Dynamics of Interfacial Pattern Formation

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A phenomenological model of dendritic solidification incorporating interfacial kinetics, crystalline anisotropy, and a local approximation for the dynamics of the thermal diffusion field is proposed. The preliminary results are in qualitative agreement with natural dendritelike pattern formation.

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The formation of complex spatial patterns in nature has long been a source of challenging problems for the theorist.[1,2] A familiar example where this occurs is the snowflake; and recently attention has focused on the mechanisms of wavelength selection in a variety of other systems far from equilibrium exhibiting periodic spatial structures, including hydrodynamic instabilities in fluids,[3] electrohydrodynamic instabilities of nematic liquid crystals,[4] cellular flame fronts,[5] autocatalytic chemical reactions,[6] and directional solidification.[2,7] These systems possess families of linearly stable stationary states of different periodicities; yet in practice a unique state is reproducibly selected, for a large class of initial conditions. Here we will be concerned with dendritic solidification, an example of propagating pattern selection[8,9] where a localized perturbation of an initially homogeneous but unstable state grows at a unique velocity, forming a stable periodic state in its wake. The crucial questions are: What is the periodicity of the stable state, and what is the velocity of the propagating front? A selection criterion which is consistent with observation is that the system somehow chooses the state of marginal stability.[10] This has not yet been given a firm basis in theory, partly because of the lack of models which are sufficiently complex to retain the essential features, yet simple enough to be mathematically and computationally tractable. The purpose of this Letter is to present a new approach to interfacial pattern formation which promises to be applicable not just in the particular example of dendritic solidification, where our preliminary results seem qualitatively to reproduce observations, but also in other situations in physics, chemistry, and biology. Our new approach permits us to explore, in a way that has not previously been possible for this class of free-boundary problems, the fully nonlinear behavior that leads to pattern formation.

The salient features of dendritic solidification are as follows. We consider a liquid of heat capacity C at temperature T_∞ below the equilibrium melting temperature T_M of its solid, in contact with a growing solidification front. In the local-equilibrium approximation, the temperature of a flat interface is T_M, but because of the work performed against surface tension, a curved interface has a temperature lower by an amount proportional to the local curvature. Newly solidifying material at the interface generates latent heat L, which is conducted away from the interface by thermal diffusion (we neglect fluid flow). This process is facilitated by a large interfacial area in contact with the liquid and a large thermal gradient at the interface, both of which may be achieved by the interface developing protrusions extending into the liquid. The surface tension acts as a stabilizing mechanism preventing the formation of deformations on an arbitrarily small length scale, by providing a coupling between the temperature of the interface and the curvature of the deformation. As first shown by Mullins and Sekerka,[11] this unstable competition between the thermal diffusion and the surface tension is the underlying mechanism for dendritic growth. The other ingredient essential for the beautiful, regular shapes observed in dendrites is the crystalline anisotropy. Apart from providing the (e.g.) sixfold symmetry seen in snowflakes it may function either as a mechanism channeling the growth of instabilities along crystallographically preferred directions, or as a triggering mechanism for instabilities. Without it, it is likely that the growing dendrite would follow a crooked path rather than propagating along a single direction, giving rise to structures similar to those observed in diffusion-limited aggregation.[12] The effects of crystalline anisotropy arise in part through the kinetics of molecular attachment, an intrinsically nonequilibrium process; its inclusion in a model of dendritic solidification marks a departure from the usual assumption of local equilibrium.

The model described above, including the com-

plete dynamics of the thermal diffusion field, defines a highly nontrivial free-boundary problem (FBP) which has so far resisted even direct numerical simulation except in relatively simple special cases. Our alternative approach presented below is to derive a boundary-layer model (BLM) for the dynamics of the interface rather than that of the diffusion field, but to take into account in a simple way the basic physical features described above. We find that models which neglect any of the above ingredients are unable to give results even qualitatively similar to those observed in dendritic solidification. In particular, the interplay between the interface motion and the diffusion field is necessary to reproduce the Mullins-Sekerka instability, and we find[13] that models where the dynamics is solely determined by the local curvature[13-15] are inadequate.

The essence of our model is the boundary-layer approximation. That is, we visualize diffusion as occurring within a boundary layer at the solidification front, which is thin compared to the local radius of curvature. For the simple case of a crystal growing in two dimensions, this replaces the full diffusion problem by one-dimensional diffusion along the interface. Diffusion normal to the interface is accounted for by variations of the thickness of the boundary layer, l, which satisfies its own dynamical equation. It is more convenient and intuitive to consider, instead of l, the heat content per unit length of the interface

$$h = u_s l, \tag{1}$$

given in terms of the dimensionless temperature of the interface

$$u_s = \Delta - d_0 K(s) - \beta(\theta) V_n, \tag{2}$$

where $K(s)$ is the curvature of the interface, d_0 is a capillary length, and $\Delta = (T_M - T_\infty)/(L/C)$. V_n is the velocity of the interface along its outward normal and $\beta(\theta)$ is a function of the angle between the normal to the interface and a fixed direction in space, chosen such that growth is relatively enhanced in crystallographically favored directions. The term $-\beta V_n$ accounts for the crystalline anisotropy and introduces the constraint that the normal velocity is proportional to the driving force of solidification, a crude nonequilibrium condition. With neglect of thermal diffusion in the solid, the rate at which liquid solidifies is the heat current entering the boundary layer from the interface. Thus

$$V_n = -D\,\partial u_s/\partial n \simeq D u_s/l, \tag{3}$$

where D is the thermal diffusion coefficient. The equation of motion for h, following a point on the interface as it moves along its outward normal, is a statement of heat balance in the boundary layer:

$$\frac{dh}{dt} = V_n(1-u_s) + D\frac{\partial}{\partial s} l \frac{\partial u_s}{\partial s} - K V_n h. \tag{4}$$

The first term is the latent heat entering the boundary layer, the factor $1-u_s$ being the fraction not retained by the new layer of solid formed from the liquid. The second term is just the lateral diffusion along the boundary layer. The final term is of purely geometric origin and arises from the change in the differential arc length as the interface grows. The phenomenological Eq. (4) is supplemented by the exact kinematic equations of the interface

$$dK/dt = -(K^2 + \partial^2/\partial s^2) V_n, \tag{5a}$$

$$dS/dt = \int_0^s ds' K V_n, \tag{5b}$$

where, as in (4), time derivatives refer to variations along the outward normal. Equations (1)–(5) completely specify the BLM.

We have been able to verify analytically that the BLM accurately reproduces special solutions of the conventional free-boundary problem and that for $\Delta \sim 1$ the boundary layer is indeed thin compared to the local radius of curvature.[16] In addition, the limit of vanishing capillary length yields a family of linearly unstable parabolic needle-crystal solutions,[17] as found by Ivantsov[18]

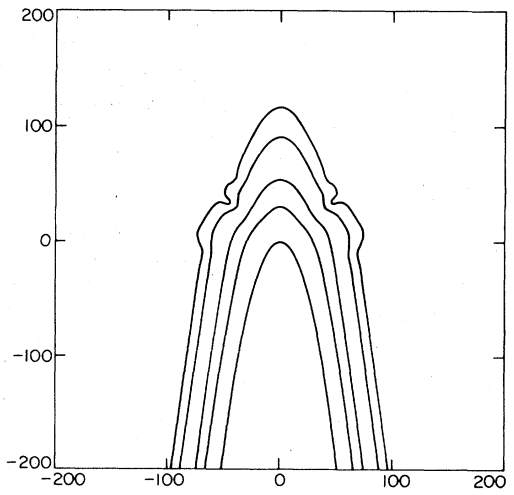

FIG. 1. Evolution of a dendritelike structure in the BLM with anisotropy. The full lines represent the interface at the times 10, 60, 111, 186, and 240.

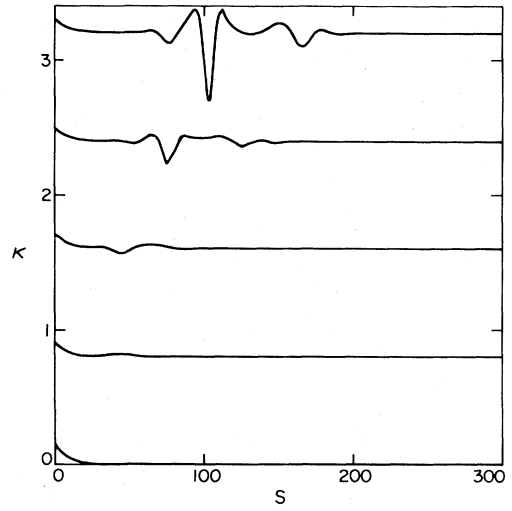

FIG. 2. Evolution of a dendritelike structure, at times (from bottom to top) 10, 60, 111, 186, and 240 plotted in (s,K) space. Each successive curve has been shifted by 0.8 from the previous one.

for the FBP. What is new, however, is that the BLM is sufficiently tractable that we can demonstrate that these solutions do not survive the inclusion of surface tension.[16] It is quite possible that this occurs in the FBP too.

Finally we present our preliminary numerical results. Starting from a parabola with the initial value of $h(s,t)$ being given by the Ivantsov solution of Eqs. (1)–(5), we find that with $\beta = 0$ the model does not generate dendritic structures for Δ in the range 0–1. Our results,[19] with the inclusion of crystalline anisotropy, are shown in Fig. 1, where fourfold anisotropy was imposed $[\beta = 0.1(1 - \cos 4\vartheta)]$ and $\Delta = 0.9$. Full details of both the analytic and the numerical results will be presented elsewhere. We verified that, at the last time shown, both the tip velocity and the tip radius reached a steady state. In Fig. 1, the formation of side branches in the wake of the moving tip is clearly visible, and in (s,K) space (Fig. 2) we see behind the tip the evolution of an increasingly complex structure. The pattern which emerges is similar to that seen during the early growth stages of real dendrites.

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