Pattern Selection in Dendritic Solidification

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We show that the dynamically selected velocity and tip radius of dendrites in the boundary-layer model of solidification have the special values which permit the existence of steady-state needle-crystal solutions. This result, in conjunction with considerations of stability, provides new insight concerning the validity of the marginal stability hypothesis.

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Simplified models of dendritic growth recently have been introduced in an attempt to discover dynamical mechanisms for pattern selection.[1,2] The boundary-layer model[2] (BLM) is a two-dimensional model of dendritic solidification in which the characteristic decay length of the diffusion field is much smaller than the local radius of curvature of the interface. In this approximation, valid when the dimensionless undercooling Δ is close to unity, thermal diffusion occurs in a thin boundary layer at the interface; variations in the boundary-layer thickness account for diffusion perpendicular to the interface. The BLM has been studied analytically and numerically in Ref. 2. At a given undercooling Δ, crystalline symmetry m, and anisotropy strength α, a unique dendritelike structure is formed for a large class of initial conditions (after the decay of initial transients). The dendrite consists of a smooth, almost parabolic tip which extends for about five tip radii before forming side branches. The tip propagates without noticeable change of shape, while the side branches appear to be generated periodically and to grow out at fixed positions in the laboratory frame, in a manner consistent with experimental observation.

From its beginning, the BLM has produced interesting surprises. For example, the continuous family of steady-state shape-preserving needle crystals, which was assumed to exist on the basis of approximate analyses of the full solidification problem,[3] does not exist in the BLM except in the manifestly unstable Ivantsov[4] limit of vanishing surface tension. Rather, for any set of growth parameters Δ, α, etc., the BLM has only one or at most a discrete set of needle-crystal solutions, each associated with its own growth velocity v_0 and tip curvature κ_0. Dendrites, with their complex time-dependent side-branching structures, are far from being needle crystals; therefore it might seem unlikely that the dynamical selection mechanism is closely tied to the existence of special stationary solutions which remain needlelike infinitely far behind the tip. The principal result to be reported here is that the tip of the dynamically selected dendritic mode in the BLM turns out to have precisely the same v_0 and κ_0 as the needle crystal. We shall see that this fact is crucial to an understanding of the dynamics of dendritic pattern selection. In particular, it leads us to a tentative explanation for the success of the marginal stability hypothesis[3,5] in predicting experimental data.

The mathematical statement of the two-dimensional BLM consists of two coupled nonlinear partial differential equations for the curvature κ and the thermal field h, as functions of the displacement s along the solidification front and of the time t.[6] We define $\theta(s)$ to be the angle of orientation of the front, so that $\kappa = d\theta/ds$. Then the BLM equations are

$$\left.\frac{\partial h}{\partial t}\right|_s = v_n(1-\Delta w) - \kappa w^2 + \frac{1}{\Delta^2}\frac{\partial}{\partial s}\frac{h}{w}\frac{\partial w}{\partial s} - \frac{\partial h}{\partial s}\int_0^s \kappa v_n \, ds', \quad (1a)$$

$$\left.\frac{\partial \kappa}{\partial t}\right|_s = -\left(\kappa^2 + \frac{\partial^2}{\partial s^2}\right) v_n - \frac{\partial \kappa}{\partial s}\int_0^s \kappa v_n \, ds', \quad (1b)$$

where the normal velocity of the interface is $v_n = w^2/h$, the scaled interface temperature is $w = 1 - \Delta^2\kappa - \beta v_n$, and the anisotropy function is $\beta = \alpha\Delta^4(1-\cos m\theta)$. We require reflection symmetry about the tip at $\theta = 0$, so that the first derivatives of h, κ, w, and v_n vanish there. In the tail of the dendrite, we have also imposed the condition that these first derivatives vanish. In our numerical solutions of (1) we have used a sufficiently long interface that side branches from the tip do not yet propagate to the boundary in the tail during the computation.

The steady-state version of (1) can be written in the following form:

$$d\theta/ds = \Delta^{-2}(1 - w - \beta v_0 \cos\theta) = \kappa, \tag{2a}$$

$$dw/ds = \lambda, \tag{2b}$$

$$\frac{d\lambda}{ds} = 2\Delta^2 v_0 \sin\theta\lambda + \frac{\Delta^2 v_0 \kappa w}{\cos\theta} - \Delta^2 v_0^2 \cos^2\theta \frac{(1-\Delta w)}{w} - \frac{\lambda^2}{w} - \lambda\kappa \tan\theta. \tag{2c}$$

Here we have used the fact that (1b) admits a first integral which satisfies the boundary conditions, namely, that $v_n = v_0 \cos\theta$, where v_0 is the velocity of the tip. In order to analyze (2) in detail we consider the phase space of θ, w, and λ. The only important fixed points are at $\lambda = w = 0$, $\theta = \pm\pi/2$; the tip is not a fixed point. A steady-state needle-crystal solution of the BLM corresponds to a trajectory in the phase space which joins the two fixed points and passes through the line $\theta = \lambda = 0$. Only those trajectories which do intersect this line satisfy the boundary conditions that we have imposed at the tip; and there is no *a priori* reason for this intersection to occur for any arbitrary value of v_0.[7] There is only a single trajectory which enters or leaves the fixed points, just as we reported earlier[2] for the BLM in the limit of small Δ and $\alpha = 0$. This trajectory is asymptotically identical to the Ivantsov solution:

$$\kappa \approx v_0(1-\Delta)\cos^3\theta, \quad \theta \to \pm\pi/2. \tag{3}$$

In order to compute these needle-crystal solutions explicitly, we have integrated (2) backwards in s from $\theta = \pi/2$, using (3) to identify initial values of θ, w, and λ as close as is numerically feasible to that fixed point. We then have looked for values of v_0 for which the trajectory reaches $\theta = 0$ at some $w = w_0$, i.e., $\kappa = \kappa_0$, with $\lambda = 0$. For example, for the parameter values $\alpha = 0.1$, $m = 4$, and $\Delta = 0.75$, we have found two eigenvalues of v_0; one at $v_0 = v_0^* = 0.2712$ with $\kappa_0 = \kappa_0^* = 0.0915$ which has exactly the shape of the dynamically selected dendrite tip, and another at $v_0 = 0.0206$ with $\kappa_0 = 0.0207$ which is a slow, flat-tipped solution of no obvious physical significance. Both v_0^* and κ_0^* are within 0.1% of the values obtained by numerical solution of the fully time-dependent Eqs. (1).

To understand the significance of this result, note first that the needle crystal, which we shall denote by $\kappa^*(s)$, is a functional fixed point of the dynamical system (1). We shall assume that the fully time-dependent solution of (1) is approaching a functional limit cycle, to be denoted by $\kappa^*(s,t)$. The shapes of both theoretical and experimental dendrites seem to repeat themselves with a well defined period when observed in a frame of reference moving with the tip. We cannot prove that our computed $\kappa(s,t)$ in the BLM will not diverge or otherwise lose periodicity at some large but finite time, nor can we discount the possibility that late-stage coarsening of side branches occurs (in real dendrites or the BLM) *via* a weak instability of the motion. But the limit cycle appears to be a good working hypothesis for the moment.

Our numerical results tell us that the limit cycle $\kappa^*(s,t)$ coexists in the dynamical function space with the fixed point $\kappa^*(s)$ and, moreover, that $\kappa^*(s,t)$ and $\kappa^*(s)$ are close to each other in the sense that they are numerically indistinguishable near the tip of the dendrite. We suggest that the simplest way to view this situation is as if $\kappa^*(s,t)$ has emerged through a single Hopf bifurcation from $\kappa^*(s)$. That is, $\kappa^*(s,t)$ and $\kappa^*(s)$ both lie on an invariant two-dimensional manifold with $\kappa^*(s)$ unstable only against perturbations in that manifold. Alternatives to this picture seem improbable. If $\kappa^*(s)$ were completely stable, it would be likely to appear as an attractor in the dynamical simulations; but we do not seem to see needle crystals without side branches. If $\kappa^*(s)$ were unstable against more than one complex-conjugate pair of deformation modes, then points initially near $\kappa^*(s)$ would not flow reliably toward a unique limit cycle. In neither case could we understand how the properties of $\kappa^*(s,t)$ are accurately determined by the needle crystal $\kappa^*(s)$. Note that, if $\kappa^*(s)$ must have exactly one conjugate pair of unstable modes, then we already have arrived at a weak form of a marginal stability principle. If the modes are part of a continuous spectrum, then $\kappa^*(s)$ must be marginally unstable; that is, dynamic trajectories must diverge from $\kappa^*(s)$ algebraically rather than exponentially. Alternatively, the modes can belong to a discrete part of the spectrum. Both of the latter possibilities imply that the selected state of dendritic growth is one in which the tip is characterized by a specially weak instability. The first possibility is consistent with the sharp statement that the needle crystal associated with dendritic motion is marginally unstable. We conjecture that this is the case.

As a first step towards testing the above-pictured crystal, we have linearized (1) about the stationary solution $\kappa^*(s)$ and have studied the eigenvalue spectrum of the resulting operator both analytically and numerically. Details of these investigations will be presented elsewhere, but the crucial result can be understood from qualitative considerations. The needle crystal has the special property that the thermal field h diverges like $s^{1/2}$ as $\theta \rightarrow \pm\pi/2$, $s \rightarrow \pm\infty$. In the BLM, this means that all deformation modes which propagate down the dendrite (as observed in the moving frame of the tip) can neither grow nor decay in a linear theory. [For example, in the planar stability spectrum reported in Eq. (4.6) of Ref. 2, ω vanishes for all q in the limit of large h and vanishing normal velocity.] It turns out that the extended states that constitute the continuous part of the dendritic deformation spectrum have the asymptotic form $\exp[iqs+\omega(q)t]$ as $s \rightarrow \pm\infty$ with $\omega(q) = -iqv_0^*$. Thus, all of these modes become stationary in the laboratory frame when observed at positions far from the tip. A similar phenomenon was observed by Müller-Krumbhaar and Langer[8] in their numerical studies of linear stability in the full diffusion problem; the spectrum seemed to flatten out at $\mathrm{Re}\omega \simeq 0$ near $q \simeq 0$. The slowing of *all* modes rather than just those of long wavelength as $h \rightarrow \infty$ is an exaggerated feature of the BLM. Our conclusion is that, in both the BLM and the full diffusion problem, any exact needle-crystal solution can be no more stable than marginally stable, and the mode (or modes) with $\mathrm{Re}\omega = 0$ must lie in a continuous part of the stability spectrum.

The remaining portion of the spectrum must consist of modes which are localized near the tip of the dendrite. Our numerical simulations of the dynamical system (1) plus preliminary results of a numerical stability analysis lead us to believe that these modes either merge with the continuum at $\mathrm{Re}\omega = 0$ or become stable with $\mathrm{Re}\omega < 0$ for sufficiently large anisotropy strength α. Because of the intrinsic marginal stability of the needle crystal, we conjecture that dendritic behavior occurs in both the BLM and the full diffusion problem throughout some nonzero range of values of α.

It is useful to compare the properties of the BLM with those of the related geometric model (GM) introduced by Brower *et al.*[1] and analyzed most recently by Kessler, Koplik, and Levine.[9] Pattern selection in the GM also turns out to be tied to the existence of special needle-crystal solutions but, because there is no analog of the thermal field, these solutions become completely stable at sufficiently strong anisotropy. Persistent side branching seems to occur only at a critical anisotropy where the needle crystal is marginally unstable; for larger anisotropy the side-branching modes are transients. It is possible that a similar stabilization occurs in the BLM, but our numerical evidence plus the special stability features of the BLM make this seem unlikely. As applied to solidification problems, the GM can now be seen to correspond to the limit of interface control, whereas the BLM is primarily a model of diffusion control. Apparently, dendritic behavior is generic only in the latter case.

We conclude with some remarks concerning the status of the marginal stability hypothesis. The way in which this hypothesis has been used in the analysis of experimental situations[3,5,10] seems to be correct but not optimal. Note that the statement of a marginal stability criterion suggested by our new results is different from previous statements in an important way: The stability requirement pertains to the needle-crystal fixed point and not to the dendritic limit cycle. This is a weakening of the hypothesis in the sense that it no longer says anything about dynamical behavior not associated with a fixed point. In another sense, however, the statement is surprisingly strong because marginal stability is simultaneously a criterion for the existence of a dendritic limit cycle and a property of the needle crystal. In practical terms, this means that if one has a family of *approximate* needle-crystal solutions, then it is reasonable to select one member of this family by testing for marginal stability, which is exactly what has been done previously. On the other hand, the optimal procedure would be to find *exact* needle crystals and use stability as a consistency requirement.

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[6]s is in units of d_0/Δ^3; t is in units of $d_0^2/D\Delta^8$. d_0 is the capillary length and D is the thermal diffusion coefficient.

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