Stable Numerical Integration of an Epitaxial Growth Model with Slope Selection

Gregory M. Seyfarth\textsuperscript{a}, Benjamin P. Vollmayr-Lee\textsuperscript{b,*}

\textsuperscript{a}Department of Physics and Astronomy, Colby College, Waterville, Maine 04901, USA
\textsuperscript{b}Department of Physics and Astronomy, Bucknell University, Lewisburg, Pennsylvania 17837, USA

Abstract
We consider a continuum phase field model for crystal growth via molecular beam epitaxy, with the goal of determining stable numerical time integration methods for the dynamics. We parametrize a class of semi-implicit methods that are linear in the updated field, which allows for efficient implementation with fast Fourier transforms. We perform unconditional von Neumann stability analysis to identify the region of stability in parameter space, and then test these predictions numerically for gradient stability. We find strong agreement between the approaches.

Keywords: Epitaxial crystal growth, slope selection, coarsening, gradient stability, unconditional von Neumann stability

1. Introduction

In growing crystal surfaces by molecular beam epitaxy (MBE), the Ehrlich-Schwoebel-Villain effect \cite{1, 2, 3} can destabilize a flat interface and lead to the formation of pyramids or mounds (see \cite{4} for a recent review). These surface features then coarsen, with their height and spatial extent growing as powers of time. Theoretical studies of MBE coarsening typically employ continuum models, justified by appeal to the large distance and slow time scales involved. The resulting field equations of motion are nonlinear, and to make progress they must be integrated numerically, a process which, unfortunately, is hampered by numerical instabilities. As such, much recent effort

\*Corresponding author
has been devoted to finding stable integration methods. In this work, we
derive a class of stable numerical integration methods that are particularly
efficient and simple to implement because the updated field can be obtained
via the fast Fourier transform (FFT).

The model we consider employs a height field \( h(x, y, t) \) that is a continuous
function of space and time, and which obeys the equation of motion

\[
\frac{\partial h}{\partial t} = -\nabla^4 h - \nabla \cdot \left\{ (1 - |\nabla h|^2) \nabla h \right\},
\]

applicable for homoepitaxial growth with isotropic slope selection. The moti-
vation for this and related models is discussed below. With these dyna-
amics, equilibrated regions of uniform gradient and unit slope form. Domains
with different slope orientations meet at edges of constant width, and as
the system evolves the edges are healed out, resulting in the growth of the
characteristic domain size. For this particular model it has been found from
theoretical analysis [5, 6, 7], simulations [6, 8], and rigorous bounds [9] that
the characteristic domain size \( L(t) \) grows with time as \( L \sim t^{1/3} \).

Numerical simulations of coarsening are useful for testing scaling and
the predicted growth laws and for measuring properties of the scaling state,
such as correlations, growth law amplitudes, autocorrelation functions, and
more (see [10] for a coarsening review). But these simulations face several
restrictions. To reach the asymptotic scaling regime, it is necessary to evolve
until \( L(t) \gg w \), where \( w \) is the width of the edges. But the lattice size \( \Delta x \)
must be sufficiently smaller than the edge width in order to resolve the edge
shape and corresponding line tension. Finally, the system size \( L_{sys} \) must be
large enough that domains can grow into the scaling regime before finite size
effects appear. To satisfy this string of conditions, \( \Delta x \ll w \ll L(t) \ll L_{sys} \),
requires lattices of very large linear size \( L_{sys}/\Delta x \), evolved to late times.

For this reason, it is desirable to use integration schemes that are accuracy-
limited rather than stability-limited. Euler integration of Eq. (1) is only
stable for time steps \( \Delta t \) smaller than a threshold determined by the lattice
spacing. In contrast, an unconditionally stable method, i.e., one with no
conditions on \( \Delta t \), would allow a time step determined by the natural time
scale of the dynamics, which turns out to be considerably more efficient. Ac-
curacy considerations require the typical distance traveled by an edge within
one step to be held fixed [11, 12], and since the characteristic edge velocity
scales as \( v_{edge} \sim \partial L/\partial t \sim t^{-2/3} \), this allows a growing time step \( \Delta t \sim t^{2/3} \).
Using \( dt/dn \sim \Delta t \), where \( n \) is the number of integration steps, it follows
that unconditionally stable methods allow accurate evolution with \( t \sim n^3 \), rather than the stability-limited \( t \sim n \). For typical simulation parameters, this provides greater than a 1000-fold increase in efficiency!

Eyre provided a general approach for generating unconditionally stable semi-implicit integration methods, based on a splitting into expansive and contractive terms [13]. Wang, Wang, and Wise used this approach for Eq. (1), as well as an MBE model without slope selection [8], and this approach has now been extended to a second-order in time method [14] and other developments [15, 16, 17]. These schemes are gradient stable, which means they preserve the energy-decreasing property of the continuous-time equation. However, these Eyre-based schemes have the drawback that usually a nonlinear term must be treated implicitly, requiring an iterative method to find the updated field, and in the worst case no guarantee of convergence or a unique solution. An alternate approach is to restrict consideration to steps with linear implicit terms that can be solved directly by FFT, determine the range of step parameters that satisfy unconditional von Neumann (UvN) stability, and then test these parameters numerically for gradient stability. This approach yielded stable, direct steps for the Cahn-Hilliard and Allen-Cahn equations [11], and it is the program we follow here for the MBE model.

Our primary results are the following: for the equation of motion, Eq. (1), there exists a class of first order, semi-implicit steps

\[
ht_{t+\Delta t} = ht + \Delta t \left[ -\nabla^4 ht - \nabla \cdot \left\{ (1 - |\nabla ht|^2) \nabla ht \right\} \right] + b_1 \Delta t \nabla^2 (ht_{t+\Delta t} - ht) + b_2 \Delta t \nabla^4 (ht_{t+\Delta t} - ht) \tag{2}
\]

that provides stable numerical integration for appropriate choice of the parameters \( b_1 \) and \( b_2 \), as shown in Fig. 1. The results of our UvN stability analysis are presented as shaded regions while our numerical tests of gradient stability are plotted as points. Although UvN stability does not ensure gradient stability, we find that it is very effective in determining the gradient stable regions, both for single- and many-domain systems. The UvN stability conditions plotted here are independent of lattice type or details of the numerical method (e.g., finite difference versus spectral methods). The difference in stability range for the single- and many-domain systems is revealed by the UvN stability analysis, which shows that the most unstable Fourier mode is that with its wavevector oriented with the local slope \( \nabla h \). In the many-domain system, each mode samples many different slope directions, which acts to suppress the instability for the parameter range \(-1 < b_1 < -1/2\).
Figure 1: (color online) Stability diagram for the parameters $b_1$ and $b_2$ in Eq. (2). The UvN stable parameter values are shaded in gray, with the darker region corresponding to a single-domain system and the combined gray regions corresponding to a many-domain system. The points represent numerical tests of gradient stability: the (blue) circles are parameter values that are stable for single-domain systems; these together with the (purple) squares are stable for multi-domain systems; and the $\times$ are parameter values that were found to be unstable.
Our results are consistent with Xu and Tang, who proved gradient stability for the parameters \( b_1 < -1 \) and \( b_2 = 1 \) [18]. Further work by one of us has led to a demonstration of gradient stability for the entire dark gray shaded region of Fig. 1 [19]. This proof will be presented elsewhere, as it is considerably more general than the model considered here, and does not distinguish the single- and many-domain cases captured by the UvN stability analysis.

While our analysis is focused on the isotropic model, Eq. (1), our results can be generalized straightforwardly to anisotropic growth, where only a discrete set of slope orientations are preferred. We demonstrate this explicitly for a model with square symmetry, appropriate for growth on (100) surface.

The remainder of the paper is as follows. In Sec. 2 we review some of the properties of the model to provide necessary background for subsequent sections. In Sec. 3 we present the UvN stability analysis, both for single- and many-domain systems. We describe the numerical tests of gradient stability in Sec. 4 as well as providing the details of our finite-difference implementation of Eq. (2). In Sec. 5 we extend our analysis to the anisotropic model with square symmetry. This is followed by a summary in Sec. 6.

2. The Continuous Time Model

In this section we provide motivation for the model we are considering and present some of its properties, showing in particular the instability to pyramid formation and the energy decreasing dynamics of the continuous time model.

The height field, \( h(x, y, t) \), is defined in a co-moving frame so that its average is zero, and obeys a continuity equation. The current \( \mathbf{J} \) has an equilibrium surface diffusion contribution equal to the gradient of the local curvature, \( \mathbf{J}_{SD} = \nabla (\nabla^2 h) \) [20], and a non-equilibrium component \( \mathbf{J}_{NE} \):

\[
\frac{\partial h}{\partial t} = -\nabla \cdot \mathbf{J} = -\nabla^4 h - \nabla \cdot \mathbf{J}_{NE}. \tag{3}
\]

A noise term is omitted as this is considered to be irrelevant for coarsening [10]. We consider the slope-selecting nonequilibrium current

\[
\mathbf{J}_{NE} = (1 - |\nabla h|^2) \nabla h, \tag{4}
\]

which gives \( \mathbf{J}_{NE} \sim \nabla h \) for small gradients, an uphill current due to the Ehrlich-Schwoebel-Villain effect [3], and \( \mathbf{J}_{NE} = 0 \) for slopes of unit magni-
itude. Inserting Eq. (4) into the continuity equation (3) yields the equation of motion, Eq. (1).

Common variations on this model include slope-selecting currents that vanish for only a discrete set of $\nabla h$ directions, reflecting the underlying crystalline structure, and models without slope selection. The physical basis and experimental evidence for these various models is described in [21, 22, 23, 5, 6] and references therein. Material parameters have been absorbed into rescaling of lateral space dimensions, height, and time.

The equation of motion, Eq. (1), can be written as a gradient flow

$$\frac{\partial h}{\partial t} = -\frac{\delta F}{\delta h}$$

for the free energy functional

$$F[h] = \int d^2x \left\{ \frac{1}{8} (\nabla h)^2 + \frac{1}{4} \left( 1 - |\nabla h|^2 \right)^2 \right\}.$$  \hspace{1cm} (6)

Gradient flow results in a monotonically decreasing free energy,

$$\frac{d}{dt} F = \int d^2x \left( \frac{\delta F}{\delta h} \right) \frac{\partial h}{\partial t} = -\int d^2x \left( \frac{\partial h}{\partial t} \right)^2 \leq 0.$$  \hspace{1cm} (7)

As first noted by Eyre [13], the essential stability criterion for discrete time steps is to preserve the energy decreasing property of the continuous-time equation. This is known as the gradient stability condition.

Next we review the the linear stability of the continuous time equation, which will be useful context for the von Neumann stability analysis in Sec. 3.

Consider a height field

$$h(x, y, t) = Cx + \eta(x, y, t),$$

which consists of small deviations $\eta$ from a uniform slope. Inserting this into Eq. (1), linearizing in $\eta$, and Fourier transforming to $\tilde{\eta}(k, t) \equiv \int d^2x \exp(i k \cdot x) \eta(x, y, t)$ gives

$$\frac{\partial \tilde{\eta}(k, t)}{\partial t} = (k^2 - k^4 - C^2k^2 - 2C^2k_x^2) \tilde{\eta}(k, t).$$

(9)

For an interface that is initially flat we set $C = 0$ and obtain the growth rate for small fluctuations in the initial conditions:

$$\frac{\partial \tilde{\eta}(k, t)}{\partial t} = k^2(1 - k^2) \tilde{\eta}(k, t).$$

(10)
Long wavelength modes with \( k < 1 \) are unstable and grow, which is exactly the instability that leads to pyramid formation. In the context of the Cahn-Hilliard equation this is the spinodal instability \[10\]. Note that the exponential growth of the mode is nevertheless accompanied by a decreasing total free energy, as required by Eq. \((7)\).

For an equilibrium interface we set the slope \( C = 1 \) to obtain

\[
\frac{\partial \tilde{\eta}(k, t)}{\partial t} = -(k^4 + k_x^2) \tilde{\eta}(k, t). \tag{11}
\]

The negative right hand side indicates that height fluctuations about the equilibrium slope decay, and the uniform slope profile is stable.

### 3. Unconditional von Neumann Stability Analysis

The goal in constructing a discrete time method is to be faithful to the physical behavior of the continuous time equation. In our case, this means our discrete step should be gradient stable, to preserve the energy-decreasing property of the continuous equation. However, in this section we analyze instead von Neumann (vN) stability, i.e. the linear stability of the discrete step, Eq. \((2)\). This analysis has certain advantages. It is relatively straightforward and, as shown in Fig. \(\[\]\) and in Ref. \[11\], it successfully predicts the parameter range for gradient stability, as judged by numerical tests. Also, the method provides insight into the dynamics of the Fourier modes, which in the present case proves useful in clarifying the distinction between the single- and many-domain systems.

We first present vN stability analysis on the Euler step, which results in conditional stability, i.e., a lattice-dependent upper bound on \(\Delta t\). Then we consider our parametrized semi-implicit step and perform unconditional vN stability analysis; that is, we seek parameter values which yield vN stable steps for any size \(\Delta t\). Note that we will only impose vN stability on the equilibrium, sloped interface and not on the flat interface, where the linear instability is part of the physical behavior of the continuum equation.

In addition to the time discretization, the spatial derivatives in our equation of motion must be treated by finite-difference or spectral methods. Without specifying the details of the scheme, we denote the Fourier transform of the two-dimensional numerical laplacian as \(\lambda(k)\). In the continuum limit, \(\lambda(k) \to -k^2\). For spatially discretized systems, \(0 \geq \lambda(k) \geq \lambda_{\text{min}}\), where the value of the lower bound \(\lambda_{\text{min}} \sim -1/\Delta x^2\) depends on the details of the
discretized laplacian. Our stability conditions will rely only on the universal upper bound of zero.

We will use $\lambda(k_x)$ to represent the Fourier transform of the numerical derivative second derivative $\partial^2 / \partial x^2$.

### 3.1. Euler Step

Our discrete time step, Eq. (2), reduces to an Euler step in the case $b_1 = b_2 = 0$. We plug in $h = x + \eta$ (i.e., slope $C = 1$), linearize in $\eta$, and Fourier transform to obtain

$$\tilde{\eta}_{t+\Delta t} = \left[ 1 + \Delta t \left\{ -\lambda(k)^2 + 2\lambda(k_x) \right\} \right] \tilde{\eta}_t. \quad (12)$$

The vN stability condition is that the square bracket term has magnitude less than unity, to ensure fluctuations die away. The negative curly bracket term in Eq. (12) has no lower bound in the continuum limit $\Delta x \to 0$, and thus the Euler step would be vN unstable for any size $\Delta t$. The situation is improved by the numerical derivative, which places a lower bound on the curly bracket terms, leading to vN stability for $\Delta t \lesssim |\lambda_{\text{min}}|^{-2} \sim \Delta x^4$. The analysis is essentially identical to what happens in the Cahn-Hilliard equation [24]. The Euler step provides an example of a lattice-dependent stability condition (relying on the lower bound of $\lambda(k)$ rather than the upper bound of zero) and it results in a fixed bound on the time step, regardless of the natural time scale of the dynamics.

### 3.2. UvN Stability for a Single Domain

We return to our parametrized discrete step, Eq. (2), but now we leave $b_1$ and $b_2$ unspecified. We seek to find ranges for the parameters which will lift any restrictions on $\Delta t$, i.e., unconditional stability. We substitute Eq. (8) with slope $C = 1$ into Eq. (2), linearize, and Fourier transform. The resulting step can be written as

$$[1 + \Delta t \mathcal{L}(k)] \tilde{\eta}_{t+\Delta t} = [1 + \Delta t \mathcal{R}(k)] \tilde{\eta}_t \quad (13)$$

with

$$\mathcal{L}(k) = b_1 \lambda(k) + b_2 \lambda(k)^2 \quad (14)$$

and

$$\mathcal{R}(k) = 2\lambda(k_x) + b_1 \lambda(k) + (b_2 - 1) \lambda(k)^2. \quad (15)$$
Before imposing the UvN stability, we note that it is necessary to have $L(k) \geq 0$ so that the square bracket on the left of Eq. (13) is non-vanishing for all $\Delta t$ and $k$. This gives the requirement that $b_1 \leq 0$ and $b_2 \geq 0$.

Next, the UvN stability condition, $|\tilde{\eta}_{t+\Delta t}| < |\tilde{\eta}_t|$ for all $\Delta t$ and $k$, will be satisfied if $L(k) > |R(k)|$. In the case that $R(k)$ is positive, this gives the condition

$$0 < L(k) - R(k) = -2\lambda(k_x) + \lambda(k)^2,$$

which is intrinsically satisfied due to the non-positivity of $\lambda(k_x)$. While here and below the $k = 0$ mode saturates the bound, we can safely ignore it since it is static.

The crucial condition, then, comes from imposing $L(k) > -R(k)$, which becomes

$$\lambda(k_x) + b_1\lambda(k) + \left(b_2 - \frac{1}{2}\right)\lambda(k)^2 > 0.$$  \hspace{1cm} (17)

The last term is positive for $b_2 > 1/2$. Next, noting that $\lambda(k_x) \geq \lambda(k)$, we have a lower bound on the remaining two terms:

$$\lambda(k_x) + b_1\lambda(k) \geq (1 + b_1)\lambda(k).$$  \hspace{.5cm} (18)

This will be positive provided that $b_1 < -1$. Thus, our conditions for UvN stability of a single-domain system are

$$b_1 < -1, \quad b_2 > 1/2,$$

which is plotted as the dark gray region of Fig. 1.

Note that for $b_1$ slightly above $-1$, in the unstable region, it is Fourier modes with $\lambda(k_x) \approx \lambda(k)$ that first violate Eq. (17). This corresponds to wavevectors $k$ that are nearly oriented along the $x$-axis, i.e. the gradient direction of the equilibrium interface.

3.3. UvN Stability for a Many-Domain System

In a many-domain system, which is the relevant case for coarsening studies, we are not free to choose the coordinate axes to align the $x$ axis with the interface gradient, since there are many facets with different gradient directions. To analyze this case, we first linearize about a single domain but with an arbitrary normal direction, parametrized by the polar coordinate $\theta$

$$h(x, y, t) = \cos(\theta)x + \sin(\theta)y + \eta(x, y, t).$$ \hspace{1cm} (20)
This follows through just as before, with the important stability condition Eq. (17) becoming

\[
\cos^2 \theta \lambda(k_x) + \sin^2 \theta \lambda(k_y) + b_1 \lambda(k) + \left(b_2 - \frac{1}{2}\right) \lambda(k)^2 > 0.
\] (21)

Now, if many domains are present in the system with essentially random orientations, then for any particular Fourier mode the above equation will be averaged over \(\theta\), giving \(\langle \cos^2 \theta \rangle = \langle \sin^2 \theta \rangle = 1/2\). Using

\[
\lambda(k_x) + \lambda(k_y) \approx \lambda(k)
\] (22)

reduces Eq. (21) to

\[
\left(b_1 + \frac{1}{2}\right) \lambda(k) + \left(b_2 - \frac{1}{2}\right) \lambda(k)^2 > 0.
\] (23)

Thus, our UvN stability condition for many-domain systems is

\[
b_1 < -1/2, \quad b_2 > 1/2,
\] (24)

which is depicted as the combined shaded regions of Fig. 1. The averaging over multiple orientations provides a greater parameter range of stability than the single-domain case.

Note that in general Eq. (22) is only an approximate relationship. It is a strict equality in the \(\Delta x \to 0\) continuum limit, and also in the common five-point stencil for the numerical laplacian on a square lattice, but for other choices of numerical derivatives it need not be exact.

4. Numerical Tests of Gradient Stability

Since the field equation of motion is nonlinear, von Neumann stability analysis is not sufficient to prove gradient stability. For that reason, we have conducted extensive numerical tests for gradient stability for a range of \(b_1\) and \(b_2\) parameter values. We present the details of the numerical derivative implementation in an appendix, but we note here two important general features such an implementation should have. First, the local conservation law should be constructed to hold exactly, not just to some order in \(\Delta x\), and second, the energy-decay property of the continuous time equation should be maintained when spatially discretizing. That is, the particular scheme of
calculating the spatially discrete analog of the free energy $F[h]$ in Eq. (6) and the equation of motion should be consistent, so that

$$\frac{d}{dt} h_{ij} = - \frac{\partial}{\partial h_{ij}} \left( \frac{F}{\Delta x^2} \right)$$

is an exact relation, not just approximate to some order in $\Delta x$.

For each $b_1$ and $b_2$ value represented as a data point in Fig. 1 we performed the following tests. We evolved a $512 \times 512$ sized lattices with lattice constant $\Delta x = 1$ out to a final time $t_{\text{max}}$. These systems were evolved using three different methods: an Euler step with $\Delta t = 0.03$ out to a $t_{\text{max}} = 10^4$, a semi-implicit step with $b_1 = 2.5$ and $b_2 = 1$ and growing time step $\Delta t = 0.03 t^{2/3}$ out to time $t_{\text{max}} = 10^6$, and the same semi-implicit parameters with a fixed time step $\Delta t = 100$ out to time $t_{\text{max}} = 10^6$. For each of these cases we analyzed multiple runs and varied between random initial conditions and sinusoidal initial conditions with long and short wavelengths.

At regular intervals during the evolution we tested a single step calculated via Eq. (2) with sizes varied between $1 \leq \Delta \leq 10^{10}$. This step was used only for energy stability testing and did not contribute to the subsequent time evolution. Any time that the free energy was found to increase, that particular set of parameter values was identified as unstable.

For the many-domain system, we used periodic boundary conditions and an initially flat interface (plus the random or sinusoidal fluctuations). For the single-domain system, we first re-write the field equation of motion, Eq. (1) in terms of deviations from the uniform slope, giving

$$\frac{\partial \eta}{\partial t} = - \nabla^4 \eta + 2 \partial_x^2 \eta + 2 \partial_x |\nabla \eta|^2 + 2 (\partial_x \eta) \nabla^2 \eta + \nabla \cdot (|\nabla \eta|^2 \nabla \eta),$$

where $\partial_x = \partial/\partial x$, and then constructed the analogous numerical implementation of this equation. This approach was necessary to eliminate sensitivity to truncation error. We imposed periodic boundary conditions on $\eta$, which corresponds to shifted periodic boundary condition on $h$.

In Fig. 1 we show the results of this testing both for the single- and many-domain systems. The (blue) circles represent parameter values that were found to be stable for the single-domain system, that is, under all our testing, there were no single incidents of energy increase. The (purple) squares are parameters values that were found to be unstable in the single-domain system, but stable for the many-domain case. The remaining $\times$ are
Figure 2: (color online) The free energy density $f = F[h]/L_{\text{sys}}^2$ as a function of time, where the time evolution utilized a growing time step, $\Delta t \sim t^{2/3}$. Simulation details are in the text. The lower (red) curve is the isotropic model, while the upper (blue) curve is for the anisotropic model with square symmetry presented in Sec. 5.

parameter values found to be unstable for both single- and many-domain systems. We find a striking degree of agreement between the predictions of UvN stability analysis and the numerical tests for unconditional gradient stability. This is one of our main results.

There is a small region for $b_2 < 1/2$ where numerical tests find gradient stability. This can be understood from Eq. (17) as a lattice-dependent stability arising from the laplacian lower bound $\lambda_{\min}$. We have emphasized instead the lattice-independent stability boundaries, as these are more widely applicable.

To illustrate the utility of these methods, we have simulated the coarsening that results from an initially flat interface, using a the stable step parameters $b_1 = -1.5$ and $b_2 = 1$ and a growing step size $\Delta t = \max(0.1, 0.01t^{2/3})$. We performed 20 independent runs on a $2048 \times 2048$ lattice with $\Delta x = 1$, out to time $t_{\max} = 10^7$.

Fig. 2 shows the decay of the free energy with time. Once equilibrated domains form, the free energy density $F$ is proportional to the amount of edge in the system, which is inversely proportional to the characteristic size of the domains. Thus the free energy should decay as $F \sim 1/L(t) \sim t^{-1/3}$. Our growing time step integration reproduces this result.

Shown in Fig. 3 are snapshots of domain configurations for various times from a single run on a $512 \times 512$ lattice, with all other parameters as given
5. Model with Square Symmetry

While the isotropic growth model, Eq. (1), provides a useful starting point for analyzing surface growth coarsening, experimental systems typically select for only a discrete set of slope orientations. For example, homoepitaxial growth on a Cu(100) surface exhibits a square symmetry with four equilibrium slope orientations [25]. This symmetry can be easily added to the phase-field model by adding a term to the free energy functional

$$F_{\text{sq}}[h] = F_{\text{iso}}[h] + \int d^2x (\partial_x h)^2 (\partial_y h)^2$$

(27)

where $F_{\text{iso}}[h]$ is the free energy of Eq. (6), and $\partial_x = \partial/\partial x$. The additional term is non-negative and vanishes for slopes oriented with the cartesian axes. We choose a prefactor of unity for this term since this results in an isotropic potential to quadratic order about any of the four equilibrium points.

Taking $\partial h/\partial t = -\delta F_{\text{sq}}/\delta h$ then gives the equation of motion

$$\frac{\partial h}{\partial t} = -\nabla^4 h - \partial_x \left\{ [1 - |\nabla h|^2 - 2(\partial_y h)^2] \partial_x h \right\}$$

$$- \partial_y \left\{ [1 - |\nabla h|^2 - 2(\partial_x h)^2] \partial_y h \right\}.$$  

(28)

We parametrize our first order accurate time step as before, with

$$h_{t+\Delta t} = h_t + \Delta t \left( \frac{\partial h}{\partial t} \right)_t + b_1 \Delta t \nabla^2 (h_{t+\Delta t} - h_t)$$

$$+ b_2 \Delta t \nabla^4 (h_{t+\Delta t} - h_t).$$

(29)

UvN stability analysis about an equilibrium slope, $h = x + \eta$, takes the same form Eq. (13), with $\mathcal{L}(\mathbf{k})$ unchanged and

$$\mathcal{R}(\mathbf{k}) = (2 + b_1)\lambda(\mathbf{k}) + (b_2 - 1)\lambda(\mathbf{k})^2.$$  

(30)

The crucial condition $\mathcal{L} + \mathcal{R} > 0$ then results in the stability region

$$b_1 < -1, \quad b_2 > 1/2,$$

(31)

with no distinction between single and multiple domain systems. We conducted numerical tests of gradient stability following the same protocol shown above.
Figure 3: (color online) Plotted is the laplacian of $h(x, y, t)$, for a system evolved with a growing time step $\Delta t \sim t^{2/3}$. Simulation details are provided in the text. Positive values (troughs) are red, negative values (peaks) are blue, and the white regions are domains of uniform slope with zero laplacian.
Figure 4: Stability diagram for the square symmetry model of Sec. 5. Squares represent \((b_1, b_2)\) parameter values which were gradient stable in our numerical tests, whereas the \(\times\) were found to be unstable. The shaded region represents UvN stable parameter values.

in Sec. 4 and again find good agreement, as shown in Fig. 4. The details of our numerical spatial derivatives are provided in the appendix.

Experiments [25] and simulations [26, 6] find \(L \sim t^{1/4}\) growth for crystal growth with square symmetry (although variants of this square symmetry model can result in \(t^{1/3}\) growth [27]). We measured the length scale via the free energy density following the same procedure as described in Sec. 4, and the results are presented in Fig. 2. For the time range simulated, we observe slightly slower than \(t^{1/4}\) growth, with an exponent around 0.22. Finally, we show in Fig. 5 snapshots of typical domain configurations from a single run on a \(512 \times 512\) lattice.

As this section demonstrates, it is straightforward to generalize the analysis of the isotropic model to the case with a discrete set of preferred slope orientations. In particular, the analysis for models with six-fold symmetry [6] and three-fold symmetry [28] should follow analogously.

6. Summary

We have parametrized a first order accurate discrete time step, Eq. (2) for MBE growth with slope selection that, unlike the Euler step, is gradient stable for appropriate choices of the parameters \(b_1\) and \(b_2\). We determined the
Figure 5: (color online) Plotted is the laplacian of $h(x, y, t)$ as described in Fig. but here for the square symmetry model.
stability range for these parameters via unconditional von Neumann stability
analysis, and then tested these predictions with numerical tests for gradient
stability, as shown in Fig. 1. We find that the UvN stability analysis serves as
an accurate proxy for unconditional gradient stability, similar to the behavior
of the Cahn-Hilliard equation [11].

Our stability analysis contained an implicit assumption that the interface
slopes do not exceed unit magnitude, which we justify by noting that the
dynamics naturally select for this slope. This came into our UvN analysis by
our choice to linearize about a unit slope domain. We note that the numerical
tests for gradient stability contained no such assumption, so the agreement
between the two approaches confirms validity of the unit slope assumption.

The increase in efficiency due to a gradient stable method is substantial.
For the simulations presented in Fig. 2, computation by Euler step, for which
the largest stable step size is $\Delta t = 0.03$, would require $3.3 \times 10^8$ time steps.
In contrast, using a stable method with step size $\Delta t = \max(\Delta t_0, At^{2/3})$ the
number of time steps required to reach some $t_{\max}$ is given by $3t_{\max}^{1/3}/A$, which
for our simulations is $6.5 \times 10^4$ steps. Each stable step involves an overhead
factor of 2.4 due to the addition of the FFT, but the net result is an overall
increase of efficiency by a factor of 2100 for the data we present! Note that
this factor will increase as computational resources allow for larger systems
to be evolved to later times.

The method of parametrizing linear semi-implicit steps, performing un-
conditional von Neumann stability analysis, and then testing the predictions
numerically for gradient stability has yielded efficient stable methods for the
Cahn-Hilliard and Allen-Cahn equations [11] and now for a class of MBE
crystal growth models. We anticipate that this procedure will prove useful
to many other phase field models.

Acknowledgments

G.M.S. was supported by NSF REU Grant PHY-1156964. B.P.V.-L. ac-
knowledges financial support from the Max Planck Institute for Dynamics
and Self-Organization and the hospitality of the University of Göttingen,
where this work was completed.
Appendix A. Finite Difference Scheme

Here we present details of the spatial discretization scheme we use in our numerical tests. We present these in a discrete-space, continuous time picture, as our goal is to ensure that the conservative dynamics and the gradient flow are exact, i.e. preserved to all orders in \( \Delta x \). The essential condition for gradient flow is that the equation of motion must be connected to a particular choice for the free energy functional such that

\[
\frac{\partial h_{i,j}}{\partial t} = -\frac{\partial}{\partial h_{i,j}} \left( \frac{F}{\Delta x^2} \right). \tag{A.1}
\]

Local conservation is imposed by ensuring that the equation of motion has the form

\[
\frac{dh_{i,j}}{dt} = -\frac{1}{\Delta x} \left[ \{J_x\}_{i+1/2,j} - \{J_x\}_{i-1/2,j} \\
- \{J_y\}_{i,j+1/2} - \{J_y\}_{i,j-1/2} \right] \tag{A.2}
\]

so that the same \( \{J_x\}_{i+1/2,j} \) flows into \( h_{i+1,j} \) and out of \( h_{i,j} \), and the same \( \{J_y\}_{i,j+1/2} \) flows into \( h_{i,j+1} \) and out of \( h_{i,j} \).

Our implementation uses an on-site finite-difference expression for \( \nabla^2 h \), for which we take the standard five-point stencil,

\[
\{\nabla^2 h\}_{i,j} = \frac{1}{\Delta x^2} [ h_{i+1,j} + h_{i-1,j} + h_{i,j+1} + h_{i,j-1} - 4h_{i,j} ], \tag{A.3}
\]

and the cell-centered expression for \( |\nabla h|^2 \),

\[
\{|\nabla h|^2\}_{i+1/2,j+1/2} = \\
\frac{1}{2\Delta x^2} \left[ (h_{i+1,j} - h_{i,j})^2 + (h_{i+1,j+1} - h_{i,j+1})^2 \\
+ (h_{i,j+1} - h_{i,j})^2 + (h_{i+1,j+1} - h_{i+1,j})^2 \right]. \tag{A.4}
\]

With these choices it is straightforward to show that

\[
\frac{\partial}{\partial h_{k,l}} \sum_{i,j} \{|\nabla h|^2\}_{i+1/2,j+1/2} = -2\{|\nabla^2 h\|_{k,l}. \tag{A.5}
\]
Our equation of motion is given by Eq. (A.1) with the choice
\[
\frac{F}{\Delta x^2} = \sum_{i,j} \left[ \frac{1}{2} (\nabla^2 h)_{i,j}^2 + \frac{1}{4} \left( 1 - \{|\nabla h|^2\}_{i+1/2,j+1/2} \right)^2 \right].
\] (A.6)

By making use of Eq. (A.5), the equation of motion can be shown to satisfy the discrete continuity equation (A.2) with current
\[
\{J_x\}_{i+1/2,j} = \{J_{x}^{SD}\}_{i+1/2,j} + \{J_{x}^{NE}\}_{i+1/2,j}
\] (A.7)
where the surface diffusion current is
\[
\{J_{x}^{SD}\}_{i+1/2,j} = \frac{\nabla^2 h}_{i+1/2,j} - \frac{\nabla^2 h}_{i,j} \Delta x
\] (A.8)
and the nonequilibrium current is
\[
\{J_{x}^{NE}\}_{i+1/2,j} = \frac{h_{i+1,j} - h_{i,j}}{\Delta x}
\times \left[ 1 - \frac{1}{2} \left( \{|\nabla h|^2\}_{i+1/2,j+1/2} + \{|\nabla h|^2\}_{i+1/2,j-1/2} \right) \right],
\] (A.9)
and analogous expressions for \{J_y\}_{i,j+1/2}. The discrete form of the free energy, Eq. (A.6), was used the numerical tests for gradient stability.

For the square symmetry model, we need additionally the cell-centered derivatives
\[
\{(\partial_x h)^2\}_{i+1/2,j+1/2} = \frac{1}{2\Delta x^2} \left[ (h_{i+1,j} - h_{i,j})^2 + (h_{i+1,j+1} - h_{i,j+1})^2 \right]
\] (A.10)
\[
\{(\partial_y h)^2\}_{i+1/2,j+1/2} = \frac{1}{2\Delta x^2} \left[ (h_{i,j+1} - h_{i,j})^2 + (h_{i+1,j+1} - h_{i+1,j})^2 \right].
\]
The free energy is given by Eq. (A.6) with the additional term
\[
\frac{F_{sq}}{\Delta x^2} = \frac{F}{\Delta x^2} + \sum_{i,j} \{(\partial_x h)^2\}_{i+1/2,j+1/2} \{(\partial_y h)^2\}_{i+1/2,j+1/2}
\] (A.11)
which corresponds to the nonequilibrium currents

\[
\{J_{x,\text{NE},sq}\}_{i+1/2,j} = \{J_{x,\text{NE}}\}_{i+1/2,j} - \frac{h_{i+1,j} - h_{i,j}}{\Delta x} \times \left(\{\left(\partial_y h\right)^2\}_{i+1/2,j+1/2} + \{\left(\partial_y h\right)^2\}_{i+1/2,j-1/2}\right),
\]

(A.12)

and

\[
\{J_{y,\text{NE},sq}\}_{i,j+1/2} = \{J_{y,\text{NE}}\}_{i,j+1/2} - \frac{h_{i,j+1} - h_{i,j}}{\Delta x} \times \left(\{\left(\partial_x h\right)^2\}_{i+1/2,j+1/2} + \{\left(\partial_x h\right)^2\}_{i-1/2,j+1/2}\right).
\]

(A.13)

References

[1] G. Ehrlich, F. G. Hudda, Atomic view of surface self-diffusion: Tungsten on tungsten, J. Chem. Phys. 44 (3) (1966) 1039. doi:10.1063/1.1726787.

[2] R. L. Schwoebel, E. J. Shipsey, Step motion on crystal surfaces, J. Appl. Phys. 37 (10) (1966) 3682. doi:10.1063/1.1707904.

[3] J. Villain, Continuum models of crystal growth from atomic beams with and without desorption, J. Phys. I France 1 (1) (1991) 19. doi:10.1051/jp1:1991114.

[4] C. Misbah, O. Pierre-Louis, Y. Saito, Crystal surfaces in and out of equilibrium: A modern view, Rev. Mod. Phys. 82 (2010) 981. doi:10.1103/RevModPhys.82.981.

[5] M. Ortiz, E. Repetto, H. Si, A continuum model of kinetic roughening and coarsening in thin films, J. Mech. Phys. Solids 47 (4) (1999) 697 – 730. doi:10.1016/S0022-5096(98)00102-1.

[6] D. Moldovan, L. Golubovic, Interfacial coarsening dynamics in epitaxial growth with slope selection, Phys. Rev. E 61 (2000) 6190. doi:10.1103/PhysRevE.61.6190.

[7] S. Biagi, C. Misbah, P. Politi, Coarsening scenarios in unstable crystal growth, Phys. Rev. Lett. 109 (2012) 096101. doi:10.1103/PhysRevLett.109.096101.
[8] C. Wang, X. Wang, S. M. Wise, Unconditionally stable schemes for equations of thin film epitaxy, Discret. Contin. Dyn. S. 28 (2010) 405. doi:10.3934/dcds.2010.28.405

[9] R. V. Kohn, X. Yan, Upper bound on the coarsening rate for an epitaxial growth model, Comm. Pure App. Math. 56 (11) (2003) 1549. doi:10.1002/cpa.10103

[10] A. J. Bray, Theory of phase ordering kinetics, Adv. Phys. 43 (1994) 357. doi:10.1080/00018739400101505

[11] B. P. Vollmayr-Lee, A. D. Rutenberg, Fast and accurate coarsening simulation with an unconditionally stable time step, Phys. Rev. E 68 (2003) 066703. doi:10.1103/PhysRevE.68.066703

[12] M. Cheng, A. D. Rutenberg, Maximally fast coarsening algorithms, Phys. Rev. E 72 (2005) 055701. doi:10.1103/PhysRevE.72.055701

[13] D. J. Eyre, Unconditionally gradient stable time marching the cahn-hilliard equation, MRS Proceedings 529 (1998) 39. doi:10.1557/PROC-529-39

[14] J. Shen, C. Wang, X. Wang, S. M. Wise, Second-order convex splitting schemes for gradient flows with ehrlichshwoebel type energy: Application to thin film epitaxy, SIAM J. Numer. Anal. 50 (2012) 105. doi:10.1137/110822839

[15] Z. Qiao, Z.-z. Sun, Z. Zhang, The stability and convergence of two linearized finite difference schemes for the nonlinear epitaxial growth model, Numer. Methods for Partial Differential Eq. 28 (6) (2012) 1893. doi:10.1002/num.20707

[16] W. Chen, Y. Wang, A mixed finite element method for thin film epitaxy, Numer. Math. 122 (2012) 771. doi:10.1007/s00211-012-0473-9

[17] W. Chen, C. Wang, X. Wang, S. M. Wise (preprint).

[18] C. Xu, T. Tang, Stability analysis of large timestepping methods for epitaxial growth models, SIAM J. Numer. Anal. 44 (2006) 1759. doi:10.1137/050628143
[19] B. P. Vollmayr-Lee (in preparation).

[20] W. W. Mullins, Flattening of a nearly plane solid surface due to capillarity, J. Appl. Phys. 30 (1) (1959) 77. doi:10.1063/1.1734979

[21] M. D. Johnson, C. Orme, A. W. Hunt, D. Graff, J. Sudijono, L. M. Sander, B. G. Orr, Stable and unstable growth in molecular beam epitaxy, Phys. Rev. Lett. 72 (1994) 116. doi:10.1103/PhysRevLett.72.116

[22] M. Siegert, M. Plischke, Slope selection and coarsening in molecular beam epitaxy, Phys. Rev. Lett. 73 (1994) 1517. doi:10.1103/PhysRevLett.73.1517

[23] M. Rost, J. Krug, Coarsening of surface structures in unstable epitaxial growth, Phys. Rev. E 55 (1997) 3952. doi:10.1103/PhysRevE.55.3952

[24] T. M. Rogers, K. R. Elder, R. C. Desai, Numerical study of the late stages of spinodal decomposition, Phys. Rev. B 37 (1988) 9638. doi:10.1103/PhysRevB.37.9638

[25] J.-K. Zuo, J. F. Wendelken, Evolution of mound morphology in reversible homoepitaxy on cu(100), Phys. Rev. Lett. 78 (1997) 2791. doi:10.1103/PhysRevLett.78.2791

[26] M. Siegert, Coarsening dynamics of crystalline thin films, Phys. Rev. Lett. 81 (1998) 5481. doi:10.1103/PhysRevLett.81.5481

[27] A. Levandovsky, L. Golubovic, Epitaxial growth and erosion on (001) crystal surfaces: Far-from-equilibrium transitions, intermediary states, and vertical asymmetry, Phys. Rev. B 69 (2004) 241402. doi:10.1103/PhysRevB.69.241402

[28] S. J. Watson, S. A. Norris, Scaling theory and morphometrics for a coarsening multiscale surface, via a principle of maximal dissipation, Phys. Rev. Lett. 96 (2006) 176103. doi:10.1103/PhysRevLett.96.176103