Boundary-Layer Formulation of Dendritic Growth: Existence of a Family of Steady-State Needle Solutions

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We develop a systematic boundary-layer–type formalism for diffusion-controlled dendritic growth, which yields an expression for the shape of steady-state needle solutions valid at large undercoolings. Both physical and analytical considerations suggest the general existence of a continuous family of steady-state needlelike solutions of the heat-flow equations. Simple modifications of the boundary-layer model of Ben-Jacob *et al.* exhibit this behavior.

Experiments show that the tip of a dendrite, freely growing into an undercooled melt, advances with a well-defined velocity V_t.[1,2] Theoretical understanding of this simple fact is, however, far from complete. Traditionally it was assumed that, for fixed experimental conditions, the heat-flow equations governing diffusion-controlled dendritic growth admit in general a continuous *family* of steady-state solutions, ranging from fat, slowly growing needles to sharp and rapidly growing ones.[3-5]

Surface-tension effects parametrized by the capillary length d_0 play an essential role in stabilizing patterns against side-branching instabilities.[5,6] However, such is the complexity of the full nonlocal heat-flow equations that the existence of a family of solutions has only been verified in an exact calculation in the artificial "Ivantsov limit,"[3,5,7] where d_0 is set equal to zero in the Gibbs-Thomson boundary condition $T_i = T_m - (L/c)d_0\kappa$ expressing the depression from the bulk melting temperature T_m of the interface temperature T_i due to interface curvature κ (L/c is the latent heat of fusion divided by the specific heat).

Recently Ben-Jacob *et al.*[8] and Brower *et al.*[9] have introduced simple *local* growth models describing the motion of the interface itself. These new models mimic many features of dendritic growth, while still permitting a more complete mathematical analysis. Both groups[8,9] find that with nonzero d_0 the Ivantsov family disappears and their models have steady-state solutions only for a discrete set of values of V_t, the largest of which also turns out to correspond to the tip velocity selected in dynamical simulations of the model.

These models are of interest in their own right, and their detailed study has stimulated many new ideas. However, when they are applied to the dendritic growth problem there are two properties of the new models that we find counterintuitive. First of all, the breakup of the family of steady-state solutions for $d_0 \neq 0$ results from the difficulty of matching the tip behavior properly with the asymptotic behavior far down in the tails of the needle.[8,9] We give below a physical argument suggesting that changes in shape near the tip could easily be accommodated in the tail region. Second, although the models do possess a family of solutions in the Ivantsov limit ($d_0 = 0$), there is a curious response to perturbations of these solutions. For example, in these models a local perturbation of the latent heat of fusion in some region along the interface produces a change in shape *only* within that region. Physically, the induced change in shape should extend at least somewhat beyond the perturbed region.

These observations led us to study the foundations of the new models and to reexamine the physical basis for the existence of a family of solutions. In this paper we sketch a systematic, and in principle exact, approach leading to a boundary-layer–type analysis in the spirit of Ben-Jacob *et al.*[8] We argue that one cannot draw general conclusions from the behavior of such models and that the emerging picture does not yield direct evidence for the breakup of the family of solutions.

The important physical parameter controlling dendritic growth is the dimensionless undercooling $\Delta \equiv (T_m - T_\infty)(L/c)^{-1}$. Here T_∞ is the temperature of the melt far from the interface. Henceforth, we consider dimensionless temperatures (normalized by L/c) measured with respect to the melt. Most real dendrites growth at small undercoolings[1,2] with $\Delta \ll 1$. Here nonlocal effects arising from long-ranged diffusion fields are very important since there is a large latent-heat surplus[10] which must diffuse away for growth to continue. Steady-state propagation at some constant velocity V_t is possible only if the interface bends back on itself in a needlelike shape, so that the excess heat can more efficiently flow to the surroundings. This is a global manifestation of the "point effect" underlying the Mullins-Sekerka instability[6] leading to side branching. Under the same conditions, the growth rate of a strictly planar interface decreases[5,8] in time t as $t^{-1/2}$, as the excess heat continually builds up in front of the interface. Likewise the growth of smooth objects (spheres, cylinders) approaches the $t^{-1/2}$ behavior as the curvature becomes small.[5]

Taken together, these results suggest that *all* needlelike solutions must become asymptotically parabolic [e.g., in two dimensions the tail of the steady-state shape $z(x,t)$ satisfies an equation of the form $z + cx^2 - V_t t$ in the laboratory frame] so that the $t^{-1/2}$ *normal* growth rate is

finally achieved in the tails where the curvature κ is small while the needle still maintains a constant growth rate in the z direction. This simple physics in the tail region holds irrespective of tip shape and of capillary effects, and it suggests to us the general existence of a family of steady-state solutions with different widths in the tails accommodating different curvatures near the tip.

To investigate these questions in more detail, we have developed a systematic boundary-layer approach in the spirit of Ben-Jacob et al.[8] (For simplicity, we consider the two-dimensional "one-sided model"[5] in which there is no heat diffusion in the solid.) We use [see Fig. 1(a)] a *time-dependent* set of orthogonal curvilinear coordinates[11,12] (u_1, u_2) chosen so that $u_1 = 0$ gives the interface position at all times, while u_2 at the interface remains constant when one follows the motion of the interface in the normal direction. This still leaves the freedom to choose the coordinates u_1, u_2 and associated scale factors[11] h_1, h_2 away from the interface. We choose them so
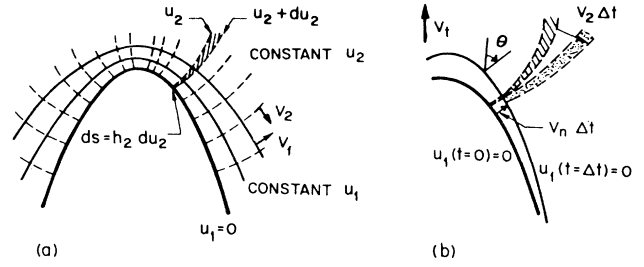

FIG. 1. The interface and the associated curvilinear coordinate fields u_1 and u_2. The interface position at all times is given by $u_1 = 0$ (heavy line). The total heat content in the tube bounded by lines of constant u_2 and $u_2 + du_2$ is $H\,ds$; v_1 and v_2 are the normal velocities of the constant-u_1 and -u_2 lines. Since $v_2 \neq 0$ and $u_1 \neq 0$, the tube deforms as a function of time, giving rise to J_{cv}, as illustrated in (b).

that the *exact* temperature field is given by

$$T(u_1, u_2, t) = T_i(u_2, t)e^{-u_1}, \tag{1}$$

with $T_i = \Delta - d_0\kappa$ from the Gibbs-Thomson boundary condition. The boundary condition for heat conservation at the interface then shows that $h_1(u_1 = 0, u_2, t)$ is equal to the boundary-layer thickness l of Ben-Jacob et al.[8]:

$$V_n = -D(\nabla T)_n = DT_i/h_1(u_1 = 0, u_2, t) \equiv DT_i/l. \tag{2}$$

Note that l is *defined* in terms of the normal interface velocity V_n, interface temperature T_i, and thermal diffusivity D by Eq. (2). Similarly H, the total heat content[8] per unit length of interface, is defined as

$$H(u_2, t) \equiv \frac{1}{h_2(u_1 = 0, u_2, t)} \int_0^\infty du_1\, h_1(u_1, u_2, t) h_2(u_1, u_2, t) T(u_1, u_2, t). \tag{3}$$

As illustrated in Fig. 1(a), $H\,ds = Hh_2 du_2$ is the total heat content in the "tube" between lines of constant u_2 and $u_2 + du_2$. Here s denotes the arc length along the interface.

Three effects contribute to the change in time of the total heat content in a tube: (i) the heat surplus generated at the interface,[10] (ii) heat diffusion across the constant-u_2 lines, and (iii) a "convective" flow associated with the motion of the tube indicated in Fig. 1(b). As a result, the dynamics of H is governed by the balance equation

$$(\partial H/\partial t)_{u_2} = V_n(1 - T_i) - \partial J_{cd}/\partial s - \partial J_{cv}/\partial s - HV_n\kappa. \tag{4}$$

The first three terms on the right-hand side are the contributions (i)–(iii) discussed above, respectively. The fourth term arises from the fact that two points on a positively curved interface grow apart in time, so that H, the heat content per unit length, decreases. While Eq. (4) has a clear physical interpretation, it can also be derived exactly from Eqs. (1)–(3), the heat-conduction equation, and exact kinematical equations describing the spatial and temporal variations of the u_1 and u_2 fields. This derivation, which will be discussed elsewhere,[13] yields

$$J_{cd} = -D \int_0^\infty ds_1\, \partial T/\partial s_2, \quad J_{cv} = -\int_0^\infty ds_1\, v_2 T. \tag{5}$$

Here $ds_1 = h_1 du_1$ is the infinitesimal arc length along a line $u_2 = \text{const}$ [above, we used $s = s_2(u_1 = 0)$; similarly $ds_2 = h_2 du_2$] and v_2 is the normal velocity of the constant-u_2 curves (see Fig. 1). Our choice of coordinates implies that v_2 and the analogous velocity v_1 satisfy at the interface $u_1 = 0$ the boundary condition $v_1 = V_n$ and $v_2 = 0$. Expressions (5) are in agreement with the physical interpretation given above; although the convective flow J_{cv} has not been considered in earlier work,[8] $J_{cv} \gg J_{cd}$ for steady states at Δ close to 1.

For simplicity we discuss here only steady-state solutions. In a steady state $V_n = V_t \cos\theta$, where θ is the angle between the normal and the growth direction [see Fig. 1(b)] and, from (2), $l = DT_i/V_t \cos\theta$. Further, one can show that[8,9] $(\partial H/\partial t)_{u_2} = -(\partial V_n/\partial \theta)(\partial H/\partial s)$. Thus for steady states (4) becomes

$$V_t\, \partial(H \sin\theta)/\partial s = V_t \cos\theta(1 - T_i) - \partial(J_{cd} + J_{cv})/\partial s. \tag{6}$$

We expect H, J_{cd}, and J_{cv} to depend on θ, $\kappa(= \partial\theta/\partial s)$, $\partial\kappa/\partial s$, and higher derivatives. Once this functional dependence

$H = H(\theta, \kappa l, l^2 \partial \kappa/\partial s, \ldots)$, etc., is known or approximated, then Eq. (6) yields a differential equation which can be solved to obtain the steady-state profile.

In general this functional dependence is difficult to determine. However, as emphasized by Ben Jacob et al.,[8] simplifications occur in the limit $\Delta \to 1^-$, where the small heat surplus requires only gently curved needles: Since steady-state solutions (if they exist!) are smooth, their curvature satisfies $\kappa^{ss}l = (1-\Delta)f(\theta) + O(1-\Delta)^2$. Thus

$$l^2 \partial\kappa^{ss}/\partial s = l^2 \kappa^{ss} \partial\kappa^{ss}/\partial\theta = O(1-\Delta)^2 \ll \kappa^{ss}l,$$

and derivatives can be ordered systematically. This allows us to expand the steady-state quantities H^{ss}, J^{ss}_{cd}, and J^{ss}_{cv} in powers of $1-\Delta$ and then, with the help of Eq. (4), determine the (asymptotic) expansion for the needle shape. In this way, we have reproduced the exact Ivantsov solution[3,7] to third order in $1-\Delta$. Similarly, for $d_1 \neq 0$, we find[13]

$$\frac{D\kappa^{ss}}{V_t} = (1-\Delta)\frac{C^3}{1-\alpha C^3} + (1-\Delta)^2 \frac{3C^3}{(1-\alpha C^3)^4}[1 - \alpha C^3(4-3C^2) + \alpha^2 C^6(1-4C^2+3C^4)]. \tag{7}$$

Here $\alpha \equiv d_0 V_t/D$ and $C \equiv \cos\theta$. This equation is consistent with the existence of a family of solutions, parametrized, e.g., by the tip velocity V_t.

However, within the boundary-layer model (BLM) of Ben-Jacob et al.[8] and the geometrical model (GM) of Brower et al.,[9] the capillary correction is a singular perturbation, and an expansion like the one above is invalid. To assess whether this might happen more generally, we first summarize the arguments for the BLM.

We can immediately obtain the BLM equation if we replace h_1, h_2, and v_2 in (3) and (5) by their values at the interface $u_1 = 0$. Then $H = T_i l = (\Delta - d_0 \kappa)l$; $J_{cd} = -Dl\partial T_i/\partial s = Dd_0 l \partial\kappa/\partial s$; $J_{cv} = 0$, and Eq. (6) yields, after rearrangement of the BLM equation,[8]

$$\kappa l - (1-\Delta)\cos^2\theta = d_0[\kappa\cos^2\theta + (l^2/\Delta)\cos\theta\sin\theta\kappa\partial\kappa/\partial s - (l^2/\Delta)\cos^2\theta\partial^2\kappa/\partial s^2]. \tag{8}$$

If we *assume* that there exists a smooth steady-state solution κ^{ss} of (8), then derivatives of κ^{ss} again order and we can ignore the last two terms on the right-hand side to first order in $1-\Delta$. From the remaining terms one gets an expression which is consistent with (8) to first order in $1-\Delta$, and which again suggests the existence of a family.

However, following Ben-Jacob et al.,[8] one can show that solutions to (8) in fact exist only for special discrete values of V_t. Equation (8) can be written as a set of first-order equations in a three-dimensional $(\theta, \kappa, \partial\kappa/\partial s)$ phase space. A steady-state solution is represented by a trajectory connecting the fixed point $(-\pi/2, 0, 0)$ to $(\pi/2, 0, 0)$. Now, if there is only one trajectory leaving the fixed point at $-\pi/2$ and, by symmetry, one flowing into the one at $\pi/2$, there is no reason to expect the two ends to connect smoothly for arbitrary V_t. Only for those discrete values of V_t where the two happen to connect is a steady-state solution to be found. This scenario actually occurs in (8), and in the GM.[8,9]

Obviously, a "counting argument" like the one sketched above for the number of stable and unstable directions near the fixed points is mathematically correct for a given model equation like (8). However, the important question is whether the physics of the problem allows the *global* flow in phase space to be modeled accurately by a simple truncation like Eq. (8). Since the driving force is large for $\Delta \lesssim 1$, one physically expects an arbitrary trajectory to change appreciably on the short length scale l, so that in general $l\partial\kappa/\partial s = O(\kappa)$. We have verified this explicitly[13] to first order in $1-\Delta$ by analyzing the flow in the neighborhood of the smooth steady-state solution, using the exact kinematical equations for h_1, h_2, v_1, and v_2 and the *full* heat-conduction equation, writing

$\kappa = (1-\Delta)(\kappa^{ss(1)} + \delta\kappa^{(1)})$ and linearizing in $\delta\kappa^{(1)}$. The result of the analysis is that $\delta\kappa^{(1)}$ either grows or decays rapidly as $e^{\pm\alpha s/l}$ with a *continuous range* of positive and negative α's possible (solutions with complex α exist as well). Thus, in a way, the *global* flow takes place in an essentially *infinite-dimensional* phase space and no natural truncation is apparent, since $l^2 \partial\delta\kappa/\partial s = O(\delta\kappa l)$, etc. Likewise, consideration of J_{cv} and the dependence of H on κ and its derivatives in (4) is essential to describe general flow in phase space, even though the truncation leading to the BLM (8) does reproduce the smooth steady-state solution to order $1-\Delta$.

Furthermore, there exist other simple, physically motivated truncations giving behavior very different from that of the BLM and the GM. In the BLM, all the terms on the right-hand side in (8) vanish for $d_0 = 0$ and the resulting *algebraic* equation fixes κ completely as $\kappa l = (1-\Delta)\cos^2\theta$. As mentioned above, this implies an infinitely fast relaxation toward the Ivantsov shape outside the range of a localized heat-source perturbation. This strange behavior can easily be modified. E.g., if we expand h_2 in (3) as

$$h_2(u_1, u_2) = h_2(0, u_2) + u_1 \partial h_2/\partial u_1$$

and use the fact that $\kappa = (h_1 h_2)^{-1}\partial h_2/\partial u_1$, we obtain $H = T_i l(1 + \kappa l)$. This approximation (with J_{cd} and J_{cv} as in the BLM) takes into account the physical effect underlying the Mullins-Sekerka instability[6] that, for a fixed l, H is larger in front of a curved interface. This new model does have a family of steady-state solutions for $d_0 \neq 0$, and perturbations relax exponentially fast toward

the Ivantsov solutions. However, it is otherwise just as arbitrary as the BLM and there is no guarantee that most other truncations behave similarly. Nevertheless, we suggest its study as a first step toward the formulation of a simple model that has the stability properties that we believe are physically called for.

Of course, strictly speaking the the above findings neither prove nor disprove the existence of a family for $d_0 \neq 0$, since arguments based on truncated boundary-layer models are inconclusive. However, our explicit calculation[13] to first order in $1 - \Delta$ does show that there are stable directions which are not included in the BLM or the GM and that the structure of the flow in phase space is hardly affected by a nonzero d_0. This similar response could be physically interpreted as a sign that a family of steady-state solutions exist for $d_0 \neq 0$ as well. Certainly the nature of the singularity, if d_0 should turn out to be a singular perturbation, is not likely to be correctly described by simple models using only a truncated phase space.

Finally, we note that while study of the $\Delta \lesssim 1$ regime is interesting in its own right,[14] one cannot necessarily carry over to the small-Δ regime results regarding the importance of possible singular perturbations[15] or anisotropy and interface kinetics at high undercooling. Glicksman[2] has already noted experimentally that the side branches "encroach on the steady-state region near the tip" for increasing driving force Δ in the range $0.002 \lesssim \Delta \lesssim 0.05$. If we extrapolate Δ to larger values, the side branches probably completely swamp the tip region before Δ is large enough for a boundary-layer approach to make sense; the growth in the $\Delta \lesssim 1$ regime thus should be quite different from the $\Delta \ll 1$ regime where the marginal-stability hypothesis of Langer[5] and Langer and Müller-Krumbhaar[4] successfully describes the data.[1,2] Nevertheless we expect that further study of the $\Delta \lesssim 1$ regime will lead to even more realistic phenomenological models in the spirit of the BLM and the GM and to a more rigorous mathematical resolution of the nature of the steady-state solutions for $d_1 \neq 0$.

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[5]See, e.g., J. S. Langer, Rev. Mod. Phys. **52**, 1 (1980).

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[10]A fraction of T_i of the latent heat is used to heat the undercooled melt to the temperature T_i. The remainder, proportional to $1 - T_i$, is the heat surplus, which must diffuse away. (For $\Delta > 1$, there is not sufficient latent heat released to satisfy the equilibrium Gibbs-Thomson boundary condition, and it is essential to take account of nonequilibrium interface kinetics. In the regime $\Delta \leq 1$ which we discuss herein, interface kinetics does not affect the qualitative conclusions we arrive at, though it may be quantitatively important.)

[11]See, e.g., F. B. Hildebrand, *Advanced Calculus for Applications* (Prentice Hall, Englewood Cliffs, N.J., 1962), p. 298.

[12]N. Goldenfield (unpublished) had earlier discussed the formulation of a boundary-layer model using a different set of time-independent curvilinear coordinates.

[13]W. van Saarloos and J. D. Weeks, unpublished.

[14]See, e.g., M. E. Glicksman and R. J. Schaefer, J. Cryst. Growth **1**, 297 (1967) for experiments in the range $0.3 \leq \Delta \leq 1.8$.

[15]Note that the effects of a singular perturbation are enhanced in the large-Δ regime. As pointed out to us by D. S. Fisher, it is conceivable that the singular effects show up only after exponentially long times [of order $\exp(l/d_0)$] for small d_0/l. In the experimentally accessible regime $\Delta \ll 1$, this ratio is smaller than 3×10^{-4} (see Refs. 1 and 2).