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Epitaxial growth kinetics with interacting coherent islands

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The Stranski-Krastanov growth kinetics of undislocated (coherent) three-dimensional islands is studied with a self-consistent mean field rate theory that takes account of elastic interactions between the islands. The latter are presumed to facilitate the detachment of atoms from the islands with a consequent decrease in their average size. Semiquantitative agreement with experiment is found for the time evolution of the total island density and the mean island size. When combined with scaling ideas, these results provide a natural way to understand the often-observed initial increase and subsequent decrease in the width of the coherent island size distribution. [S0163-1829(99)50728-5]

Heteroepitaxy begins with the formation of a thin, lattice-matched wetting layer if the energy gain from substrate-adlayer adhesion exceeds the elastic energy cost from lattice constant misfit $\delta a/a$. As deposition continues, two-dimensional (2D) islands nucleate on top of the wetting layer. These islands contribute to the buildup of elastic strain and, for this reason, the system does not tolerate their growth, coalescence, and renucleation indefinitely. Instead, at large misfit, coherent (undislocated) three-dimensional (3D) islands form that are lattice matched near their base but are largely strain relieved near their top and sidewalls. Further deposition leads to their growth and eventual coalescence. This is the so-called *Stranski-Krastanov* growth mode.[1]

A coherent island is the source of strain fields because it elastically distorts the wetting layer and substrate in its immediate vicinity. Early on, several experimental groups observed a significant decrease in the mean size of the coherent islands at relatively early stages of growth and suggested the possible role of long-range strain fields. For example, Ponchet *et al.*[2] presented data for the InAs/InP(001) system and pointed out that elastic interactions should cause islands to *destabilize* one another because their interactions are mutually repulsive. Kobayashi *et al.*[3] identified several other features of the island-island interaction as a basis for understanding their experiments on the InAs/GaAs(001) system. Theoretical work on island interactions has been limited to *equilibrium* considerations[4] up to the present time.

In this paper, we present a theoretical analysis of Stranski-Krastanov growth kinetics that generalizes previous work by Dobbs *et al.*[5] to take acccount of island interactions and atom detachment from 3D islands. Dobbs *et al.* employed a mean-field theory for the density of adatoms n_a, the density of 2D islands n_2, their average size s_2, the density of 3D coherent islands n_3, and their average size s_3. A rate equation was derived for each based on the physical processes of adatom deposition, surface diffusion, attachment and detachment of adatoms from the islands, etc. In brief, an incident flux F contributes directly to the increase of adatom popuation. The adatoms diffuse on the surface with a diffusion constant $D=\omega a^2\exp(-E_s/k_BT)$ where ω is an attempt frequency, a is the lattice constant, E_s is the energy barrier for diffusion, k_B is the Boltzmann constant, and T is the temperature. Diffusing adatoms that meet bond together to form small 2D islands but thermal fluctuations can cause them to break apart if the island size is too small. There is a critical island size i such that islands of size i and less are unstable.

An island grows by capturing adatoms from both the vapor and the substrate. The rates for these processes are $F\kappa$ and $D\sigma n_a$, where κ is the direct capture number and σ is the diffusion capture number. To relieve strain, 2D islands convert into 3D islands at a rate γ_2. We assume that atoms that detach from the edges of a 2D island do not leave the island but instead migrate to the top of the island. On the other hand, we suppose that atoms *do* detach from 3D islands (at a rate $1/\tau_3$) when interactions become significant. A fraction m_2 of these attach to 2D islands. The remaining fraction m_a contributes to the adatom population. In this work, we approximate m_2 by the areal coverage of 2D islands.

Rate equations that incorporate all of these elementary processes are

$$\begin{aligned}
\dot{n}_a &= F[1-(i+1)\kappa_i n_i-\kappa_2 n_2-\kappa_3 n_3]-D[(i+1)\sigma_i n_i \\
&\quad +\sigma_2 n_2+\sigma_3 n_3]n_a+m_a n_3/\tau_3, \\
\dot{n}_2 &= F\kappa_i n_i+D\sigma_i n_i n_a-\gamma_2 n_2, \\
\dot{n}_3 &= \gamma_2 n_2, \qquad (1) \\
(s_2\dot{n}_2) &= F[(i+1)\kappa_i n_i+\kappa_2 n_2]+D[(i+1)\sigma_i n_i+\sigma_2 n_2]n_a \\
&\quad +m_2 n_3/\tau_3-\gamma_2 s_2 n_2, \\
(s_3\dot{n}_3) &= F\kappa_3 n_3+D\sigma_3 n_3 n_a+\gamma_2 s_2 n_2-n_3/\tau_3.
\end{aligned}$$

The suffixes i, 2, and 3 for κ and σ denote critical nuclei (of size i), 2D and 3D islands, respectively.

We assume that 2D islands are circular with radius r and 3D islands are truncated pyramids with base length l, height h, and base angle ϕ. The radius of a 2D island is $r=\sqrt{s_2/\pi}$. We assume that 3D islands very quickly achieve their equilibruim shape and that the angle ϕ does not change significantly during growth. For a given island size s_3, h and l are found by minimizing the energy expression derived for a 3D coherent island by Tersoff and Tromp.[6]

The direct capture number κ is given by the surface area of the island normal to the incident flux, i.e., $\kappa_2 = s_2$ and $\kappa_3 = l^2$. The diffusion capture number σ measures the efficiency with which an island captures adatoms from the surface. We compute σ using the prescription of Bales and co-workers,[7] which relates it to the size of the diffusional depletion zone ξ that surrounds each island. For a circular island of radius r with no barrier to adatom attachment we solve self-consistently

$$\sigma = 2\pi \frac{rK_1(r/\xi)}{\xi K_0(r/\xi)}, \tag{2}$$

$$\xi^{-1} = \sqrt{F\kappa_i/D + (i+1)\sigma_i n_i + \sigma_2 n_2 + \sigma_3 n_3}, \tag{3}$$

where $K_n(x)$ is the modified bessel function of order n. We use Eq. (2) for circular 2D islands and for σ_3 as well (with r replaced by $l/2$) because the details of the island shape should not affect the results significantly.

Conversion of a 2D island to a 3D island occurs when a sufficient density of atoms is present on its top (due to strain-driven detachment from its perimeter and upward migration) to nucleate a new island at its center. The requisite nucleation rate is[5]

$$\gamma_2 = \pi r^2 D \exp[(E_i - (i+1)E_d(r))/(k_B T)], \tag{4}$$

where E_i is the binding energy of critical nuclei and $E_d(r) = E_0 \ln(r/a)/(r/a)$ is a size-dependent energy barrier for the detachment of atoms from the 2D island. The form of γ_2 as a function of r is such that 2D islands barely convert at all until they reach a size s^* after which most of them convert very rapidly.

The escape rate of an atom from a 3D island is

$$\frac{1}{\tau} = \frac{D}{a^2} \exp\left(-\frac{E_b}{k_B T}\right), \tag{5}$$

where E_b is the energy barrier for detachment. Elasticity theory[5] predicts that the change in the barrier due to strain is $\Delta E_b \approx (\eta_u - \eta_s)\epsilon$, where ϵ is the local strain and $\eta_s(\eta_u)$ is the local surface stress at the binding site (transition state) configuration. The predicted linearity with strain has been confirmed by first-principles calculations.[8] The strain field due to a misfitting island is proportional to the size of the island and varies as d^{-3} for distances d far from the island.[9] We therefore put

$$E_b = E_b^0 - 2\pi\alpha s_3 \left(\frac{a}{d_3}\right)^3, \tag{6}$$

where E_b^0 is the strain-independent part of the energy barrier and $d_3 = 1/\sqrt{n_3}$ is the average 3D island separation. We treat α as an adjustable parameter because the surface stress difference discussed above is difficult to estimate. The factor of 2π is, in this model, the mean number of islands that are nearest neighbors to a given island.

The rate equations were integrated numerically using an algorithm suited for systems of stiff differential equations.[10] We used values of the parameters typical of those found in experiments, $T = 900$ K, $F = 0.1$ ML/s, $a = 3.0$ Å , $i = 4$, $E_0 = 3.5$ eV, $E_i = 0.5$ eV, $E_s = 1.0$ eV, $E_b^0 = 0.7$ ev, $\phi = 25°$, and $\delta a/a = 0.05$. Our results for the time (coverage) evolution of the 3D island density and mean size are shown in Figs. 1(a) and 2(a). For comparision we have plotted the experimental results of Kobayashi *et al.* for InAs/GaAs(001)[3] in Figs. 1(b) and 2(b). The sizes were estimated from the published experimental distributions of island heights and island widths. Note also that we have shifted the theoretical curves to align the rapid island density onsets because the precise onset position is related to alloying[11] that we do not attempt to model.

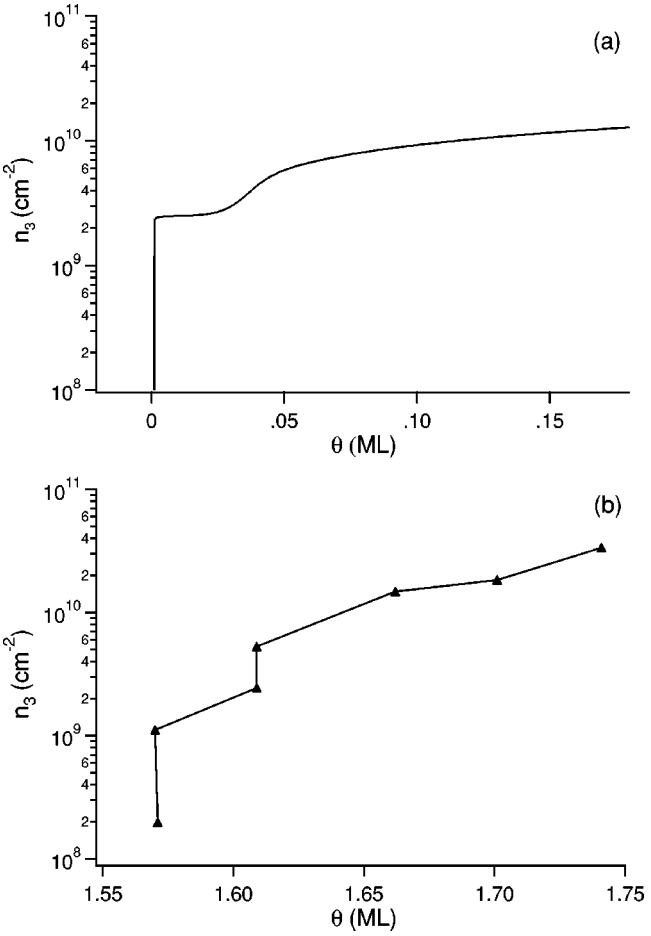

FIG. 1. 3D island densities: (a) from the present theory, (b) from the data of Kobayashi *et al.* (Ref. 3).

The 3D island density initially rises very rapidly due to the fast conversion of 2D islands to 3D islands. It then tends to saturate because, as a result of conversion, the average 2D island size decreases below s^*. During this time the average 3D island size continues to grow. Soon the interactions become important and significant detachment of atoms from the 3D islands begins. This results in the very rapid decrease of s_3 seen in Fig. 2(a). The detached adatoms that reattach to 2D islands increase the average size of the latter to s^*, which, in turn, leads to more 2D to 3D conversion. That is why the 3D island density increases again. The same trend is seen in the experimental data although we do not obtain quantitative agreement between our model and the data.

The results shown correspond to $\alpha = 120$ eV, which is three orders of magnitude greater than typical elastic energies. This large number arises in our model because the rapid decrease in 3D island size seen in the data of Kobayashi *et al.*[3] occurs when the experimental mean island separation

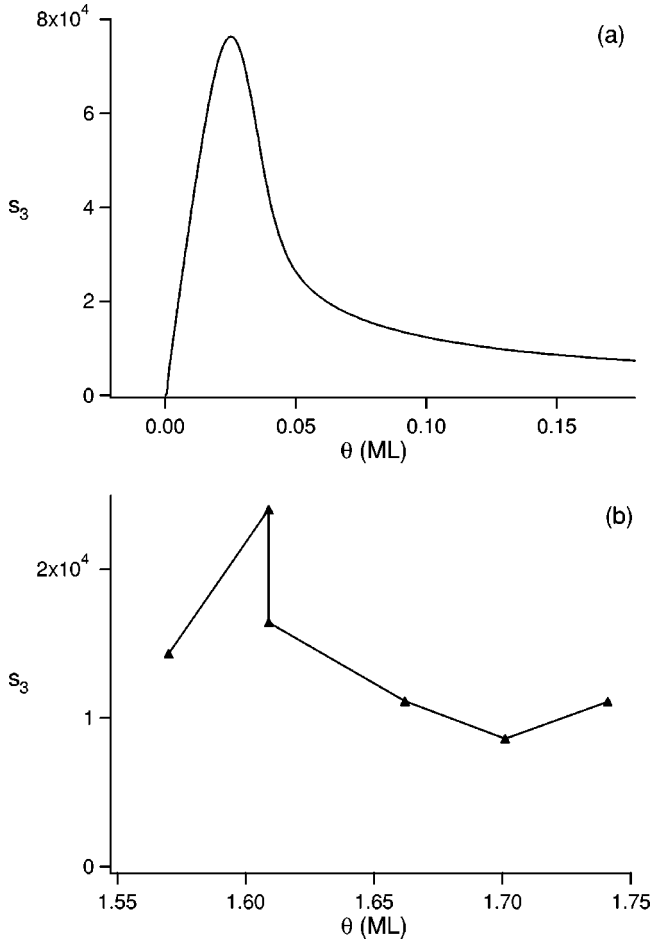

FIG. 2. Average 3D island size: (a) from the present theory, (b) estimated from the data of Kobayashi *et al.* (Ref. 3).

is ten times larger than the mean island radius! Of course, the real system has many islands at much closer distances than our simple mean theory can describe, but it remains the case that detachment effects seem to set in far earlier than simple elasticity estimates would suggest. The detailed origin of this behavior is an outstanding open question and our simple form (6) must be regarded as a convenient parametrization.

In principle, the entire island size distribution can be gotten from a rate equation analysis. In practice however, it is prohibitively difficult to solve the tens of thousands of equations so generated. This theoretical problem is ameliorated for the case of 2D *homoepitaxy* because the island size distribution shows scaling behavior.[12] It is therefore highly significant that Ebiko *et al.* have shown that the 3D coherent island size distribution for the InAs/GaAs(001) system also shows scaling.[13] Their data fits remarkably well to an analytic scaling form suggested for 2D homoepitaxy.[14] In detail, the number of islands of size s, n_s takes the form

$$n_s = \frac{\theta_c}{\langle s \rangle^2} f\left(\frac{s}{\langle s \rangle}\right), \tag{7}$$

where $\theta_c = \Sigma_s s n_s$ and

$$f(u) = 1.1u \exp(-0.27u^{3.7}). \tag{8}$$

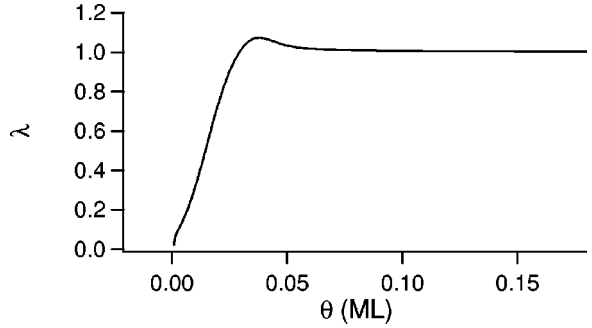

FIG. 3. Coverage dependence of λ, the detachment rate to attachment rate ratio for a 3D island.

It is surprising that an island distribution that works well for 2D islands works equally well for 3D islands. Even more puzzling is the fact that Eq. (8) applies only to situations where atom detachment from 2D islands is strictly forbidden ($i=1$) whereas the coherent islands studied here shrink precisely due to copious detachment.

This can be understood if we parametrize the island size distribution not by a fictitious ''critical island size'' but by the ratio of the net detachment rate from an island to the net attachment rate to an island,[15] namely,

$$\lambda = \frac{1/\tau}{(Fk + D\sigma n_a)}. \tag{9}$$

Monte Carlo simulations of 2D homoepitaxy show that λ parametrizes a continuous family of scaling functions.[16] When $\lambda \sim 1$ or less, the island size distribution fits Eqs. (7) and (8) very well even when significant detachment is present. The computed time evolution of λ for our model is shown in Fig. 3. Note that its value exceeds unity when the island interactions are most important but only barely so. This is *not* inconsistent with the rapid decrease in the aver-

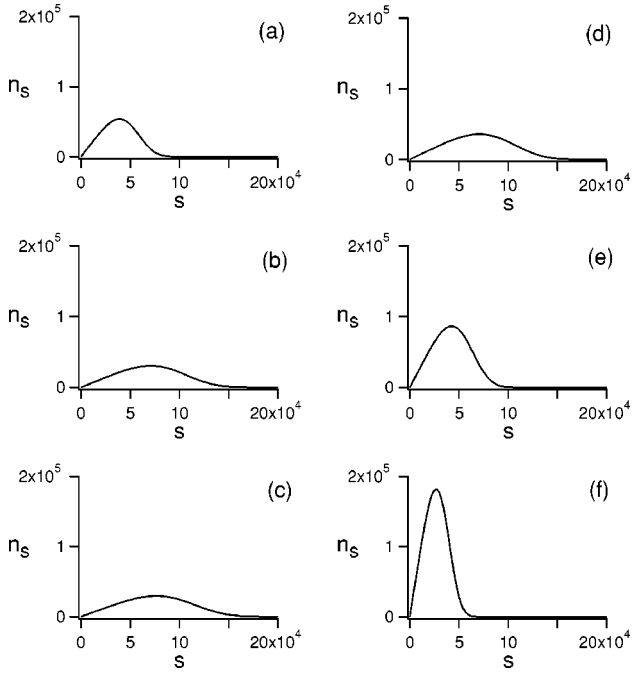

FIG. 4. Evolution of the island size distribution: (a) $\theta=0.01$, (b) $\theta=0.02$, (c) $\theta=0.025$, (d) $\theta=0.03$, (e) $\theta=0.04$, (f) $\theta=0.05$ (ML).

age coherent island size seen in Fig. 2 because the rate equation for this quantity in Eq. (1) involves the *difference* (rather than the ratio) of the attachment and detachment rates which can be large. These considerations provide a rationale for the fitting procedure used by Ebiko *et al.*[13]

We conclude that island interactions strongly influence the average island size but not the island size distribution scaling function. This is important because it means that we can ''synthesize'' the time dependence of the entire island size distribution merely from knowledge of the time dependence of the average size. This is shown in Fig. 4. As expected, the island size distribution broadens and its peak position moves to the right as the coverage increases from zero. But as a consequence of Eqs. (7) and (8), the decrease of s_3 when interactions become imporant induces a narrowing of the distribution and a shift back to the left. Precisely this behavior is seen in the experimental island size distributions.[2,3]

In summary, we have generalized the theory of Dobbs *et al.*[5] to take account of island-island elastic interactions that are presumed to induce atom detachment from 3D coherent islands. Semiquantitative agreement was found with experimental results for InAs/GaAs(001) but the large value for the interaction parameter needed to model the data suggests that we still lack a good understanding of the energy barriers to detachment for this problem. In conjunction with a scaling ansatz, the results could be used nonetheless to rationalize the ubiquitous ''narrowing'' of the full island size distribution seen in experiment. An interesting and open question is to establish the veracity of this scaling assumption in a theoretical framework.

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