Theory of strained-layer epitaxial growth near step flow

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(Received 13 June 1994)

A reaction-diffusion theory of homoepitaxial growth on a stepped surface introduced by Myers-Beaghton and Vvedensky [Phys. Rev. A **44**, 2457 (1991)] and Fuenzalida [Phys. Rev. B **44**, 10 835 (1991)] is generalized to the case of heteroepitaxy by taking account of misfit strain relief in the growing epilayer. For deposition conditions where two-dimensional (2D) islands nucleate on the terraces before individual adatoms can migrate to vicinal step edges, analysis of the distribution of island sizes resolved in both space and time reveals a strain-driven transition from the familiar layer growth mode dominated by 2D island coalescence to a form of step flow where moving steps engulf very many very small 2D islands. The latter mode of growth is predicted to persist until three-dimensional island formation disrupts layer growth altogether. Both with and without strain, it is found that the island size distribution averaged over a terrace very rapidly approaches a steady-state form, even when kinematic theory predicts oscillations in the signal from diffraction measurements.

I. INTRODUCTION

It has been understood on theoretical grounds for at least forty years that epitaxial crystal growth onto vicinal surfaces involves a competition between the nucleation, growth, and coalescence of two-dimensional (2D) islands on terraces and the direct capture of deposited adatoms by the steps that bound the terraces. Explicit discussion of the first of these dates back to Volmer's authoritative study of the kinetics of phase change[1] while the latter so-called *step-flow* mode of growth was introduced in a seminal paper by Burton, Cabrera, and Frank (BCF).[2] Nonetheless, convincing experimental confirmation of these ideas for vapor-phase growth of single crystals has been achieved only rather recently by the use of *in situ* electron and x-ray diffraction techniques during homoepitaxy of high-quality semiconductor materials by molecular-beam epitaxy (MBE),[3] metal-organic molecular-beam epitaxy (MOMBE),[4] and organometallic vapor-phase epitaxy (OMVPE).[5] The diffraction signal is time independent in the step-flow regime but develops periodic oscillations in the nucleation regime reflecting cyclical variations in the morphology of the growth front.

These experiments reveal that a smooth transition from the nucleation-dominated growth regime to the step-flow growth regime can be achieved either by an increase in substrate temperature or by a decrease in the deposition flux. Detailed study of this transition evidently falls within the purview of fundamental studies of the kinetics of growth.[6,7] But it is important as well for process control in the fabrication of epitaxial architectures such as quantum wires,[8,9] where maintenance of step flow is essential. With this motivation, both Monte Carlo computer simulations[10] and numerical solutions of reaction-diffusion equations[11-13] have been brought to bear on the mode transition problem with notable success.

The purpose of the present paper is to demonstrate how this transition can be effected solely by the presence of epitaxial misfit strain. Our motivation is twofold. First, there have been recent demonstrations that the step-flow mode of growth can be maintained during heteroepitaxy.[14-17] Second, it is a challenging theoretical problem to incorporate strain effects into models of epitaxial *kinetics*. To our knowledge, all previous theoretical studies of this general question fall into one of two categories. Computer simulations that involve Monte Carlo[18-26] or molecular-dynamics[27-31] technology are popular but perforce are restricted severely by computer time considerations. This problem does not exist with continuum modeling,[32,33] but atomistic effects are completely lost with this intrinsically long-wavelength approach to the problem. In any event, all these studies have been concerned with singular (no miscut) surfaces and focus either on defect generation or three-dimensional island formation. As will become evident, neither of these issues is germane to the mode transition question.

Our analysis generalizes to the case of heteroepitaxy a previously developed reaction-diffusion theory of homoepitaxial growth on vicinal surfaces.[11-13] The latter itself generalizes the usual BCF theory of step flow to include 2D island nucleation, growth, and dissolution at the level of rate equation treatments[34] usually employed to analyze growth onto singular surfaces. This theory neglects desorption and the mobility of all island species except single atoms (as is likely for the growth of semiconductors by MBE) but includes free parameters that describe the rates of kinetic processes such as deposition, adatom surface diffusion, adatom attachment to 2D islands and step edges, and adatom detachment from 2D islands and step edges. To incorporate epitaxial misfit into this formalism, we suppose here that strain alters these rate constants in a well-prescribed manner. Since we shall do so *without* recourse to the computation of individual interatomic separations, our approach to the rate constants most nearly resembles that adopted in some computer experiments devoted to this problem.[21,26]

Our principal result is that, so long as layer-by-layer growth is maintained, epitaxial strain induces the dissociation of 2D islands and thus promotes a form of step

flow. No account is taken of possible strain-relieving reconstructions *within* the advancing terraces[15-17] but a careful analysis of the evolution of the island size distribution function as a function of time and position along the terraces reveals that strain-driven "step flow" is *not* identical to temperature-driven or flux-driven step flow. The former obliterates larger islands much more effectively than the latter and leads to a steady-state dominated by very many very small 2D islands. This surface morphology should be detectable in "quench and look" scanning tunneling microscopy (STM) experiments as has been done previously[35] for strained layers on nominally singular substrates. We also find that the island size distribution function integrated over the terrace width very rapidly achieves a time-independent shape. This is true even as one moves out of step flow and the *spatial* distribution of islands varies periodically in time and thus yields periodic structure in the kinematic diffraction signal. *Post facto* justification thus is provided for our earlier brief discussion of the present problem[36] that *assumed* the existence of such steady-state behavior.

The organization of the remainder of this paper is as follows. Section II reviews the basic reaction-diffusion theory for homoepitaxy and presents some results for the time evolution of the 2D island size distribution function not addressed in previous studies. The extensions to the theory required to take account of misfit strain are described in Sec. III while Sec. IV is devoted to a discussion of the results. Section V contains a summary and some suggestions for experimental tests of our predictions.

II. THE BASIC MODEL FOR HOMOEPITAXY

In this section we review the generalized BCF theory of homoepitaxial growth onto vicinal surfaces in the time-dependent formulation of Myers-Beaghton and Vvedensky.[12] As in the original BCF discussion,[2] the substrate is assumed to consist of a perfectly uniform staircase of terraces of width l (the lattice constant is set equal to 1) bounded by straight steps of unit height. The premise of straight steps is equivalent to the assumption that the density of kinks is sufficiently great that the diffusion problem on the terraces may be regarded as one dimensional. The areal density of mobile adatoms across the terrace, $n(x,t)$, and the areal density of immobile 2D islands composed of i atoms, $n_i(x,t)$, thus should be regarded as *averaged* with respect to the spatial coordinate parallel to the steps.[13] Complications due to crystalline anisotropy, reconstruction, and other chemical details are neglected.

In a frame of reference that moves with the steps, the defining equations of motion are

$$\frac{\partial n}{\partial t} = D\frac{\partial^2 n}{\partial x^2} + J + v\frac{\partial n}{\partial x} - (2Dn + Jm_1)n$$
$$- \sum_{j=2}^{N} n_j(nD + Jm_j) + 2k_2n_2 + \sum_{j=3}^{N} k_jn_j ,$$

$$\frac{\partial n_i}{\partial t} = v\frac{\partial n_i}{\partial x} + Dn(n_{i-1} - n_i) + J(m_{i-1}n_{i-1} - m_in_i)$$
$$+ k_{i+1}n_{i+1} - k_in_i \quad (i \geq 2) ,$$

(1)

where J is the deposition flux and D is the adatom surface diffusion jump rate. The instantaneous step velocity v appears in convection terms that must be retained[37] for growth rates typical of vapor-phase epitaxy (~ 1 ML/sec). The quantities k_i denote the rates at which single atoms *detach* from islands of size i. These constants appear in first-order terms while, in this Smoluchowski-type treatment,[38] the rates at which adatoms *attach* to islands of any size are treated as second-order processes controlled by the surface diffusion constant D. The terms proportional to the factors Jm_i count the effect of deposition events where an arriving atom lands either immediately adjacent to or on top of an existing island of size i. In the latter case, we suppose that the freshly deposited atom rapidly diffuses to the island edge and incorporates. This implies that the ratio J/D cannot be arbitrarily large.

Mass conservation and flux balance considerations at the step edges are sufficient to complete the specification of the problem.[2,12] The most general boundary condition for the monomers is obtained if both attachment (k_+) and detachment (k_-) processes at such edges are allowed:

$$D\frac{dn}{dx}\bigg|_{x=0} = k_+n(0) - k_-n_S ,$$

$$D\frac{dn}{dx}\bigg|_{x=l} = -k_+n(l) + k_-n_S .$$

(2)

The number density of kinks along the step n_S is assumed to be constant but with a value that depends upon the deposition conditions. The requirement that no islands form atop a freshly formed step yields the condition

$$n_i(l,t) = 0 \quad (i \geq 2) ,$$

(3)

while the step velocity is determined from

$$v = D\frac{dn}{dx}\bigg|_{x=0} - D\frac{dn}{dx}\bigg|_{x=l} + v\sum_{j=2}^{N} jn_j(0,t) .$$

(4)

Note that v is obtained *self-consistently* as growth proceeds since (4) requires knowledge of the densities $n_j(0,t)$ computed from (1). This may be contrasted with the steady-state situation[11,13,36] where the step velocity $v = Jl$ for the case of no desorption studied here.

To make progress, we must assign values to the various kinetic parameters defined above. The choices to follow mostly conform to those of Ref. 12, to which we refer the reader for further discussion and justification. Accordingly, we set $J = 1$ and parametrize $D = v\exp(-E_D/k_BT)$ where $v \sim 10^{13}$/sec and E_D is an adatom surface migration barrier. The choices $k_+ = \frac{1}{4}D$, $k_i = k_+\exp(-E_N/k_BT)$, and $k_-n_S = k_+\exp(-2E_N/k_BT)$ establish E_N as an effective *lateral* bond energy between nearest-neighbor surface atoms. This is so because we identify doubly coordinated kink site atoms as those that detach from a step, whereas almost all small compact islands exhibit singly coordinated species on a square lattice. For most of the results to be discussed below we set $E_N/E_D = 0.4$ eV and $l = 20$.

For an island of size j, the integer m_j is equal to the number of atomic sites occupied by the island ($=j$) *plus* the number of unoccupied sites immediately adjacent to the island. The latter can be counted explicitly for very small islands and varies approximately as $4\sqrt{j}$ for larger islands.

Figure 1 illustrates typical results for $n_i(x)$ obtained by numerical integration of the rate equations for $E_D/k_BT = 18.2$. Observe that the spatial distributions of island densities across the terrace rapidly approach time-invariant shapes that exhibit a distinct skewing of the distributions toward the trailing step edge ($x = 0$). Although small for our choice of deposition flux, this skewing effect is present even for the monomer concentration and is well understood as a consequence of the motion of the coordinate system.[37] Such shapes have been obtained previously in studies[11,13] that adopted a steady-state approximation to the rate equations. The lack of spatial variation in the total number density of island species after the first fraction of a monolayer has been grown is made particularly evident from a calculation of the intensity of scattering expected from this surface in the kinematic approximation:[39]

$$I(t) \propto \left| \int_0^l \left[1 - 2 \sum_{j=1}^{N} j n_j(x,t) \right] e^{i\pi x} dx \right|^2 . \tag{5}$$

This formula may be expected to approximate the signal obtained from a reflection high-energy electron-diffraction (RHEED) or thermal-energy atom-scattering (TEAS) experiment performed under "out-of-phase" conditions where scattering from terraces separated by one atomic step interferes destructively.

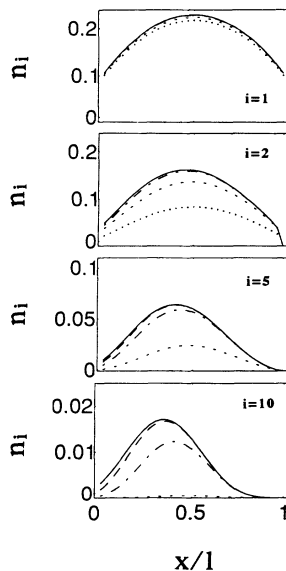Figure 2 illustrates $I(t)$ as computed for several values of E_D/k_BT which, for discussion purposes, can be re-

FIG. 2. Kinematic scattering intensity oscillations computed from (5) for the step-flow region ($E_D/k_BT = 18.2$; dashed line), the transition region ($E_D/k_BT = 18.5$; dotted line), and the nucleation region ($E_D/k_BT = 19.0$; solid line).

garded as obtained at successively lower temperatures from top to bottom. The uppermost trace (corresponding to Fig. 1) decreases monotonically and does indeed achieve a time-independent value after about $\frac{1}{2}$ ML of growth. At the lowest temperature shown, the initial decrease in scattering intensity is largest and well-defined oscillations are evident. As is well known,[7] this transition from a time-invariant signal to an oscillatory signal is the signature that step-flow growth has been supplanted by the nucleation, growth, and coalescence of 2D islands on the terraces.[40] The oscillation period is the monolayer completion time and the larger initial drop in scattering intensity is related to the increased surface roughness associated with the nucleation of islands.

A particularly useful way to quantify this growth mode transition makes use of island size distribution functions obtained by integrating concentration profiles such as those in Fig. 1 over the length of the terrace.[12] Two closely related distributions of this sort are presented in Fig. 3 at a coverage of $\Theta = 0.75$ ML. The upper panel is

FIG. 1. Spatial distribution of islands of size i. The different curves represent results at coverages 0.01 (dotted), 0.05 (short dashed), 0.25 (dash dotted), 0.5 (long dashed), and 0.75 ML (solid).
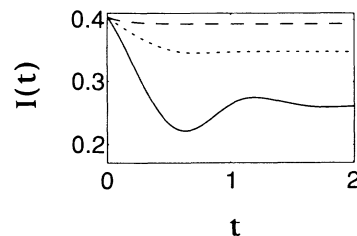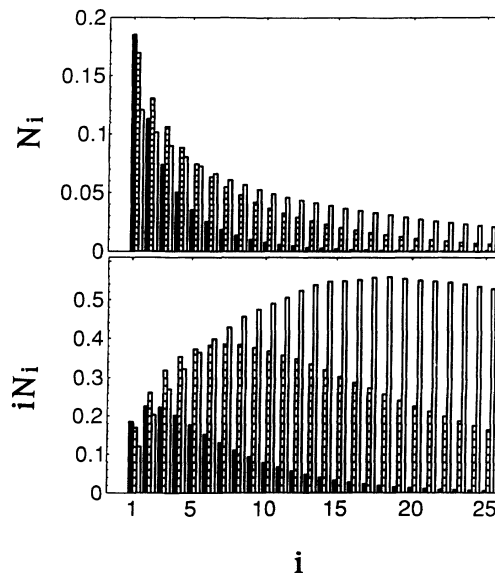

FIG. 3. Number of islands $\mathcal{N}_i$ (upper panel) and total number of atoms $i\mathcal{N}_i$ as a function of the island size i. Bar graphs shown represent results for $F = 1.0$ and $E_D/k_BT = 18.2$ (black), 18.5 (cross hatched), and 19.0 (white).

a plot of $\mathcal{N}_i = \int_0^i n_i(x)dx$ vs i while the lower panel shows $i\mathcal{N}_i$ vs i. The latter is the total number of atoms in islands of size i,[12,13,36] and is useful as a representation of the numerical data in so far as it accentuates some of the effects of interest. In particular, the competition between island nucleation and adatom capture by vicinal step edges favors monomers (large islands) as the dominant surface species at high (low) temperature. A similar result is obtained at constant temperature for small (large) values of the deposition flux (Fig. 4). All of these observations are consistent with simple physical arguments based on homogeneous nucleation theory.[41]

Interestingly, our calculations show that the *shapes* of the integrated island size distributions are very nearly time invariant after a short transient. This is obvious for the step-flow regime given the behavior illustrated in Fig. 2 (dashed line). But it turns out to be true for the nucleation regime as well (Fig. 5), where the *spatial* distribution of islands varies in time so as to produce the oscillations in the kinematic intensity shown in Fig. 2 (solid line). This observation provides some justification for previous studies that focused exclusively on steady-state solutions to the rate equations.[11,13,36] On the other hand, the simple reaction-diffusion methodology we use becomes unreliable far into the nucleation regime, since the real-space effect of finite island sizes is not taken into account properly. For this reason, the present study is restricted to the *transition regime* which we define as centered at the temperature at which $I(t)$ first ceases to be monotonic upon cooling. Although somewhat arbitrary, this definition has the virtue that it is easy to apply to actual experimental data. Accordingly, for the parameters adopted here, the transition occurs very near $E_D/k_B T = 18.5$ for the case of homoepitaxy (cf. Fig. 2).
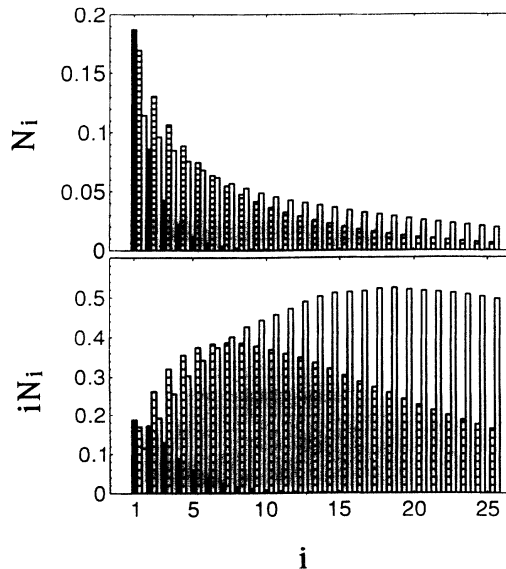

FIG. 5. Number of islands $\mathcal{N}_i$ as a function of the island size i in the nucleation region ($E_D/k_B T = 1.0$). The different panels show results at coverages $\Theta = 0.25$ (a), 0.75 (b), and 1.25 ML (c).

III. EXTENSION TO STRAINED-LAYER GROWTH

In this section, we generalize the rate equations introduced above to take account of the elastic strain that must develop when there is lattice misfit between the deposited material and the substrate material. Other, purely chemical effects associated with heteroepitaxy on vicinal surfaces are ignored here but have been discussed by us elsewhere.[42] The purpose of the present calculation is to demonstrate that such strain induces a mode transition from 2D island nucleation and coalescence to a variant of step flow. The results are expected to be valid until the accumulated elastic strain energy in the growing film forces the system to abandon layer-by-layer growth in favor of three-dimensional islanding.[43] The film thickness when this occurs depends on both the material parameters and the deposition conditions.[25,44]

The basic premise of this work is that the effect of strain on the kinetics of layer growth can be captured adequately by suitable modification of the energy barriers that appear in the Arrhenius rate coefficients for (i) surface diffusion and adatom attachment and (ii) detachment from islands and step edges. Consider first the isolated effect of strain on the rate of single-atom surface diffusion. To our knowledge, only three molecular-dynamics simulation investigations have been addressed to this question specifically. The first of these[45] studied

FIG. 4. Number of islands $\mathcal{N}_i$ (upper panel) and total number of atoms $i\mathcal{N}_i$ in an island of size i as a function of the island size i. Bar graphs shown represent results for $E_D/k_B T = 18.5$ and $F = 0.5$ (black), 1.0 (cross hatched), and 2.0 (white).
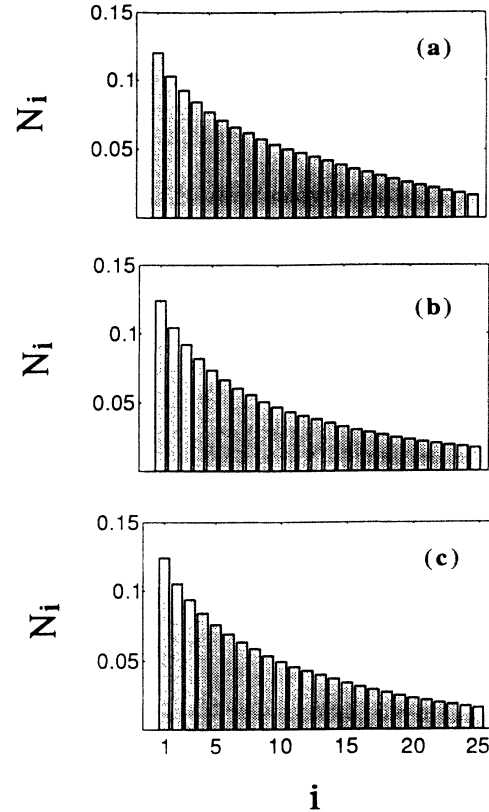
gallium atom motion in the vicinity of a step on GaAs(100). Although some systematic effects of strain were reported, the empirical potential used in this work subsequently was shown[46] to yield a gallium-terminated surface that is unstable at low temperature. The conclusions of Ref. 45 thus cannot be accepted with great confidence. By contrast, extensive simulations of silicon atom diffusion on strained and unstrained Si(100) surfaces have been performed[47,48] that make use of a well-tested empirical potential that has been used with considerable success in kinetic Monte Carlo simulations of epitaxial growth on this surface.[49] Among other things, it is found that the barrier to surface diffusion in the channels parallel to the dimer rows of the reconstructed surface is *reduced* by about 10% for either 2% tensile stress or 3% compressive stress but that the barrier to diffusion in the same direction but atop the dimer rows is *increased* by about 10% under these same conditions. Clearly, the effects of strain are nontrivial and even the *sign* of changes to the diffusion barriers is not easily guessed on the basis of chemical intuition for this covalently bonded system.

Given the foregoing, we believe it is appropriate to regard the strain dependence of the barrier to adatom surface diffusion as a function to be determined on a case-by-case basis. But it turns out that we can explore the qualitative consequences of any physically reasonable choice for this function without any additional calculations. This is so because our parametrization of the rate coefficients guarantees that every term in (1) depends linearly on either the adatom diffusion constant D or the deposition flux J. Hence an increase (decrease) of E_D due to misfit strain corresponds *exactly* to an appropriate increase (decrease) in flux. The distribution functions in Fig. 4 can be reinterpreted accordingly if no other strain-dependent effects are operative. When strain increases the rate of surface diffusion, adatom mobility[50] increases so that more atoms reach the step edge before dimer formation can ensue. The system is driven away from the transition regime and toward the step-flow regime. Conversely, when strain decreases the rate of surface diffusion, the probability increases that any given atom will encounter a critical nucleus on the terrace before it reaches the step edge. The system is driven away from the transition regime and toward the nucleation regime.

We turn now to the isolated effect of epitaxial strain on the rate coefficients for adatom attachment and detachment from islands and step edges. Part of the answer has been given already since, in our treatment, all these quantities are directly proportional to the surface diffusion constant. For simplicity, we will suppose that any additional strain-induced changes to the barrier to adatom *attachment* are sufficiently small and can be neglected. But changes to the barrier to adatom *detachment* must be consistent with the response of the system to the fact that the local free energy of every atom in the interior of a strained island or terrace exceeds that of an unstrained island or terrace. We assert[36] that the kinetic consequences of this fact are described most properly by the renormalizations

$$k_i \rightarrow k_i \exp\left[\frac{E_{\text{strain}}(i)}{k_B T}\right] , \tag{6a}$$

$$k_- \rightarrow k_- \exp\left[\frac{E_{\text{strain}}(\infty)}{k_B T}\right] , \tag{6b}$$

where $E_{\text{strain}}(i)$ is the strain energy per atom of a 2D epitaxial island composed of i atoms. Our choice (6a) is consistent with the kinetic schemes advanced for the closely related problem of strain-induced 3D islanding in the analytic work of Stoyanov,[51] Srolovitz,[32] and Spencer et al.[33] but differs somewhat from the choices made by Orr et al.[25] and Grandjean and Massies[26] in their Monte Carlo simulations.

In principle, the function $E_{\text{strain}}(i)$ for a given island is not independent of the disposition of neighboring islands (due to the long range of elastic strain fields) and proper atomistic approaches to its calculation are not difficult to imagine.[18,25] But since the computational time requirements are not consistent with the survey nature of the study proposed here, we adopt instead a simple approximation that we believe captures the essential features of the problem. More precisely, we suppose that

$$E_{\text{strain}}(i) \simeq 2E_{\text{FK}}(\sqrt{i}) \tag{7}$$

where $E_{\text{FK}}(N)$ is the total energy per atom of a *one-dimensional* chain of N harmonically coupled atoms in contact with a rigid sinusoidal potential. The latter will be recognized as a finite-chain version of the familiar Frenkel-Kontorova model[52] of epitaxy.[53] For the 1D problem, this model has the great virtues that it can be solved analytically[54] (in the continuum limit) and that it correctly reproduces the important and experimentally verified[55] phenomenon of lateral strain relaxation at the edges of epitaxial islands. Very small islands are essentially unstrained. As one might expect, $E_{\text{FK}}(N)$ increases quadratically with misfit and increases rapidly and then saturates as N increases. The choice (7) implies (i) that the islands are compact and (ii) that the energy density of a square island is well approximated by summing the energy density from two orthogonal *noninteracting* chains of atoms. The first of these is equivalent to the assumption that diffusion along island step edges is rapid.[56] The second point is implicit in the numerical results obtained by Snyman and Van der Merwe[57] in their analysis of a fully coupled 2D Frenkel-Kontorova model. In this paper, we use a parametrization of $E_{\text{FK}}(N)$ appropriate to semiconductor materials used previously by us[44] in a study of the Stranski-Krastanov epitaxial morphology. It turns out that the exponential factor in (6a) increases sublinearly with i for all island sizes of interest in this paper.[58] We appeal to these earlier calculations also for the result that 2D islands are thermodynamically stable against 3D island formation and misfit dislocation injection for $i \leq 10^4$ at 2% misfit and for $i \leq 10^3$ at 4% misfit.

IV. RESULTS AND DISCUSSION

As for the case of homoepitaxy, numerical integration of the strain-modified rate equations demonstrates that

the distribution of islands in both space and time quickly achieves a time-invariant shape in the step-flow and transition regimes. We thus ignore all transients and focus exclusively on the asymptotic steady state. Also, in light of the discussion above regarding surface diffusion, we present results first assuming that epitaxial misfit acts only to modify the adatom detachment rate parameters. Figure 6 illustrates the spatial distribution of islands for three values of strain at $\Theta = 0.75$ ML and $E_D/k_B T = 18.5$, i.e., at the transition temperature for homoepitaxy (cf. Fig. 2). Although only subtle changes from the zero-strain case are evident at 2% misfit, a clear rearrangement has occurred at 4% misfit. Closer examination reveals that (i) almost all the change is associated with the portion of the terrace that is adjacent to the trailing step edge; (ii) the change involves a transfer of atoms from large islands to small islands; and (iii) the total number of atoms on the surface is not conserved. The first of these is associated with the effects of convection as before and represents a prediction that can be checked by microscopy. The other points are best appreciated upon integration over the spatial degree of freedom.

It is immediately clear from plots of either $\mathcal{N}_i$ or $i\mathcal{N}_i$ (Fig. 7) that strain systematically eliminates larger islands from the surface and increases the number of smaller islands. This is expected from (6a) and the qualitative behavior of $E_{\text{strain}}(i)$ noted above. Indeed, atomistic Monte Carlo simulations based on the same assumptions used in the present work confirm the strain-induced narrowing of the island size distribution evident from Fig. 7.[58] The fact that the total number of atoms present on the surface changes as a function of misfit leads to no inconsistencies so long as the total coverage on each terrace in the moving frame remains less than 1 ML. This is so because mass conservation between the island populations and the growing solid is built into the self-consistent
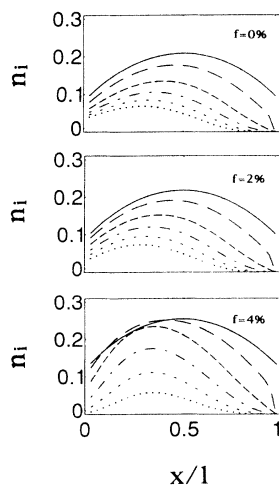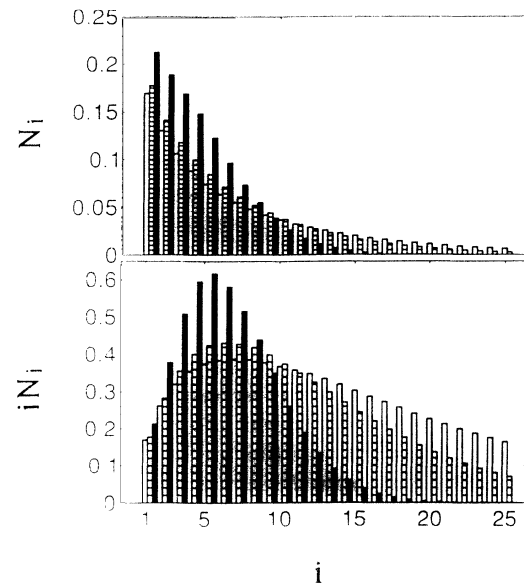

FIG. 7. Number of islands $\mathcal{N}_i$ (upper panel) and total number of atoms $i\mathcal{N}_i$ in an island of size i as a function of the island size i for $E_D/k_B T = 18.5$, $F = 1.0$, and misfit $f = 0\%$ (white), 2% (cross hatched), and 4% (black).

expression for the step velocity (4). What matters for the step velocity is not the total number of various island species but their distribution in space. Thus, in principle, the surface could be flooded with island species at large strain since, from (6b), the barrier to adatom attachment is most reduced precisely at the (concomitantly) slowly advancing vicinal step edges that bound the strained terraces. In the steady state,[36] the requisite strain-independent value $v = Jl$ is achieved by a strain-dependent partitioning of the monomer and immobile island contributions to v in (4).

Inspection of Fig. 7 also reveals that for each value of misfit there exists a critical island size i^* such that $i\mathcal{N}_i$ increases (relative to the zero-strain case) when $i \le i^*$ and $i\mathcal{N}_i$ decreases when $i > i^*$. Observe that it is not the case that the monomer population is the primary beneficiary of the detachment of single atoms from strained islands. This is so because almost all new monomers are eliminated by the rapid formation of dimers, trimers, and other small islands that do not break up at an accelerated rate since they are nearly unstrained. We thus predict a mode of "step flow" whereby steps advance mostly by engulfing very many very small islands. It would be very interesting to conduct a series of in situ TEAS or RHEED measurements or "quench and look" STM experiments to check this prediction.

Comparing Fig. 7 with Fig. 3, one sees that the changes in the island size distributions that occur at zero strain when one passes from the transition regime to the step-flow regime either by raising the substrate temperature or by lowering the deposition flux[59] are only qualitatively similar to the foregoing. As noted above, the most

FIG. 6. Spatial distribution of islands of size $i = 1$ (solid), 2 (long dashed), 4 (medium dashed), 6 (dash dotted), 8 (short dashed), and 10 (dotted) for different values of misfit f in the transition region ($E_D/k_B T = 18.5$ and $F = 1.0$).
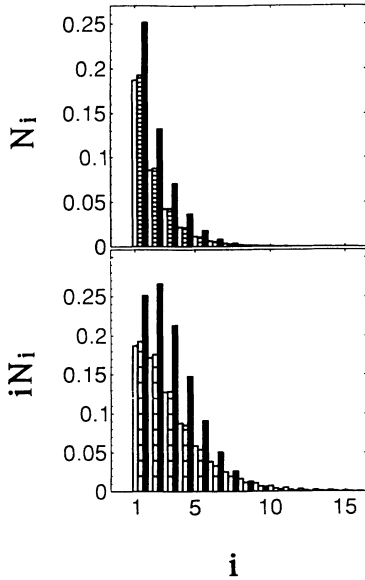
FIG. 8. Number of islands $\mathcal{N}_i$ (upper panel) and total number of atoms $i\mathcal{N}_i$ in an island of size i as a function of the island size i for $E_D/k_B T = 18.5$, $F = 0.5$, and misfit $f = 0\%$ (white), 2% (cross hatched), and 4% (black).

obvious quantitative difference is the fact that the island size distribution is significantly narrower when step flow is achieved by strain. This observation may be significant if these 2D structures form the nuclei for subsequent 3D coherent island formation since size uniformity is a figure of merit for quantum dots formed in this way.[60] It is also the case that the variation of the critical island size i^* with misfit for the case of heteroepitaxy differs considerably from the variation with temperature and flux of a corresponding quantity defined for the case of homoepitaxy. Both of these quantitative differences from the no-misfit case ultimately arise from the island size dependence of $E_{\text{strain}}(i)$, although the detailed behavior of i^* may be expected to depend strongly on the symmetry of the substrate.

It is not difficult to combine the isolated effect of strain on the adatom surface diffusion rate with the isolated effect of strain on the adatom detachment rate. For this purpose, Fig. 8 illustrates the effect of lattice misfit on the integrated island size distributions at a lower value of flux than in Fig. 7 so that conventional step flow occurs at zero strain. Now suppose that a system is prepared in the transition region without strain (Fig. 7, white bars). If misfit strain *increases* the rate of adatom diffusion, the system is driven toward the distribution depicted by the white bars in Fig. 8. The additional effect of strain on island dissolution then yields final island populations more nearly like the black bars in Fig. 8. Conversely, if strain *decreases* the rate of adatom diffusion, the two effects we have discussed tend to cancel one another and the system remains in the transition region, albeit with an increased population of small islands.

V. SUMMARY AND PROSPECTS FOR EXPERIMENT

In this paper, we have generalized a reaction-diffusion theory of homoepitaxial growth on a vicinal surface to the case of heteroepitaxy as a model for strained-layer growth near step flow. The effect of strain was introduced by modification of the Arrhenius energy barriers to adatom surface diffusion and adatom detachment from two-dimensional islands and step edges. A suitably modified Frenkel-Kontorova model was used to compute the relevant energies while direct numerical integration of coupled nonlinear equations of motion yielded the densities of the various island species in space and time. No account was taken of possible strain-relieving reconstructions within the advancing terraces.

Both with and without strain, we discovered that the island size distributions integrated over a full terrace width always rapidly evolve to a time-independent form even when the intensity of scattering from the surface in the kinematic approximation exhibits oscillations. For deposition conditions where 2D island nucleation is favored, strain was found to promote the fragmentation of large islands into small (essentially) unstrained islands. But the monomer population is smaller and the size distribution narrower than that obtained when island dissolution is induced by thermal detachment of atoms from the edges of unstrained islands. The resulting mode of strained layer growth is best characterized as a variant of step flow where vicinal terrace edges advance by the incorporation of very many very small islands rather than by the incorporation of monomers. This behavior will persist until the accumulated strain in the epilayer is sufficient to nucleate 3D island formation and thus destroy layer growth altogether.

Many of the predictions of the foregoing theory are amenable to direct experimental test. The distribution of island sizes probably is best addressed by surface-sensitive microscopy. Since the resolution of current low-energy electron microscopes[61] probably is insufficient to resolve the small islands predicted here, recent progress with scanning tunneling microscopy performed *in situ* during growth is very encouraging.[62] On the other hand, studies of the distribution of 2D island sizes for Fe(100) homoepitaxy at submonolayer coverages[63] demonstrate that the popular "quench and look" approach to STM can still be extremely valuable.

Surface-sensitive diffraction appears best suited for trend studies such as a search for systematic behavior as a function of misfit. Both RHEED and TEAS have been used to study the mode transition question. But caution must be exercised since, as we have seen, the various effects of strain do not always act in concert morphologically. Thus, for example, a mode transition study for GaAs growth onto various $\text{In}_x\text{Ga}_{1-x}\text{As}$ substrates is to be preferred to a study of $\text{In}_x\text{Ga}_{1-x}\text{As}$ on GaAs so that the fact that In atoms and Ga atoms have very different surface diffusion rates does not confuse the issue.[64]

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[64]The authors are indebted to Dr. David Eaglesham for this remark.
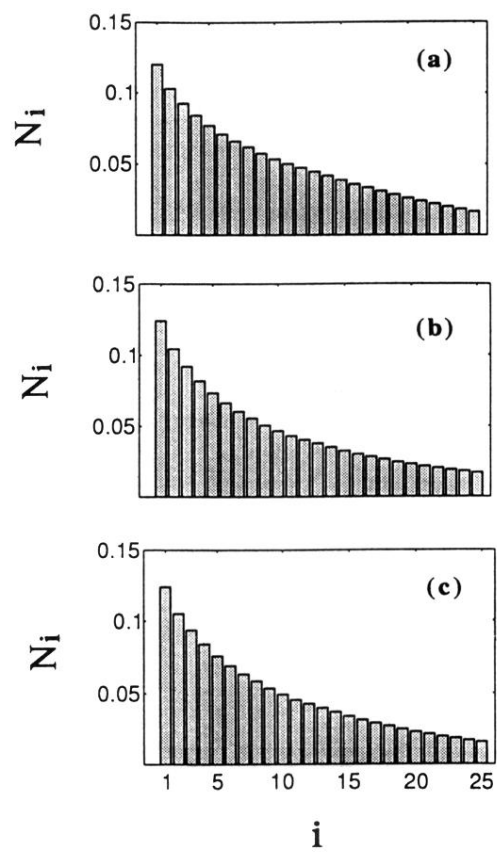
FIG. 5. Number of islands $\mathcal{N}_i$ as a function of the island size i in the nucleation region ($E_D/k_B T = 1.0$). The different panels show results at coverages $\Theta = 0.25$ (a), 0.75 (b), and 1.25 ML (c).