The Level-Set Method for Modeling Epitaxial Growth

C. Ratsch, M. Petersen, and R.E. Caflisch

Department of Mathematics, University of California
Los Angeles, CA 90095-1555

ABSTRACT

A level-set model for the simulation of epitaxial growth is described. In this model, the motion of island boundaries of discrete atomic layers is determined by the time evolution of a continuous level-set function φ. The adatom concentration is treated in a mean field manner. Thus, fast events (such as diffusion or detachment of adatoms from island boundaries) can be described without extra computational cost. We discuss results for the scaled island size distributions in the submonolayer aggregation regime and compare them to those obtained from atomistic KMC simulations and experiments. The level-set method can naturally be extended to describe multilayer growth. Roughening and coarsening of the surface will be discussed.

Keywords: Level-Set, Epitaxial Growth, Aggregation Phenomena

1 INTRODUCTION

Epitaxial growth and many other phenomena of practical interest in materials sciences occur on time and length scales that span many orders of magnitude. The most basic physical processes that occur during epitaxial growth occur on the atomic scale, i.e. on length scales of the order of Ångstroms, and time scales that reflect the typical atomic vibration frequencies (i.e. 10^{-13} s). On the other hand, a typical opto-electronic device might be up to several microns in size, and its growth can take minutes or even hours. Thus, the challenge in modeling epitaxial growth is to span these vastly different time and length scales.

The models that are typically used to describe epitaxial growth are either completely stochastic or completely deterministic. Mean field rate equations that were introduced to this problem [1] almost 30 years ago are a set of coupled ordinary differential equations. Continuum models based on partial differential equations (PDE's) are appropriate mainly at large time and length scales [2], [3]. An alternative to completely analytic approaches are atomistic models that are typically implemented in the form of molecular dynamics (MD) [4] or kinetic Monte Carlo (KMC) [5] simulations.

We have introduced a new model to describe epitaxial growth [6], [7], the island dynamics model, that might be considered a hybrid model between continuum, PDE-based methods, and atomistic, stochastic methods. The numerical solution of the model is based on the level-set method [8], [9], which is a general technique for simulating the motion of moving boundaries. This model allows us to describe epitaxial growth as continuous in the plane of the surface, yet it also allows us to discretely resolve each atomic layer [11]. Moreover, different sources of fluctuations can be isolated and studied individually [14].

2 THE MODEL

The main component of our model is that a (zero thickness) boundary curve Γ_k, such as the boundary of an island of height $k+1$, can be represented by the set $\varphi = k$, called the *level-set*, of a smooth function φ, called the *level-set function*. The boundaries of islands of height k then correspond to the set of curves $\varphi = k-1$. A schematic representation of this idea is given in Fig. 1, where two islands on a substrate are shown. Growth of

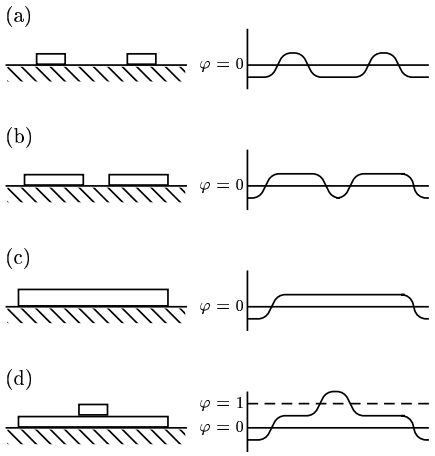

Figure 1: A schematic representation of the level-set formalism. Shown are island morphologies (left side), and the level-set function φ (right side) that represents this morphology.

these islands is described by a smooth evolution of the

function φ (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. Islands on top of islands are also easily described, as illustrated in Fig. 1 (d).

For a given boundary, the level-set function φ evolves according to

$$\frac{\partial \varphi}{\partial t} + \mathbf{v} \cdot \nabla \varphi = 0 \quad , \tag{1}$$

where $\mathbf{v}$ is the boundary velocity. 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 $\mathbf{v} \cdot \nabla \varphi = v_n |\nabla \varphi|$. The boundary velocity is computed by solving the diffusion equation for the adatom concentration ρ

$$\frac{\partial \rho}{\partial t} = F + D\nabla^2 \rho - 2\frac{dN_{\rm nuc}}{dt} \quad , \tag{2}$$

where F is the deposition flux, D is the surface diffusion constant, and the last term on the right hand side is the rate of nucleation of new islands on the surface. The velocity of the island boundaries is determined by the flux of adatoms to the island boundaries, and is given by

$$v_n = a^2 D(\mathbf{n} \cdot \nabla \rho^- - \mathbf{n} \cdot \nabla \rho^+) + D_{\rm edge}(\kappa - \kappa_{av}) \quad , \tag{3}$$

The superscripts $(+)$ and $(-)$ label the contributions from above and below the island boundary, and a is the lattice constant. $D_{\rm edge}$ relates to the edge diffusion rate, and κ and κ_{av} are the local and average curvature of the island. We note that edge diffusion smoothes the island edges. However, because of the mean-field treatment of the adatom densities, islands are compact even without edge diffusion [15]. Thus, we will include edge diffusion only in the multilayer regime, where the smoothing of island edges after merger is important (see below).

In order to solve the diffusion equation (2), a boundary condition needs to be specified. For the case of irreversible aggregation, in which all atoms are adsorbed by the boundary, the standard continuum (absorbing) boundary condtion is

$$\rho(\mathbf{x}, t) = 0 \quad \text{for all } \mathbf{x} \text{ with } \varphi(\mathbf{x}, t) = 0, 1, 2, ... \quad . \tag{4}$$

We have shown that this boundary condition is valid strictly only in the limit $D/F \longrightarrow \infty$. However, a correction that accounts for the fact that one needs $\rho = 0$ in a region around the island boundary that is (at least) one lattice constant wide can easily be implemented. [10]

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

$$\frac{dN_{\rm nuc}}{dt} = D\sigma_1 \langle \rho^2 \rangle \quad , \tag{5}$$

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

$$\sigma_1 = \frac{4\pi}{\ln[(1/\alpha)\langle \rho \rangle D/F]} \tag{6}$$

is the adatom capture number as derived by Bales and Chrzan [12]. Please note that the nucleation density $N_{\rm nuc}$ is slightly larger than the island density N, and that the two only agree before coalescence. The parameter α reflects the island shape, and $\alpha \simeq 1$ for compact islands. Expression (5) for the nucleation rate implies that the time of a nucleation event is chosen deterministically. Whenever $N_{\rm nuc} L^2$ passes the next integer value, 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. The choice of the location however needs to be determined by including a stochastic element. We found that the probability for a new island to be seeded at location $\mathbf{x}$ needs to weighted by the local value of $\rho(\mathbf{x})^2$. Details of the numerical implementation have been given elsewhere [11].

3 RESULTS

3.1 Submonolayer Growth for Irreversible Aggregation

The scaled island size distribution as obtained from our model in comparison to the one obtained from an atomistic KMC simulation, and also in comparison to experimental data for Fe/Fe(001) [13] is shown in Fig. 2. The agreement of the data is excellent. This demon-

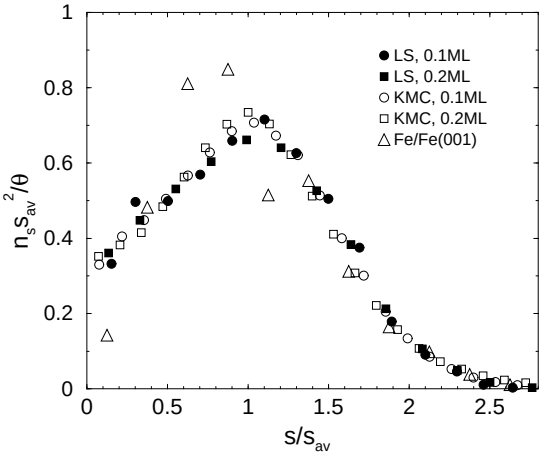

Figure 2: Scaled islands size distribution for $D/F = 10^6$ obtained with the level set method (closed symbols), in comparison to data obtained from a KMC simulation (open circles and squares), and experimental data for Fe/Fe(001) [13] (open triangles). The quantities n_s, s_{av}, and θ denote the density of islands of size s, average island size, and surface coverage, respectively.

strates that our model that is based on coupled partial differential equations does indeed capture all the essential physics, without resolving explicitly the motion of each individual atom, as it is done in the atomistic simulation.

We have shown in Ref. [14] that the spatial fluctuations in the seeding style are essential to obtain the correct island size distribution. We have also tested the effect of the island shape on the size distribution. The size distribution for square or circular shaped islands is essentially indistinguishable [15]. The reason is that the distribution of the capture areas as discussed in Ref. [14] determines the distribution of the island sizes.

3.2 Multilayer Growth for Irreversible Aggregation

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). We have found that the surface roughness also crucially depends on edge diffusion, as represented by the parameter $D_{\rm edge}$ in eq. (3).

In particular, we find that the surface roughness increases as the edge diffusion increases. This is illustrated in Fig. 3. This behavior is somewhat surprising, since

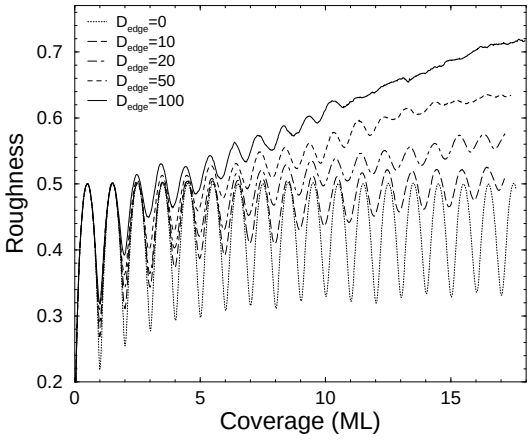

Figure 3: Surface roughness for different values of the edge diffusion parameter.

one would initially expect that the overall effect of edge diffusion is a smoothing of the surface. The explanation for this effect is the following: 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 [15].

3.3 Extension to Reversible Aggregation

We have extended our model to include the effects of adatom detachment from island boundaries. Details of the model and its implementation are given in [16]. The main idea of this extension is that the velocity of the island boundaries is modified, by adding a (negative) shrink velocity $v_{\rm shrink}$. This quantity $v_{\rm shrink}$ is a function of a microscopic detachment rate $D_{\rm det}$, and also includes some information about the local environment of each islands. More precise, we accout for the effect that the effective escape rate from an isolated island is smaller then the one for an island that has many other islands in its proximity.

The important consequence of our approach is that frequent atomistic events (such as detachment and subsequent re-attachment of atoms from island boundaries) are not resolved explicitly. Rather, they are treated in a mean-field approach, without a need to change the numerical timestep. Thus, fast events can properly be included without an increase in computational time, as is illustrated in Fig. 4. This is in contrast to atomistic

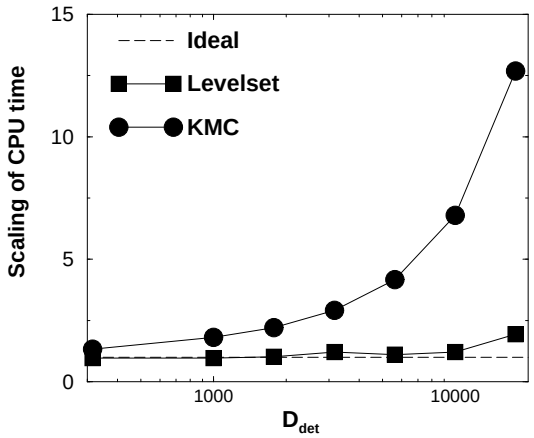

Figure 4: Scaling of CPU time for different values of the detachment rate.

methods (such as KMC), where every event is resolved explicitly. We have shown in [16] that the results obtained with this approach are essentially the same as those obtained from the atomistic KMC model.

4 DISCUSSION AND CONCLUSION

We have described a basic model for homoepitaxy on an isotropic substrate. Anisotropy in the substrate can be readily included at the level of surface diffusion and

in the attachment rates, the latter through the velocity function [7]. Moreover, the coupling to other external continuous fields can also be carried out within this general framework. At every time step, the velocity of all island boundaries is calculated from the integration of the diffusion equation. The solution of other global fields might influence the velocity as well. For example, the solution of an elastic field can be applied to modeling the strain relaxation in heteroepitaxial systems.

The mean-field treatment of fast events, such as detachment and re-attachment of atoms from island boundaries, allows us to properly accout for these processes without explicitly resolving them. Thus, they can be included without any extra computational costs. Therefore, our model is ideally suited for other processes and applications where the rates of the relevant events span a large scale. This is in contrast to a typical atomistic simulation, where such a large span in the rates makes an atomistic simulation compuationally extremely expensive. Additionally, it is rather straightforward in our model to accout for the presence of multiple diffusing species. In the simplest case, the effect of the additional species can be subsumed by solving a separate diffusion equation.

REFERENCES

[1] J. Venables, Philos. Mag. **27**, 697 (1973).

[2] J. Villain, J. Phys. I **1**, 19 (1991).

[3] J. Krug, Adv. Phys. **46**, 139 (1997).

[4] M. Schneider, I. K. Schuller, and A. Rahman, Phys. Rev. B **36**, 1340 (1987); M. H. Grabow and G. H. Gilmer, Surf. Sci. **194**, 333 (1988); B. W. Dodson, CRC Crit. Rev. Sol. State and Mater. Sci. **16**, 115 (1990).

[5] J. D. Weeks and G. H. Gilmer, Adv. Chem. Phys. **40**, 157 (1979); S. Clarke and D. D. Vvedensky, Phys. Rev. Lett. **58**, 2235 (1987); A. Madhukar and S. V. Ghaisas, CRC Crit. Rev. Sol. State and Mater. Sci. **14**, 1 (1988); H. C. Kang and W. H. Weinberg, J. Chem. Phys. **90**, 2824 (1989); H. Metiu, Y.-T. Lu, and Z. Y. Zhang, Science **255**, 1088 (1992).

[6] M.F. Gyure, C. Ratsch, B. Merriman, R.E. Caflisch, S. Osher, J.J. Zinck, and D.D. Vvedensky, Phys. Rev. E **58**, R6927 (1998).

[7] R.E. Caflisch, M.F. Gyure, B. Merriman, S. Osher, C. Ratsch, D.D. Vvedensky, and J.J. Zinck, Appl. Math. Lett. **12**, 13 (1999).

[8] S.J. Osher and J.A. Sethian, J. Comput. Phys. **79**, 12 (1988).

[9] S. Chen, B. Merriman, S.J. Osher, and P. Smereka, J. Comput. Phys. **135**, 8 (1997).

[10] C. Ratsch, M. Kang, and R.E. Caflisch, Phys Rev. E **64**, 020601 (2001).

[11] S. Chen, B. Merriman, M. Kang, R.E. Caflisch, C. Ratsch, L.-T. Cheng, M. Gyure, R.P. Fedkiw, C. Anderson, and S. Osher, J. Comput. Phys. **167**, 475 (2001).

[12] G.S. Bales and D.C. Chrzan, Phys. Rev. B **50**, 6057 (1994).

[13] J.A. Stroscio and D.T. Pierce, Phys. Rev. B **49**, 8522 (1994).

[14] C. Ratsch, M.F. Gyure, S. Chen, M. Kang, and D.D. Vvedensky, Phys. Rev. B **61**, R10598 (2000).

[15] C. Ratsch, M.F. Gyure, R.E. Caflisch, F. Gibou, M. Petersen, M. Kang, J. Garcia, and D.D. Vvedensky, submitted to Phys. Rev. B.

[16] M. Petersen, C. Ratsch, R.E. Caflisch, and A. Zangwill, Phys. Rev. E **64**, 061602 (2001).