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Geometrical Approach to Moving-Interface Dynamics

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A general class of models is introduced which relate the motion of a phase boundary to properties of the local interfacial geometry. These systems can undergo successive destabilizations as they grow, possibly giving rise to nonequilibrium spatial patterns. This formalism has applications to a wide variety of physical problems, especially including dendritic solidification.

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Many problems of physical interest involve the question of how a nonlinear system dynamically evolves in time. This is a particularly fascinating issue whenever the initial state is prepared in a configuration which has domains that are not absolute minima of the free energy. In this case, the interface motion is necessarily a nonequilibrium problem which has the possibility of giving rise to either chaotic final states or to intricate patterns. A characteristic feature of these problems is the competition between long-wavelength instabilities and a stabilization mechanism acting at the shortest scales.

A prototypical situation is the formation of dendritic ice crystals from a supercooled melt. The equations which govern the growth of the solid phase are the heat-diffusion equation for which the interface motion, through emission of latent heat, acts as a source, and the thermodynamic boundary condition that surface tension reduces the interfacial temperature by an amount proportional to the curvature. This problem has been studied extensively by Langer and co-workers[1,2] over the past few years. There exists evidence for the operation of a pattern-selection mechanism wherein the growing dendrite undergoes a series of successive destabilizations and subsequent restabilizations by the emission of "side branches." The associated hypothesis that this sort of nonlinear limit cycle causes the system to operate at the marginal stability point seems consistent with experimental results.[2] However, the detailed workings of this mechanism as well as side-branch spacings and global symmetries are not understood. Some other systems that exhibit similar competition between stabilizing and destabilizing forces are directional solidification,[3] fluid flow in a porous material,[4] and biological growth.[5]

The purpose of this work is to introduce a sim-

ple class of models in which we believe some of the essential features of the moving-interface problem may be isolated and explored. The key idea is to reduce the complicated dynamics of the entire system which is responsible for producing the domain structure to an equation for the interface motion which is a function only of the local geometry of the interface itself. In what follows, we will show that this simplification to a geometrical surface dynamics still preserves a rich and varied phenomenology. In particular, we examine a subset of models whose behavior bears a striking resemblance to several features of the full solidification problem.

Specifically, we assume equations of the general form

$$\hat{n}\cdot\partial\vec{x}/\partial t=\mathcal{F}(\kappa,\nabla\kappa,\ldots), \tag{1}$$

where $\vec{x}$ is the interface position, κ is the local curvature, $\hat{n}$ is the outward unit normal, and ∇ is the covariant derivative on the surface. $\mathcal{F}$ is a general function of κ and its covariant derivatives. For simplicity, we will restrict the discussion here to the two-dimensional problem where the phase boundary is just a curve. Then ∇ is just $\partial/\partial s$, where s is the arclength. An attractive choice for $\mathcal{F}$ is

$$\mathcal{F}=F(\kappa)+\gamma\,\partial^2\kappa/\partial s^2, \tag{2}$$

where the second term stabilizes the system at short wavelengths for $\gamma>0$.

Equation (1) has the important property of reparametrization invariance. This means that the curve $\vec{x}$ can be arbitrarily parametrized without affecting the physical content of the time evolution. In fact, the phase boundary is a purely geometrical, stringlike[6] entity, totally independent of any coordinate grid used to describe it. One consequence of this is that the tangential component of $\partial\vec{x}/\partial t$ is not determined by the physics (as contained in $\mathcal{F}$), but instead may be chosen for convenience. An often useful choice is the orthogonal gauge,[6] $\partial\vec{x}(t,\sigma)/\partial t=\hat{n}\mathcal{F}$, where σ parametrizes the curve.

It is useful to rewrite the time-evolution equation (1) in terms of the curvature and the arclength. The arclength is related to the parametrization σ by $ds=g^{1/2}d\sigma$, where the metric $g=\vec{x}'\cdot\vec{x}'$, and the prime means derivative with respect to σ. Defining the curvature by $\kappa=\hat{n}\cdot\partial^2\vec{x}/\partial s^2$, one can easily derive

$$\dot{\kappa}=-(\partial^2/\partial s^2+\kappa^2)\mathcal{F}(\kappa),\quad \dot{g}=2g\kappa\mathcal{F}. \tag{3}$$

A full discussion of these equations as well as the generalization to three dimensions will be presented elsewhere.[7]

In order to motivate a choice for F in (2), we turn to the physics of the solidification problem. It is easy to argue that the growth rate will be enhanced by increasing the curvature of the surface. Schematically, a local outward bulge will allow latent-heat diffusion to occur more rapidly than a more planar interface. Moreover, an absolutely planar interface cannot move at all, other than for certain special values of the undercooling. Thus, F must vanish at the origin. The undercooling leads to an asymmetry between positive and negative curvature, suggesting a positive term quadratic in κ. Finally, the presence of a minimum nucleation size constrains F to become negative at large κ. This leads us to suggest that solidification could be modeled by our geometrical surface dynamics with F of the form

$$F(\kappa)=\kappa+\alpha\kappa^2-\beta\kappa^3. \tag{4}$$

We view this resulting model as a suggestion in the spirit of a Landau-Ginzburg mean-field approach.

Our model allows for analytic treatment directly parallel to that applied to the solidification problem, though with significantly greater ease. As there, an exact solution of the above model is a circle whose radius obeys

$$\dot{R}_0(t)=F(1/R_0). \tag{5}$$

Because of our choice of F, large enough circles will always expand. Consider, then, the stability of such a solution. To leading order, we can set

$$R(\theta)=R_0+\textstyle\sum_m\delta_m(t)\cos(m\theta), \tag{6}$$

where the initial circle is parametrized by the polar angle θ. A standard analysis yields the instantaneous growth rate

$$\dot{\delta}_m=\frac{m^2-1}{R_0^2}\left[F'\left(\frac{1}{R_0}\right)-\frac{\gamma m^2}{R_0^2}\right]\delta_m. \tag{7}$$

With $\gamma=0$, stability depends on the sign of F'. This is our analog of the Mullins-Sekerka instability.[8] Nonzero γ acts as a short-distance cutoff on the fluctuation spectra, making the dominant instabilities occur at intermediate length scales. A comparison of this expression with the results of the full solidification problem[2] shows that γ is analogous to the surface tension.

The full solidification problem is known to have a family of exact solutions in the limit of zero surface tension.[8] These dendritelike structures translate at a constant velocity related to their

tip radius. Similarly, for $\gamma = 0$ our equations also possess such solutions. For the case $F = \kappa$, the profile of the solution is given by

$$x = \rho\theta, \quad y = \rho \ln(\cos\theta), \tag{8}$$

where the parameter θ is just the angle that the normal vector makes with the $\hat{y}$ axis. Near the tip, this curve looks parabolic with arbitrary radius ρ. The velocity of translation along the $\hat{y}$ axis is given by $1/\rho$. A stability analysis along the lines of Muller-Krumbhaar and Langer[9] leads to the eigenvalue equation

$$-\cos^2\theta(\delta'' + \delta') = \omega\delta \tag{9}$$

for the distortion $\delta(\theta)$ normal to the interface. This problem can be solved exactly by transforming it to a Schrödinger equation in the presence of a kink potential.[10] The eigenfunctions are given by $\delta \sim (\cos\theta)^{1/2} P_{1/2}{}^{ik}(\sin\theta)$, $\omega = \frac{1}{4} + k^2$, where P is an associated Legendre polynomial. All modes grow in time, representing the instability of the dendrite to tip splitting in the absence of a surface-tension term.

For nonzero γ, the situation is very different. Perturbation theory seems to predict a relatively insignificant steady-state shape correction. However, a direct integration of Eqs. (4) shows that perturbation theory breaks down at large distances from the tip.[11] This manifests itself in the presence of increasingly rapid oscillations in $\kappa(\theta)$ as θ approaches $\pm\frac{1}{2}\pi$. In the strict mathematical sense, there cannot exist uniformly translating solutions. This leads us to conjecture that a similar breakdown occurs in the solidification problem, contrary to earlier expectations.[12] Nonetheless, we believe it still may be useful, even for nonzero γ, to consider the linear mode analysis around the $\gamma = 0$ solution. It is straightforward to show that there is a critical value of γ/ρ^2 above which all modes become stable. This point of marginal stability may be relevant for the long-time behavior of dendrites in our model, in analogy with Langer's suggestions for the solidification problem.[9] Perhaps the lack of a steady-state solution provides a new mechanism to drive the system to the marginally stable point.

One of the most useful features of our approach is the ease of doing computer experiments. The full nonlocal diffusion-limited boundary motion is extremely difficult to treat numerically, and this has been carried out to only a limited extent.[13] Our models, on the other hand, immediately lend themselves to simulations. We have done this by integrating the curvature equations (3) and then reconstructing the curve. Our programs were checked by varying the number of points and also by comparing the results to direct integration of Eq. (1). We will discuss the detailed results of our simulation studies elsewhere,[6] but we wish to point out here that this class of models can indeed give rise to interesting structures, including dendritic growth patterns. In Fig. 1, for example, the initial pattern stabilizes to a circle as it grows, but is then destabilized by (computer) noise and emits irregular dendrites. Figure 2 exhibits tip splitting and in Fig. 3 the dendrite initiates a side-branch excitation. We emphasize that (a) these curves represent the *initial* stages of growth and that (b) we do not claim that *all* observed features of solidification are reproduced by our specific choices of $\mathfrak{F}$ and its parameters.

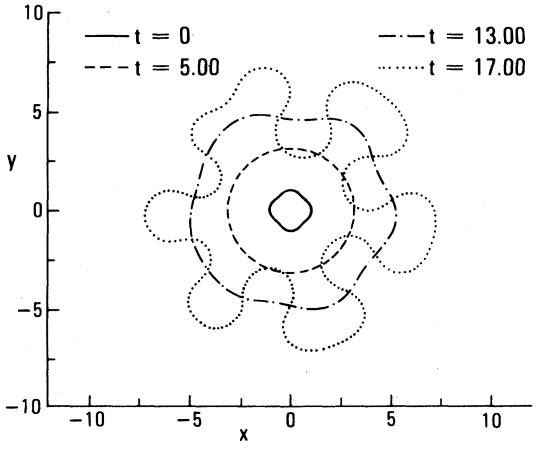

FIG. 1. Emergence of dendrites: solution of (4) with $\alpha = 0$, $\beta = \frac{1}{3}$, $m = 0.2$.

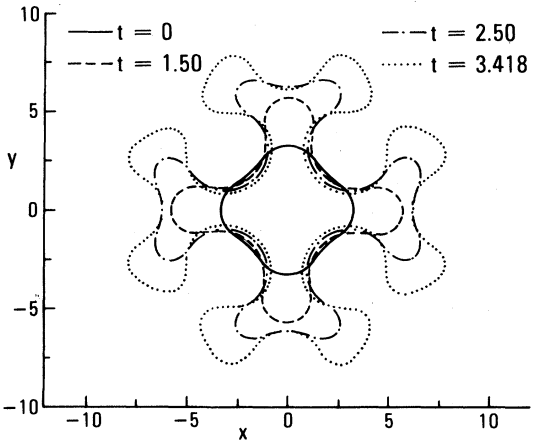

FIG. 2. Tip splitting; $\alpha = 1$, $\beta = 0.25$, $\gamma = 0.25$.

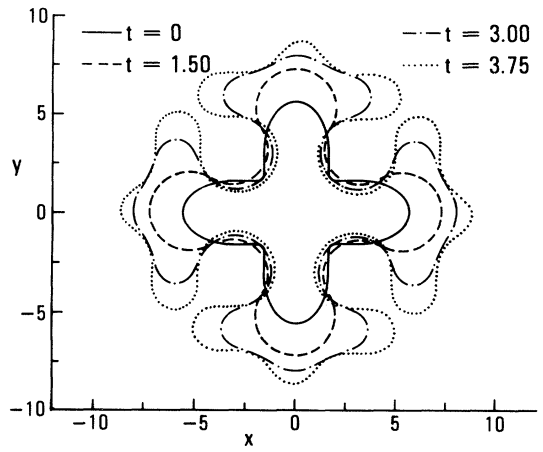

FIG. 3. Side branching; $\alpha = 1$, $\beta = 0.25$, $\gamma = 0.35$.

In fact, a crucial question in solidification is the importance of crystalline anisotropy, which may be required for repeated side branching.

The ice-crystal problem led us to consider $\mathcal{F}$'s of the form given in (2). If we turn instead to the oscillations of a bubble of one liquid immersed in a second, immiscible liquid (e.g., oil in water), we must incorporate the fact that the area of the enclosed liquid cannot change, at least insofar as the fluid is incompressible. This will indeed be the case if we choose $\mathcal{F}$ of the form $\partial V(\kappa)/\partial s + \gamma\, \partial^2\kappa/\partial s^2$, where V is some function of curvature. Likewise, a system whose phase boundary moves even for planar interfaces, such as a water flood displacing oil in a reservoir,[14] would necessitate a term in $\mathcal{F}$ which would be nonzero at $\kappa = 0$. Finally, one could attempt to formulate more realistic models by allowing at least some nonlocality in $\mathcal{F}$. Stable crystal growth has been studied in this manner,[15] and recently a model of this sort for dendritic growth has been independently proposed by Langer.[16]

In summary, our geometrical surface dynamics can be used as a starting point for investigating the question of nonequilibrium pattern formation in a variety of systems. We have demonstrated that for solidification, these models reproduce the Mullins-Sekerka instability and the tip-splitting behavior for the dendrite. We also conjecture that the lack of a steady-state solution for nonzero γ could lead to a new explanation for the marginal-stability–selection mechanism. Finally, some simple numerical experiments have convinced us that there is much to be understood here in the way of nonlinear phenomena. While this class of models cannot hope to reproduce all of the intricate details inherent in a full nonlocal approach (such as the lack of self-intersections), we are optimistic that a detailed study of these phenomena will provide much needed insight into the process of pattern formation.

We would like to thank Jim Langer for generously sharing with us his insight into the physics of these systems as well as some of his unpublished results on models which are quite similar (and in some cases identical) to the ones studied here.

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