Phase-field modeling of step dynamics

J.S. Lowengrub[1], Zhengzheng Hu[1], S.M. Wise[1], J.S. Kim[1], A. Voigt[2]

[1]Dept. Mathematics,

University of California, Irvine, CA 92697

[2]The Crystal Growth Group

Caesar Research Center, Friedensplatz 16, 53111, Bonn, Germany.

ABSTRACT

A phase-field model is used to simulate the dynamics of step-flow during epitaxial growth. An efficient numerical method is developed using a second order accurate fully implicit time discretization together with a second order accurate finite difference spatial discretization. To test the algorithm, we focus on simulations of kinetic instabilities that arise due to the anisotropy of adsorption (attachment) rates at steps from the upper and lower terraces during growth. When the attachment rate from the lower terrace is larger than that from the upper terrace (Ehrlich-Schwoebel effect), step meandering occurs. When the opposite is true of the rates, step bunching occurs. Step-step interactions are seen to reduce the meandering instability.

INTRODUCTION

At the nanoscale, the surface of a crystalline solid is composed of atomic steps that separate terraces differing in height by a single lattice spacing. During crystal growth, steps provide the kink cites at which new atoms are incorporated into the crystal. During epitaxial growth, atoms are fluxed onto the surface of a crystal from a vapor for example by molecular beam expitaxy. At the surface, atoms are adsorbed (adatoms), desorbed and diffused. Epitaxial growth is a means to produce almost defect-free, high quality single crystals that have a wide range of applications including semiconductor technologies. Growth however is subject to instabilities that can lead to irregular or wavy surface patterning (e.g. [1]).

In this paper, a phase-field model is used to simulate the dynamics of step-flow during epitaxial growth. Advantages of using a phase-field approach include the automatic capture of topological changes such as step merger or island formation. In addition, it is straightforward to include additional physical effects such as nucleation, edge-diffusion, anisotropy and elastic interactions within a phase-field framework. There has been significant recent work on the development of phase-field models for step motion, e.g. see [2, 3, 4, 5]. Kinetic Monte Carlo methods have also been used to study step dynamics (e.g. see [6, 7]) although the time scales over which these simulations can be performed is limited. For the related problem of island dynamics, adaptive sharp interface methods have been used by E. Bänsch et al. [8] and level-set methods have been developed (e.g. see Ratsch et al. [9]).

The classical approach of Burton, Cabrera and Frank [10] (BCF) is to formulate a step-flow model in which the growth direction is discrete but the lateral direction is continuous. Let $\rho_i = \rho_i(x, y, t)$ be the adatom density on a terrace, Ω_i where $i = 0, \ldots N$ is the discrete height of the layers. Then,

$$\partial_t \rho_i - \nabla \cdot (D \nabla \rho_i) = F - \tau^{-1} \rho_i, \quad \text{in } \Omega_i(t), \tag{1}$$

where D is the diffusion constant, F is the deposition flux rate and τ^{-1} is the desorption rate. At the step edges Σ_i, the adatom density satisfies the kinetic boundary conditions

$$-D \nabla \rho_i \cdot \mathbf{n}_i = k_+ \left(\rho_i - \rho^* \left(1 + \sigma \kappa_i \right) \right) \tag{2}$$
$$D \nabla \rho_{i-1} \cdot \mathbf{n}_i = k_- \left(\rho_{i-1} - \rho^* \left(1 + \sigma \kappa_i \right) \right) \tag{3}$$

where $\mathbf{n}_i$ is the unit normal pointing from the upper to lower terrace, k_+, k_- are the kinetic attachment rates from the upper and lower terraces respectively, ρ^* is the equilibrium value of the adatom density for a straight step, κ_i is the curvature of Σ_i and σ is the capillary length. The normal velocity of a step-edge is given by

$$V_i = -D \nabla \rho_i \cdot \mathbf{n}_i + D \nabla \rho_{i-1} \cdot \mathbf{n}_i + \nu \Delta_{s_i} \kappa_i \tag{4}$$

where ν is the surface mobility and $\Delta_{s_i} = (I - \mathbf{n}_i \mathbf{n}_i) \nabla \cdot (\mathbf{I} - \mathbf{n}_i \mathbf{n}_i) \nabla$ is the surface Laplacian. See, for example, [10, 11, 12, 1] for details.

In this paper, we will focus on the case in which the evolution is quasi-steady. That is, then the time scale on which the density relaxes is much smaller than the time scale for step motion: $l_T^2 / D << 1/F$ where l_T is a typical terrace width. We also assume that the time for an adatom to reach a step is much smaller than the time scale to desorb: $l_T^2 / D << \tau$ and that the equilibrium density $\rho^* << F\tau << 1$. This implies that the desorption term may also be neglected.

PHASE-FIELD MODEL

In the phase-field approach, each step is treated as a diffuse interface with thickness ϵ. Each terrace can be viewed as a phase of the system and accordingly is described by a phase-field variable ϕ^ϵ. The phase function ϕ^ϵ also can be interpreted as a continuous height function of the growing film. The phase-field model we consider is a combination of the models in [2, 4, 3] and results in a viscous Cahn-Hilliard equation:

$$\partial_t \phi^\epsilon = \nabla \cdot \left(M \left(\phi^\epsilon, \epsilon \right) \nabla \rho^\epsilon \right) + F, \tag{5}$$
$$\alpha \epsilon^2 \partial_t \phi^\epsilon = \epsilon^2 \Delta \phi^\epsilon - G'(\phi^\epsilon) + \frac{\epsilon}{\rho^* \sigma} \left(\rho^\epsilon - \rho^* \right), \tag{6}$$

where ρ^ϵ is the approximate adatom density, $M(\phi, \epsilon)$ is the (anisotropic) mobility, α is a phase-field parameter, and $G(\phi)$ is the multi-well free energy function:

$$G(\phi) = 18 \left(\phi - i \right)^2 \left(i + 1 - \phi \right)^2, \quad \phi \in [i, i+1] \tag{7}$$

where $i = 0, 1, \ldots, N - 1$. Note that $G(\phi + 1) = G(\phi)$ for $\phi \in [0, N - 1]$ and G has the symmetry $G(i + 1/2 + \phi) = G(i + 1/2 - \phi)$. To account for the Erhlich-Schwoebel barrier, the mobility is asymmetric:

$$M(\phi, \epsilon) = \frac{D}{1 + \epsilon^{-1}\zeta(\phi)} + \epsilon^{-1}\eta(\phi) \tag{8}$$

where $\zeta(\phi) = f(\phi)G(\phi)$, with $f(\phi) = \gamma(\phi - i)^p$ when $\phi \in [i, i+1]$ and $i = 0, \ldots, N - 1$. Further, $p = 2k_-/k_+ - 2$, $\gamma = \dfrac{D}{3k_+}(p+3)(p+4)$. The other function in the mobility is $\eta(\phi) = 2\dfrac{\nu}{\rho^*\sigma}G(\phi)$. Following the asymptotic analysis in [2, 3] it can be shown that if the parameter α is taken to be

$$\alpha = \frac{1}{D\rho^*\sigma} \int_0^1 \frac{\zeta(\phi)}{\sqrt{2G(\phi)}}(1 - \phi)\phi \, d\phi, \tag{9}$$

then the phase-field model described above reduces to the classical quasi-steady BCF model without desorption at leading order in an asymptotic expansion in ϵ.

NUMERICAL METHOD

We solve the system (5)-(6) using nonlinear multigrid methods originally developed for Cahn-Hilliard equations by Kim, Kang and Lowengrub [13, 14]. These algorithms are based on splitting the fourth order Cahn-Hilliard equation into two second order equations and solving for the concentration and chemical potential simultaneously using second order accurate discretizations in time and space. The spatial discretizations use centered differences and are conservative. The time discretizations are based on generalizations of the Crank-Nicholson algorithm and are fully implicit, thus eliminating the high (fourth) order time step restrictions (i.e. $\Delta t \leq C\Delta x^4$). In certain cases, it is possible to rigorously prove that the resulting numerical schemes converge [14] and that the schemes inherit a discrete version of the continuous energy functional for any time and space step sizes [13, 14].

To solve the nonlinear discrete system, a nonlinear full approximation storage (FAS) method is used. The convergence of our multigrid algorithms is achieved with $\Delta t \leq \Delta t_0$ where Δt_0 depends only on the physical parameters and not on the mesh size. Typically we find that our algorithms are 1000 times faster than corresponding explicit methods.

RESULTS

Step-flow, as described by the phase field model (5) and (6), depends on three nondimensional parameters:

$$q = k_-/k_+, \quad \xi_+ = D/(k_+ l) \quad \text{and} \quad \tilde{\epsilon} = \epsilon/l, \tag{10}$$

where l is the length scale $l = (D\rho^*\sigma/F)^{1/3}$. At this length scale, the variations in adatom density due to curvature and flux balance one another. As a time scale, we use the deposition flux time scale $t_{dep} = F^{-1}$.

To simulate an infinite step train, quasi-periodic boundary conditions are used: ρ^ϵ is assumed to be periodic in both coordinate directions while ϕ^ϵ is periodic in the y-direction and in the x-direction $\phi^\epsilon(x_{left}, y) = \phi^\epsilon(x_{right}, y) + N$ where N is the total number of steps. That is, as a step moves out of the domain at the right, it re-enters the domain from the left and grows in height by one atomic level.

In Figure 1[a], the evolution of a single step is shown under the Ehrlich-Schwoebel (ES) effect with $q = 11$, $\xi_+ = 10$ and $\tilde{\epsilon} = 0.1$. The (nondimensional) computational domain is $[0, 2\pi]^2$ and 256 nodes are used in each coordinate direction corresponding to a mesh size of $\Delta x = 0.025$. There are approximately 8 computational nodes across the step edge. The (nondimensional) time step $\Delta t = 10^{-5}$ and the times shown are $t = 0$, $t = 1$ and $t = 2$ respectively (from left-to-right).
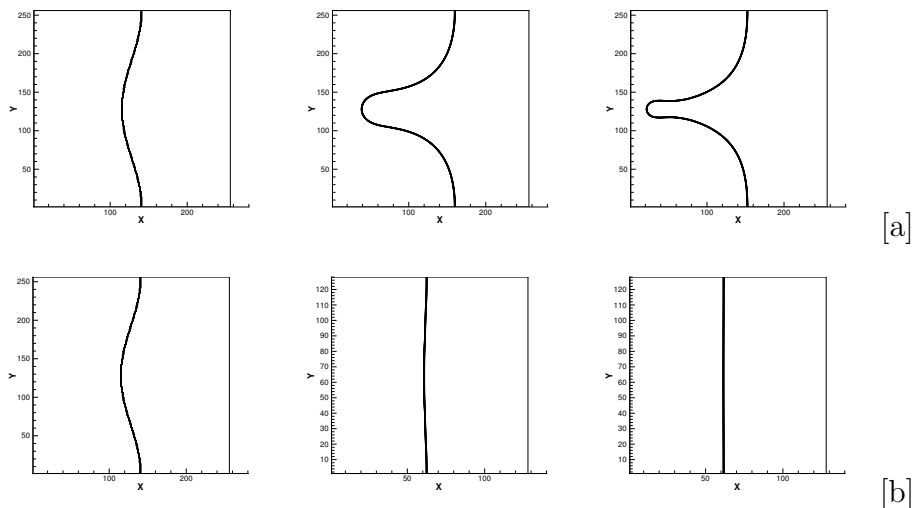

Figure 1: Time evolution of a single step. [a] Ehrlich-Schwoebel (ES) effect with $k_-/k_+ = 11$ and $D/(k_+ l) = 10.0$. [b]. Local equilibrium $k_-/k_+ = 1$ and $k_+ \to \infty$. The finger grows under the ES effect due to the Bales-Zangwill instability [15]. Perturbations decay if the step is in local equilibrium.

The step in Figure 1[a] moves from left-to-right with the upper terrace being on the left of the step. As seen from the figure, the step undergoes the Bales-Zangwill instability [15]. The perturbation grows forming an elongated, narrow finger that penetrates the upper terrace. In Figure 1[b], the same initial perturbation of the step decays leading to the development of flat step. In this case, the attachment rate is symmetric, $q = 1$, and we have taken the limit $k_+ \to \infty$ so that the system is in local equilibrium. This is consistent with the results of linear stability theory that show that steps are always stable in this regime.

In Figure 2, the time evolution of 10 steps is shown. The initial data is a monotone step train where the step heights differ by one atomic unit with the upper terrace being on the left of the step. The step train moves through the domain from left-to-right. Here, $\tilde{\epsilon} = 0.2$, the computational domain is $[0, 2\pi]^2$, 128 grid points are used in each coordinate direction and $\Delta t = 5 \times 10^{-5}$. The solutions are shown at (nondimensional) times $t = 0$, $t = 5$ and $t = 10$.

In Figure 2[a], the steps move under the ES effect with $q = 11$ and $\xi_+ = 10$. Step-step interactions are seen to reduce the Bales-Zangwill instability as the initially perturbed steps tend to straighten under conditions that would result in instability for a single step (cf. Fig. 1[a]). In addition, there is step-step repulsion [16, 17] with the result being the formation of a step train consisting of equally-spaced steps. In contrast, in Figure 2[b], the steps move under the inverse ES effect with $q = 1/11$ and $\xi_+ = 10/11$. As seen from the figure, the steps straighten and bunch together. This is consistent with the linear theory of Bales-Zangwill [15] as the decrease in terrace width limits the action of the diffusion field.
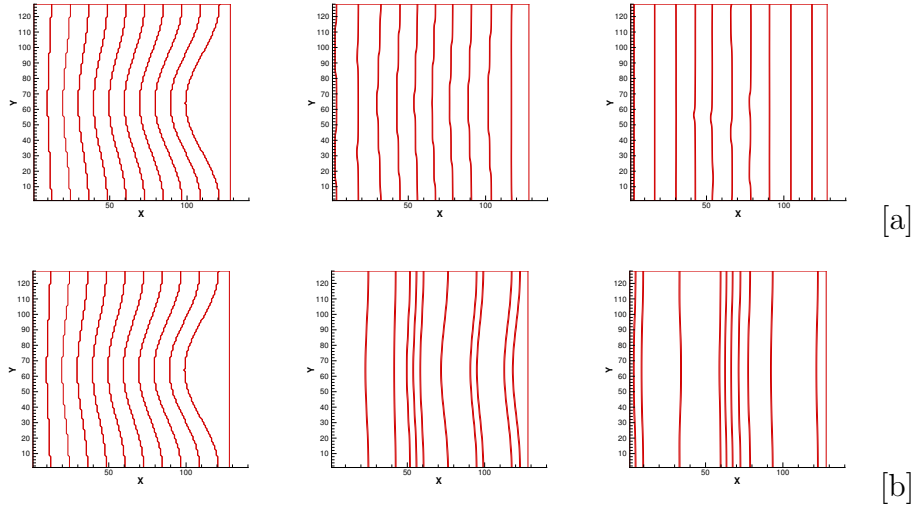

Figure 2: Time evolution of 10 steps. [a] The Ehrlich-Schwoebel (ES) effect with $k_-/k_+ = 11$ and $\xi_+ = 10.0$. [b]. The Inverse Ehrlich-Schwoebel (IES) effect with $k_-/k_+ = 1/11$ and $\xi_- = 10/11$. The steps tend to straighten and repel under the ES effect leading to a step train consisting of nearly straight, equidistant steps. Under the IES the steps straighten and bunch.

CONCLUSIONS AND FUTURE WORK

An efficient, second order accurate finite difference scheme was used to simulate a phase-field model of the dynamics of step-flow during epitaxial growth. Step meandering and bunching was studied. Step-step interactions were seen to reduce the meandering instability. In future work, we will investigate the effects of edge diffusion, anisotropy and elastic interactions as these phenomena can play a critical role in step meandering and bunching instabilities [1].

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