E_b$ (or at $45° + \Delta\theta$ if $E_a < E_b$). θ_m moves farther away from 45° with increasing island size, i.e., the strain relaxation makes the aspect ratio of the islands increase continuously with increasing island size rather than stay equal to the step free energy ratio. At small size, strain drives the islands toward a more isotropic shape, making the island aspect ratio smaller than the step free-energy ratio; at large size, strain makes the island aspect ratio larger than the step free-energy ratio.

Figure 3 demonstrates the relationship between the island shape (aspect ratio) and the step free-energy ratio under the influence of strain. Figure 3a shows the dependence of the island aspect ratio a/b on the step free-energy ratio E_a/E_b for different island sizes, for a fixed value of the ratio of average step free energy and strain energy, $\alpha = \sqrt{E_a E_b}/E_s = 5$. At a given value of E_a/E_b, a/b increases with increasing island size, D. Figure 3b shows



(a) (b)

FIG. 3. Island aspect ratio vs step free-energy ratio, demonstrating the strain effect on island aspect ratio. (a) At a given value of α for different island sizes. Note that for $E_a/E_b = 1$, $a/b = 1$ for small island sizes, but becomes larger than 1 at $D = 64$, as the island becomes larger than the critical size D_c defined in Eq. (6) (see Fig. 2a). (b) At a given island size for a different value of α.

a/b as a function of E_a/E_b for different values of α for a fixed island size ($D = 6$). The horizontal dashed line ($\alpha = 0$) marks the island's aspect ratio defined solely by strain energy relaxation, i.e., when the step energies are zero. The inclined dashed line corresponds to $\alpha = \infty$, i.e., there is no strain energy, and the island's aspect ratio equals the step free-energy ratio. All the curves pass through the same point, at which the island aspect ratio defined by minimizing the strain energy coincides with that defined by minimizing the step free energy. Below this point, strain relaxation drives the island toward an anisotropy higher than the step free-energy ratio; above this point, the reverse is true, although it is never possible for strain to force an isotropic island shape. This point will, of course, shift if the size D of the island is changed.

If a substrate surface stress anisotropy is present, as in the Si(001) surface, the above conclusion obtains even for homoepitaxy. The surface stress anisotropy introduces a force monopole [10] along the periphery of the 2D island similar to that introduced by misfit strain (Fig. 1b), the only difference being that the force monopoles on the two a sides point in directions opposite to those in heteroepitaxy (compare Fig. 1b to Fig. 1a), leading to a slightly different geometry factor $G(c)$. In Eq. (4), the last term is replaced by $4[(1 - 4\nu)(c + \frac{1}{c}) - 2(1 - 3\nu)\sqrt{c^2 + \frac{1}{c^2}}\,]$.

It has been a common practice to derive the step free-energy ratio on an anisotropic surface from the equilibrium aspect ratio of 2D islands at a given temperature [6–9]. We have shown that, for islands under stress, the aspect ratio of 2D islands does not simply equal the step free-energy ratio, but becomes dependent on the island size and on the ratio of the strengths of the step free energy and strain energy. Because both the parameter α and the step free-energy ratio β vary with temperature, there is no way to determine the step free-energy ratio at different temperatures from only one data point of island shape at each temperature. In addition, the strain-induced island-island interaction also influences island shape, further complicating the problem.

To derive the step free-energy ratio from the shape of strained 2D islands, we propose an experiment to observe [e.g., by using low-energy electron microscopy (LEEM)] the changing shape of a single "isolated" island during growth at a *fixed* temperature. As the temperature is fixed, both the step free-energy ratio and α remain constant. By carefully measuring the increasing island aspect ratio with increasing equilibrium island size (i.e., very slow growth or interrupted growth), one can uniquely derive both the step free-energy ratio and α (for that particular temperature) with a best fit of the theoretical curve to experimental data. If the unit strain energy (E_s), i.e., the misfit strain or surface stress anisotropy, is also known, one can further determine the individual step free energies. A recent experiment indeed confirms quantitatively our prediction [16], in which the equilibrium aspect ratio of a single isolated Si island grown on a large Si(001) surface was observed to increase continuously with increasing size.

In conclusion, we have demonstrated a strain-induced shape instability in 2D islands that challenges the conventional view of the relationship of island shape and step free energy. The equilibrium shape of 2D islands under stress is determined by both island step free energies and strain energies. Strain makes the island shape size dependent; the magnitude of the effect depends on the relative strengths of step free energies and strain energies. Thus, we have provided a theoretical framework for deriving the step free energies from island shape taking into account the effect of strain.

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