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Phase-field modeling of epitaxial growth: Applications to step trains and island dynamics

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ABSTRACT

In this paper, we present a new phase-field model including combined effects of edge diffusion, the Ehrlich–Schwoebel barrier, deposition and desorption to simulate epitaxial growth. A new free energy function together with a correction to the initial phase variable profile is used to efficiently capture the morphological evolution when a large deposition flux is imposed. A formal matched asymptotic analysis is performed to show the reduction of the phase-field model to the classical sharp interface Burton–Cabrera–Frank model for step flow when the interfacial thickness vanishes. The phase-field model is solved by a semi-implicit finite difference scheme, and adaptive block-structured Cartesian meshes are used to dramatically increase the efficiency of the solver. The numerical scheme is used to investigate the evolution of perturbed circularly shaped small islands. The effect of edge diffusion is investigated together with the Ehrlich–Schwoebel barrier. We also investigate the linear and nonlinear regimes of a step meandering instability. We reproduce the predicted scaling law for the growth of the meander amplitude, which was based on an analysis of a long wavelength regime. New nonlinear behavior is observed when the meander wavelength is comparable to the terrace width. In particular, a previously unobserved regime of coarsening dynamics is found to occur when the meander wavelength is comparable to the terrace width.

1. Introduction

Thin film growth through for example, molecular beam epitaxy (MBE), liquid phase epitaxy (LPE) and chemical vapor deposition (CVD) is a modern technology of growing single crystals that inherit atomic structures from a substrate [1]. Epitaxial growth produces almost defect-free, high quality crystals, which have wide ranges of applications in electronic, optical and magnetic materials. For example, epitaxial growth is useful in the manufacture of reflective or anti-reflective coatings for optics, and is important in the fabrication of layers of insulators and semiconductors for integrated circuits (e.g. see [2]). Moreover, epitaxial growth can be used to create structures on much smaller length scales through self-assembly, that is the nanostructure emerges spontaneously, rather than structures produced by top-down methods. Examples include quantum dots and quantum wires which have very good transport and optical properties, and therefore have specific importance in the development of diode lasers, amplifiers, biological sensors and etc. (e.g. see [3,4]). The fundamental problem in epitaxial growth is to understand growth processes so that one can develop techniques to control nanostructure formation and promote self-assembly of spatially ordered nanostructures.

During epitaxial growth, physical and chemical processes occur at widely varying length and time scales. Examples of such phenomena include, the chemical interaction of the film and substrate; the heteroepitaxial misfit between substrate and film; the formation of defects, such as dislocations and grain boundaries; the extreme elastic heterogeneity of the system; the strong elastic and surface anisotropies; interface kinetic effects; epilayer deposition; edge diffusion; substrate topographical patterning and subsurface implant patterning. These processes interact and compete to form complex thin film morphologies, such as step structures, and faceted quantum dots and wires. Given such a complex multi-scale problem, modeling epitaxial growth presents an enormous challenge to mathematicians, theoretical physicists and materials scientists. Since the macroscopic evolution of the growing film is directly related to the movements of adatoms

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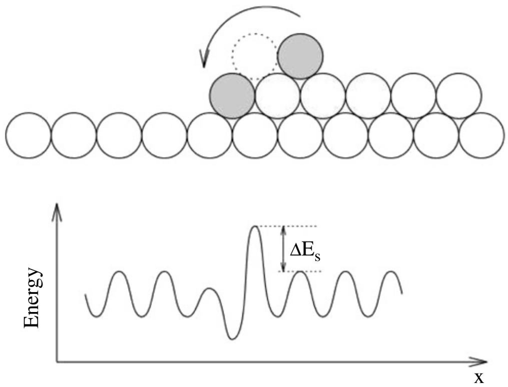

Fig. 1. Schematic of Ehrlich–Schwoebel barrier. An adatom detaching from a step edge is experiencing an additional energy ΔE_s [30].

(absorbed atoms) on surfaces and their various interactions, it is appealing to use atomic scale simulations for a theoretical description of epitaxial growth (e.g., ab-initio [5] molecular dynamics [6] and kinetic Monte Carlo [7–9] models). However, the length and time scales that can be achieved by the atomic scale simulations are limited, thus semi-discrete step-flow models (e.g., [10–15]) and continuum models (e.g., [16–27]) may be used in order to study various applications at larger scales.

Below the roughening temperature, steps as long-lived surface defects are suitable as a basis for the description of the surface morphology. Atomic steps separate exposed lattice terraces that differ in height by a single lattice spacing and provide the kink sites at which new atoms are incorporated into the crystal. The growth of a crystal surface can thus be reduced to the advancement of existing steps, the nucleation and growth of new closed step loops (i.e., atomic height islands), the annihilation of steps by the merging of islands and terraces. Burton, Cabrera and Frank (BCF) [28] first introduced a semi-discrete model, in which the growth direction is discrete but the lateral direction is continuous, to describe the diffusion of adatoms and the motion of steps during epitaxial growth of thin films. The BCF model, supplemented with later modifications and extensions, has been used to study the stability of step trains and islands.

A fundamental investigation to predict the meandering wavelength for the step meandering instability using linear stability analysis, was performed by Bales and Zangwill [10]. Their analysis shows that step meandering is an instability that may arise as a result of a terrace Ehrlich–Schwoebel barrier, see Fig. 1, which characterizes the preference of adatoms to attach to an ascending step, i.e., $k_+ < k_-$ in Eqs. (2) and (3). More recently, following Avignon and Chakraverty [13], Mullins and Sekerka [14] and Li et al. [15], a comprehensive morphological stability analysis of small circularly shaped islands was performed by Hu et al. in [29]. In this work, Hu et al. demonstrated the existence of a naturally stabilizing radius of the growing island, so that beyond this radius, the growth is always stable. Up to this radius, taking fluxes larger than a critical flux results in unstable growth. The explicit form of the dispersion relation is given in a supplementary document. Hu et al. also suggested a way of controlling the shape of an island using the deposition flux and far-field flux as control parameters. However, in the nonlinear regime, mathematical modeling and efficient numerical algorithms are essential and it remains to determine whether shape control may be achieved in the nonlinear regime.

In this paper, a new phase-field model for step flow accounting for Ehrlich–Schwoebel barrier, edge diffusion, a far-field flux, deposition and desorption, is presented and used to investigate instabilities during epitaxial growth. To accurately and efficiently capture the dynamics when the deposition flux is large, we propose a different free energy function from that used by Rätz et al. [31] and Otto et al. [32]. An analysis using matched asymptotic expansions is performed to show that the phase-field model reduces to classical sharp interface models of BCF type when the interfacial thickness vanishes. Advantages of using a phase-field approach include the automatic capture of topological changes such as island formation, coalescence and coarsening. In addition, other physical effects such as nucleation and elastic interactions may be included. Previously, front tracking methods have been used to study the combined effects of edge diffusion and the Ehrlich–Schwoebel during island growth [33–35]. In phase field simulations [36,37] the edge diffusion term is typically neglected. Only in [2] was this effect considered, however attachment and detachment processes were neglected. In [38], a level-set method was used, where the edge diffusion term in the normal velocity was approximated by the deviation of the curvature from the averaged curvature. But none of these methods considered the combined effects of edge diffusion, the Ehrlich–Schwoebel barrier and desorption.

Moreover, to demonstrate the versatility of the proposed phase-field model, we also investigate the linear and nonlinear regimes for the step trains concentrating on the step meandering instability. A similar study has been performed by Haußer and Voigt [39] in which a front tracking method based on linear adaptive finite elements is used. In this work, we confirm some of their nonlinear results and identify a new regime of coarsening with different sets of parameters. In particular, we reproduce the predicted scaling law for the growth of the meander amplitude, which was based on an analysis of a long wavelength regime [40–42]. New nonlinear behavior is observed when the meander wavelength is comparable to the terrace width.

This paper is organized as follows: In Section 2, we describe the BCF model. In Section 3, the new phase-field model is presented. In Section 4, we briefly discuss the numerical solution of the phase-field model. In Section 5, we present and discuss our numerical results applied on both island dynamics and step trains. We give some concluding remarks and suggest some future work in Section 6. In Appendix A, a formal matched asymptotic analysis is performed to show the reduction of the phase-field model to the classical sharp interface BCF model for step flow when the interfacial thickness vanishes. The computation of the surface Laplacian is outlined in Appendix B. We present the initial condition used in the simulation of the growth under a constant flux (Section 5.1.1) in Appendix C. In Appendix D, the dispersion as appeared in [10] is presented. A supplementary document contains details of the linear stability analysis.

2. The BCF model

2.1. The BCF model for island dynamics

We consider a domain Ω on a plane containing a sequence of steps and terraces. To model island dynamics, the steps are described by closed curves Γ_i, which divide Ω into terraces Ω_i where $i = 0, \ldots, N$ denotes the discrete height of the layers, cf. Fig. 2.

Let $\rho_i = \rho_i(x, y, t)$ be the adatom concentration on a terrace Ω_i, with $i = 0, \ldots, N$. Then, the BCF model [28] is

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

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

$$-D\nabla \rho_i \cdot \mathbf{n}_i = k_+ \left(\rho_i - \rho^* \left(1 + \sigma \kappa_i\right)\right) \quad \text{on } \Gamma_i, \tag{2}$$

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