University of Massachusetts Amherst

From the Selected Works of Ashwin Ramasubramaniam

2004

Three-dimensional simulations of self-assembly of hut shaped Si-Ge quantum dots

Ashwin Ramasubramaniam, *University of Massachusetts - Amherst*
V. B Shenoy

Available at: https://works.bepress.com/ashwin_ramasubramaniam/4/

Three-dimensional simulations of self-assembly of hut-shaped Si-Ge quantum dots

A. Ramasubramaniam and V. B. Shenoy

Citation: *J. Appl. Phys.* **95**, 7813 (2004); doi: 10.1063/1.1751640

View online: <http://dx.doi.org/10.1063/1.1751640>

View Table of Contents: <http://jap.aip.org/resource/1/JAPIAU/v95/i12>

Published by the [American Institute of Physics](#).

Related Articles

Optical properties of multi-stacked InGaAs/GaNAs quantum dot solar cell fabricated on GaAs (311)B substrate
J. Appl. Phys. **112**, 064314 (2012)

Optical polarization properties of a nanowire quantum dot probed along perpendicular orientations
Appl. Phys. Lett. **101**, 111112 (2012)

Electroluminescence from quantum dots fabricated with nanosphere lithography
Appl. Phys. Lett. **101**, 103105 (2012)

Nucleation features and energy levels of type-II InAsSbP quantum dots grown on InAs(100) substrate
Appl. Phys. Lett. **101**, 093103 (2012)

Highly luminescing multi-shell semiconductor nanocrystals InP/ZnSe/ZnS
Appl. Phys. Lett. **101**, 073107 (2012)

Additional information on J. Appl. Phys.

Journal Homepage: <http://jap.aip.org/>

Journal Information: http://jap.aip.org/about/about_the_journal

Top downloads: http://jap.aip.org/features/most_downloaded

Information for Authors: <http://jap.aip.org/authors>

ADVERTISEMENT

The advertisement banner features a green background with abstract, flowing lines. At the top center, the text "AIPAdvances" is displayed in a green, sans-serif font, with a series of orange dots forming an arc above it. Below this, the text "Special Topic Section:" is written in white, followed by "PHYSICS OF CANCER" in large, bold, white capital letters. At the bottom left, the phrase "Why cancer? Why physics?" is written in a yellow-green font. On the bottom right, there is a blue button with the text "View Articles Now" in white.

Three-dimensional simulations of self-assembly of hut-shaped Si-Ge quantum dots

A. Ramasubramaniam and V. B. Shenoy^{a)}

Division of Engineering, Brown University, Providence, Rhode Island 02912

(Received 4 February 2004; accepted 25 March 2004)

This article presents the results of three-dimensional modeling of heteroepitaxial thin film growth with the objective of understanding recent experiments on the early stages of quantum dot formation in SiGe/Si systems. We use a continuum model, based on the underlying physics of crystallographic surface steps, to study the growth of quantum dots, their spatial ordering and coarsening behavior. Using appropriate parameters, obtained from atomistic calculations, the (100) orientation is found to be unstable under compressive strains. The surface energy now develops a minimum at an orientation that may be interpreted as the (105) facet observed in SiGe/Si systems. This form of the surface energy allows for the growth of quantum dots without any barrier to nucleation—dots are seen to start off via a surface instability as shallow stepped mounds, which steepen continuously to reach their low energy orientations. During the very initial stages of growth, mounds are seen to grow in a dense array with several of them impinging on each other and subsequently coalescing to form larger mounds. This behavior occurs due to the competition between surface energy which seeks to minimize the free-energy by the formation of islands with side-walls at the strain stabilized orientations and repulsive elastic interactions between such closely spaced islands. Using simple analytical calculations, we show the existence of a critical island size for this coalescence behavior. A key result of our analysis is the *inverse scaling* of this critical size with the misfit strain in the film. While energetic analyses may be used to obtain useful insights, the growth of quantum dots is essentially a nonequilibrium process and requires a fundamental understanding of the kinetics. Numerical studies show that the growth kinetics has a profound effect on surface morphology: arrays of well-separated islands or, alternatively, intersecting ridges are obtained in different kinetic regimes. We also study an alternative model of a stable but nonfacet (100) orientation and point out the inconsistencies of this assumption. © 2004 American Institute of Physics.
[DOI: 10.1063/1.1751640]

I. INTRODUCTION

Nanoscale semiconductor devices offer promise for a wide range of applications in electronics and information technology. The phenomenon of strain-driven self-assembly of nanostructures is attractive as an inexpensive and effective manufacturing process and has been an area of extensive experimental and theoretical research over the past several years. Reliable production of such spatially ordered features with narrow size distributions, however, still remains a challenge.

A viable approach for obtaining a large number of self-assembled nanostructures appears to be via the Stranski-Krastanov growth instability. In this process, the surface of a film becomes unstable after a few layers of pseudomorphic growth, resulting eventually in the formation of three-dimensional islands. The driving force for this process arises essentially from the mismatch strain due to heteroepitaxy. The formation of an island can then be viewed as a competition between surface energy and strain energy—the former being proportional to the surface area of the island and the latter to its volume.

The study of surface stability and evolution has received considerable attention in the past. Asaro and Tiller,¹ Grinfeld,² and Srolovitz³ showed independently by a two-dimensional, plane-strain analysis that a flat surface of a stressed solid is unstable if the surface energy is orientation independent. A three-dimensional analysis for this problem was provided by Freund⁴ for small fluctuations. Numerical investigations of large fluctuations were provided by Yang and Srolovitz⁵ in two-dimensions and by Zhang and Bower⁶ in three-dimensions. In all these investigations, the surface energy was assumed to be isotropic and independent of surface strain. A recent study by Shenoy and Freund⁷ presented a departure from this viewpoint. Their model used the underlying physics of crystallographic steps to provide a basis for the orientation and strain dependence of surface energy. Shenoy and Freund showed that, with an appropriate choice of parameters, the (100) orientation becomes unstable under a compressive mismatch strain, and the surface energy attains a minimum at an orientation which can be interpreted as the (105) facet observed in SiGe/Si films. A natural outcome of this result is the absence of a nucleation barrier for the formation of hut- or pyramid-shaped islands. The experimental studies of Sutter and Lagally⁸ and Tromp *et al.*⁹ on the growth of Si_{1-x}Ge_x alloys on Si(100) substrates provide clear evidence for the absence of any nucleation barrier to

^{a)}Electronic mail: shenoyv@engin.brown.edu

the growth of epitaxial islands. Similar observations are reported in experiments on Ge films grown on Si(001) substrates by Vailionis *et al.*¹⁰ and Rastelli *et al.*¹¹ In general, when the Ge concentration of the films exceed 20%, growth of islands starts out via an array of shallow stepped mounds which evolve continuously to form faceted islands. More recent work of Sutter and coworkers¹² provides a clearer picture of the early stages of island evolution, where stepped mounds with atomically flat (100) terraces and single-height steps are found to act as precursors to the (105) faceted islands.

While the work of Shenoy and Freund provides a framework for understanding the nucleationless growth of islands, the restriction to two-dimensions imposes some limitations on the features of the growth process that can be studied. An important issue that cannot be addressed is whether two-dimensional ridges or three-dimensional islands appear in the initial stages of growth. Another aspect of interest is the possibility of coarsening of an island array or, alternatively, the formation of ridges via impingement of islands, depending upon the kinetics of the growth process. The contrasting morphologies that are observed in these situations cannot be distinguished from each other in two-dimensions. An adequate treatment of such issues necessitates a full three-dimensional formulation which we undertake in this work. This formulation will also form the basis for future work on strain-patterned surfaces which are of increasing experimental and theoretical interest.

This article is organized in the following way. In Sec. II, we derive expressions for the free-energy and surface chemical potential of a film deposited on a lattice mismatched substrate. A numerical method to model the surface evolution equations is outlined in Sec. III. Section IV provides a discussion of the results of our numerical studies on the growth and coarsening of quantum dots. The Appendix provides detailed derivations for the surface evolution equations.

The key results of this article can be summarized as follows:

(1) In general, during the initial stages of the growth the deposited material gathers into an array of impinging mounds which coalesce subsequently to establish a distribution of larger, widely spaced mounds. This behavior occurs due to the competition between surface energy which seeks to minimize the free-energy by the formation of islands with side-walls at the strain stabilized orientations and repulsive elastic interactions between such closely spaced islands. We show that there is a size-dependent energetic barrier to coalescence which can provide an explanation for the dense array of impinging mounds observed in the very initial stages of film growth by Koch *et al.*¹³ A key outcome of our calculation is the *inverse scaling* of critical island size with the misfit strain.

(2) The process of islanding is essentially a nonequilibrium process and is significantly influenced by the growth kinetics. We provide a discussion of the relation of growth kinetics to the formation and spatial ordering of morphological features. At low growth rates, sparse arrays of islands are observed while at high growth rates, the arrays are seen to be dense with islands impinging on each other to form elon-

FIG. 1. Schematic of a thin film deposited on a lattice mismatched substrate. The film is in a state of equibiaxial strain— σ_0 is the corresponding stress. V represents the volume of the film and substrate and S is the traction-free surface of the film. $\mathbf{t}_1$ and $\mathbf{t}_2$ are unit tangent vectors and $\mathbf{n}$ is the unit normal on the surface.

gated ridges. The ridges that we observe in our simulations closely resemble the kinetically limited structures observed in recent experiments of Floro and co-workers.¹⁴

(3) Numerical studies are also conducted to identify potential mechanisms of island coarsening—we find that both contact-coarsening as well as Ostwald ripening are possibly operative in dense island arrays.

(4) Finally, we analyze a recently proposed alternative model of a stable but nonfacet (100) orientation that has been suggested by Tersoff *et al.*¹⁵ based on two-dimensional studies. Our calculations in three-dimensions, both analytical and numerical, find the consequences of this assumption to be at variance with experimental observations.

II. FREE ENERGY OF A HETEROEPIITAXIAL FILM

Consider a lattice-mismatched film whose thickness is denoted by $h(x_1, x_2, t)$, where x_i and t are the spatial coordinate and time, respectively, as shown in Fig. 1. It is assumed that the film is periodic along the coordinate directions with period λ , so that $h(x_1, x_2, t) = h(x_1 \pm \lambda, x_2 \pm \lambda, t)$. The film experiences a homogeneous strain ϵ_{ij}^0 ($i, j = 1, 2$) owing to lattice mismatch and thus the strain energy density of a planar film is $U^0 = 1/2 C_{ijkl} \epsilon_{ij}^0 \epsilon_{kl}^0$ where C_{ijkl} is the array of elastic constants. For simplicity, the elastic constants of the film and the substrate are assumed to be the same. In this section, we will derive the total surface free energy of the film and the strain energy of the film-substrate system. As noted earlier, the surface energy density of the strained film depends on both the local orientation and the strain; in what follows, we will provide an expression for the surface energy density based on the physics of surface steps.

Using ϵ_{ij} to denote the strain tensor in the film, the limiting value of the strain on the surface of the film provides a definition of the surface strain tensor field $\tilde{\epsilon}_{ij}$ —the surface stress $\tilde{\tau}_{ij}$ is the corresponding work-conjugate stress measure. Following the work of Shenoy and Freund,⁷ the dependence of the surface energy density on strain and the local surface slope can be written as

$$\gamma(h, \tilde{\epsilon}_{ij}) = (\gamma^0 + \tilde{\tau}_{ij}^0 \tilde{\epsilon}_{ji}) \sqrt{1 + |\nabla_s h|^2} + (\beta_1 + \tilde{\beta}_1 \tilde{\epsilon}_{kk}) |\nabla_s h| + \beta_3 \frac{|\nabla_s h|^3}{1 + |\nabla_s h|^2}, \quad (1)$$

where γ^0 is the surface energy density of the nominally flat surface, β_1 is related to the formation energy of a step, β_3 is related to the interaction energy of two steps, $\tilde{\tau}_{ij}^0 = (\gamma^0 + \partial\gamma^0/\partial\tilde{\epsilon}_{ij})$ is the surface stress of the flat surface at the current level of strain, $\tilde{\beta}_1 = (\partial\beta_1/\partial\tilde{\epsilon}_{ij})\delta_{ij}$ is the strain sensitivity of the formation energy of a step, and ∇_s denotes the surface gradient operator. The parameters β_1 , $\tilde{\beta}_1$, β_3 , and $\tilde{\tau}_{ij}^0$ are assumed to be isotropic to keep the discussion simple—this is not necessary in general. A consequence of these assumptions is that the equilibrium island shape is conical as opposed to pyramidal.

The total free energy of the system is written as the sum of the elastic energy and the surface energy

$$E = \frac{1}{2} \int_V C_{ijkl} \epsilon_{ij} \epsilon_{kl} dV + \int_S \gamma(h, \tilde{\epsilon}_{ij}) dS, \quad (2)$$

where the volume integral is over the film and the substrate. Note that the free energy depends both on the height of the film $h(x_1, x_2, t)$ and the strain field in the film-substrate system. However, since the change in the shape of the film due to surface diffusion takes place over time scales which are much larger than the time scales required for the establishment of mechanical equilibrium, the conditions for mechanical equilibrium can be enforced at a fixed surface shape to obtain the surface strain fields, which can then be used to derive the chemical potential for surface mass transport. If the deviation of the surface of the film from the nominally flat orientation is small, the strain field can be derived from the surface displacements

$$u_i(x_1, x_2) = \int_{-\infty}^{\infty} dx'_1 \int_{-\infty}^{\infty} dx'_2 \times G_{ij}(x_1 - x'_1, x_2 - x'_2) f_j(x'_1, x'_2), \quad (3)$$

where G_{ij} is the half-space elastic Green's function and the surface tractions are given by

$$\begin{aligned} f_1(x_1, x_2) &\equiv C_{13kl} u_{l,k} = C_{11kl} \epsilon_{kl}^0 h_{x_1} + C_{12kl} \epsilon_{kl}^0 h_{x_2} \\ &\quad + \tilde{\beta}_1 \frac{h_{x_1} h_{x_1 x_1} + h_{x_1} h_{x_1 x_2}}{\sqrt{h_{x_1}^2 + h_{x_2}^2}}, \\ f_2(x_1, x_2) &\equiv C_{23kl} u_{l,k} = C_{21kl} \epsilon_{kl}^0 h_{x_1} + C_{22kl} \epsilon_{kl}^0 h_{x_2} \\ &\quad + \tilde{\beta}_1 \frac{h_{x_1} h_{x_1 x_2} + h_{x_2} h_{x_2 x_2}}{\sqrt{h_{x_1}^2 + h_{x_2}^2}}, \\ f_3(x_1, x_2) &\equiv C_{33kl} u_{l,k} = \tilde{\tau}^0 \kappa, \end{aligned} \quad (4)$$

where κ is the mean curvature at the surface point in consideration and $h_{x_i} = \partial h / \partial x_i$. Simple physical interpretations to these boundary conditions can be obtained as follows: The first terms on the right-hand sides of the first and second equalities of Eq. (4) correspond to the tractions applied to the

FIG. 2. Plot of the surface energy, $\gamma = \gamma_0 + \hat{\beta}_1 |\nabla h| + \beta_3/3 |\nabla h|^3$, for a compressively strained film ($\hat{\beta}_1 < 0$). The minimum is at $|\nabla h|^* = \sqrt{-\hat{\beta}_1/\beta_3}$.

surface to keep it traction-free during relaxation of the bulk stress, $\sigma_{ij}^0 = C_{ijkl} \epsilon_{kl}^0$, via surface reorientation. This term can also be interpreted in terms of the distribution of force-monopoles at the step-edges (refer to Appendix A of Ref. 7). The remaining terms on the right-hand sides of the first and second equalities of Eq. (4) represent the shear tractions due to variation of the surface stress along the surface; the right hand side of the third equality of Eq. (4) represents the normal traction due to the Laplace pressure arising from the surface stress. These terms also correspond to the distribution of the force dipoles at the step-edges.⁷

Using the strain fields derived in the preceding paragraph, the total free energy of the system can now be recast in the form

$$E = E^0 + \int_S \left(\hat{\beta}_1 (h_{x_1}^2 + h_{x_2}^2)^{1/2} + \frac{\beta_3}{3} (h_{x_1}^2 + h_{x_2}^2)^{3/2} - \frac{1}{2} f_i u_i \right) dS, \quad (5)$$

where $\hat{\beta}_1 = \beta_1 + \tilde{\beta}_1 \epsilon_{kk}^0$ and E^0 is the free energy of a nominally flat film. The first term under the integral sign in Eq. (5) corresponds to the formation energy of the surface steps, while the second and third terms correspond to the interaction between the monopoles and the dipoles at the step-edges. Also, while the first and second terms under the integral sign depend only on the local slope of the film, the third term is nonlocal, so that the surface energy density at any point is influenced by the overall shape of the film. It is worth noting here that under compressive strains, with an appropriate choice of parameters, the term $\hat{\beta}_1$ can take on negative values, which means that the nominally flat surface becomes unstable leading to spontaneous formation of steps. While stepped surfaces are preferred for sufficiently large compressive strains, with increasing step density, the dipolar repulsion between steps [third term in Eq. (5)] becomes significant. At this point, the competition between strain induced lowering of the step formation energy and the step interactions leads to an optimum slope as shown in Fig. 2. This effect is crucial in our calculations since it allows for the growth of stepped mounds without any nucleation barrier. As we will see, such islands start out as shallow mounds

and proceed to evolve towards their optimal slopes corresponding to minimum surface energy as more material is deposited. Recent atomistic calculations by Shenoy *et al.*¹⁶ on biaxially stressed Si and Ge surfaces provide evidence for strain-induced lowering of step-formation energy leading to optimal orientations such as (105) facets.

III. NUMERICAL METHOD

We now proceed to outline a numerical procedure to model the process of heteroepitaxial film growth. Since the surface shape is periodic, it can be expressed in terms of a Fourier series as

$$h(\mathbf{x}, t) = \sum_{p, q=-N}^N A_{pq}(t) e^{ik(px_1 + qx_2)}, \quad (6)$$

where $k = 2\pi/\lambda$ and the wavelength of the cell λ is chosen to be large enough to accommodate several features of interest. Note that the number of terms included in this series is finite, but otherwise unrestricted. In what follows, we will derive first-order ordinary differential equations for the Fourier coefficients $A_{pq}(t)$. If the shape of the surface is known at $t = 0$, these equations can be numerically integrated to obtain the surface shape for $t > 0$.

The kinetics of surface mass transport is assumed to be terrace diffusion-limited, so that the surface mass flux j_i is related to the surface chemical potential $\chi[h, t]$ through

$$j_i = -D \frac{\partial \chi[h, t]}{\partial x_i}, \quad (7)$$

where D is a constant related to the mobility of the diffusing atoms. For simplicity, the surface diffusion process is assumed to be isotropic in the present work. Using the free energy derived in the previous section [Eq. (5)], the surface chemical potential can be written as

$$\chi[h, t] = U - \frac{\partial}{\partial x_i} \left(\frac{\partial \gamma}{\partial h_{x_i}} \right), \quad (8)$$

where $U = 1/2 C_{ijkl} \epsilon_{ij} \epsilon_{kl}$ is the strain-energy density, which can be computed from the strain fields derived in Sec. II. Mass conservation allows us to make the connection between surface shape and the mass flux as

$$\frac{\partial h}{\partial t} + \frac{\partial j_i}{\partial x_i} = f, \quad (9)$$

where f represents a deposition flux. Using Eqs. (7) and (8) in Eq. (9) and employing the variational approach developed in Ref. 17, we can obtain the following coupled first-order ordinary differential equations (ODEs) for the Fourier coefficients

$$\dot{A}_{pq} = -\iota \frac{Dk^4}{2\pi^2} (p^2 + q^2) \frac{\partial E(A_{pq})}{\partial A_{pq}}, \quad (10)$$

where E is the free energy of the film given in Eq. (5). The details of the derivation are given in the Appendix. As noted earlier, if the initial shape of the film is given, the subsequent evolution of the shape due to deposition or annealing can be determined by numerically integrating the above ODEs.

We also note a couple of points regarding the free energy of the film that requires special consideration due to the presence of the substrate. First, since the mismatch strain is present only in the film, ϵ_{ij}^0 in Eq. (5) should be replaced with a spatially dependent strain $\epsilon_{ij}^0 H(h)$ where $H(\cdot)$ is the Heaviside step function which is unity if $h > 0$ and vanishes when $h \leq 0$. Next, it is possible that there is a region close to the substrate where the formation energy of steps is different from that in the film due to the presence of a “wetting layer.” Experimental observations indicate that the overall roughness of the regions of the substrate that are covered by only the wetting layer remains low. We therefore choose the step-formation energy in the substrate and in the wetting layer to be large so that any undulation penetrating the substrate decays rapidly. The surface energy is now rewritten as

$$\gamma(h) = \hat{\beta}_1 [H(h) - \eta H(-h)] (h_{x_1}^2 + h_{x_2}^2)^{1/2} + \frac{\beta_3}{3} (h_{x_1}^2 + h_{x_2}^2)^{3/2}, \quad (11)$$

where $\eta (\gg 1)$ is a phenomenological parameter. We recall from before that $\hat{\beta}_1$ is a negative number hence implying that the step formation energy in the substrate, given by the product $-\eta \hat{\beta}_1$, is positive. It is thus evident that the surface waviness can penetrate into the substrate only at a high energetic cost.

The effect of η on the *energetics* of the island growth process is expected to be small since any mode that grows into the substrate must eventually decay. The rate at which this decay occurs is directly influenced by the value of η and hence, it is expected to play a role in the *kinetics* of the growth process—a qualitative understanding can be obtained by carrying out an analysis for a single Fourier mode.^{7,18}

IV. RESULTS AND DISCUSSION

To aid in the discussion of our calculations, we introduce a set of nondimensional parameters

$$\tilde{x}_i = kx_i, \quad \tilde{A}_{pq} = kA_{pq}, \quad \tilde{t} = \frac{D\beta_3 k^4}{\pi^2} t, \quad \tilde{f} = \frac{\pi^2}{D\beta_3 k^3} f, \quad (12)$$

which allows us to write Eq. (10) in the functional form

$$\frac{d\tilde{A}_{pq}}{d\tilde{t}} = -\iota(p^2 + q^2) F\left(\tilde{A}_{mn}, \frac{\hat{\beta}_1}{\beta_3}, \frac{\sigma_0^2}{\mu k \beta_3}\right), \quad (13)$$

where σ_0 is the stress corresponding to the mismatch strain ϵ^0 and μ is the shear modulus of the system. This functional form is suggestive of an interplay of various system parameters which will be examined later in this section. The values chosen for the parameters in the present work are $\lambda = 400$ nm, $\gamma_0 = 1$ J/m², $\hat{\beta}_1 = -0.43$ J/m², $\beta_3 = 10.2$ J/m², $\sigma_0 = 2$ GPa, and $\mu = 80$ GPa from which the nondimensional parameters can be computed. The values for $\hat{\beta}_1$ and β_3 were estimated from the atomistic calculations of Shenoy *et al.*¹⁶ for Si–Ge systems. Using these values, the surface energy is found to attain a minimum at a slope $|\nabla h|^* = 0.2$ which is that of a (105) facet.

FIG. 3. Schematic of (a) two islands of volume $V/2$ which can potentially coalesce to form (b) a larger island of volume V . (All the islands are two dimensional.) The island side-walls are assumed to be at optimal slope θ^* . Since volume is conserved during coalescence, the base-width of the large island is $2\sqrt{2}L$, where $2L$ is the base-width of the individual small islands.

With the form of surface energy density plotted in Fig. 2, a planar film would be unstable to formation of a dense array of islands with side-walls at orientations that correspond to the minimum in the surface energy density. If the elastic interactions between the islands are ignored, the free energy of the system can be lowered by maximizing the total surface area of the islands (note that under compressive strain, the surface energy of the optimum orientation is *lower* than the surface energy of the planar orientation at $|\nabla h|=0$). This observation would imply that the film would break up into an array of closely spaced islands, whose sizes increase as more material is deposited. Since the repulsive elastic interactions between impinging islands are proportional to their size, these interactions become comparable in magnitude to the surface energy gained by the formation of the island at a certain critical size of the island. As elastic interactions favor large widely spaced islands to small closely spaced islands, coarsening of the island array should then be expected beyond this critical size. In what follows, using simple analytical arguments, we will show that the critical island size scales inversely with the misfit strain in the film.

A. Analytical results for island coalescence

In our analytical analyses, we restrict the discussion to two dimensions to keep the mathematical details simple. Consider the scenario illustrated schematically in Fig. 3 where two islands, each of volume $V/2$, grow close enough to each other so that they can potentially coalesce and form a larger island of volume V . The side-walls of all the islands are assumed to be at optimal slope $\theta^* = \sqrt{-\hat{\beta}_1/\beta_3}$. Using a Green's function approach (e.g., Appendix B of Ref. 7), the elastic contributions to the self-energies and the interaction energy of the two islands can be computed in closed form. The result of this calculation is shown in Fig. 4(a) where the total elastic energy of two islands, both at optimal slopes, is plotted as a function of their separation d . As long as the two islands remain separated, the interaction energy is proportional to $1/d^2$ which is the expected result for repulsive dipolar interactions. Once the two islands combine, the exact details of the elastic energy curve depend upon the assumptions made for the island shape—we have assumed the island side-walls to remain at optimal slope under the constraints of symmetry and mass conservation. In any case, it is clear that there is an energetic barrier to island coalescence owing to greater elastic interactions between closely spaced islands.

To understand whether coalescence is favorable, we also need to take into account the surface energy contribution.

FIG. 4. Plot of (a) elastic energy relieved and (b) total surface energy of two islands as a function of the ratio d/L . The separation of the islands at various stages of coalescence are shown in a schematic manner. As the two islands approach each other, the interaction energy increases at first followed by a decrease beyond a critical distance. This leads to a barrier to island coalescence as seen in (a).

The net surface energy of two islands as a function of separation is shown in Fig. 4(b). Until the two islands combine, the total surface energy is merely the sum of their individual energies. Thereafter the surface energy increases owing to the decrease in net surface area until it reaches the limiting value of surface energy for the single large island. Combining the surface and elastic energy contributions, the total energies for the two situations illustrated in Fig. 3 are

$$E^{(a)} = 4L\Delta\gamma(\theta, \epsilon_0) - \frac{4\pi\sigma_0^2}{\bar{E}}L^2|\theta^*|^2\log 4 + \frac{L^2}{d^2} \frac{2\pi\sigma_0^2}{\bar{E}}L^2|\theta^*|^2\log 4, \quad (14)$$

where $\Delta\gamma(\theta, \epsilon_0)$ is the difference in the surface density between the optimum orientation at θ^* and the (001) orientation, $\bar{E} = 2\mu/(1-\nu)$ is the plane strain elastic modulus in which the Poisson ratio is taken to be $\nu=0.3$ and the last term corresponds to island interactions, and

$$E^{(b)} = 2\sqrt{2}L\Delta\gamma(\theta, \epsilon_0) - \frac{4\pi\sigma_0^2}{\bar{E}}L^2|\theta^*|^2\log 4. \quad (15)$$

For impinging islands ($d=2L$), coalescence becomes favorable when $E^{(a)} > E^{(b)}$, or if the size of the island satisfies the condition

$$L > L^* \equiv \frac{(2 - \sqrt{2})}{\pi |\theta^*| \log 4} \frac{\bar{E} |\Delta \gamma(\theta, \epsilon_0)|}{\sigma_0^2} \approx 11 \text{ nm}. \quad (16)$$

Scaling of the critical island size as a function of the mismatch strain can be obtained if the strain dependence of the quantity $\Delta \gamma(\theta, \epsilon_0)$ is known. Atomistic calculations¹⁶ show an approximately linear variation of the difference in the surface energy density between the (100) and (105) orientation with the mismatch strain. When this result is used in Eq. (16), we find the critical island size to be inversely proportional to the misfit strain. If an additional dimension were to be included, the numerical estimates obtained should be of the same order as those from the two-dimensional (2D) calculation since the relative magnitudes of the individual terms scale in a identical manner.

The above analysis shows that as long as $L < L^*$, it is possible to have a dense array of impinging islands without coarsening driven by coalescence events. This conclusion provides a possible explanation for the experimental observations by Koch and coworkers¹³ on the surface morphology during the initial stages of growth of $\text{Si}_x\text{Ge}_{1-x}/\text{Si}(001)$. For misfit strains of 1%–2%, their scanning tunnel microscope images clearly show the presence of a dense distribution of 3D islands of an average diameter of 5 nm at a low mean film thickness of around 1 nm. These islands subsequently merge, possibly owing to kinetic limitations, at film thicknesses of 2–3 nm to form a broad continuous layer. In the simple analytical analysis, we assumed the coarsening events to take place at the optimum slope. In the following section, we show using numerical simulations that coarsening can take place even before the islands reach their optimum slope, so that the spacing of the islands that are at the optimum slopes can be much larger than the critical length predicted in Eq. (16). As we will see, the island spacing can also be influenced by growth conditions, in particular the growth flux and the mobility of adatoms that is determined by the growth temperature.

B. Numerical results

We now proceed to present the results of numerical simulations of the growth of strained islands. The effect of growth kinetics on surface morphology can be studied by varying the nondimensional growth flux $\tilde{f} = (\pi^2/D\beta_3 k^3)f$. The functional form of $\tilde{f}$ allows for the modification of growth conditions by varying the deposition flux f or by changing the deposition temperature which manifests itself through the mobility parameter D . The effect of raising (lowering) the growth temperature is then analogous to decreasing (increasing) the deposition rate.

The starting configuration of the system consists of a very small layer of strained film on a substrate. The film surface is subjected to a small random perturbation and the deposition process is started. Subsequent evolution of the film surface is kept track of using approximately 350 Fourier modes. Numerical integration of the ODEs in Eq. (13) is carried out using a standard fourth-order Runge–Kutta routine with adaptive time-stepping. Before analyzing the re-

FIG. 5. (Color) Evolution sequence of a cluster of islands. (a) The islands start off as shallow mounds which (b) continue to grow in volume accompanied by an increase in the side-wall slopes. Island 5 is not favored since it is located between 3 and 4 leading to greater elastic interactions. Islands 3 and 4 grow at the expense of 5 instead. (c) Island 1 is now at its optimal slope. (d) Both Islands 1 and 3 are at optimal slopes now. Some distortion in the shape of Islands 1 and 2 is seen owing to elastic interactions between them.

sults of various growth conditions individually, we note some observations that are consistent across all the cases that we have considered.

At the very outset, the instability in the surface energy causes deposited material to organize rapidly into small, closely-spaced, impinging clusters. As these clusters grow in size, the repulsive elastic interactions between them increase hence making this configuration of closely spaced clusters energetically unfavorable. Consequently, impinging clusters combine to form larger, widely spaced islands which have less elastic interactions overall. Further evolution of a typical group of islands, thus formed, is shown in Fig. 5. While the sizes and spacing of the islands depend on the growth conditions, the islands always start off as shallow mounds [Figs. 5(a) and 5(b)] which proceed to evolve toward their optimal orientation in a *continuous* manner as more material is deposited. Island 1 in Fig. 5(c) and Islands 1 and 3 in Fig. 5(d) are examples of islands that have reached their optimal slope. The subsequent growth of such islands occurs in a self-similar manner; this can be seen more clearly in a sparse array of islands where elastic interactions with other islands is minimal. When islands are close to each other, i.e., roughly within an island spacing of each other, as is the case with Islands 1 and 2 in Fig. 5(d), deviations from a conical shape due to elastic interactions are clearly observed. The effect of growth kinetics on the sizes and spacing of islands is considered next.

1. Kinetic effects: Islands versus ridges

To study the role of growth kinetics, two contrasting cases corresponding to a low deposition flux ($\tilde{f}=1$) and a

FIG. 6. (Color) Evolution sequence for a film grown at a low deposition flux ($\tilde{f}=1$). (a) Initial random perturbation of the surface. (b) Array of mounds which (c) continue to evolve toward their optimal slopes. Some of the larger islands are already at or close to the optimal slope. (d) All the islands are now at their optimal slopes. The average island size is 50 nm, while the average spacing is 110 nm.

high deposition flux ($\tilde{f}=6$) are examined. Snapshots of different stages of evolution are illustrated for the former case in Fig. 6 and for the latter in Fig. 7. In general, increasing the deposition flux leads to an increase in the density of islands. This is readily understood via a comparison of the rate of arrival of atoms on the surface with the rate of diffusion of atoms along the surface toward existing clusters. Within our

modeling approach, this can be viewed as an interplay between the destabilizing effect of the surface which promotes fast-growing, short-wavelength modes and the stabilizing effect of the substrate which causes the rapid decay of such modes. At low deposition rates, the short-wavelength modes grow rapidly, encounter the substrate, and are suppressed. The long-wavelength modes then dominate and establish a

FIG. 7. (Color) Evolution sequence for a film grown at a high deposition flux ($\tilde{f}=6$). (a) Random initial perturbation of the film surface. (b), (c) Mounds begin to form and evolve toward their optimal slope. Some of the larger islands in (c) are at optimal orientations. Contrast the density of islands as well as the relative sizes with those in Fig. 6. (d) Continued deposition leads to the impingement of islands and the formation of ridgelike structures. These ridges bear a resemblance to the SEM images of kinetically limited ridges of Floro *et al.* (Ref. 14).

sparse array of large mounds. These mounds evolve to their optimal slopes and we obtain a sparse distribution of large islands. Conversely, at high deposition rates, the short-wavelength modes are able to grow rapidly without encountering the substrate. The dominance of these modes then leads to a dense array of small mounds which evolve to their optimal slopes to form a dense array of small islands.

The kinetics of the growth process is thus seen to play an important role in establishing the sizes and spatial distributions of islands. A key point to be noted is that the observed spacing of the islands does not reflect an equilibrium property of the growing surface. This observation is in qualitative agreement with the experimental work of Tromp *et al.*⁹ which shows that the roughness does not develop at an equilibrium length-scale during growth and is subject to considerable kinetic constraints. Their conclusions are based on the fact that the island spacing continues to increase when the deposition is abruptly stopped and the islands are allowed to evolve. As we will see later, our calculations also show a qualitatively similar effect—if deposition is stopped and the film is allowed to evolve, coarsening of the islands is observed. The role of kinetics on morphological evolution during deposition is further illustrated in the following paragraph by considering the impingement of islands in the presence of large growth flux.

On continued deposition at high flux, the islands eventually impinge on each other to form elongated ridgelike structures [Fig. 7(d)]. Since these ridges are less effective in relaxing the misfit strain when compared with three-dimensional islands, they are formed as a result of kinetic constraints. A similar kind of island growth and coalescence phenomenon has been reported by Floro and coworkers^{14,19} during low temperature growth of SiGe films. Their *in situ* stress measurements show that the average strain in the film, which initially decreases due to the formation of 3D islands, attains a minimum beyond which the instantaneous stress increases due to formation of ridge like structures shown in Fig. 7(d) (refer to Figs. 8 and 9 of Ref. 14).

Analytical calculations for the dominant Fourier modes in the system at a given stage of growth are intractable due to the inherent non-linearity of the problem. However, in our numerical calculations, we can keep track of the frequency spectrum of the evolving surface shape at any given time. Figure 8 is an example of such a sequence of the frequency spectrum corresponding to the time sequence of Fig. 6. The initial stages show the presence of several high frequency modes in the system. These modes rapidly decay once they encounter the substrate and the spectrum is seen to shift toward lower frequencies. This would then justify retaining a finite number of terms in the Fourier representation for surface shape since our interest is in capturing the long time behavior of the system and not the very initial stages of growth. Figures 6 and 7 also indicate that the islands show some tendency to order on a square lattice prior to coalescence events. Furthermore, the sizes of islands at optimal slope are found to be rather similar—in other words, the size distribution is rather narrow. The frequency spectra, which show distinct peaks after a short duration of deposition and

FIG. 8. Frequency spectrum corresponding to the evolution sequence in Fig. 6. (a) Initial random perturbation. (b), (c) The initial stages of growth show the presence of high frequency modes as well as some noise. (d) Well-defined peaks, indicated by the arrows, corresponding to the dominant modes are now visible. These peaks remain on continued deposition until coalescence events begin to occur.

prior to island coalescence events, provide additional corroboration of this fact.

2. Coarsening behavior

It is also possible, via simulations, to study coarsening of an array of islands (excluding for now the issue of accompanying shape-transitions). As an example, we grow a film at a low deposition flux until a few islands just begin to impinge on each other [Fig. 9(a)]. The flux is now turned off and the subsequent behavior studied; the evolution of island morphology is shown in Figs. 9(b)–9(d). Two types of islands can be identified in the initial configuration shown in Fig. 9(a): islands that are well separated from other islands and the ones that are in contact with their neighbors. The coarsening behavior observed in each of these cases is discussed in the following paragraph.

As the coarsening process progresses, islands that are in contact are seen to coalesce and form larger islands [Figs. 9(b) and 9(c)] whose side-walls are once again close to the optimal orientation. We note that the islands in contact in Fig. 9(a) are large enough (≈ 50 nm in diameter) to overcome the energetic barrier to coalescence discussed previously. Further-more, during the intermediate stages of coalescence, island surfaces can deviate significantly from their optimal slopes and this can drive them into additional surface energy minima, if provided for. While contacting islands coalesce quickly, isolated islands tend to be relatively stable and undergo coarsening at a much slower rate. This can be readily understood by noting that in the latter case, coarsening can only take place through communication via excess material (or adatoms) along the flat surface and requires mass transport over large distances. It can be seen that all the isolated islands in Fig. 9(a) shrink in size, though the rate depends on their size and the distance from larger islands. The small islands in the vicinity of large ones are in commu-

FIG. 9. (Color) Annealing of an array of islands. (a) The initial configuration is obtained by growth at a low flux ($\tilde{f}=1$) until a few islands establish contact with each other. (b), (c) Various stages in the annealing process. Islands that are in contact are seen to coalesce over long time-scales and form large islands. The small islands in the vicinity of large ones can at times be in communication via excess material along the flat surface, (not visible due to the contour scalings), thereby allowing for mass transfer from the small islands to the larger ones. This can result in some small islands being consumed entirely by their large neighbors. More often though, the small islands lose some mass but never disappear altogether.

nication via excess material along the flat surface and are consumed entirely by their large neighbors. More often though, the isolated islands lose some mass but never disappear altogether on the scale of the simulation.

Theoretical studies of the coarsening process in dense island arrays utilize models that are based solely on either contact-coarsening^{21,22} or Ostwald ripening mediated by adatoms.²⁰ Our numerical work shows that both of these mechanisms could be operative in dense arrays of impinging islands observed by Ross *et al.*,²³ Floro *et al.*,²⁰ Sutter and Lagally,⁸ and Tromp *et al.*⁹ A quantitative comparison with the island size evolution reported in experiments²⁰ would require simulations on larger samples, so that the statistics of the distribution of islands sizes can be reliably extracted. Results of these numerically intensive calculations will be the subject of a future publication.

C. Consequences of a stable (100) orientation for island formation

As noted earlier, the functional form for the surface energy density used in this work is based on the physics of crystallographic surface steps. It was demonstrated that under sufficiently large compressive strains, the (100) facet becomes unstable to the formation of steps. Recently, on the basis of a two-dimensional model, Tersoff and coworkers¹⁵ have suggested that the initial stages of Si-Ge island formation can be explained by assuming the (100) orientation to be a stable but nonfacet orientation. Below, we study the consequences of this assumption in a three-dimensional setting. Our analytical and numerical calculations show that a stable

(100) orientation without a cusp in the surface energy, leads to the formation of elongated ridges rather than two-dimensional mounds that are observed during the early stages of growth.

To analyze island growth with a stable (100) orientation, we take the surface energy density to be of the form

$$\gamma = \gamma_0 + a|\nabla h|^2 + b|\nabla h|^3 + c|\nabla h|^4, \quad (17)$$

with phenomenological parameters $a = 0.11 \text{ J/m}^2$, $b = -1.1 \text{ J/m}^2$, and $c = 2.5 \text{ J/m}^2$. With this choice of parameters, the (100) orientation ($|\nabla h| = 0$) is stable (refer to Fig. 10) while the global minimum is located at $|\nabla h| = 0.23$, which is close to the (105) orientation.

We first discuss the results of numerical simulations of film deposition and island growth, carried out using the variational approach described in the Appendix. With the material parameters used in Sec. IV ($\sigma_0 = 2 \text{ GPa}$ and $\mu = 80$

FIG. 10. Plot of the surface energy density, $\gamma = \gamma_0 + a|\nabla h|^2 + b|\nabla h|^3 + c|\nabla h|^4$. The minimum is at $|\nabla h| = 0.23$ which is close to a (105) orientation.

FIG. 11. (Color) Evolution sequence for a film grown at a low deposition flux ($f=1$) under the assumption that the (100) is a stable but nonfacet orientation. (a) Initial random perturbation of the surface. (b) The surface morphology consists of ridgelike features at the outset. (c) These ridges grow as a result of the instability and (d) eventually break up into distinct islands.

GPa), the critical wavelength for the strain driven surface instability^{1–3} is estimated to be approximately 80 nm. The simulation cell size is taken to be $\lambda = 10\lambda_{cr}$. A time sequence of the initial stages of growth with a deposition flux of $\tilde{f} = 1$ is shown in Fig. 11. The surface morphology at the very outset is characterized by intersecting elongated ridgelike structures [Fig. 11(b)]. As more material is deposited, these ridges eventually break up into three-dimensional islands, with slopes close to the (100) orientation [Figs. 11(c) and 11(d)]. As the islands grow in size, the slopes of their side-walls increase gradually until they reach the orientation that corresponds to the local maximum in the surface energy. Beyond this point, the side-walls of the islands rapidly evolve to the optimum slope at $|\nabla h| \approx 0.2$. A key point to be noted here is that the island growth starts in the form of ridges, rather than isolated mounds that are observed in experiments. The formation of ridges depends only on the fact that (100) is a nonfacet orientation with a quadratic dependence of the surface energy on the angle. While the subsequent evolution of the ridges will depend on the details of the form of the surface energy between the (100) and (105) orientations, the wavelength of the ridges is determined by the quadratic term. This is illustrated in the following paragraph by considering the instability of a two-dimensional surface perturbation of a strained film.

Consider a surface height perturbation of a strained epitaxial film of the form $\alpha \cos(k_1 x_1) \cos(k_2 x_2)$, where the ratio of the wave numbers k_1/k_2 gives a measure of the aspect ratio of the perturbation. When the amplitude of the perturbation is small, its growth is determined by a competition between the surface energy cost of creating the perturbation and the elastic energy that can be relaxed by this perturbation. Using the surface energy density given in Eq. (10), to

first order in α , the amplitude of the perturbation decreases as

$$\dot{\alpha}^{(1)} = -2D(k_1^2 + k_2^2)^2 \alpha \alpha, \quad (18)$$

while the contribution from elasticity leads to a growth in the amplitude given by

$$\dot{\alpha}^{(2)} = D(k_1^2 + k_2^2)^{3/2} (1 - \nu) \frac{\sigma_0^2}{2\mu} \alpha. \quad (19)$$

This competition between the two effects in Eqs. (18) and (19), provides a discriminating condition that leads to a critical wavelength in the system

$$\lambda_{cr} = \frac{4\pi a \bar{E}}{\sigma_0^2}. \quad (20)$$

The condition for growth of a perturbation can be written in terms of the critical wavelength as

$$\frac{\lambda_{x_1}}{\lambda_{cr}} > \sqrt{1 + \left(\frac{\lambda_{x_1}}{\lambda_{x_2}} \right)^2}, \quad (21)$$

where $\lambda_{x_1} = 2\pi/k_1$ and $\lambda_{x_2} = 2\pi/k_2$. The result in this form clearly indicates that perturbations with large aspect ratios ($\lambda_{x_2} \gg \lambda_{x_1}$) have a greater tendency for instability than those with aspect ratios close to one. In the very early stages of growth, we should therefore expect to see elongated ridge-like features, which could then subsequently evolve into two-dimensional mounds. This expectation is indeed borne out by our numerical simulations as illustrated in Fig. 11. We are thus led to conclude from our analytical and numerical cal-

culations that the growth mode obtained under the assumption of a stable (100) orientation is at variance with experimental evidence.^{8,9,12,13}

It should also be pointed out that our calculations show that the formation of islands with the side-walls at the strain-stabilized orientation is a continuous process that starts from shallow stepped mounds, without any first-order transitions in the shape. This observation is in agreement with the recent experimental work of Sutter *et al.*,¹² who find clear evidence for the precursor mounds with smooth (001) facets and single-height steps prior to the formation of (105) facets. However, the experiments reported by Tersoff *et al.*¹⁵ suggest that the islands have to grow to a certain size before they transform to a pyramidal shape through a first-order transition. With the form of the surface energy given in Figs. 2 and 10, the equilibrium shape of strained islands should be pyramidal, without any (001) facets, independent of their size. One of the possibilities for the observation of islands in the work of Tersoff *et al.*¹⁵ which do not show pyramidal shapes is that these islands have not achieved their equilibrium configuration.

V. SUMMARY

In conclusion, we have developed a three-dimensional continuum model to study the initial stages of hut-shaped Si-Ge quantum dot growth. The surface energy of a stepped vicinal surface is suitably modified to account for the nature of the extensional strain due to lattice mismatch. For suitable values of compressive strain, the (100) orientation becomes unstable and a minimum is obtained at the (105) orientation. This effect allows for the nucleationless formation of stepped mounds which gradually evolve towards their optimal orientations.

We have formulated a Green's function²⁴ based Fourier spectral method to model the surface evolution equations—detailed derivations are included in the Appendix. The computational advantages of this approach are significant since it does not entail expensive front-tracking and adaptive remeshing procedures. Our method is applied to study the effect of growth kinetics on surface morphology. Arrays of distinct islands or, alternatively, intersecting ridges are observed in different kinetic regimes. We have also studied the process of island impingement and coarsening both analytically and via numerical simulations. Our results are in good correspondence with experimental studies on SiGe/Si systems.

In this work, the functional form of the surface energy depended only upon the magnitude of the local slope. Work is currently underway to include the effects of anisotropy in step-energies. This would allow us to obtain faceted pyramids, which are observed in reality, as opposed to conical ones. The inclusion of additional minima in the surface energy, which would allow us to model shape-transitions, is also being explored. Our method can be easily extended to model strain-patterned substrates and is a subject of current research. Since Si-Ge films are binary alloy films, it might be possible to obtain better insight into their morphological evolution by incorporating alloy segregation effects.

ACKNOWLEDGMENTS

The authors thank Professor L. B. Freund and Dr. C. V. Ciobanu for useful discussions. The research support of the National Science Foundation through Grant Nos. CMS-0093714 and CMS-0210095 and the Brown University MR-SEC program through Grant No. DMR-0079964 is gratefully acknowledged.

APPENDIX: DERIVATION OF EVOLUTION EQUATIONS

To formulate a numerical scheme, we introduce a variational functional¹⁷

$$\Phi[j_i] = \int_S -\chi \frac{\partial j_i}{\partial x_i} dS + \int_S \frac{j_i^2}{2D} dS, \quad (\text{A1})$$

which is readily recognized as the weak form of Eq. (7). We also recall the following expressions from Sec. III

$$h(\mathbf{x}, t) = \sum_{p, q=-N}^N A_{pq}(t) e^{ik(px_1 + qx_2)} \quad (\text{surface shape}),$$

$$j_i = -D \frac{\partial \chi(\mathbf{x}, t)}{\partial x_i} \quad (\text{diffusion-limited kinetics}), \quad (\text{A2})$$

$$\frac{\partial h(\mathbf{x}, t)}{\partial t} = -\nabla \cdot \mathbf{j}(\mathbf{x}, t) + f \quad (\text{mass conservation}).$$

The chemical potential is obtained from Eq. (8) and has contributions from the surface energy as well as the elastic energy. We consider first the role of surface energy. For a constant flux f , $A_{00}(t)$ evolves as $A_{00} = ft$. Using the third equality in Eq. (A2), the mass flux $\mathbf{j}$ can be written as

$$(j_1, j_2) = i \sum_{p, q=-N}^N \left(\frac{\alpha_{pq}}{pk}, \frac{\beta_{pq}}{qk} \right) e^{ik(px_1 + qx_2)} \quad (p, q \neq 0), \quad (\text{A3})$$

where $\alpha_{pq} + \beta_{pq} = \dot{A}_{pq}$. This can now be substituted in the variational functional [Eq. (A1)] to obtain

$$\begin{aligned} \Phi[\alpha_{ij}, \beta_{ij}] = & \sum_{p, q=-N}^N i(\alpha_{pq} + \beta_{pq}) \mathcal{H}_{pq} \\ & + \frac{2\pi^2}{Dk^4} \sum_{p, q=-N}^N \left(\frac{\alpha_{pq}\alpha_{-p, -q}}{p^2} \right. \\ & \left. + \frac{\beta_{pq}\beta_{-p, -q}}{q^2} \right), \end{aligned} \quad (\text{A4})$$

where

$$\mathcal{H}_{pq} = \int_0^{\lambda_1} \int_0^{\lambda_2} \left(pk \frac{\partial \gamma}{\partial x_1} + qk \frac{\partial \gamma}{\partial x_2} \right) e^{ik(px_1 + qx_2)} dx_1 dx_2. \quad (\text{A5})$$

Minimizing this expression with respect to α_{pq} and β_{pq} and adding the two equations thus obtained, we get

$$\dot{A}_{pq} = -i \frac{Dk^4}{2\pi^2} (p^2 + q^2) \mathcal{H}_{-p, -q} \quad (p, q \neq 0). \quad (\text{A6})$$

Equations for $\dot{A}_{p0}$ and $\dot{A}_{0q}$ can be derived similarly.

The strain energy contribution to the chemical potential is accounted for by considering the surface evolution equation in the strong form

$$\frac{\partial h(\mathbf{x}, t)}{\partial t} = D \frac{\partial^2}{\partial x_i^2} U(\mathbf{x}, t), \quad (\text{A7})$$

where the strain energy density, $U(\mathbf{x}, t)$ is

$$U(\mathbf{x}, t) = \frac{1}{2} C_{ijkl} (\epsilon_{ij} + \epsilon_{ij}^0) (\epsilon_{kl} + \epsilon_{kl}^0) \approx \sigma_{ij}^0 \epsilon_{ij} = \sigma_{ij}^0 u_{i,j}, \quad (\text{A8})$$

to first order in surface slope. Computing the discrete Fourier transform of Eq. (A7), we obtain

$$\frac{dA_{pq}}{dt} = -\iota D k^3 (p^2 + q^2) (\sigma_{11}^0 p \hat{u}_1^{(pq)} + \sigma_{22}^0 q \hat{u}_2^{(pq)}), \quad (\text{A9})$$

where $\hat{f}$ is used hereafter to denote the discrete Fourier transform of $f(\mathbf{x}, t)$. The displacement field $\mathbf{u}(\mathbf{x})$ due to a surface traction $\mathbf{f}(\xi)$ can be computed using surface Green's functions $\mathbf{G}(\mathbf{x} - \xi)$ as

$$u_\alpha(\mathbf{x}) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} G_{\alpha\beta}(\mathbf{x} - \xi) f_\beta(\xi) d\xi, \quad (\text{A10})$$

which can then be Fourier transformed to obtain the terms required in Eq. (A9).

The Fourier transforms of the Green's functions are trivial to compute analytically while the transforms of the surface tractions have to be computed numerically using a fast Fourier transform algorithm. Denoting the transform of $f_i(\mathbf{x}, t)$ by $\hat{f}_i^{(pq)}$ and that of $G_{\alpha\beta}(\mathbf{x}, t)$ by $\hat{G}_{\alpha\beta}^{(pq)}$, we can rewrite the evolution equations [Eq. (A9)] as

$$\begin{aligned} \dot{A}_{pq} = -\iota D k^3 (p^2 + q^2) & \left[\frac{\sigma_{11}^0}{\mu} p (\sigma_{11}^0 \hat{G}_{11}^{(pq)} \hat{f}_1^{(pq)} \right. \\ & + \sigma_{22}^0 \hat{G}_{12}^{(pq)} \hat{f}_2^{(pq)}) + \frac{\sigma_{22}^0}{\mu} q (\sigma_{11}^0 \hat{G}_{21}^{(pq)} \hat{f}_1^{(pq)} \\ & \left. + \sigma_{22}^0 \hat{G}_{22}^{(pq)} \hat{f}_2^{(pq)}) \right]. \end{aligned} \quad (\text{A11})$$

This equation can now be combined with Eq. (A6) to obtain a set of ordinary differential equations for the Fourier coefficients of the surface shape.

¹R. J. Asaro and W. A. Tiller, *Metall. Trans.* **3**, 1789 (1972).

²M. A. Grinfeld, *Sov. Phys. Dokl.* **31**, 831 (1986); *Dokl. Akad. Nauk. (SSSR)* **265**, 868 (1982).

³D. J. Srolovitz, *Acta Metall.* **37**, 621 (1989).

⁴L. B. Freund, *Int. J. Solids Struct.* **32**, 911 (1995).

⁵W. H. Yang and D. J. Srolovitz, *Phys. Rev. Lett.* **71**, 1593 (1993).

⁶Y. W. Zhang and A. F. Bower, *J. Mech. Phys. Solids* **47**, 2273 (1998).

⁷V. B. Shenoy and L. B. Freund, *J. Mech. Phys. Solids* **50**, 1817 (2002).

⁸P. Sutter and M. G. Lagally, *Phys. Rev. Lett.* **84**, 4637 (2000).

⁹R. M. Tromp, F. M. Ross, and M. C. Reuter, *Phys. Rev. Lett.* **84**, 4641 (2000).

¹⁰A. Vailionis, B. Cho, G. Glass, P. Desjardins, D. G. Cahill, and J. E. Greene, *Phys. Rev. Lett.* **85**, 3672 (2000).

¹¹A. Rastelli, M. Kummer, and H. von Känel, *Phys. Rev. Lett.* **87**, 256101 (2001).

¹²P. Sutter, P. Zahl, and E. Sutter, *Appl. Phys. Lett.* **82**, 3454 (2003).

¹³R. Koch, G. Wedler, J. J. Schulz, and B. Wasserman, *Phys. Rev. Lett.* **87**, 136104 (2001).

¹⁴J. A. Floro, E. Chason, L. B. Freund, R. D. Twisten, R. Q. Hwang, and G. A. Lucadamo, *Phys. Rev. B* **59**, 1990 (1999).

¹⁵J. Tersoff, B. J. Spencer, A. Rastelli, and H. von Känel, *Phys. Rev. Lett.* **89**, 196104 (2002).

¹⁶V. B. Shenoy, C. V. Ciobanu, and L. B. Freund, *Appl. Phys. Lett.* **81**, 364 (2002).

¹⁷V. B. Shenoy, A. Ramasubramaniam, and L. B. Freund, *Surf. Sci.* **529**, 365 (2003).

¹⁸A. Ramasubramaniam, Ph.D. thesis, Brown University, 2004.

¹⁹J. A. Floro, E. Chason, M. B. Sinclair, L. B. Freund, and G. A. Lucadamo, *Appl. Phys. Lett.* **73**, 951 (1998).

²⁰J. A. Floro, M. B. Sinclair, E. Chason, L. B. Freund, R. D. Twisten, R. Q. Hwang, and G. A. Lucadamo, *Phys. Rev. Lett.* **84**, 701 (2000).

²¹N. Pinto, R. Murri, and R. Rinaldi, *Thin Solid Films* **336**, 53 (1998).

²²F. Family and P. Meakin, *Phys. Rev. Lett.* **61**, 428 (1988).

²³F. M. Ross, R. M. Tromp, and M. C. Reuter, *Science* **286**, 1931 (1999).

²⁴We have restricted attention in this work to the initial stages of the growth of quantum dots. Surface features are shallow with orientations ranging from 0–11°, i.e., a flat surface to a (105) facet, which then justifies a Green's function approach.