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Abstract

We adapt the level set method to simulate the growth of thin films described by the motion of island boundaries. This island dynamics model involves a continuum in the lateral directions, but retains

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atomic scale discreteness in the growth direction. Several choices for the island boundary velocity are discussed, and computations of the island dynamics model using the level set method are presented.

1 Introduction

The performance of many modern electronic and optoelectronic devices depends critically on the ability to fabricate high-quality semiconductor thin films and interfaces. These structures are produced by epitaxial growth techniques, whereby atoms and molecules are deposited onto a heated substrate and assemble into layers that have near-perfect registry with the substrate, resulting in the extremely low defect densities required for optoelectronic applications. A comprehensive theoretical description of epitaxial growth must accurately incorporate growth from submonolayer coverages with atomic resolution to the multilayer regime on lateral scales of at least several microns (typical device sizes). This presents enormous challenges to theoretical physicists and applied mathematicians attempting to model this process. No single model or computational scheme currently exists that is capable of bridging the desired length and time scales.

The basic processes during epitaxial growth are as follows. For simplicity, we consider growth only on so-called "singular" (or perfectly flat) surfaces. Atoms arrive at the surface from a ballistic beam and diffuse over this surface by hopping from one lattice site to another, typically an adjacent site. When two adatoms collide, they can form additional bonds and become a stable entity, called a nucleus, which is usually assumed to be immobile. This nucleus can grow laterally by capturing migrating atoms and the epitaxial film evolves by the formation and growth of many of such islands. As these islands grow, they eventually coalesce, leading to the completion of an atomic layer (but islands can also nucleate on top of existing islands, leading to multilayer growth—a process known as kinetic roughening). For the growth of planar heterostructures it is desirable that growth proceeds in an approximately layer-by-layer manner, which means that one layer is nearly completed before the next one is started. The extent to which this is attained depends on the sizes and density of the islands which, in turn, is

determined essentially by the ratio of the surface diffusion constant D to the deposition flux F, both of which are experimentally-controllable quantities. This quantity determines whether the growth condition allow an adatom to incorporate into an existing island (large D and/or small J) before it forms it collides with another atom to form a nucleus for a new island (small D and/or large J).

Several models have been used to describe epitaxial growth; we distinguish between analytic and atomistic (or particle) models. Analytic models, such as phenomenological rate equations [11] or coarse-grained continuum equations of motion [12], are formulated in terms of differential equations for densities and heights, respectively. Continuum equations provide some analytic understanding of stability, scaling, and steady-state behavior, but are only valid for rough surfaces with a large step density. Such surfaces are, almost without exception, extremely undesirable for device applications. Moreover, they do not allow *atomic* height resolution of the growing film. Rate equations in their typical form do not contain any spatial information, and are thus not amenable to describing processes such as the coalescence of islands. While it is in principle possible to incorporate the required spatial information into rate equations [1, 7], an analytic procedure for doing so has yet to be developed.

An alternative to analytical models are atomistic or particle models which are based on simulations using kinetic Monte Carlo or molecular dynamics methods. These methods are based on the kinetics or dynamics of interacting particles and can describe epitaxial growth in a wide range of circumstances. While providing atomic-level detail that complements the information provided by analytical approaches, the simulation of particle models is computationally demanding and it is therefore difficult to reach length and time scales of engineering interest.

In this Letter we introduce a new model that describes epitaxial growth continuously from the submonolayer to multi-layer regime. This model involves coarse-graining in the lateral directions but retains atomic resolution discreteness in the growth direction. It is particularly appropriate for describing growth of very thin layers (10-100 Å). The essential component to our model is knowledge about the island boundaries $\Gamma_i(t)$, whose positions are determined within the framework of the level set method and whose motion is described by their normal velocity $v(x,t)$. This general description of an epitaxial surface can easily accommodate the inclusion of many additional

processes, such as adatom diffusion, and we describe two possible choices for $v(x,t)$. The first, which we refer to as the "uniform density model," assumes the adatom density ρ is spatially uniform, and the island boundary velocity is proportional to ρ. The second, which we call the "irreversible aggregation model," assumes that the adatom density varies in space and satisfies a diffusion equation with an absorbing boundary condition at island edges. This implies that any adatom striking the boundary attaches irreversibly and, hence, the velocity is then proportional to the net flux to the boundary. Numerical computations for these two models are presented.

In general, surfaces are not perfectly flat, but instead have a certain density of steps on the surface that is due to a miscut with respect to a low index plane. Growth on these vicinal surfaces can be described by our method as well, because the step edges can be interpreted as the boundaries of (large) islands.

2 The Level Set Method

The general idea behind the level set method [8] is that a boundary curve $\Gamma = \Gamma_i(t)$ is represented as the level set $\varphi = 0$ of a smooth function φ. For a given boundary velocity $\boldsymbol{v}$, the equation for φ is then

$$\frac{\partial \varphi}{\partial t} + \boldsymbol{v} \cdot \nabla \varphi = 0 \tag{2.1}$$

in which $\boldsymbol{v}$ has been extended in an arbitrary way off of Γ. Since $\nabla \varphi = \boldsymbol{n} |\nabla \varphi|$ (i.e. $\boldsymbol{\tau} \cdot \nabla \varphi = 0$, in which $\boldsymbol{\tau}$ is the tangent vector), then $\boldsymbol{v} \cdot \nabla \varphi = v|\nabla \varphi|$, in which $v = \boldsymbol{n} \cdot \boldsymbol{v}$ is the normal component of $\boldsymbol{v}$, and $\boldsymbol{n}$ has been chosen to point in the direction of $\nabla \varphi$. Thus (2.1) becomes

$$\frac{\partial \varphi}{\partial t} + v|\nabla \varphi| = 0. \tag{2.2}$$

In this approach the interface Γ is captured by merely locating the set for which $\varphi = 0$ (or $\varphi = n$, $n = 1, 2, 3, \ldots$, see below). This deceptively simple statement is of great significance for numerical computation, because topological changes such as breaking and merging are well-defined and easily

performed. The effectiveness of level set computations is greatly enhanced by a number of techniques as described in [4, 9, 10]

For the application to island dynamics, several additional innovations are required. Overhangs and "undercuts" are prevented by using a single level set function φ instead of different level set functions for different layers. Islands of different heights k are represented by the level sets $\varphi = k$. Nucleation of new islands requires insertion of "peaks" within the level set function φ. These peaks are one unit high and their width corresponds to the size of the nucleus.

3 Uniform Density Model

The simplest form of island dynamics assumes that the adatom density $\rho = \rho(t)$ is spatially uniform. Then the velocity of the island boundaries is proportional to the flux f of adatoms toward an island boundary which is independent of the size of the island, and we assume

$$v = a^2 f = \lambda a^3 \rho \tag{3.1}$$

where λ is an incorporation rate and a^2 is the area per particle.

The total flux to all of the islands in a unit area is then $Lf = L\lambda a\rho$, in which $L = L(t)$ is the boundary perimeter per unit area (i.e. the step edge density). It follows that

$$\frac{d}{dt}\rho = F - L\lambda a\rho - 2\kappa a^2\rho^2 \ . \tag{3.2}$$

The last term in (3.2) describes the loss of adatoms due to random nucleation of new islands where κ is the nucleation rate. This happens because two atoms come together and form an entity which is stable against dissociation into two adatoms. When first nucleated, the islands are chosen to consist of a small number of grid points and their subsequent growth is found to be approximately independent of this choice. There are no spatial interactions between the islands before they coalesce, so that the exact location of the islands is irrelevant to the dynamics in the pre-coalescent phase. In fact, this

model is equivalent to a rate equation formulation (for a particular choice of capture numbers) in the pre-coalescence regime [6]. This comparison to rate equations provides a physical interpretation and a method for calculating λ and κ.

Figure 1 shows the island boundaries computed by this method for a typical choice of parameters at various values of the coverage. In these computations, islands are nucleated as circles and stay circular until they start to coalesce. The corresponding variation of adatom density $\rho(t)$ is shown in Figure 2. This quantity is seen to drop precipitously after the onset of growth, due to the nucleation of new islands, but then enters a "steady-state" phase, until the first layer is near completion, whereupon it rises again. Because the nucleation of the second layer is spread over time, the variation of $\rho(t)$ is not as dramatic as that during the nucleation of the first layer. This smoothing in the behavior of $\rho(t)$ continues as more layers are deposited due to kinetic roughening.

A variation of the uniform density model is to let the attachment rate λ depend on the angle θ of the normal at a point on the island boundary. The resulting steady shape is determined through the kinetic Wulff construction. Figure 3 shows the island boundaries at two values of the coverage, from a computation using a choice of $\lambda(\theta)$ for which the steady shapes are triangles.

4 Irreversible Aggregation Model

A more realistic island dynamics model involves the solution of the diffusion equation for ρ away from the island boundaries. The adatom diffusion equation is

$$\frac{\partial}{\partial t}\rho - D\nabla^2\rho = -\tau^{-1}\rho + F - M. \tag{4.1}$$

in which D is the adatom diffusion coefficient, F is the deposition flux of adatoms to the surface, M is the rate of loss of adatoms due to nucleation of new islands, and τ^{-1} is the desorption rate of adatoms from the surface (which is often negligible). For simplicity, additional mechanisms such as cluster diffusion are ignored, although it could easily be included if needed.

Further specification of the model requires a choice of the boundary conditions for ρ. The simplest choice is

$$\rho = 0 \tag{4.2}$$

which implies that any adatom striking the boundary is immediately absorbed with no possibility of subsequent detachment.

The normal velocity v of the boundary is then proportional to the net particle flux to the boundary, i.e. $v = a^2(f_+ + f_-)$, in which f_+ and f_- are fluxes from the upper terrace and the lower terrace respectively. These are given by

$$\begin{aligned} f_+ &= -\rho_+ v - Dn \cdot \nabla \rho_+ \\ f_- &= \rho_- v + Dn \cdot \nabla \rho_- \end{aligned} \tag{4.3}$$

in which normal vector n is pointing out of the island. Previous accounts of epitaxial growth (e.g. [5]) have often omitted the convective term ρv in this flux. It follows that

$$v = -(1 + a^2[\rho])^{-1} a^2 Dn \cdot (\nabla \rho_+ - \nabla \rho_-) \quad . \tag{4.4}$$

The upper panels in Figure 4 show the island boundaries at two values of the coverage. Although the islands are initially circular, the presence of neighboring islands causes a distortion in the island shape, which results from the position dependence of $\rho(x,t)$. This position dependence also prevents two islands from nucleating too close together, the absorbing boundary condition leads to a "denuded" zone of adatoms around each island, which reduces the nucleation probability of new islands.

The lower panels in Figure 4 show the value of adatom density $\rho(x,t)$ as x varies along a horizontal line through the middle of the region. Note the strong variation of $\rho(x,t)$ caused by the islands and the huge decrease of $\rho(x,t)$ at 50% coverage, because almost all the atoms that are deposited get incorporated into existing islands (cf. Figure 2).

Currently, we are developing a theory [3] to determine the island boundary velocity using a kinetic approach that is a generalization of the theory of Burton, Cabrera and Frank [2]. In the resulting model, the irreversible aggregation condition (4.2) will be replaced by kinetic equations for the microscopic state of the boundary.

5 Summary

We have formulated an island dynamics model for epitaxial growth and implemented a level set method for its simulation. Computations are presented for two simple models which specify the behavior of the adatom density ρ, and, hence, the island boundary velocity: the uniform density model and the irreversible aggregation model. These computational results show island morphologies that are qualitatively realistic. The two choices for the island boundary velocity illustrate the capabilities of island dynamics and the level set method. This approach easily extends to include more realistic physics such as edge diffusion and other microscopic details of the boundary evolution, multiple species diffusion, and anisotropic diffusion and attachment. With these additions, we expect that the island dynamics approach will be an effective tool for simulation of epitaxial growth in regimes of engineering interest, providing predictive capability and a basis for design of control methods.

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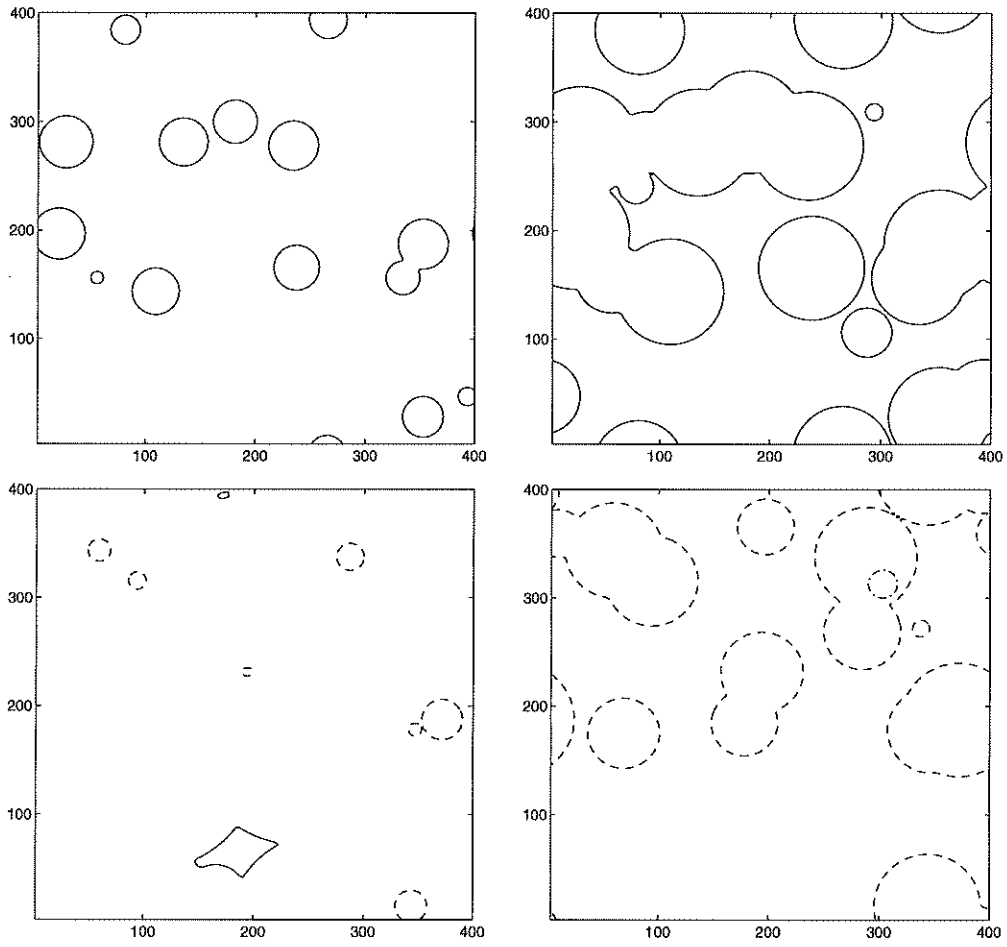

Figure 1: Uniform density model with circular islands: Islands of height 1 (solid), 2 (dashed) and 3 (dash-dot). Coverages of 10% (upper left), 50% (upper right), 100% (lower left) and 130% (lower right).

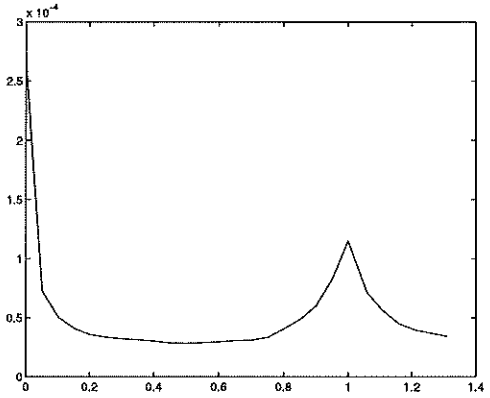

Figure 2: Adatom density ρ vs. time t for the uniform density model with circular islands.

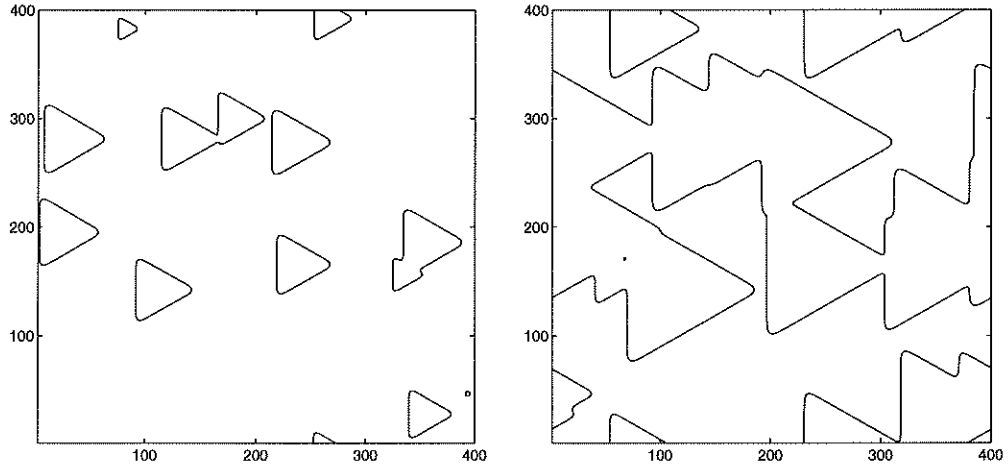

Figure 3: Uniform density model with triangular islands Island boundaries at coverage of 10% (left) and 50% (right).

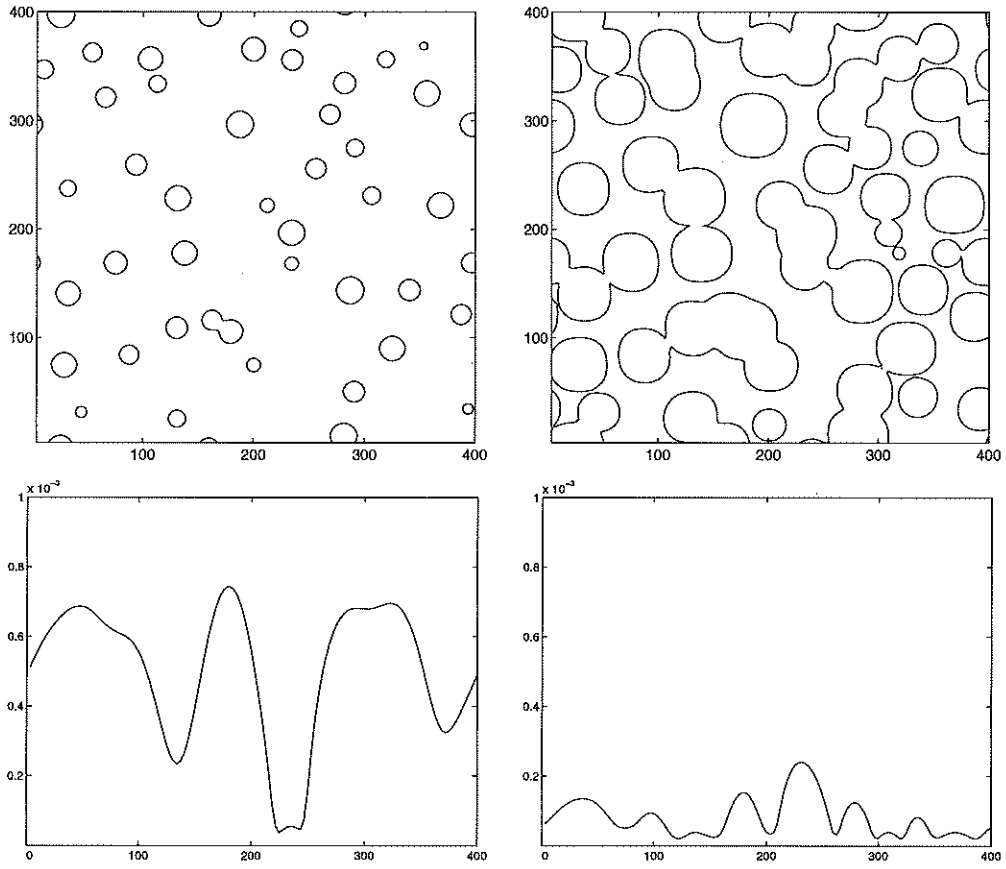

Figure 4: Irreversible Aggregation Model: Island boundaries (upper) and adatom densities at horizontal midline (lower) at coverages of 10% (left) and 50% (right).