

## 7.15

# LEVEL SET METHODS FOR SIMULATION OF THIN FILM GROWTH

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The level set method is a general approach to numerical computation for the motion of interfaces. Epitaxial growth of a thin film can be described by the evolution of island boundaries and step edges, so that the level set method is applicable to simulation of thin film growth. In layer-by-layer growth, for example, this includes motion of the island boundaries, merger or breakup of islands, and creation of new islands. A system of size  $100 \times 100$  nm may involve hundreds or even thousands of islands. Because it does not require smoothing and or discretization of individual island boundaries, the level set method can accurately and efficiently simulate the dynamics of a system of this size. Moreover, because it does not resolve individual hopping events on the terraces or island boundaries, the level set method can take longer time steps than those of an atomistic method such as kinetic Monte Carlo (KMC). Thus the level set approach can simulate some systems that are computationally intractable for KMC.

## 1. The Level Set Method

The level set method is a numerical technique for computing interface motion in continuum models, first introduced by [11]. It provides a simple, accurate way of computing complex interface motion, including merger and pinchoff. This method enables calculations of interface dynamics that are beyond the capabilities of traditional analytical and numerical methods. For general references on level set methods, see the books [12, 21].

The essential idea of the method is to represent the interface as a level set of a *smooth* function,  $\phi(x)$  – for example the set of points where  $\phi=0$ . For numerical purposes, the interface velocity is smoothly extended to all points

$x$  of the domain, as  $v(x)$ . Then, the interface motion is captured simply by convecting the values of the smooth function  $\phi$  with the smooth velocity field  $v$ . Numerically, this is accomplished by solving the convection equation

$$\frac{\partial \phi}{\partial t} + v \nabla \phi = 0 \quad (1)$$

on a fixed, regular spatial grid.

The main advantage of this approach is that interface merger or pinch off is captured without special programming logic. The merger of two disjoint level sets into one occurs naturally as this equation is solved, through *smooth* changes in the function  $\phi(x, t)$ . For example, two disjoint interface loops would be represented by a  $\phi$  with two smooth humps, and their merging into a single loop is represented by the two humps of  $\phi$  smoothly coming together to form a single hump. Pinch off is the reverse process. In particular, the method does not involve smoothing out of the interface.

The normal component of the velocity  $v_n = \mathbf{n} \cdot \mathbf{v}$  contains all the physical information of the simulated system, where  $\mathbf{n}$  is the outward normal of the moving boundary and  $\mathbf{v} \cdot \nabla \phi = v_n |\nabla \phi|$ .

Another advantage of the method is that the local interface geometry – normal direction,  $n$ , and curvature,  $\kappa$  – can be easily computed in terms of partial derivatives of  $\phi$ . Specifically,

$$n = \frac{-\nabla \phi}{|\nabla \phi|} \quad (2)$$

$$\kappa = \nabla n \quad (3)$$

provide the normal direction and curvature at points on the interface.

## 2. Epitaxial Growth

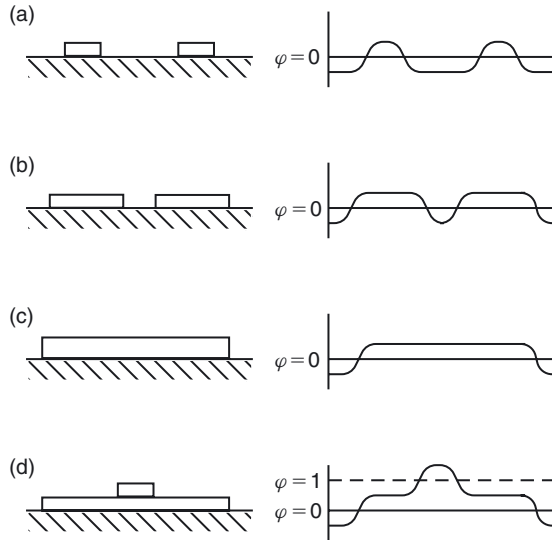
Epitaxy is the growth of a thin film on a substrate in which the crystal properties of the film are inherited from those of the substrate. Since an epitaxial film can (at least in principle) grow as a single crystal without grain boundaries or other defects, this method produces crystals of the highest quality. In spite of its ideal properties, epitaxial growth is still challenging to mathematically model and numerically simulate because of the wide range of length and time scales that it encompasses, from the atomistic scale of Ångströms and picoseconds to the continuum scale of microns and seconds.

The geometry of an epitaxial surface consists of step edges and island boundaries, across which the height of the surface increases by one crystal layer, and adatoms which are weakly bound to the surface. Epitaxial growth involves deposition, diffusion and attachment of adatoms on the surface. Deposition is from an external source, such as a molecular beam. The principal

dimensionless parameter (for growth at low temperature) is the ratio  $D/(a^4 F)$ , in which  $a$  is the lattice constant and  $D$  and  $F$  are the adatom diffusion coefficient and deposition flux. It is conventional to refer to this parameter as  $D/F$ , with the understanding that the lattice constant serves as the unit of length. Typical values for  $D/F$  are in the range of  $10^4$ – $10^8$ .

The models that are typically used to describe epitaxial growth include the following: *Molecular dynamics* (MD) consists of Newton's equations for the motion of atoms on an energy landscape. A typical *Kinetic Monte Carlo* (KMC) method simulates the dynamics of the epitaxial surface through the hopping of adatoms along the surface. The hopping rate comes from an Arrhenius rate of the form  $e^{-E/kT}$  in which  $E$  is the energy barrier for going from the initial to the final position of the hopping atom. *Island dynamics and level set methods*, the subject of this article, describe the surface through continuum scaling in the lateral directions but atomistic discreteness in the growth direction. *Continuum equations* approximate the surface using a smooth height function  $h = h(x, y, t)$ , obtained by coarse graining in all directions. *Rate equations* describe the surface through a set of bulk variables without spatial dependence.

Within the level set approach, the union of all boundaries of islands of height  $k + 1$ , can be represented by the level set  $\varphi = k$ , for each  $k$ . For example, the boundaries of islands in the submonolayer regime then correspond to the set of curves  $\varphi = 0$ . A schematic representation of this idea is given in Fig. 1, where two islands on a substrate are shown. Growth of these islands is



*Figure 1.* A schematic representation of the level-set formalism. Shown are island morphologies (left side), and the level-set function  $\varphi$  (right side) that represents this morphology.

described by a smooth evolution of the function  $\varphi$  (cf. Figs. 1 (a) and (b)). The boundary curve  $\Gamma(t)$  generally has several disjoint pieces that may evolve so as to merge (Fig. 1(c)) or split.

Validation of the level set method will be detailed in this article by comparison to results from an atomistic KMC model. The KMC model employed is a simple cubic pair-bond solid-on-solid (SOS) model [24]. In this model, atoms are randomly deposited at a deposition rate  $F$ . Any surface atom is allowed to move to its nearest neighbor site at a rate that is determined by  $r = r_0 \exp\{-(E_S + nE_N)/k_B T\}$ , where  $r_0$  is a prefactor which is chosen to be  $10^{13} \text{ s}^{-1}$ ,  $k_B$  is the Boltzmann constant, and  $T$  is the surface temperature.  $E_S$  and  $E_N$  represent the surface and nearest neighbor bond energies, and  $n$  is the number of nearest neighbors. In addition, the KMC simulations include fast edge diffusion, where singly bonded step edge atoms diffuse along the step edge of an island with a rate  $D_{\text{edge}}$ , to suppress roughness along the island boundaries.

### 3. Island Dynamics

Burton, Cabrera and Frank [5] developed the first detailed theoretical description for epitaxial growth. In this “BCF” model, the adatom density solves a diffusion equation with an equilibrium boundary condition ( $\rho = \rho_{\text{eq}}$ ), and step edges (or island boundaries) move at a velocity determined from the diffusive flux to the boundary. Modifications of this theory were made, for example in [9], to include line tension, edge diffusion and nonequilibrium effects. These are “island dynamics” models, since they describe an epitaxial surface by the location and evolution of the island boundaries and step edges. They employ a mixture of coarse graining and atomistic discreteness, since island boundaries are represented as smooth curves that signify an atomistic change in crystal height.

Adatom diffusion on the epitaxial surface is described by a diffusion equation of the form

$$\partial_t \rho - D \nabla^2 \rho = F - \frac{2dN_{\text{nuc}}}{dt} \quad (4)$$

in which the last term represents loss of adatoms due to nucleation and desorption from the epitaxial surface has been neglected. Attachment of adatoms to the step edges and the resulting motion of the step edges are described by boundary conditions at an island boundary (or step-edge)  $\Gamma$  for the diffusion equation and a formula for the step-edge velocity  $v$ .

For the boundary conditions and velocity, several different models are used. The simplest of these is

$$\begin{aligned} \rho &= \rho_* \\ v &= D \left[ \frac{\partial \rho}{\partial n} \right] \end{aligned} \quad (5)$$

in which the brackets indicate the difference between the value on the upper side of the boundary and the lower side. Two choices for  $\rho_*$  are  $\rho_* = 0$ , which corresponds to irreversible aggregation in which all adatoms that hit the boundary stick to it irreversibly, and  $\rho_* = \rho_{\text{eq}}$  for reversible aggregation. For the latter case,  $\rho_{\text{eq}}$  is the adatom density for which there is local equilibrium between the step and the terrace [5].

Line tension and edge diffusion can be included in the boundary conditions and interface velocity as in

$$\begin{aligned} \frac{\partial \rho}{\partial n_{\pm}} &= D_T(\rho_{\pm} - \rho_*) - \mu\kappa, \\ v &= D_T \mathbf{n} \cdot [\nabla \rho] + \beta \rho_{*ss} + \left( \frac{\mu}{D_E} \right) \kappa_{ss}, \end{aligned} \quad (6)$$

in which  $\kappa$  is curvature,  $s$  is the variable along the boundary, and  $D_E$  is the coefficient for diffusion along and detachment from the boundary.

Snapshots of the results from a typical level-set simulation are shown in Fig. 2. Shown is the level-set function (a) and the corresponding adatom concentration (b) obtained from solving the diffusion Eq. (4). The island boundaries that correspond to the integer levels of panel (a) are shown in (c). Dashed (solid) lines represent the boundaries of islands of height 1. Comparison of panels (a) and (b) illustrates that  $\rho$  is indeed zero at the island boundaries (where  $\varphi$  takes an integer value).

Numerical details on implementation of the level set method for thin film growth are provided in [7]. The figures in this article are taken from [17] and [15].

#### 4. Nucleation and Submonolayer Growth

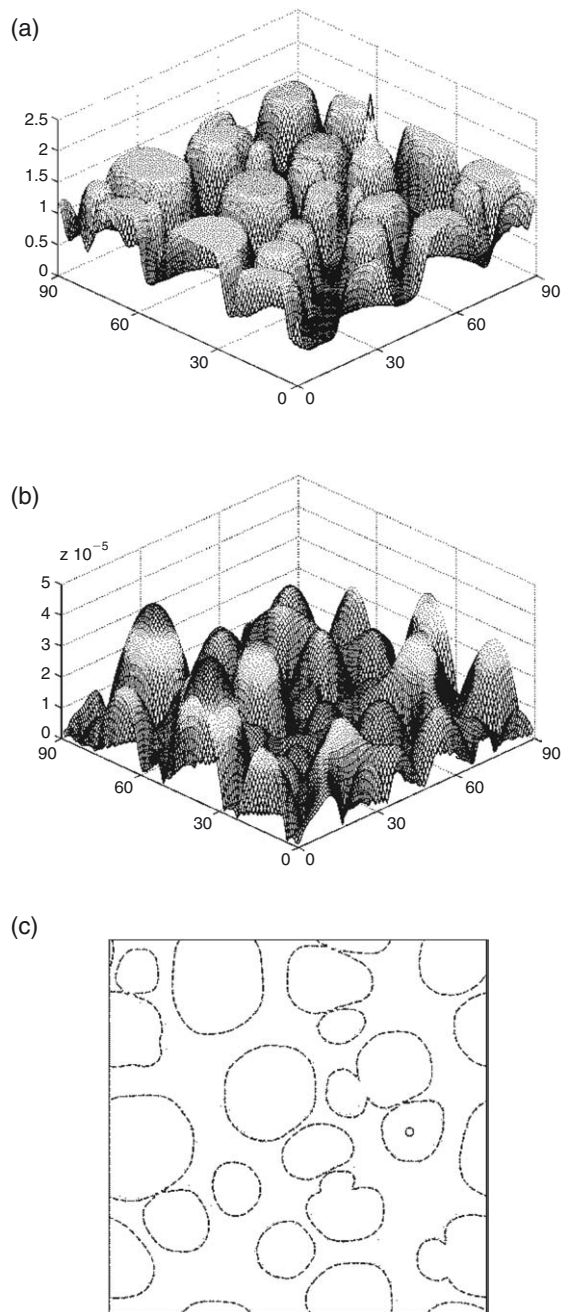
For the case of irreversible aggregation, a dimer (consisting of two atoms) is the smallest stable island, and the nucleation rate is

$$\frac{dN_{\text{nuc}}}{dt} = D\sigma_1 \langle \rho^2 \rangle, \quad (7)$$

where  $\langle \cdot \rangle$  denotes the spatial average of  $\rho(\mathbf{x}, t)^2$  and

$$\sigma_1 = \frac{4\pi}{\ln[(1/\alpha)\langle \rho \rangle D/F]} \quad (8)$$

is the adatom capture number as derived in [4]. The parameter  $\alpha$  reflects the island shape, and  $\alpha \simeq 1$  for compact islands. Expression (7) for the nucleation rate implies that the time of a nucleation event is chosen deterministically. Whenever  $N_{\text{nuc}} L^2$  passes the next integer value ( $L$  is the system size), a new island is nucleated. Numerically, this is realized by raising the level-set function to the next level at a number of grid points chosen to represent a dimer.



*Figure 2.* Snapshots of a typical level-set simulation. Shown are a 3D view of the level-set function (a) and the corresponding adatom concentration (b). The island boundaries as determined from the integer levels in (a) are shown in (c), where dashed (solid) lines correspond to islands of height 1 (2).

The choice of the location of the new island is determined by probabilistic choice with spatial density proportional to the nucleation rate  $\rho^2$ . This probabilistic choice constitutes an atomistic fluctuation that must be retained in the level set model for faithful simulation of the epitaxial morphology. For growth with compact islands, computational tests have shown additional atomistic fluctuations can be omitted [18].

Additions to the basic level set method, such as finite lattice constant effects and edge diffusion, are easily included [17]. The level set method with these corrections is in excellent agreement with the results of KMC simulations. For example, Fig. 3 shows the scaled island size distribution (ISD)

$$n_s = \frac{\Theta}{s_{av}^2} g\left(\frac{s}{s_{av}}\right), \quad (9)$$

where  $n_s$  is the density of islands of size  $s$ ,  $s_{av}$  is the average island size, and  $g(x)$  is a scaling function. The top panel of Fig. 3 is for irreversible attachment; the other two panels include reversibility that will be discussed below. All three panels show excellent agreement between the results from level set simulations, KMC and experiment.

## 5. Multilayer Growth

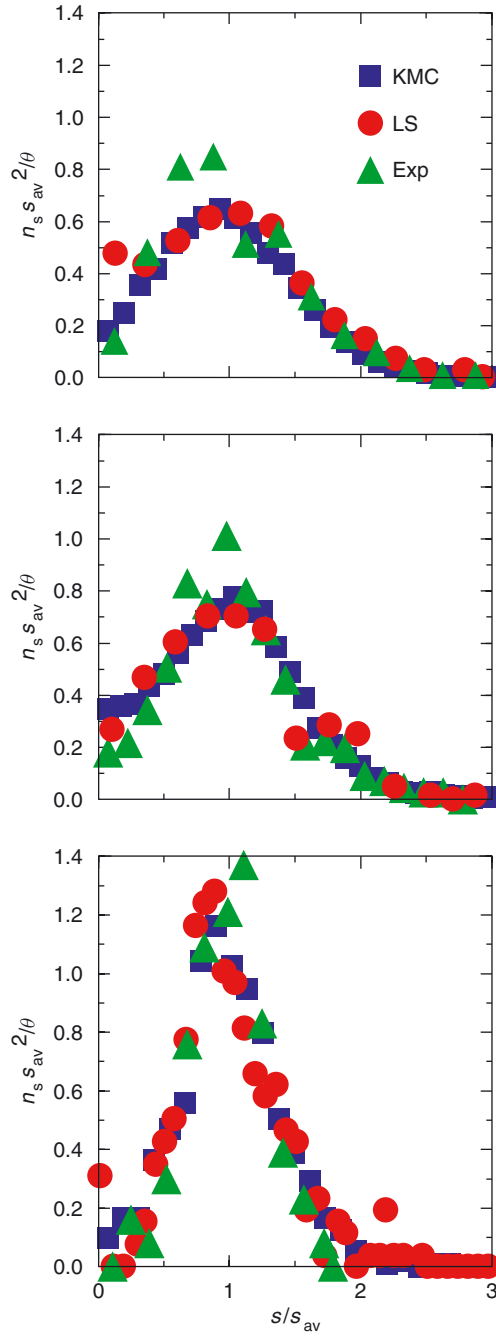
In ideal layer-by-layer growth, a layer is completed before nucleation of a new layer starts. In this case, growth on subsequent layers would essentially be identical to growth on previous layers. In reality, however, nucleation on higher layers starts before the previous layer has been completed and the surface starts to roughen. This roughening transition depends on the growth conditions (i.e., temperature and deposition flux) and the material system (i.e., the value of the microscopic parameters). At the same time, the average lateral feature size increases in higher layers, which we will refer to as coarsening of the surface.

These features of multilayer growth and the effectiveness of the level set method in reproducing them is illustrated in Fig. 4 that shows the island number density  $N$  as a function of time for two different values of  $D/F$  from both a level set simulation and from KMC. The results show near perfect agreement. The KMC results were obtained with a value for the edge diffusion that is 1/100 of the terrace diffusion constants. The island density decreases as the film height increases which implies that the film coarsens.

The surface roughness  $w$  is defined as

$$w^2 = \langle (h_i - \langle h \rangle)^2 \rangle, \quad (10)$$

where the index  $i$  labels the lattice site. Figure 5 shows the increase of surface roughness for various different values of the edge diffusion, which implies that edge diffusion contributes to roughening, as also observed in KMC studies.



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*Figure 3.* The island size distribution, as given by KMC (squares) and LS (circles) methods, in comparison with STM experiments (triangles) on Fe/Fe(001) [23]. The reversibility increases from top to bottom.



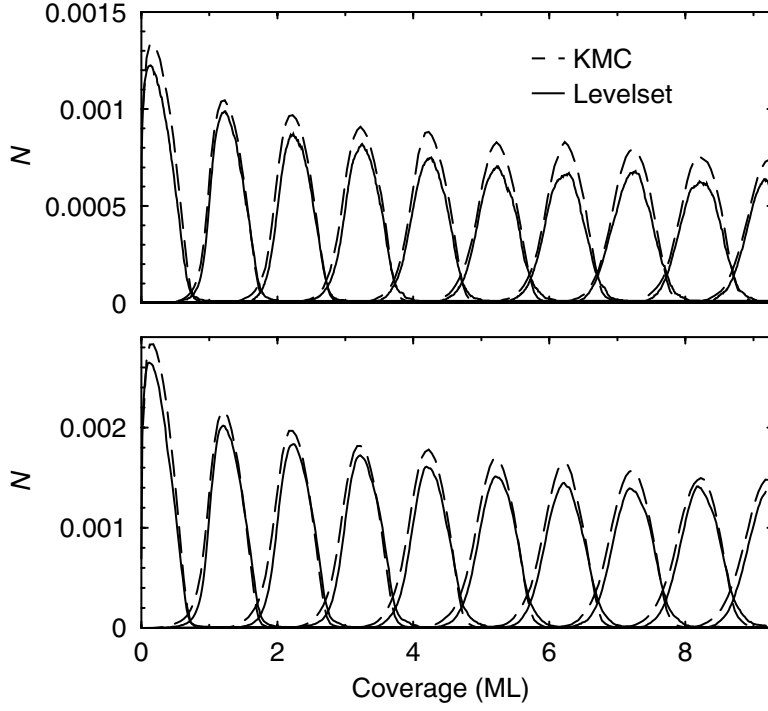


Figure 4. Island densities  $N$  on each layer for  $D/F=10^6$  (lower panel) and  $D/F=10^7$  (upper panel) obtained with the level-set method and KMC simulations. For each data set there are 10 curves in the plot, corresponding to the 10 layers.

It suggests that faster edge diffusion leads to more compact island shapes, and as a result the residence time of an atom on top of compact islands is extended. This promotes nucleation at earlier times on top of higher layers, and thus enhanced roughening. Effects of edge diffusion were included in these simulations through a term of the form  $\kappa - \langle \kappa \rangle$  rather than  $\kappa_{ss}$  as in (6).

## 6. Reversibility

The simulation results presented above have been for the case of irreversible aggregation. If aggregation is reversible the KMC method must simulate a large number of events that do not affect the time-average of the system: Atoms detach from existing islands, diffuse on the terrace for a short period of time and reattach to the same island most of the time. These processes can slow down KMC simulations significantly. On the other hand, in a level set simulation these events can directly be replaced by their time average and therefore the simulation only needs to include detachment events that

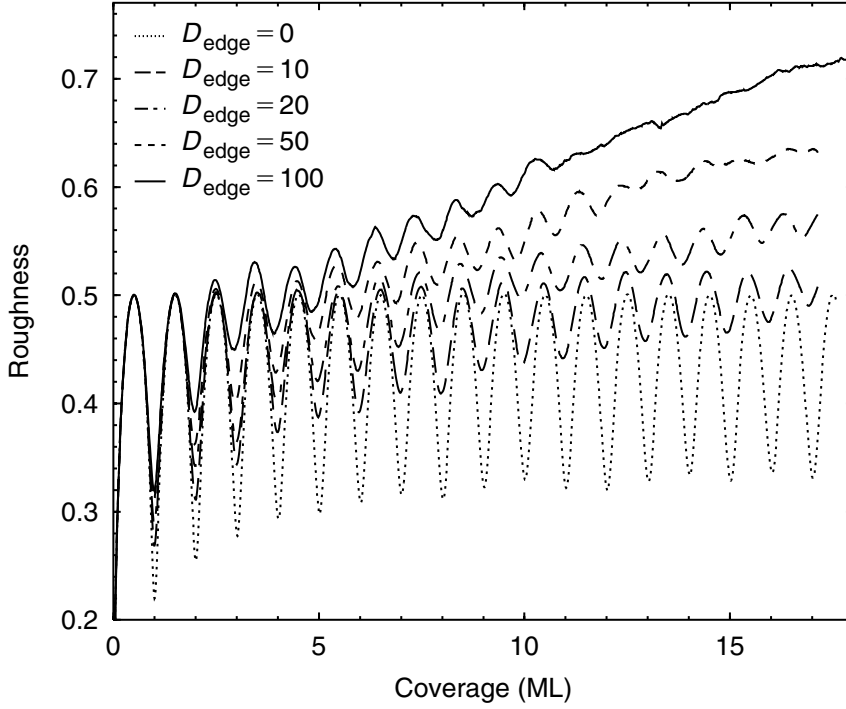


Figure 5. Time evolution of the surface roughness  $w$  for different values of edge diffusion  $D_{\text{edge}}$ .

do not lead to a subsequent reattachment, making the level set method much faster than KMC.

Reversibility does not necessarily depend only on purely local conditions (e.g., local bond strength) but often on more global quantities such as strain or chemical environment. To include these kind of effects in a rather hard task in a KMC simulation but can be quite naturally included in a mean field picture.

Reversibility can be included in the level set method using the boundary conditions (5) with  $\rho_* = \rho_{\text{eq}}$  in which  $\rho_{\text{eq}}$  depends on the local environment of the island, in particular the edge atom density [6]. For islands consisting of only of a few atoms, however, the stochastic nature of detachment becomes relevant and is included through random detachment and breakup for small islands, as detailed in [14].

Figure 3 shows that the level set method with reversibility reproduces nicely the trends in the scaled ISD found in the KMC simulations and experiment. In particular, the scaled ISD depends only on the degree of reversibility, and it narrows and sharpens in agreement with the earlier prediction of [19].

In [15], the level set method with reversibility was used to determine the long time asymptotics of Ostwald ripening. A similar computation was

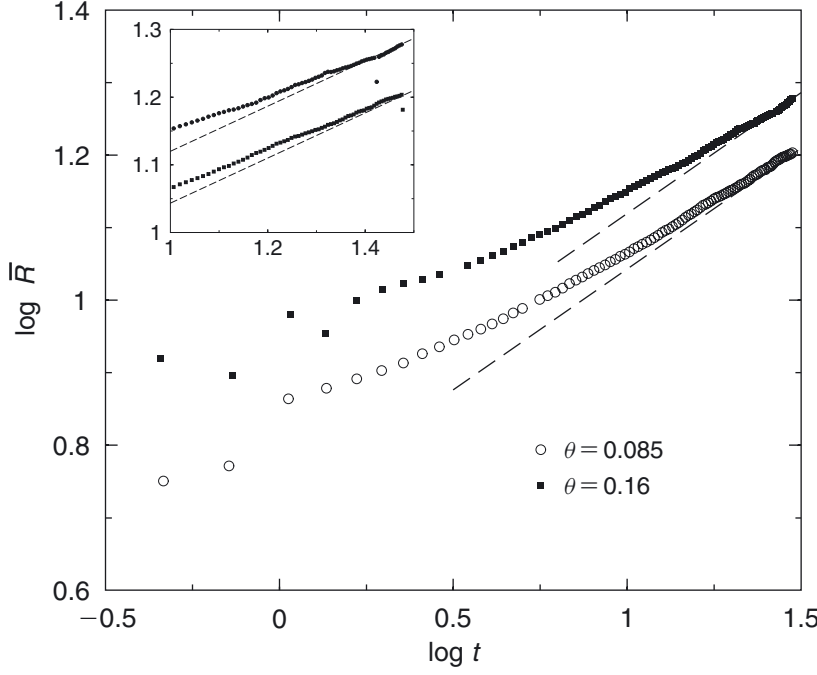


Figure 6. Time dependence (in seconds) of the average island radius  $\bar{R}$  (in units of the lattice constant) for two different coverages  $\Theta$  on a log-log plot. The straight lines have slope  $1/3$ , which was the theoretical prediction.

performed in [8]. Figure 6 shows that the average island size  $\bar{R}$  grows as  $t^{1/3}$ , which was an earlier theoretical prediction. Because reversibility greatly increases the number of hopping events and thus lowers the time step for an atomistic computation, KMC simulations have been unable to reach this asymptotic regime. The longer time steps in the level set simulation give it a significant advantage over KMC for this problem.

## 7. Hybrid Methods and Additional Applications

As described above, the level set method does not include island boundary roughness or fractal island shapes, which can be significant in some applications. One way of including boundary roughness is by including additional state variables  $\phi$  for the density of edge atoms and  $k$  for the density of kinks along an island boundary or step edge. A detailed step edge model was derived in [6] and used in determination of  $\rho_{\text{eq}}$  for the level set method with reversibility. While adequate for simulating reversibility, this approach will not extend

to fractal island shapes. A promising alternative is a hybrid method that combines island dynamics with KMC; e.g., the adatom density is evolved through diffusion of a continuum density function, but attachment at island boundaries is performed by Monte Carlo [20]. In a different approach [10], where diffusion is described and the adatom density is evolved by explicit solution of the master equation, the atoms are resolved explicitly only once they attach to an island boundary. While these methods do not use a level set method, it is sufficiently similar to the method discussed here to warrant mention in this discussion.

Level set methods have been used for a number thin film growth problems that are related to the applications described above. In [22] a level set method was used to describe spiral growth in epitaxy. A general level set approach to material processing problems, including etching, deposition and lithography, was developed in [1], [2] and [3]. A similar method was used in [13] for deposition in trenches and vias.

## **8. Outlook**

The simulations described above have established the validity of the level set method for simulation of epitaxial growth. Moreover, the level set method makes possible simulations that would be intractable for atomistic methods such as KMC. This method can now be used with confidence in many applications that include epitaxy along with additional phenomena and physics. Examples that seem promising for future developments include strain, faceting and surface chemistry: Elastic strain is generated in heteroepitaxial growth due to lattice mismatch between the substrate and the film. It modifies the material properties and surface morphology, leading to many interesting growth phenomena such as quantum dot formation. Strained growth could be simulated by combining an elasticity solver with the level set method, and this would have significant advantages over KMC simulations for strained growth. Faceting occurs in many epitaxial systems, e.g., corrugated surfaces and quantum dots, and can be an important factor in the energy balance that determines the kinetic pathways for growth and structure. The coexistence of different facets can be represented in a level set formulation using two level set functions, one for crystal height and the second to mark the boundaries between adjacent facets [16]. Determination of the velocity for a facet boundary, as well for the nucleation of new facets, should be performed using energetic arguments. Similarly, surface chemistry such as the effects of different surface reconstructions could in principle be represented using two level set functions.

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## **Author Queries**

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