

Equilibrium Shapes and Properties of Epitaxially Strained Islands

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We calculate the equilibrium morphology of an epitaxial strained layer which wets the substrate (Stranski-Krastanow growth), in a two-dimensional continuum model. The layer coalesces into a single discrete island, with zero contact angle to the film wetting the substrate. Small islands have a minimum width, and hence an arbitrarily small aspect ratio. Very large coherent islands have a shape that approaches a ball sitting atop the wetting layer. [S0031-9007(97)04757-1]

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Much effort has been devoted to understanding epitaxial layers that have a different lattice constant than the substrate, and are therefore strained. Such layers are inherently unstable or metastable against various modes of relaxation. In some cases relaxation can be suppressed, and unrelaxed layers are already used in commercial electronic devices. In other cases, elastic relaxation drives the material to rearrange as discrete islands on the substrate, and there is wide speculation that such islands could be used as “quantum dots” in a variety of future nanoscale devices [1].

Yet despite their importance, our understanding of strained layers is in its infancy. Even the equilibrium of a strained layer is not fully understood. If the layer is assumed flat, one can study the equilibrium distribution of dislocations [2]. But in fact, a flat strained layer is morphologically unstable [3–5] or metastable [6], and can “roughen” (i.e., become nonplanar) even without the formation of dislocations. There have been many studies of the initial morphological instability, assuming a pre-existing strained layer [3–5], and a number of studies have examined behavior beyond the initial instability [5,7–13].

However, in the case of an unfaceted surface [14] there is no barrier to roughening. As a result, at low growth rates the material would never form a continuous layer, but would grow from the outset as discrete islands. In this case the most fundamental issue is not the instability, but the equilibrium morphology. Only a few studies have even touched on this issue [10,12,15–18].

Here we study the equilibrium of a coherent strained layer, i.e., in the absence of dislocations. We consider the common case of Stranski-Krastanow growth, where the strained layer wets the substrate. We find that the equilibrium morphology consists of a single island whose edges meet the wetting layer with zero contact angle. Very small islands have a fixed asymptotic width and a vanishingly small aspect ratio. With increasing volume, the island initially grows by vertical thickening. At a larger volume it widens as well, but the aspect ratio continues to increase.

Sufficiently large islands appear to meet the wetting layer at a finite contact angle, although close inspection reveals a smooth zero-angle contact at a small length scale. The “apparent contact angle” increases continuously with size. In fact, a sufficiently large island overhangs its base, and approaches a ball shape except near its edge. This has been suggested previously [18,19], but we are not aware of any previous calculation of the actual equilibrium structure.

We consider a two-dimensional, fully isotropic, epitaxially strained system with the film/substrate interface given by $y = 0$ and the film surface described by $y = h(x)$. The islands we describe in two dimensions are equivalent to elongated island “ridges” in three dimensions [15,20]. However, we expect all of the *qualitative* results here to carry over to the full three-dimensional case.

A difference in lattice parameters between the film and substrate generates a misfit strain in the film. We take the film and substrate to be linearly elastic materials with identical elastic constants, which is a reasonable approximation for a number of strained-layer systems of technological interest, such as Ge/Si and InAs/GaAs. In this case, as long as $h(x) > 0$, the elasticity problem is equivalent to that for a stressed semi-infinite solid. The stress \mathbf{T} is given by $\nabla \cdot \mathbf{T} = 0$, with the boundary conditions $\mathbf{n} \cdot \mathbf{T} = 0$ on $y = h(x)$, and $\mathbf{T} \rightarrow \mathbf{T}_0$ as $y \rightarrow -\infty$. Here \mathbf{T}_0 is the uniaxial (misfit) stress for a uniform film, and \mathbf{n} is the unit normal to the film surface. The stresses in the film are then given by \mathbf{T} , and the stresses in the substrate are given by $\mathbf{T} - \mathbf{T}_0$.

Over some range of temperature, evaporation and bulk diffusion may be neglected. Then mass transport occurs only by surface diffusion in response to surface gradients in the chemical potential μ . (We consider the case of a single-component film.) The equilibrium morphology thus has a constant chemical potential along the free surface. The chemical potential at the surface has three contributions, $\mu = \gamma\kappa + S + w(h)$. The first two terms are the usual surface energy and strain energy contributions (see, for example, [3]) where γ is the

surface energy of the film, κ is the local curvature of the film surface, and S is the strain energy density on the film surface given by $S = \frac{1}{2} \text{Tr}(\mathbf{TE}) = \frac{1}{2} T_{ij} E_{ij}$, where \mathbf{E} is the strain tensor. The last term $w(h)$ represents the contribution to the chemical potential due to the interaction of the film surface with the substrate (a wetting energy). For thick films $w(h)$ is negligible, but not for thin films.

An important issue in modeling island morphologies is the physics of the thin wetting layer. Previous analyses of island morphologies have used a boundary layer model to account for the wetting effects which become important for very thin films [12,17]. Since this boundary layer is really atomically thin, we consider the limiting case of the boundary layer thickness going to zero. From an asymptotic analysis of a boundary layer model for the surface energy, the equations for the equilibrium island and wetting layer are [21]

$$\begin{aligned} \mu &= \gamma\kappa + S, \quad \text{for } h(x) > 0 \text{ (on the island)}; \\ h'(x) &= 0, \quad \text{at the island edge}; \\ h(x) &= 0, \quad \text{otherwise (on the wetting layer)}. \end{aligned} \quad (1)$$

This set of equations for the film morphology corresponds to constant chemical potential μ everywhere on the surface of the film and the thin wetting layer. On the island surface, $w(h)$ is negligible and μ is a balance of surface energy and strain energy terms. On the wetting layer, in the thin boundary layer limit, h approaches zero, with only the infinitesimal deviations in h needed to equilibrate μ via the rapidly varying $w(h)$. Thus, in the limit of zero boundary layer thickness, $h = 0$ to leading order in the wetting layer, and is independent of the specific form of $w(h)$. Finally, the zero slope condition at the island edge is a necessary matching condition between the island and wetting layer.

The strained film has a characteristic strain energy density $S_0 = \frac{1}{2} \text{Tr}(\mathbf{T}_0 \mathbf{E}_0)$, where \mathbf{T}_0 and \mathbf{E}_0 are the stress and strain tensors for a uniform film. The equilibrium island shape represents a balance between surface energy and strain energy terms. As a result, it has a characteristic length $l = \gamma/S_0$. In the results that follow, we scale all lengths by l , all energy densities by S_0 , and all stresses by the misfit stress T_0 . Also, the equilibrium morphology consists of a single isolated island, so we focus on the corresponding limit of large spacing between islands. In this case *the island shape depends only on the (dimensionless) volume V* .

We begin by considering the limit of small island volume, i.e., the earliest stage of strained-layer growth. In the limit of a single isolated island with $V \rightarrow 0$, the island shape can be described by a small slope theory [16]. Let W be the island width, with the island occupying $-W/2 \leq x \leq W/2$. For $h(x) \ll W$, the leading order island shape is described by the nondimensional equation

$$\mu - 1 = -h''(x) + \frac{4}{\pi} \int_{-W/2}^{W/2} \frac{1}{x_0 - x} h'(x_0) dx_0. \quad (2)$$

The island width appears as an eigenvalue. The energy minimizing equilibrium solution is obtained for $W = 1.73$, with the island height $H = h(0)$ arbitrary, $\mu = 1 - 2.98H/W$, and $V = 0.839H$. As shown in Fig. 1, the numerical solution to (2) is approximated very well by the function

$$h(x) \approx H \cos^2 \frac{\pi x}{W} \quad \text{for } |x| \leq W/2. \quad (3)$$

These results demonstrate that very small islands all have a fixed width and the same basic shape. To leading order in H/W , islands of different sizes differ only by a vertical scaling factor corresponding to the island height. As seen below, larger islands have larger widths, so the width $W = 1.73$ represents a *minimum width for an isolated island*.

To determine the island shape at larger V , we modify the numerical method of [9] to describe an island on a wetting layer [21]. Briefly, islands with periodic spacing L are described by a symmetric distribution of N Chebyshev nodes distributed with respect to surface arclength. The equilibrium conditions (1) are enforced at each node point on the film surface. To treat an isolated island, L is increased until the results converge to the asymptotic value. The numerical solution to the coupled free boundary and elasticity problem determines the island

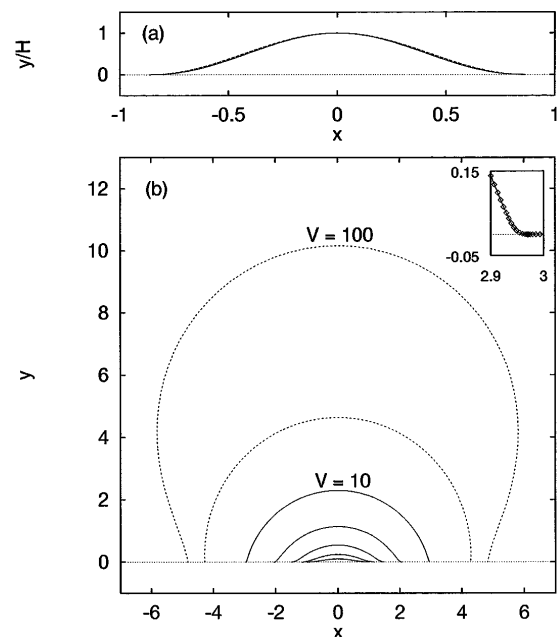


FIG. 1. Equilibrium island shapes: (a) asymptotic solution for thin island (solid curve). The dashed curve is the cosine approximation Eq. (3). For small islands the width is fixed and the height scale is arbitrary. (b) Numerical solution for successive volumes differing by factor $\sqrt{10}$, from $V = 0.1$ to $V = 100$. Solid curves indicate that the shape is converged in N and L . Dotted curves for the shape indicate that the numerical results are not fully converged. The inset (upper right) resolves the details of the island edge for $V = 10$ showing the transition to a zero contact angle (symbols indicate computational nodes).

shape and chemical potential as a function of the island volume V .

Figure 1 shows the equilibrium island shape for a range of island volumes. For small islands the shape is described by the asymptotic thin-island solution. As the island volume increases the island aspect ratio increases monotonically, as does the width. For $V \sim 20$ or larger, the island shape displays overhanging sides. Our results suggest that as $V \rightarrow \infty$ the island shape approaches that of a ball sitting on top of the wetting layer.

In Fig. 1, large islands appear to attach to the substrate at finite contact angle. If the contact angle were finite, however, there would be a singularity in the strain energy density at the island edge [22]. Shown in the inset of Fig. 1 is a detail of the solution for $V = 10$, which displays a thin transition region, in which the singularity is smoothed out and the tangent angle varies from the apparent contact angle to the necessary zero contact angle at the wetting layer. As the island size increases, this apparent contact angle increases, and the transition region is compressed towards the island edge. The resulting “near singularity” has a large stress localization and large curvature.

The increasing contact angle and stress concentration with increasing V is important for understanding the ultimate relaxation by introduction of dislocations. LeGoues *et al.* [23] suggested that such an effect could allow the growing island to incorporate a dislocation (even a sessile one) at its edge, which could be the dominant process at a low temperature when nucleation and glide of dislocations are suppressed.

A calculation of the total energy for the island supports the conclusion that the large V shape is that of a ball sitting atop the wetting layer. The energy of an island morphology is

$$E = \int_0^V \mu(V) dV, \quad (4)$$

with μ given by Eq. (1). This is compared with the energies of a uniform film $E_f = V$ and a detached ball $E_b = \sqrt{4\pi V}$ in Fig. 2. At zero volume the planar film and the island have the same energy. As the volume increases the energy of the island approaches that of the ball, and can be approximated by the simple formula $1/E = 1/E_f + 1/E_b$ as shown in Fig. 2. While we are not able to continue the calculations to larger V , the trend suggests that the island tends toward a ball, with its contact area being a diminishing fraction of its total area.

The chemical potential $\mu(V)$ shown in Fig. 3 is a monotonically decreasing function of V . Thus any distribution of islands will coarsen, with larger islands growing at the expense of smaller ones, until the system reaches equilibrium with a single island containing all the material (except what is bound in the wetting layer). Including island-island interactions should not affect this conclusion (see also [17]).

Figure 3 also shows the apparent contact angle for the island, which we define as the maximum tangent angle

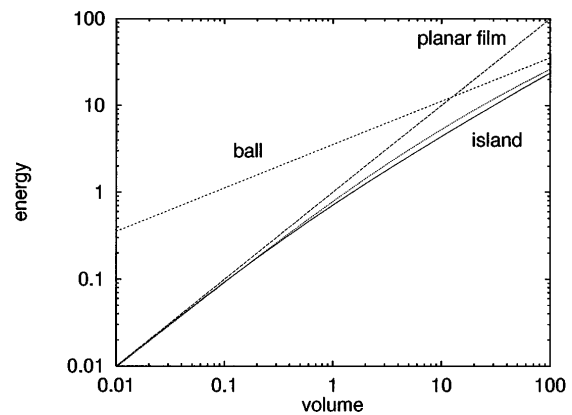


FIG. 2. Island energy E vs volume, compared with energy E_b of ball and E_f of uniform film. The computation parameters are $L = 20$ and $N = 160$. Convergence of energies suggests that the island approaches the ball morphology at large volumes. The dotted line is the approximation $1/E = 1/E_f + 1/E_b$ for the island energy.

of the surface with respect to the horizontal. The results indicate that the island reaches an overhanging shape (maximum angle $>90^\circ$) for a large enough island volume.

The maximum strain energy density increases steeply at large island volumes, as shown in Fig. 3. Mirroring this behavior, the minimum radius of curvature R on the island surface decreases steeply for large islands. When this radius reaches atomic dimensions, any continuum model must break down. For sufficiently large misfits, this could happen at quite modest island volumes. The physical magnitude of this cutoff is directly related to the length scale and is hence very sensitive to the misfit stress; however, a typical length scale for a system with a relatively large misfit might be $l = 160$ nm, which gives a typical atomic spacing cutoff of $R = 0.04$ and corresponds to $V \approx 5$. The implication is that islands with a cross

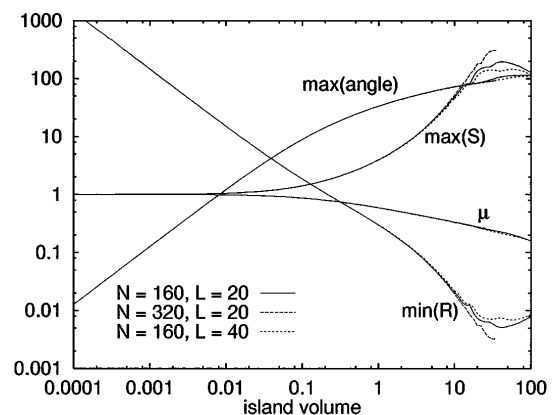


FIG. 3. Island properties as a function of island volume. Shown are the maximum strain energy density on the island surface S , the minimum radius of curvature R , the apparent contact angle (in degrees), and the chemical potential μ . For $V \leq 10$, the $N = 160, L = 20$ curve is converged in N and corresponds to an “isolated” island.

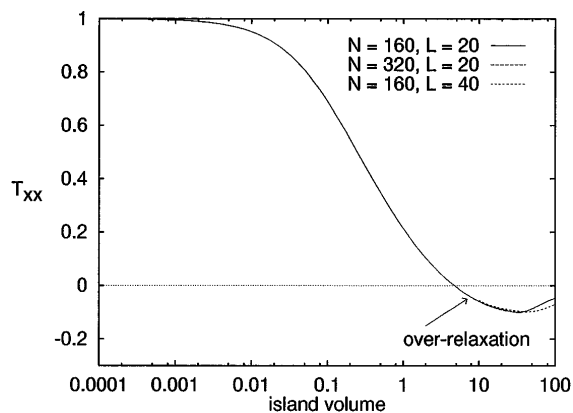


FIG. 4. Stress component T_{xx} at top of island. Islands display overrelaxation for $V > 4.76$.

section larger than about $0.1 \mu\text{m}^2$ in this system would have a finite contact angle on the atomic scale.

Figure 4 shows the stress component T_{xx} at the top of the island. As the island volume increases, the stress is relaxed. For large enough island volumes ($V > 4.76$) there is actually *overrelaxation* at the top of the island, with the stress having opposite sign from the misfit stress. As the island volume increases further, the overrelaxation reaches a maximum value and then appears to decrease towards zero as the island approaches a ball morphology. The overrelaxation phenomena is consistent with the observations of overrelaxation of the lattice constant in Ge islands on Si(111) [24]. Overrelaxation has also been calculated for islands of assumed (nonequilibrium) shape [25], but here we show that *overrelaxation occurs even on unfaceted equilibrium island shapes*.

Finally, we note that a film can never reach the perfect equilibrium state of one large island. Since any island can gather up only material within some characteristic "diffusion length," there are initially many islands, and the island distribution coarsens over time. Such coarsening becomes slower as it proceeds, so that the shape of an island at any given time should be very close to the equilibrium shape for that volume.

This equilibrium island shape evolves continuously with size, from an arbitrarily thin sinusoidal shape of fixed width, to an overhanging ball. While the contact angle remains zero, the apparent contact angle and the curvature at the edge increase rapidly with island size, as does the concentration of stress at the island edge. Eventually this can lead to the introduction of dislocations, or to the breakdown of a continuum picture.

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