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Instability of needle crystals in anisotropic dendritic growth

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We investigate the linear stability of steady-state needle crystals in dendritic growth in the presence of anisotropies in both surface tension and interfacial kinetics. The needle crystals are linearly unstable for certain ranges of values of the surface tension and kinetic coefficients. This instability results in complex tip-splitting and sidebranching events that lead to morphological transitions.

In this paper, we consider dendritic growth[1,2] with anisotropy in *both* surface tension *and* interfacial kinetics, using the boundary-layer model (BLM) of dendritic solidification.[3] The BLM is a model of solidification at large undercoolings, where diffusion may be thought of as occurring in a thin boundary layer around the solidification front. The BLM ceases to be a good approximation when the solidification front becomes sufficiently complex that diffusion no longer occurs predominantly along the interface. Nevertheless, for the steady-state and small-amplitude behavior, the BLM has proved to be a reliable guide to the behavior of the standard, fully nonlocal model for solidification. Previously, dendritic growth has been investigated when only a single direction of growth is favored by the crystalline anisotropy.[4-6]

The situation of interest in this paper is when surface tension anisotropy and anisotropy in interfacial kinetics favor *different* directions of growth. This possibility can arise because the kinetic coefficient in physical units is proportional to the ratio of the surface free energy σ to the width W of the solidification front.[7] In physical units, the temperature at the solidification front, T_s is given by

$$T_s = T_m - \frac{L}{C}[\sigma(\theta)+\sigma''(\theta)]k - \frac{B\sigma(\theta)}{W(\theta)}v_n , \qquad (1)$$

where T_m is the melting temperature, L is the latent heat of melting, C is the heat capacity of the melt, k is the curvature of the front, v_n is its normal velocity along the direction θ with respect to a crystallographic axis, and B is a constant independent of θ. As written this expression is valid in two dimensions. For a system with fourfold symmetry,

$$\sigma(\theta) = \sigma_0(1-\epsilon\cos 4\theta) \qquad (2)$$

and

$$\sigma+\sigma'' = \sigma_0(1+15\epsilon\cos 4\theta) . \qquad (3)$$

The direction of growth is determined by the minima of the coefficients of k and v_n in Eq. (1). The variation of W with θ is model and system dependent, but if the ratio $\sigma(\theta)/W(\theta)$ has minima at the same locations as those of $\sigma(\theta)$, then the surface tension term $(\sigma+\sigma'')$ and the kinetic term (σ/W) in Eq. (1) will favor growth directions separated by $\pi/4$ radians. Empirical evidence that this can occur is provided by viscous fingering experiments in anisotropic Hele-Shaw cells,[8,9] where a transition from growth along the direction of minimum surface tension to growth along the direction of maximum surface tension is observed as the driving force (pressure) is increased. This morphological transition has also been observed in crystal-growth experiments[10] and in time-dependent simulations of crystal growth using a cell-dynamical scheme approach.[11]

These observations may be interpreted as arising from the presence of two types of steady states: surface-tension needle crystals (SN's) growing in the direction of minimum surface tension, and kinetic needle crystals (KN's) growing in the direction of maximum surface tension. At low undercooling only SN's exist; at high undercooling both SN's and KN's can exist. It has been proposed[8] that the morphology observed at a given driving force corresponds to the fastest-growing needle crystal at that driving force.

The purpose of this paper is to perform a linear stability analysis of the surface-tension and kinetic needle crystals. We establish that at large undercooling, only kinetic needle crystals are linearly stable, and that for an intermediate range of undercooling, both types of steady states become linearly unstable to tip-splitting and sidebranching modes. Previously,[6] it was shown that needle crystals are stable in the presence of either kinetic anisotropy or surface-tension anisotropy *alone*. Thus our new results show that the morphology selection and the presence of the dense-branching structure at intermediate undercooling may be understood within the framework of linear stability analysis.

We consider the boundary-layer model of solidification in scaled, dimensionless units.[3] The dimensionless temperature w at the interface satisfies the modified Gibbs-Thomson condition:

$$w = 1-\Delta^2\alpha(\theta)k - \Delta^4\beta(\theta)v_n , \qquad (4)$$

where $0<\Delta<1$ is the dimensionless undercooling. A detailed description of the model and the equation of motion may be found in Ref. 6.

Surface tension and kinetic anisotropy are accounted for by the angular dependences of $\alpha(\theta)$ and $\beta(\theta)$:

$$\alpha(\theta) = 1-\epsilon_s\cos 4\theta , \qquad (5)$$

$$\beta(\theta) = 1-\epsilon_k\cos 4(\theta-\theta_d) , \qquad (6)$$

where $\epsilon_s, \epsilon_k \geq 0$ are anisotropy strengths and $\theta_d = \pi/4$ is the offset angle between the minima of α and β. With the definitions above, SN's grow along the $\theta = 0$ direction, while KN's grow along the diagonal $\theta = \pi/4$ direction. We search for the needle-crystal solutions by numerically integrating steady-state equations of motion of the BLM.[6] Physical boundary conditions impose a solvability condition[4] that can be satisfied only for certain growth velocities v_0. We were unable to find a steady-state solution for any choice of $\theta_d \neq \pi/4$.

Figure 1 shows the selected steady-state velocity v_0 as a function of undercooling Δ for fixed anisotropy strength $\epsilon_s = \epsilon_k = 0.40$.[12] It is found that while SN's exist for arbitrary Δ, KN's only exist above a critical undercooling $\Delta_c = 0.7351(2)$. (The parentheses denote the uncertainty in the last digit quoted.) Below Δ_c, the mismatch function in the solvability condition fails to vanish. Across Δ_c, there is an abrupt jump in the growth velocity. Above this critical undercooling, steady states corresponding to both SN's and KN's are possible, with different velocities. At $\Delta_0 = 0.7359(2)$, the growth velocities of the two kinds of needle crystals coincide. These features of steady-state velocity selection have been reported in earlier studies of the BLM,[8] and later in the symmetric model.[13] Motivated by experiments in Hele-Shaw cells, Ben-Jacob *et al.* hypothesized[8] that well above Δ_0, only the morphology with largest velocity (here kinetic dendrites) will be observable in real experiments. They also pointed out the possibility of a region of tip-splitting and sidebranching growth near Δ_0 due to the competition between the two anisotropies.

We shall now show, via explicit stability analysis, that the presence of a tip-splitting region is due to the loss of linear stability of the steady-state needle crystals. We consider the evolution of infinitesimal perturbations of the form $\delta k(s,t) \sim \exp(\lambda t)\delta k(s)$ around the steady states. Linearizing equations of motion around the steady-state solutions gives a linear stability operator which can be discretized into matrix form and then numerically diagonalized. The details of our method are described in Ref. 6. The eigenvalue spectrum $\{\lambda\}$ determines the linear stability of the needle crystal under examination: the presence of any $\mathrm{Re}\lambda > 0$ implies instability in the comoving frame of the needle crystal. The numerical accuracy of our solution is checked by the convergence of a zero mode due to the reparameterization symmetry.[14]

First we examine the linear stability of surface-tension needle crystals. It is found that they are linearly stable for all $\Delta < \Delta_s = 0.6750(2)$. As Δ is increased above Δ_s, a complex conjugate pair of discrete eigenvalues moves across the imaginary axis and acquires a positive real part, making the needle crystal linearly unstable. Figure 2(a) is the eigenvalue spectrum of a SN at $\Delta = 0.7250$, $\epsilon_s = \epsilon_k = 0.4$, for symmetric perturbations. Apart from the two unstable discrete eigenvalues at $\lambda = (0.527, \pm 2.20)$, the spectrum also has a stable continuum of eigenvalues and a well-converged zero mode at -2.63×10^{-8}. Eigenvalues are measured in units of v_0/ρ_0, where v_0 is the needle-crystal velocity and ρ_0 is the tip radius. We find that the unstable eigenvalue $\mathrm{Re}\lambda$ increases from zero as Δ increases across Δ_s while $\mathrm{Im}\lambda$ does not.

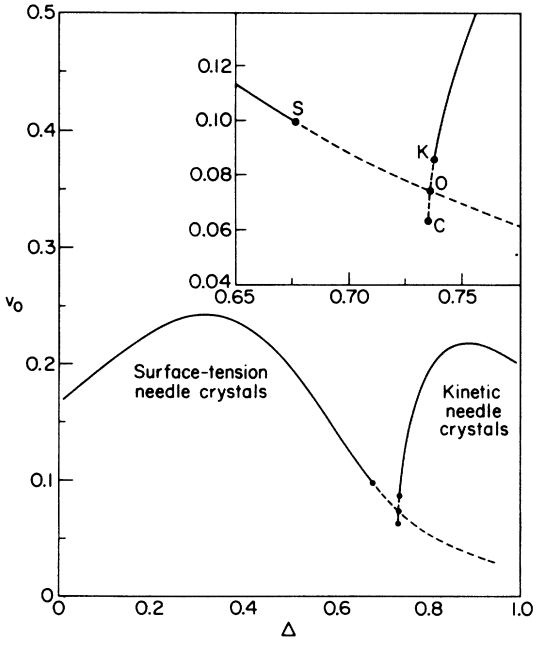

FIG. 1. Velocities of the steady-state needle crystals as a function of undercooling. Solid (dashed) lines represent regions where needle crystals are stable (unstable). Kinetic needle crystals (KN) only exist above $\Delta_c = 0.7351$. Inset is a partial amplification of the graph.

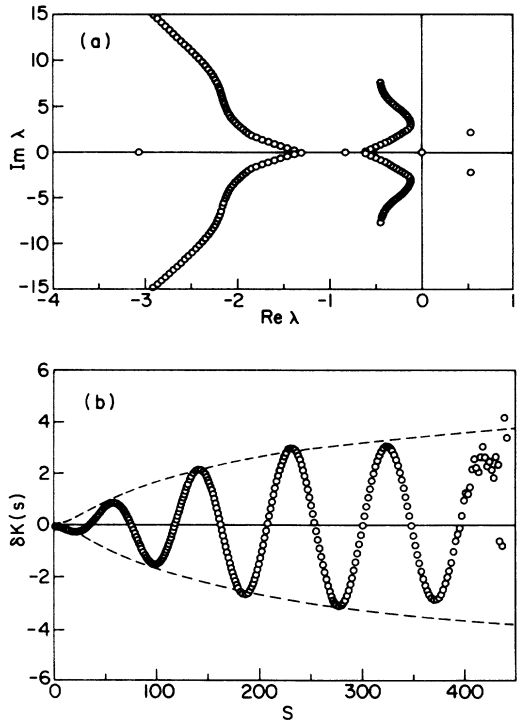

FIG. 2. (a) Eigenvalue spectrum for a surface-tension needle crystal at $\Delta = 0.7250$, $\epsilon_s = \epsilon_k = 0.4$. (b) Real part of the eigenfunction for the unstable mode in (a) with eigenvalue (0.537, 2.20). The arclength from the needle-crystal tip is denoted by s. The dashed curves are the fit of the envelope with the form $a\exp(-b/s^{1/2})$.

We have also observed two discrete unstable modes of the linear stability operator for antisymmetric perturbations.

The real part of the eigenfunction for the unstable eigenvalue at (0.527, 2.20) in Fig. 2(a) is plotted in Fig. 2(b). The envelope of the spatially oscillatory eigenfunction $\delta k(s)$ can be well fitted with a form $A(s) \propto \exp(-b/s^{1/2})$, where b is a constant. The fit is shown as the dashed curves in Fig. 2(b). Notice that the amplitude of the envelope monotonically increases and saturates to a constant at large s; this is very different from the envelope of the eigenfunction for the Ivantsov parabolic solutions,[6] where the envelope is spatially localized near the tip. On the other hand, it is also distinct from the asymptotic $\exp(s^{1/4})$ form predicted on the basis of the WKB approximation.[15] We do not yet know the implication, if any, of our finding for the generation of sidebranches. In general, the initial growth of perturbations will be dominated by the eigenmode with the largest $\mathrm{Re}\lambda$. By combining the approximate form of the eigenfunction and its time dependence, we obtain for the evolution of the fastest growing modes,

$$\delta k(s,t) \propto \exp\left[\mathrm{Re}\lambda \frac{v_0 t}{\rho_0}\right] \exp\left[-\frac{b}{s^{1/2}}\right] \times \cos\left[qs - \mathrm{Im}\lambda \frac{v_0 t}{\rho_0}\right], \quad (7)$$

which represents perturbations moving away from the needle-crystal tip with velocity $\mathrm{Im}\lambda(v_0/\rho_0 q)$ in the comoving frame of the needle crystal. Here q is the wave number of the oscillation of the eigenfunction. For the mode we are considering in Fig. 2(b), $\rho_0 = 35.0$, $q = 2\pi/2.7\rho_0$; thus the velocity $\approx 0.94 v_0$, i.e., the disturbances are almost stationary in the laboratory frame. The complex modes are not detectable in the WKB approximation,[16] since the unstable eigenfunctions are not localized.

The analysis for kinetic needle crystals is similar: there is also a critical undercooling $\Delta_k = 0.7382(2)$ below which KN's become unstable, as shown in Fig. 1. The envelopes of the complex unstable eigenfunctions have the same form as for the SN's. However, an additional unstable *real* eigenvalue is found for the symmetric linear stability operator. Figure 3(a) shows this real eigenvalue at $(7.20\times10^{-2}, 0)$ in the eigenvalue spectrum of a KN at $\Delta = 0.7360$, $\epsilon_s = \epsilon_s = 0.4$. This unstable discrete mode corresponds to a symmetric tip-splitting instability, since its eigenfunction is spatially localized and nonoscillatory, as shown in Fig. 3(b). Surprisingly, the spectrum for antisymmetric perturbations does not possess this additional real mode. Such a linear instability, if restabilized by nonlinearities, could generate sidebranches by the "solvability induced" sidebranching mechanism.[17]

Calculations on several sets of values of the parameters ϵ_s, ϵ_k confirm the generality of the above results. We have invariably found that $\Delta_s < \Delta_0 < \Delta_k$, which excludes the possibility of the coexistence of stable surface tension *and*

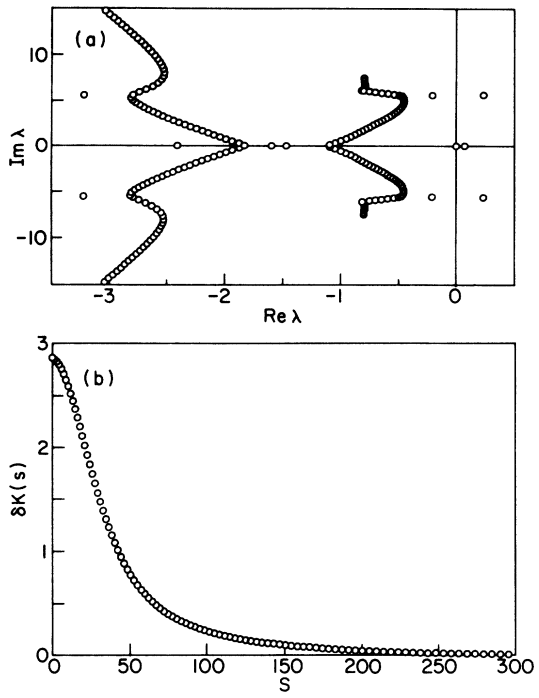

FIG. 3. (a) Eigenvalue spectrum for a kinetic needle crystal, with $\Delta = 0.7360$, $\epsilon_s = \epsilon_k = 0.4$. (b) The eigenfunction for the real mode in (a) with eigenvalue (0.072, 0).

kinetic needle crystals.[18] Thus we have demonstrated the nonexistence of linearly stable needle crystals when $\Delta_s < \Delta < \Delta_k$. This conclusion is supported by time-dependent simulations of the BLM where complex tip-splitting and sidebranching behavior is observed in the above parameter range. However, in our simulations, we have not been able to observe the extra real unstable mode associated with kinetic needle crystals.

To summarize, we have demonstrated that the competition between the anisotropy in surface tension and that in interfacial kinetics can destroy the linear stability of needle crystals. These results account for the presence of a morphological transition and lend further justification for the existence of a dense-branching structure. We expect that the results presented in this paper are more general than our calculation. It would be useful to perform the corresponding calculation for the full diffusion problem and for the Saffman-Taylor fingers. Our findings may also have implications for directional solidification when the direction of drawing does not coincide with a crystallographic axis.[19]

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