

Lecture 13 Strain-induced self-assembly

The innovation

Semiconductor particles in the size range 1-100 nm have special optoelectronic properties dictated by the quantum mechanics of the potential well. These particles are known as quantum dots. Fabricating structures in this size range has been a great challenge of our time. Self-assembly has become an attractive method to fabricate quantum dots. By 1990, it was known that when Ge was deposited on Si substrate, cube on cube, the Ge film is flat up to a few monolayers, and then forms three-dimensional islands. This mode of growth, from layer-by-layer to three-dimensional islands, is known as the Stranski-Krastanov growth. Many other heteroepitaxial semiconductor films also grow this way. For a while people were studying, both experimentally and theoretically, why this happens and how to grow flat thin film. One man's trash is another man's treasure. In 1993, Pierre Petroff, of the University of California at Santa Barbara (UCSB), and his co-workers showed that these 3D islands had the optoelectronic properties of quantum dots. This innovation combined two known facts: heteroepitaxial films are susceptible to Stranski-Krastanov growth, and small semiconductor dots have desirable optoelectronic properties. Since then, the world has been studying how to grow 3D islands.

Strained epitaxy

Both Si and Ge have the diamond crystalline structure. Si has lattice constant 0.357 nm, and Ge has lattice constant 0.357 nm. The lattice constants of the two crystals differ by 4%. When a Ge thin film grows on a thick silicon substrate, one Ge unit cell on one Si unit cell, without any defect, Ge film must be *compressed* by a 4% strain. This is an *enormous* strain, corresponding to several GPa stress. (By comparison, a window glass breaks at 0.1 GPa stress, and an aluminum rod yields at 0.1 GPa stress.) A small crystal can sustain a large stress without

fracture or plastic deformation. At an elevated temperature, the stored elastic energy can motivate mass diffusion, breaking the film to islands.

Surface diffusion driven by elastic energy variation is difficult to analyze. The stress field has to be solved as an elasticity boundary value problem for every surface shape during evolution, which is analytically intractable for most problems of technical interest. The high order differential equation, as listed below, requires great care in the numerical analysis. In many situations, the stress is partially relieved by misfit dislocations; linear elasticity is inadequate. Due to these difficulties, only a few idealized problems have been solved.

Differential equations to evolve the surface of a strained body

Consider a surface with isotropic surface tension, γ . No normal or shear stress acts on the surface. The solid is stressed by the misfit strain. The elastic energy per unit volume on the surface, W , may vary from point to point on the surface.

Consider the chemical potential. The reference body is a large crystal, with a flat surface and under no stress. The given body has a curved surface and is subject to a stress field. The chemical potential on the surface, μ , is the increase in the free energy of the combined system when one atom is transferred from the reference body to a given point on the surface of the given body. The chemical potential consists of two parts: that due to the curvature, and that due to the stress, namely,

$$\mu = \Omega\gamma(K_1 + K_2) + \Omega W.$$

Here Ω is the volume per atom. The curvatures of the surface are positive for a solid sphere, and negative for a spherical cavity in a solid. Both the curvatures and the elastic energy density are taken at the points to which the atom is transferred.

This stress effect differs from what we learned before. When an atom is transferred from the reference body to the surface of a body subject to a stress normal to the surface, σ_n , this normal stress does work through the volume of the atom, giving the chemical potential $\mu = \Omega \sigma_n$. In the previous lectures, we have neglected the elastic energy, treating the body as being rigid. We have justified this negligence by showing that the effect of the elastic energy is small. In the present situation, the surface is subject to no normal stress. When the atom is transferred to a point on the surface, the atom picks up the local strain energy density, giving the chemical potential $\mu = \Omega W$. The two chemical potentials have very different magnitudes. Recall that the elastic energy scales with the stress times the strain, and that the strain is less than a few percent.

The chemical potential gradient drives the atomic flux on the surface:

$$\mathbf{J} = -M \nabla \mu.$$

The atomic flux is a vector defined on the surface. The mobility relates to the surface diffusivity as usual, $M = D_s \delta_s / \Omega kT$. The gradient is taken on the surface.

Consider a part of the surface S bounded by a closed curve l . Let dl be an element of the curve, and \mathbf{m} be the unit vector in the surface, normal to the curve element, pointing to the outside of the surface area S . By definition, $\mathbf{J} \cdot \mathbf{m} dl$ is the number of atoms per unit time goes out of S across the curve element dl . Consequently, the integral over the closed curve, $\int \mathbf{J} \cdot \mathbf{m} dl$, is the number of atoms per unit time going out of the surface areas. Let dS be the surface area element. When the element gains atoms, the solid extends, and the surface moves at the velocity v_n normal to the element. Mass conservation requires that

$$\int v_n dS + \int \mathbf{J} \cdot \mathbf{m} dl = 0.$$

The first integral extends over the surface area S , and the second integral extends over the curve enclosing the area. When the surface area S is made infinitesimal, the above equation relates the velocity normal to the free surface, v_n , to the divergence of the atomic flux, \mathbf{J} :

$$v_n + \Omega \nabla \cdot \mathbf{J} = 0.$$

The divergence is taken on the surface.

The algorithm to evolve the surface is as follows. Start with a given surface shape. Calculate the curvatures at all points on the surface. Solve the elasticity boundary value problem, which gives the strain energy density at all points on the surface. The curvatures and the strain energy density give the chemical potential, the gradient of which gives the atomic flux on the surface. The divergence of the atomic flux gives the velocity of the normal surface. This velocity updates the surface shape for a small time increment. Repeat the above steps for the new surface shape. Once again, in this loop, the essential difficulty is to solve the elasticity boundary value problem for each body shape.

Elasticity boundary value problem

Theory of Elasticity is a serious subject. I took three courses on the subject, once as an undergraduate student and twice as a graduate student, from teachers who had used the theory of elasticity extensively. I also taught the course several times and used the results in my own research. I'm still learning. It also gives me a model of a good theory, guiding me to construct my own models. Here we will not have time to go over the material covered in a normal elasticity course. The following synopsis serves as a reminder for those who have taken a course on elasticity, or as a comfort for those who like partial differential equations. Perhaps you have solved Laplace equation before, or had some unpleasant experience with the Maxwell equations. Just as you will not readily see how van der Waals interaction arises from the Maxwell

equations, you will not readily see how quantum dots self-assemble from the elasticity equations. Establishing a set of equations is just an appetizer. Understanding what the equations say is the main course.

Let's have the appetizer now. Consider a three-dimensional body. Identify each material particle by its coordinates (x_1, x_2, x_3) in the undeformed configuration. The body deforms under load. Let $u_i(x_1, x_2, x_3)$ be the displacement vector of the particle (x_1, x_2, x_3) . The strain field $\varepsilon_{ij}(x_1, x_2, x_3)$ relates to the displacement field as

$$\varepsilon_{ij} = \frac{1}{2} \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right).$$

Let $\sigma_{ij}(x_1, x_2, x_3)$ be the stress field in the body. The stress and strain tensors are related by the generalized Hooke's law:

$$\sigma_{ij} = c_{ijpq} \varepsilon_{pq}.$$

The stiffness tensor c_{ijpq} has been measured for many materials, and can be found in handbooks.

The repeated Latin index implies the summation over 1,2,3.

Force balance requires that

$$\frac{\partial \sigma_{ij}}{\partial x_j} = 0.$$

The above field equations govern the displacement field, the strain field, and the stress field. You can see this clearly by substituting the strain-displacement relation into the stress-strain relation, and then substituting into the force balance equation. This results in three second-order partial differential equations for the displacement fields. Once the displacement field is solved, you can then obtain the strain and the stress fields.

The partial differential equations are solved subject to the boundary conditions. One can prescribe the displacement vector at one part of the boundary, and the force vector on another part of the boundary. In the latter case, consider a surface element with the unit normal vector n_i . The surface element is subjected to the force per unit area t_i , known as the traction vector. Force balance requires that

$$\sigma_{ij}n_j = t_i.$$

Thus, the prescribed force vector set the value of $\sigma_{ij}n_j$.

Once the boundary value problem is solved, the strain energy per unit volume is given by

$$W = \frac{1}{2} \sigma_{ij} \varepsilon_{ij}.$$

It is this quantity that enters the chemical potential of atoms on the surface.

Asaro-Tiller instability

Consider a large piece of crystal with a flat surface, subject to a stress parallel to the surface. Asaro and Tiller (1971) showed that the flat surface is unstable: a small-amplitude perturbation amplifies if its wavelength exceeds a critical value. Asaro and Tiller considered this instability in the context of nucleating a crack from a flat surface by mass transport. The phenomenon was not taken seriously for a long time, because the stress needed to induce this instability is much higher than the stress typically applied to load-bearing materials. In early 1990s, this instability was linked to the film strained by a substrate. The undulation may break the film into islands.

You can understand this instability from the following qualitative considerations. Compare the free energy of the two configurations. The first configuration comprises a flat film on a substrate. The second configuration comprises a wavy film on the substrates. The two

films have the identical number of atoms. The film and the substrate have dissimilar lattice constants, so that a stress field exists in the composite. The free energy of the system, G , consists of the surface energy U_S , and the elastic energy stored in the solid U_E :

$$G = U_S + U_E.$$

The surface energy of the second configuration is higher than the first. However, the elastic energy of the second system is lower than the first. Consequently, if the mismatch strain is large enough, the elastic energy reduction will prevail over the surface energy increase, causing the flat surface to undulate.

You can also understand the instability from a consideration of the chemical potential. The surface energy causes a negative chemical potential at the trough, and a positive chemical potential at the crest. Consequently, the surface energy tends to motivate atoms to diffuse from the crest to the trough, stabilizing the flat surface. The elastic field has the opposite effect. The magnitude of the stress is higher at the trough than the crest. Consequently, the elastic energy difference drives atoms to diffuse from the trough to the crest, destabilizing the flat surface. In the following, we put the qualitative understanding into a quantitative form.

Energetics determines the length scale

To focus on main ideas, we treat a thin film on a semi-infinite elastic crystal. When the film is flat, it is subject to a uniform stress, σ , parallel to the free surface of the crystal. The surface tension of the crystal, γ , is isotropic. Perturb the flat surface by a wave of amplitude q and period λ :

$$y(x,t) = q(t) \cos \frac{2\pi x}{\lambda}.$$

Here y is the height of the perturbed free surface from the initial flat surface, x -axis coincides with the flat surface, and t is the time. The amplitude q is the generalized coordinate in this problem. We will carry out a linear stability analysis, with $q / \lambda \ll 1$. The energies will be calculated to the leading order in $q / \lambda \ll 1$, relative to the energies of the stressed crystal with the flat surface. Because the total area of the film/substrate interface is fixed, we will calculate the energy divided by the interface area.

The surface energy is

$$U_s = \frac{\gamma}{\lambda} \int_0^\lambda \sqrt{1 + (dy/dx)^2} dx - \gamma$$

A direct calculation shows that the undulation increases the surface energy by

$$U_s = \frac{\pi^2 \gamma}{\lambda^2} q^2$$

Because a change in the sign of q leaves the curve length unchanged, U_s is proportional to q^2 to the leading order in q . The term λ^2 fixes the dimension.

For a similar reason, the elastic energy variation U_E is proportional to q^2 to the leading order in q . In addition, linear elasticity dictates that U_E be proportional to σ^2 / E , where E is Young's modulus. Note that σ^2 / E has the dimension of energy per unit volume. A dimensional analysis shows that

$$U_E = -\beta \frac{\sigma^2}{E \lambda} q^2.$$

where β is a dimensionless number of order unity. The negative sign shows that the undulation reduces the elastic energy. An elasticity problem of the wavy surface can be solved analytically to the leading order in $q / \lambda \ll 1$, giving $\beta = \pi$. This boundary value problem has an intermediate

difficulty, which we will not solve here. All it provides is the value of the dimensionless number.

The free energy difference between the solid with a wavy surface and the solid with a flat surface:

$$G = \left(\gamma - \frac{\sigma^2}{\pi E} \lambda \right) \left(\frac{\pi q}{\lambda} \right)^2.$$

The solid with the wavy surface has a lower free energy than the solid with a flat surface when the quantity in the bracket is negative. Consequently, the perturbation amplifies when the wavelength exceeds a critical value, given by

$$\lambda_c = \pi \frac{E\gamma}{\sigma^2}.$$

Because the elastic energy is quadratic in the applied stress, the flat surface undulates under both tension and compression. Take $E = 10^{11} \text{ Pa}$, $\gamma = 1 \text{ J/m}^2$, $\sigma = 10^9 \text{ Pa}$, and we find the critical wavelength $\lambda_c \approx 300 \text{ nm}$. The critical wavelength decreases as the stress increases. The critical wavelength gives us an approximate estimate of the spacing between the quantum dots.

If you know the Griffith theory of fracture, you may recall that a similar group of parameters appear in his theory. Of course, the two phenomena (fracture and surface undulation) both involve the competition between the surface energy and the elastic energy. Because the surface energy scales with the area, and the elastic energy scales with volume, the dimensional difference introduces a length scale. In the theory of fracture, the length is the crack diameter. In the theory of surface instability, the length is the wavelength.

In the problem of cavity growth by diffusion, we also identified a length scale. The critical cavity radius is given by $R_c = 2\gamma/\sigma$. The length scale is also a consequence of the competition between the surface energy and stress. However, in this phenomenon, atoms

relocate from the cavity surface to the external surface, and the stress normal to the external surface does work through the volume of the atoms. Consequently, for the same stress level, R_c is only a small fraction of λ_c : they differ by a factor on the order of elastic strain.

To highlight this difference, I will call these two lengths the Laplace length and the Griffith length, respectively. The Laplace length results from the competition between the surface energy and the stress-volume work. The Griffith length results from the competition between the surface energy and the strain energy.

Kinetics determines the time scale

To the first order in the undulation amplitude q , the curvature of the wavy surface is

$$K = -\frac{\partial^2 y}{\partial x^2} = \left(\frac{2\pi}{\lambda}\right)^2 q \cos\left(\frac{2\pi x}{\lambda}\right).$$

The curvature is negative at the trough, and positive at the crest. This difference in the curvature motivates atoms to diffuse from the crest to the trough, decreasing the undulation amplitude and flattening the surface.

By solving the elasticity boundary value problem, one obtains the elastic field in the body. In particular, the strain energy density on the film surface is given by

$$W = W_0 - \frac{4\pi}{\lambda} \frac{\sigma^2}{E} q \cos\left(\frac{2\pi x}{\lambda}\right),$$

where W_0 is the elastic energy density in the flat film. Once again, the form of the expression can be determined by elementary considerations, without solving the boundary value problem. The exact solution of the boundary value problem gives the coefficient 4π . The perturbation increases the strain energy density at the trough, but decreases the strain energy density at the

crest. The difference in the elastic energy density motivates atoms to diffuse from the trough to the crest, increasing the undulation amplitude.

Combining the two contributions, the chemical potential on the surface is

$$\mu = \Omega\gamma K + \Omega W = \Omega W_0 + \Omega \left(\gamma - \frac{\lambda\sigma^2}{\pi E} \right) \left(\frac{2\pi}{\lambda} \right)^2 q \cos\left(\frac{2\pi x}{\lambda} \right).$$

The chemical potential is nonuniform along the surface. The surface energy and the elastic energy compete. The elastic energy prevails when the wavelength exceeds the critical value $\lambda_c = \pi\gamma E / \sigma^2$. This reproduces the result we obtained by comparing the free energy of the flat and wavy configurations.

The chemical potential gradient causes the atomic flux:

$$J = -M \frac{\partial \mu}{\partial s}.$$

For a small-amplitude perturbation, the curve length s can be replaced by x . Substituting the chemical potential into the above, we obtain the atomic flux field:

$$J = M\Omega \left(\gamma - \frac{\lambda\sigma^2}{\pi E} \right) \left(\frac{2\pi}{\lambda} \right)^3 q \sin\left(\frac{2\pi x}{\lambda} \right).$$

When $\lambda > \lambda_c$, the elastic energy prevails, and atoms diffuse from the trough to the crest. When $\lambda < \lambda_c$, the surface energy prevails, and atoms diffuse from the crest to the trough.

Mass conservation relates the normal velocity of the surface to the divergence of the atomic flux:

$$v_n = -\Omega \frac{\partial J}{\partial s}.$$

Substituting the atomic flux into the above, we obtain the surface velocity:

$$v_n = -M\Omega^2 \left(\gamma - \frac{\lambda \sigma^2}{\pi E} \right) \left(\frac{2\pi}{\lambda} \right)^4 q \cos \left(\frac{2\pi x}{\lambda} \right)$$

When $q / \lambda \ll 1$, velocity normal to the surface is simply $v_n = \partial y / \partial t$, so that

$$v_n = \frac{dq}{dt} \cos \left(\frac{2\pi x}{\lambda} \right).$$

A comparison of the two expressions of the surface velocity gives

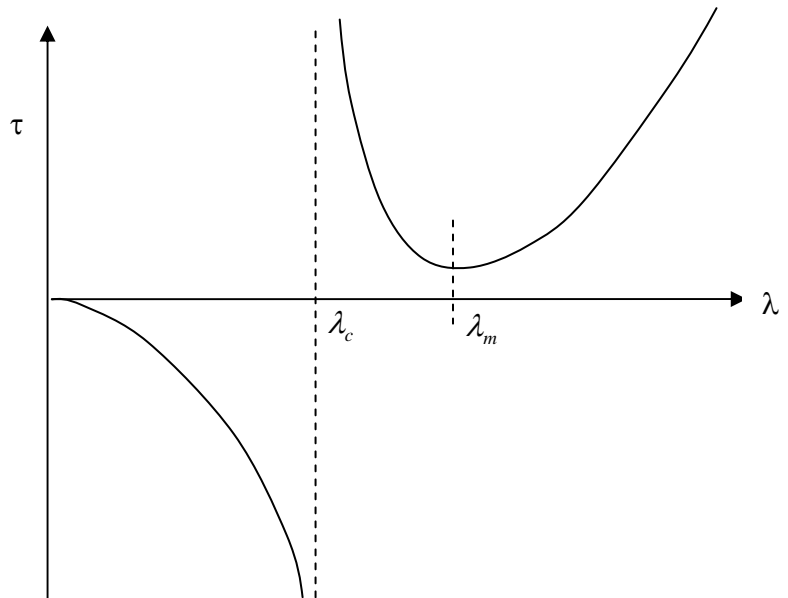
$$\frac{dq}{dt} = \frac{q}{\tau}$$

with the characteristic time being

$$\tau = \frac{\lambda^4}{16\pi^4 M\Omega^2 \gamma} \left(\frac{\lambda}{\lambda_c} - 1 \right)^{-1}.$$

The solution is $q(t) = q(0)\exp(t / \tau)$, where $q(0)$ is the wave amplitude at $t = 0$ (i.e., the amplitude of the initial imperfection).

Sketch the characteristic time τ as a function of the wavelength λ . The trend of the curve is understood from the elementary energetic and kinetic considerations. When $\lambda < \lambda_c$, the surface energy prevails, $\tau < 0$, and the perturbation diminishes with the



time. When $\lambda > \lambda_c$, the elastic energy prevails, $\tau > 0$ and the perturbation grows with the time.

In the range $\lambda > \lambda_c$, the characteristic time τ is large when $\lambda \rightarrow \lambda_c^+$, because the elastic energy

barely overcomes the surface energy to amplify the undulation. The characteristic time τ is also large when the wavelength is very long, because atoms have to diffuse over a long distance to amplify the undulation. τ minimizes at the wavelength $\lambda_m = 4\lambda_c / 3$. If perturbations of all wavelengths have an identical initial amplitude, the perturbation of wavelength λ_m grows most rapidly at $t = 0$.

Insert the wavelength of the fastest growing mode into the characteristic time, and we obtain that

$$\tau_m = \frac{16\lambda_c^4}{27\pi^4 M\Omega^2 \gamma}.$$

This gives a time scale for the surface to develop undulation. When the stress decreases by an order of magnitude, the critical wavelength increases by 2 orders of magnitude, and the relaxation time increases by 8 orders of magnitude.

Island coarsening

Despite intense efforts over the last decade, it remains a challenge to fabricate uniform quantum dots by the Stranski-Krastanov growth. Because islands nucleate at random times and sites, the initial islands are uneven in size and spatial arrangement. On annealing, the islands coarsen: some islands grow and others shrink; over time, the mean island size increases, but the number of the islands decreases. Random nucleation and coarsening are basic causes for island nonuniformity. In practice the two processes, deposition and annealing, can be concurrent. Here we focus on annealing by itself. That is, after a certain amount of matter is deposited on the substrate, the flux is stopped, but atoms diffuse on the surface to transport matter among islands.

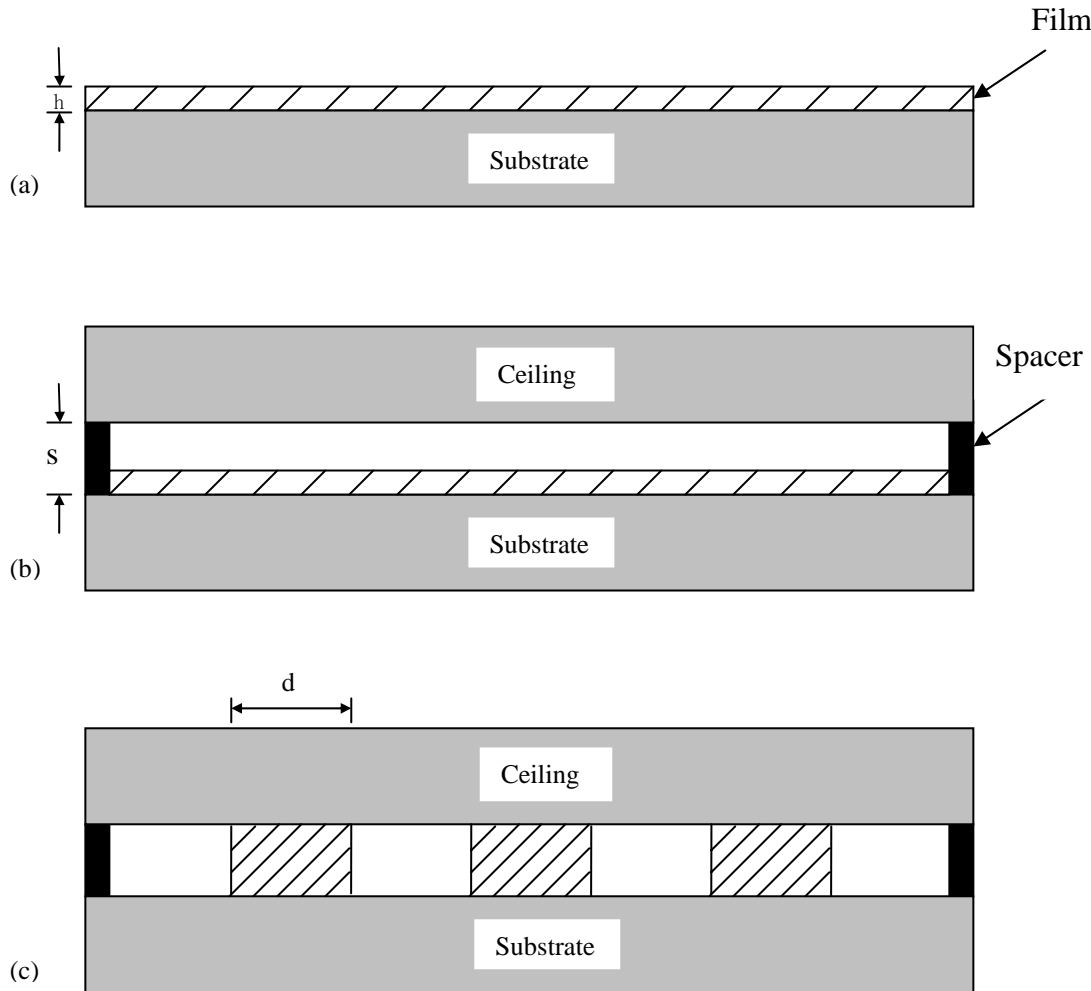
It is well known that the surface energy drives coarsening. Coarse islands reduce the collective surface area, and thereby reduce the total surface energy. It is less appreciated that, in

the Stranski-Krastanov growth, elastic energy also drives coarsening. This is understood as follows. Let the average thickness of the deposited material be h . To simplify the argument, imagine that the islands are identical and form a periodic lattice. Let the base diameter of each island be d , and the spacing between neighboring islands be l . When the islands coarsen, both d and l increase, but the islands are assumed to maintain a similar three dimensional shape and a similar periodic lattice. Volume conservation requires that $d^3 = \beta l^2 h$, where β is a dimensionless number depending on the island shape and the lattice type. Consequently, $l/h = \beta(l/d)^3$. When the islands coarsen, l/h increases, and so does the spacing-to-diameter ratio l/d . Both the film and the substrate are taken to be cubic crystals, with lattice constants a_f and a_s , respectively. The mismatch strain is $\varepsilon_M = (a_s - a_f)/a_s$. Because the total volume of the islands remains constant, we need only consider the average elastic energy (i.e., the total elastic energy stored in the system divided by the volume of the deposited matter). The average elastic energy takes the form

$$U_E = E \varepsilon_M^2 g(l/d),$$

where E is a representative elastic modulus, and $g(l/d)$ is a dimensionless function, which also depends on ratios of other elastic moduli to E . The elastic energy can be calculated from a unit cell containing a single coherent island on a substrate of the shape of a long prism, with zero displacement normal to the sides of the prism, as required by the periodic boundary conditions. For a fixed island diameter d , the larger the cell size l is, the more compliant the system is, and so can the more elastic energy be relaxed. Consequently, the function g decreases as l/d increases. This trend has been shown by the finite element calculation using an axisymmetric unit cell (Woon and Thouless, 1995). Combining the above considerations of volume conservation and elastic relaxation, we conclude that the total elastic energy decreases when the

islands coarsen. Islands sometimes change shape during annealing, which does not appear to stop coarsening. The shape change is ignored. We also neglect energy change of other origins, such as surface stress and line tension.



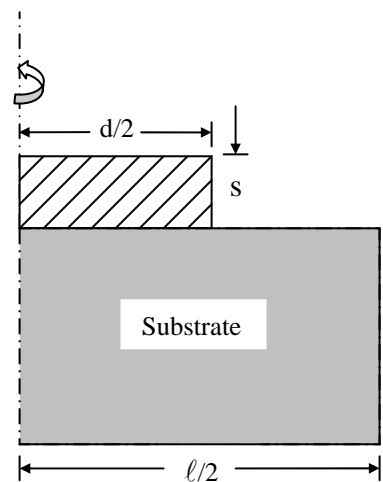
Stable island arrays by height-constrained Stranski-Krastanov growth

Liang and Suo (2001) suggested the height-constrained Stranski-Krastanov growth, inspired by the discovery of lithographically induced self-assembly (LISA) of polymers (Chou and Zhuang 1999). First deposit a crystalline film on a crystalline substrate. The two crystals have different lattice constants, but are coherent. The film need not be perfectly flat, h being the average thickness. Place a stiff ceiling at a spacing s from the substrate surface, e.g., by wafer-

bonding the ceiling to a spacer fabricated on the substrate. The ceiling can even be a functional part of the final device. The film is then annealed to break into islands. After contacting the ceiling, the islands grow in two dimensions. The materials are so selected that, during annealing, atoms of the film diffuse, but atoms of the substrate and the ceiling remain immobile. The growing islands remain coherent with the substrate. It may be necessary that the ceiling be amorphous, or of the same crystal symmetry, so that the ceiling does not affect the crystalline perfection of the islands. From now on we will only consider the process after the islands contact the ceiling.

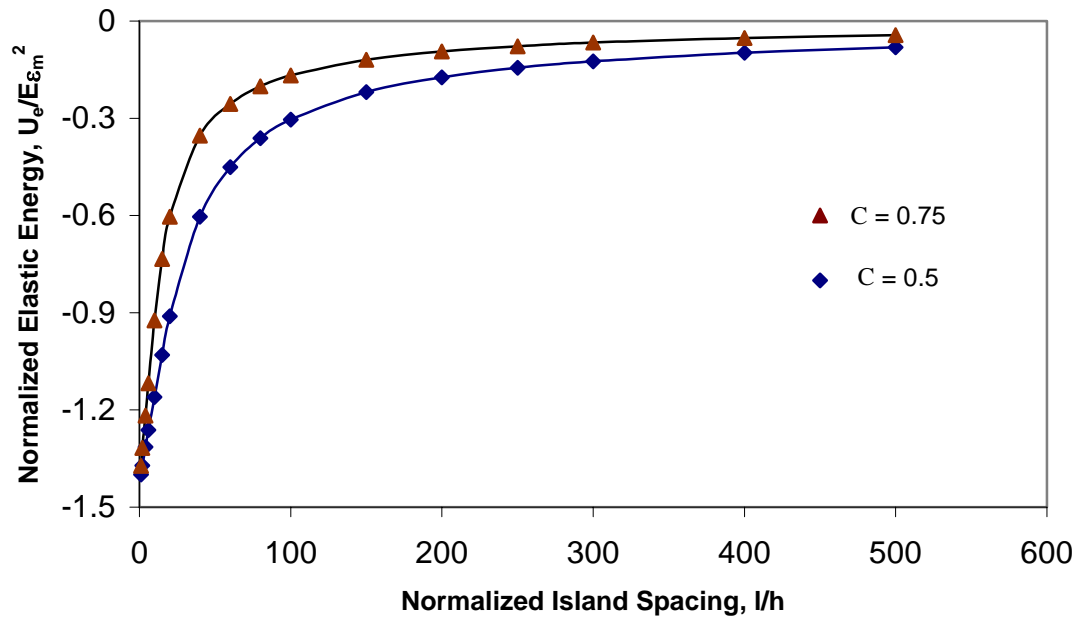
The ceiling reverses the role of elasticity: the elastic energy now drives the islands to refine, rather than coarsen. This trend is readily understood by considering two limits: pancakes and needles. When the islands are pancakes ($d/s \gg 1$), the stress state approaches that of the biaxial stress in the blanket film. When the islands are needles ($d/s \ll 1$), the stress in the d/s system vanishes. One can also see readily that the total surface energy decreases when the islands coarsen. It is the competition between the refining driven by elastic energy and coarsening driven by surface energy that results in an equilibrium island size.

To estimate the equilibrium island size, we now analyze an axisymmetric model, where a circular disk of diameter d lies on an infinitely long cylinder of diameter l . They represent an island and a unit cell of the substrate. Cylinders do not fill space, unlike prisms. The elastic energy stored in an axisymmetric cell, however, should not differ too much from that in a prismatic cell, so long as the two cells have the same volume. Thus, l represents the effective spacing between the neighboring islands. The ratio



$C = h/s$ sets the volume fraction of the matter in the gap between the substrate and the ceiling,

and remains constant during annealing. Volume conservation requires that $d = l\sqrt{C}$. As islands coarsen the spacing-to-diameter ratio l/d remains constant, in contrast to the three dimensional islands considered above. The curved surface of the substrate has no radial displacement and shear stress. The ceiling exerts a capillary pressure on the top surface of the island. The capillary pressure is much smaller than the typical stress caused by the lattice mismatch, and should have a small effect on elastic energy, so that we take the top surface to be traction free. This Letter will not consider the case that the island is also coherent with the ceiling, although the principle remains the same.



Assume that the film and the substrate have identical elastic constants and are isotropic, with Young's modulus E and Poisson's ratio ν . The average elastic energy now takes the form

$$U_E = E\epsilon_M^2 f(C, l/h).$$

The dimensionless function f also depends on Poisson's ratio. We will take the blanket film as the energy reference state, so that $f(C, \infty) = 0$ for pancakes, and $f(C, 0) = -1/(1 - \nu)$ for needles. When the islands are between the two limits, the stress field cannot be determined analytically. We use the finite element package ABAQUS instead. The mismatch strain ε_M is represented by assigning different coefficients of thermal expansion to the two crystals, and then subjecting the system to a temperature rise. Poisson's ratio is assigned to be $\nu = 0.3$. The figure shows the normalized elastic energy as a function of the normalized island spacing for $C = 0.5$ and $C = 0.75$. Indeed, the functions increase monotonically. Both approach the analytic value as $l/h \rightarrow 0$, and to zero as $l/h \rightarrow \infty$.

Next consider the change in the surface energy during annealing. In the idealized model, the edge of the islands are taken to be vertical. As the islands coarsen laterally, the area is constant for each of the following interfaces: islands-substrate, islands-ceiling, substrate-vacuum, and ceiling-vacuum. Consequently, the change in the surface energy is the surface energy density γ times the total area of the island edge. The change in the surface energy per unit volume is

$$U_s = \frac{4\gamma}{l\sqrt{C}}.$$

The surface energy decreases as the islands coarsen.

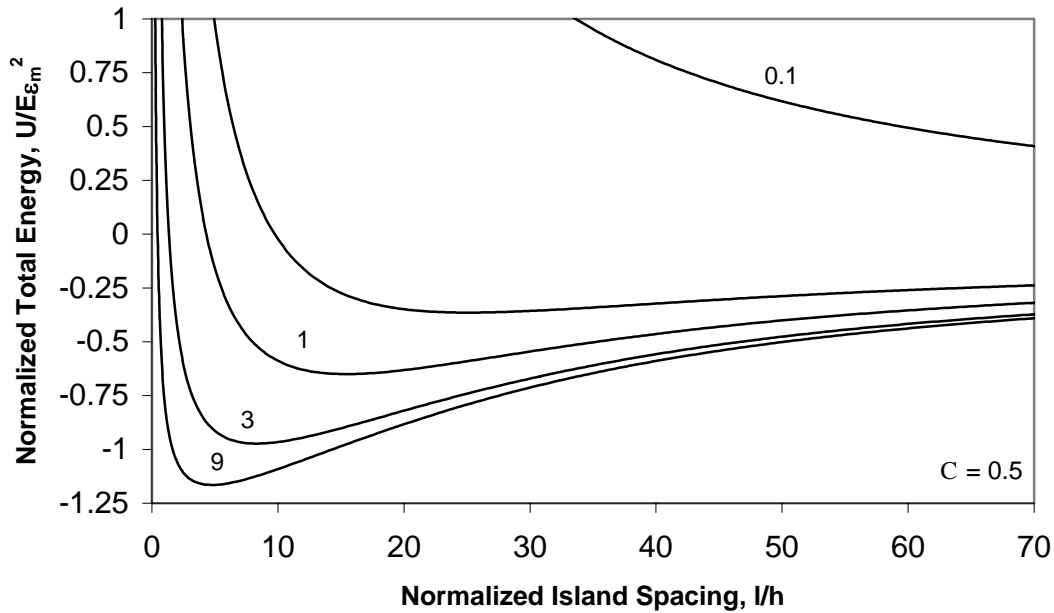
The combined surface energy and elastic energy, $U = U_s + U_e$, takes the dimensionless form

$$\frac{U}{E\varepsilon_M^2} = \frac{4}{(l/h)\Lambda\sqrt{C}} + f(C, l/h).$$

The Griffith number,

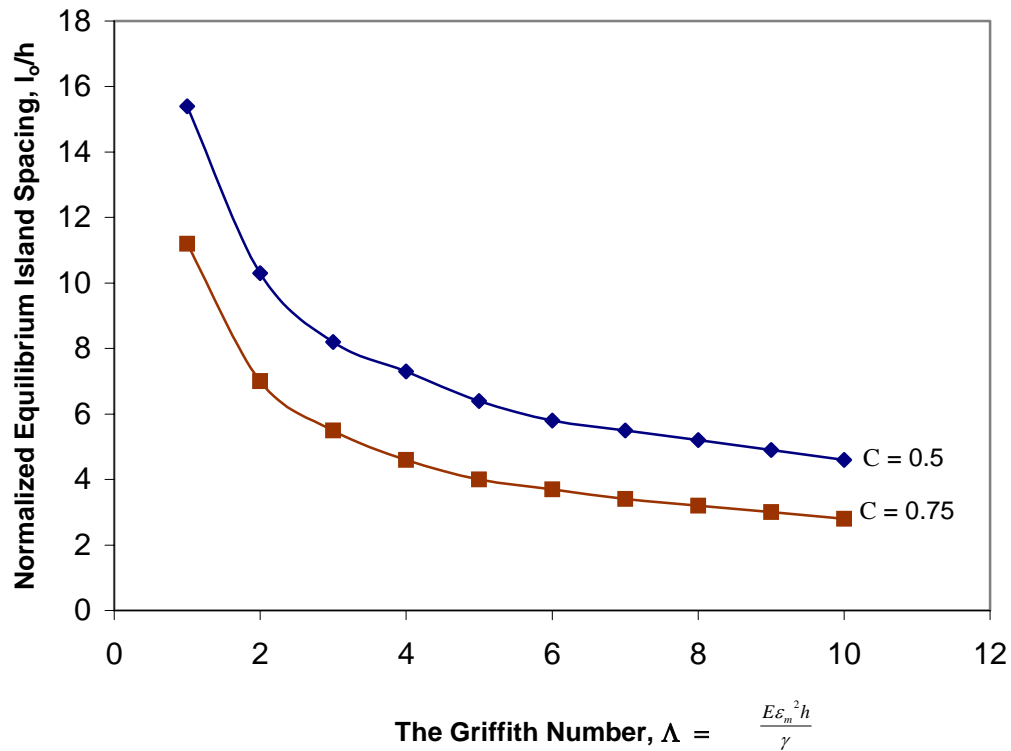
$$\Lambda = hE\varepsilon_M^2 / \gamma,$$

measures the relative importance of the elastic energy and the surface energy. Taking representative values of the various quantities, we find that the Griffith number falls in the range $\Lambda = 1-10$.



The combined energy as a function of the normalized island spacing for $C = 0.5$ is plotted. When the Griffith number is small, the surface energy prevails, and the combined energy decreases monotonically as the island spacing increases, so that the islands coarsen indefinitely. When the Griffith number is sufficiently large, the combined energy reaches a minimum, selecting an equilibrium island size. The following figure plots the equilibrium island spacing, l_0 , as a function of the Griffith number, at two values of the volume fraction. The equilibrium island diameter is given by $d_0 = l_0 \sqrt{C}$. The height-constrained growth offers means to tune the island size and spacing. The average film thickness h is controlled by the amount of deposited matter. The volume fraction C is controlled by the gap between the substrate and the ceiling.

The Griffith number can be varied by varying the mismatch strain, or by varying the surface energy with a surfactant.



Readings

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