# Needle-Crystal Solution in Three-Dimensional Dendritic Growth

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(Received 29 July 1993)

The whole needle-crystal solution for three-dimensional (3D) dendritic growth is described analytically. Its construction involves the existing 3D selection theory for the tip of the dendrite (M. Ben Amar and E. A. Brener [Phys. Rev. Lett. **71**, 589 (1993)]) plus matching of the tail to this tip. This is exhibited here. Both intermediate and final asymptotics of the tail shape are given. This shape, which deviates strongly from an Ivantsov paraboloid, is in qualitative agreement with experimental observations.

PACS numbers: 44.30.+v, 68.45.–v, 81.30.Fb

Solvability theory has been very successful in predicting certain properties of the selected pattern in two-dimensional (2D) dendritic growth and a number of related phenomena [1]. In the dendritic case, the basic approach is as follows. One attempts to model the tip of a growing dendrite by a needle crystal, that is, a shape-preserving steady-state growth shape which is a solution to the equations of motion governing heat diffusion in the neighborhood of a solidification front. This needle crystal is assumed to be close in shape to the parabolic Ivantsov solution [2]. If anisotropic capillary effects are included, then a single dynamically stable solution is found for any external growth conditions. This unique needle crystal is believed to be a stable attractor for the (noise-free) system. Confirmation that the steady-state solution is actually an attractor for the fully dynamical system was obtained in two-dimensional time-dependent numerical simulations [3,4]. In addition, these calculations exhibit sidebranching of the dendritic tail (due to inherent numerical noise). We note that capillarity is a singular perturbation and the anisotropy of surface energy is a prerequisite for the existence of the solution. Solvability theory provides a very natural explanation for the fact that dendrites grow only in directions parallel to crystalline axes of symmetry.

This theory becomes extremely difficult, however, for three-dimensional (3D) anisotropic crystals. A simple extrapolation of the 2D case, where the surface energy is averaged in the azimuthal direction (axisymmetric approach [5,6]), is very important in order to have some qualitative predictions. But any physical anisotropy will give rise to a nonaxisymmetric shape of the crystal. A numerical approach to the nonaxisymmetric problem was presented by Kessler and Levine (KL) [7], who pointed out the following aspect of the problem. In the 2D case or in the axisymmetric case selection of the growth velocity follows from the solvability condition of smoothness of the dendrite tip. In the 3D nonaxisymmetric case a solvability condition must be satisfied for each of the azimuthal harmonics. KL made several approximations and performed only a two-mode calculation, but the crucial point of their analysis is that they found enough degrees of freedom to satisfy all solvability conditions.

Recently, an analytic theory of three-dimensional dendritic growth has been developed by Ben Amar and Brener [8]. In the framework of asymptotics beyond all orders, they derived the inner equation in the complex plane for the nonaxisymmetric shape correction to the Ivantsov paraboloid. The solvability condition for this equation provides selection of both the velocity of the dendrite and the interface shape. The selected shape can be written as

$$z(r,\phi) = -\frac{r^2}{2} + \sum A_m r^m \cos(m\phi), \tag{1}$$

where all lengths are reduced by the tip radius of curvature $\rho$. Solvability theory [8] predicts that the numbers $A_m$ are independent of the anisotropy strength $\alpha$, in the limit of small $\alpha$. For example, the first nontrivial term for a cubic symmetry corresponds to $m = 4$ and $A_4 = 1/88$ is only numerically small (note that in [8] lengths are scaled by $2\rho$ rather than $\rho$). Therefore, the shape correction [Eq. (1)], in units of the tip radius of curvature, depends mostly on the crystalline symmetry and is almost independent of the material and growth parameters.

An important aspect of Eq. (1) is that the shift vector $r^m \cos m\phi$ grows at a faster rate than the underlying Ivantsov solution. This means that only the tip region, where the anisotropy correction is still small, can be described by the usual approximation [7,8], a linearization around the Ivantsov paraboloid. This is the crucial difference between the 3D nonaxisymmetric case and the 2D case. In the latter, small anisotropy implies that the shape of the selected needle crystal is close to the Ivantsov parabola everywhere; in the former, strong deviations from the Ivantsov paraboloid appear for any anisotropy.

Several of the following important questions arise: How can the tail of the dendrite be described? Is it possible to match the nonaxisymmetric shape (1) in the tip region to the asymptotic shape in the tail region? What is the final needle-crystal solution? The aim of this paper is to answer these questions. I will show that the tail shape *follows* from that of the tip after solution of the selection

0031-9007/93/71(22)/3653(4)$06.00

problem for the tip region [8], where the needle crystal is still close to the Ivantsov solution and linearization is possible, and that asymptotically it deviates strongly from the Ivantsov paraboloid.

The basic idea is that the nonaxisymmetric shape correction, generated in the tip region, should be used as an "initial" condition for a time-dependent *two-dimensional* problem describing the motion of the *cross section* of the interface in the tail region. This two-dimensional problem will be defined in detail below. In this reduced description, the role of time is played by the coordinate $z$ for steady-state growth in the $z$ direction. The deviation from the isotropic Ivantsov solution remains small only during the initial period of the evolution. This initial deviation can be handled by a linear theory starting from a mode expansion which takes the same form as Eq. (1) with, in principle, arbitrary coefficients. These amplitudes then have to be chosen according to the predictions of selection theory [8] in order to provide the matching to the tip region. As "time" goes on, the deviation increases due to the Mullins-Sekerka instability and a nonlinear theory must take over. This two-dimensional time-dependent problem of a moving interface with anisotropic surface tension was considered in [9] in the framework of a Laplacian approximation. After some transition time the system shows an asymptotic behavior which is independent of the initial conditions: Four well-developed arms are formed (for fourfold symmetry, which I will assume from now on). The length of the arms increases in time as $t^{3/5}$ and their width increases as $t^{2/5}$. Translated to our case, this means that the length and width of the arms increase as $|z|^{3/5}$ and $|z|^{2/5}$, respectively, where $|z|$ is the distance from the dendritic tip. This shape, which is very different from the Ivantsov paraboloid, should hold as long as the two-dimensional diffusion length is larger than the size of the arms, before a subsequent crossover to the classical constant velocity regime of the two-dimensional dendrite. In this last regime the length of the arms increases as $|z|$. This brief description comprises, in a qualitative way, the entire solution to the needle-crystal problem in three dimensions.

Let us now describe the tail region more precisely. We start by writing down the equations of motion for the aforementioned 2D problem. We use the terminology and notation pertaining to thermally driven solidification. The equations are

$$D\nabla^2 u = \partial u/\partial t, \tag{2}$$

$$v_n = D\mathbf{n}\cdot(\nabla u_S|_{\text{int}} - \nabla u_L|_{\text{int}}), \tag{3}$$

$$u|_{\text{int}} = \Delta - d_0(1-\alpha\cos 4\tilde{\phi})K. \tag{4}$$

$L$ and $S$ refer to the liquid and solid, respectively, and $u = (T-T_\infty)c_p/L$ is the rescaled temperature field, measured from the temperature at infinity $T_\infty$. The specific heat $c_p$ and the thermal diffusion constant $D$ are considered to be the same in both phases, $L$ is the latent heat. In terms of these parameters, $\Delta = (T_M - T_\infty)c_p/L$ is the dimensionless undercooling ($T_M$ is the melting temperature), $d_0 = \gamma T_M c_p/L^2$ is a capillary length proportional to the isotropic part of the surface energy $\gamma$.

The physics underlying Eqs. (2)–(4) is quite simple. A solidifying front releases latent heat which diffuses away as expressed by (2); requiring heat conservation at the interface gives (3) ($\mathbf{n}$ is the normal to the interface and $v_n$ is the normal velocity). Equation (4) is an anisotropic local-equilibrium condition which takes into account curvature corrections ($\alpha$ is the strength of crystalline anisotropy, the angle $\tilde{\phi}$ is the orientation of the normal relative to a fixed direction such as the $x$ axis and $K$ is a two-dimensional curvature).

We note that Eqs. (2)–(4) in this general form also describe 3D growth. The only difference is that the anisotropic capillary correction in Eq. (4) has a much more complicated form. In order to reduce the description to a 2D equation for the cross section of the tail, we must neglect the second derivative with respect to $z$ in (2), the $z$ component of the normal velocity in (3), and 3D curvature effects in (4). I will justify these approximations later. Note also that the time derivative in (2) turns into a $z$ derivative due to the stationary character of the solution we are seeking.

To solve the problem, we need the following additional information which comes from 3D selection theory [8]. The dendritic tip with the radius of curvature $\rho$ moves at a constant velocity $v$. The Péclet number $P = \rho v/2D$ is related to the undercooling $\Delta$ by the 3D Ivantsov formula [2] which reads for small $\Delta$

$$P(\Delta) = -\Delta/\ln\Delta. \tag{5}$$

The stability parameter $\sigma$,

$$\sigma \equiv d_0/P\rho = \sigma^*(\alpha), \tag{6}$$

where $\sigma^*(\alpha)$ is given in [8] and $\sigma^*(\alpha) \propto \alpha^{7/4}$ for small $\alpha$. Relation (6) together with the Ivantsov relation (5) selects both $v$ and $\rho$. The interface shape in the tip region is described by Eq. (1) with the amplitudes $A_m$ given by 3D selection theory [8].

First of all, Eqs. (2)–(4) have a simple solution,

$$r_0^2(t) = 4P(\Delta)Dt, \tag{7}$$

which describes a growing circle. The field outside of the circle can be written as

$$u_0(r,t) = -2P(\Delta)\ln\frac{r}{(Dt)^{1/2}} \tag{8}$$

for $r_0 < r \ll (Dt)^{1/2}$, and this condition can be fulfilled for small $\Delta$. In this region the field satisfies the Laplace equation but for larger $r$ it decays exponentially. Actually, Eq. (7) describes the Ivantsov paraboloid solution. Indeed, replacing $t$ by $|z|/v$ and reducing all lengths by $\rho$, we find $r^2 = 2|z|$.

For a slightly deformed circle we can look for the interface shape in the form

$$r(\phi,t) = r_0(t) + \delta_m(t)\cos m\phi$$

and for the field in the form

$$u(r,\phi,t) = u_0(r,t) + B_m(t) r^{-m}\cos m\phi,$$

which satisfies the Laplace equation. To linear order in $\delta_m$, Eqs. (2)–(4) give

$$\delta_m(t) \sim t^{(m-1)/2}.$$

Again, replacing $t$ by $|z|/v$ and reducing all lengths by $\rho$, we find

$$r(z,\phi) = (2|z|)^{1/2} + \sum A'_m (2|z|)^{(m-1)/2}\cos m\phi, \quad (9)$$

where the amplitudes $A'_m$ are arbitrary at this stage. An important point is that this shape can be precisely written in the form of Eq. (1), since the deviation from the Ivantsov paraboloid is small. The matching between the tip region [Eq. (1)] and the tail region [Eq. (9)] therefore just requires setting $A'_m = A_m$.

Up to now, we have neglected the capillary correction in Eq. (4) because it just gives a small correction, proportional to $\sigma$, to the result (9). As "time" goes on, the deviation from the Ivantsov paraboloid increases and a nonlinear theory must be applied. Actually, we can guess what the long-time behavior of the system will be [10]. Indeed, the two-dimensional problem, defined by Eqs. (2)–(4), is precisely the same as the one that leads to two-dimensional dendritic structures [1]. Four well-developed arms (for cubic symmetry) grow with a constant speed in the directions which are favored by the surface energy anisotropy. Each arm has a parabolic shape, its growth velocity, $v_2 = 2DP_2^2(\Delta)\sigma_2^*(\alpha)/d_0$, and the radius of curvature of its tip, $\rho_2 = d_0/P_2\sigma_2^*$, are given by 2D selection theory [1], where the anisotropic surface energy again plays a crucial role. The Péclet number $P_2 = \rho_2 v_2/2D$ is related to the undercooling $\Delta$ by the 2D Ivantsov formula [2] which for small $\Delta$ gives

$$P_2(\Delta) = (\pi\Delta)^{1/2}. \quad (10)$$

The selected stability parameter $\sigma_2 = d_0/P_2\rho_2$ depends on the strength of the anisotropy $\alpha$, and $\sigma_2^*(\alpha) \propto \alpha^{7/4}$ for small $\alpha$. Replacing $t$ by $|z|/v$ and reducing all lengths by $\rho$, we can present the shape of one quarter of the interface (except very close to the dendritic backbone) in the form

$$x(y,z) = |z|\frac{v_2}{v} - \frac{y^2}{2}\frac{\rho}{\rho_2}. \quad (11)$$

This asymptotics describes the strongly anisotropic interface shape far behind the tip and does not match, for small $\Delta$, to the shape described by Eq. (9). In this case an important intermediate asymptotics exists.

If the size of the 2D pattern is still much smaller than a diffusion length $(Dt)^{1/2}$, the field $u$ far away from the pattern is described by Eq. (8). This 2D Laplacian problem (with a fixed flux from the outside) was solved recently, both numerically and analytically, by Almgren, Dai, and Hakim [9], who were interested in anisotropic Hele-Shaw flow. They found that after some transition time the system shows an asymptotic behavior which is independent of the initial conditions and involves the formation of four well-developed arms. The length of these arms increases in time as $t^{3/5}$ and their width increases as $t^{2/5}$ (see the figures in [9]). The basic idea which explains these scaling relations is that the stability parameter $\sigma = 2Dd_0/\rho_2^2 v_2$ is supposed to be equal to $\sigma_2^*(\alpha)$ even though both $v_2$ and $\rho_2$ depend on time. Moreover, the growing self-similar shape of the arms was determined in [9]. In the terms of our problem it reads

$$y(x,z) = (5|z|/3)^{2/5}\left(\frac{\sigma^*}{\sigma_2^*}\right)^{1/5}\left(\frac{x}{x_{\text{tip}}}\right)^{2/3} \times \int_{x/x_{\text{tip}}}^{1}\frac{ds}{s^{2/3}\sqrt{1-s^4}}, \quad (12)$$

where the tip position of the arm $x_{\text{tip}}$ is given by

$$x_{\text{tip}}(z) = (5|z|/3)^{3/5}(\sigma_2^*/\sigma^*)^{1/5}. \quad (13)$$

The ratio $\sigma_2^*(\alpha)/\sigma^*(\alpha)$ is independent of $\alpha$ in the limit of small $\alpha$. This means that the shape (12) in the tail region is almost independent of the material and growth parameters, as well as the shape (1) in the tip region (if the all lengths are reduced by $\rho$). This shape which differs strongly from the Ivantsov paraboloid should hold as long as the two-dimensional diffusion length is larger than the size of the arms,

$$1 \ll |z| \ll \Delta^{-5}, \quad (14)$$

before subsequent crossover (at $|z| \sim \Delta^{-5}$) to the constant velocity regime (11) described above.

Our 2D approximation for the tail description requires just $|z| \gg 1$. This condition allows us to neglect 3D curvature effects and the $z$ component of the normal velocity. Furthermore, it results in a negligible second derivative with respect to $z$ *in the vicinity* of the interface. Of course, for the *far field*, our 2D approximation breaks down. But this turns out not to be important for the interface evolution. Rigorously speaking, the linearization in the tip region and in the matching region requires the amplitudes $A_m$ to be small. As mentioned in Ref. [8] the used model with cubic anisotropy contains only a numerically small (nontunable) parameter providing the smallness of $A_m$. The above linearization can be made mathematically rigorous introducing an artificial (tunable) small parameter which guarantees smallness of the azimuthal anisotropy and, hence, of the $A_m$.

In conclusion, the results of this paper together with the results of 3D selection theory [8] constitute an analytic solution to the needle-crystal problem for 3D dendritic growth. Some details, such as the crossover regimes and structure of the backbone, can be worked out numerically by existing codes [3,4,9] for the 2D time-dependent problem.

The term "needle crystal" came from the idea that the solution should be close to the Ivantsov paraboloid and that anisotropic surface energy plays an important role for the tip velocity selection. I find that the shape of the needle crystal is close to the Ivantsov one only in the tip region [Eq. (1)] and deviates strongly in the tail region [Eqs. (11)–(13)]. Surface-energy anisotropy is important for the shape selection in the tail region as well as in the tip region.

Clearly, the needle-crystal solution does not account for noise-induced sidebranching behavior. The description of this behavior necessitates the solution of a time-dependent problem for the noise-induced perturbation around the needle-crystal shape [11]. Perhaps, a theory which takes into account the actual nonaxisymmetric shape of the needle crystal will be able to resolve the puzzle that experimentally observed sidebranches have much larger amplitudes than explicable by thermal noise in the framework of the axisymmetric approach [11]. Indeed, for a shape $x \sim |z|^\beta$ with $\beta > 1/2$, the amplification of noise is larger than for the parabola, which is a change in the right direction. With the present 3D solution in hand, hopefully the difficult and interesting problem of the sidebranching activity of 3D dendrites can be attacked more successfully than in the past. Another direction of research, where further progress should be possible, concerns the stability analysis of 3D dendrites based on this new solution.

Sidebranching activity leads to the appearance of new primary 3D dendritic branches far behind the main tip (at distances of the order of a 3D diffusion length from the tip). This effect could completely suppress the long-distance asymptotic behavior (11), but the intermediate asymptotics (12),(13) should still be detectable. Recently, experiments on 3D dendrites have been performed [12] in an attempt to determine the scaling relation between the length $|z|$ of a dendrite and the width $w(z)$ measured across its main arms. The result $w(z) \sim |z|^\beta$ with $\beta = 0.74 \pm 0.06$ at least does not seem inconsistent with the theory presented here.

I thank M. Ben Amar, H. Müller-Krumbhaar, D. Temkin, and K. Kassner for valuable discussions and the latter, in particular, for a critical reading of the manuscript and various suggestions to improve its readability.

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[1] For a review see J.S. Langer, in *Chance and Matter*, Proceedings of the Les Houches Summer School, Session XLVI (Elsevier, New York, 1987); D. Kessler, J. Koplik, and H. Levine, Adv. Phys. **37**, 255 (1988); E.A. Brener and V.I. Melnikov, Adv. Phys. **40**, 53 (1991); Y. Pomeau and M. Ben Amar, in *Solids Far From Equilibrium*, edited by C.Godréche (Cambridge University Press, Cambridge, 1992).

[2] G.P. Ivantsov, Dokl. Akad. Nauk SSSR **58**, 567 (1947).

[3] Y. Saito, G. Goldbeck-Wood, and H. Müller-Krumbhaar, Phys. Rev. Lett. **58**, 1541 (1987).

[4] T. Ihle and H. Müller-Krumbhaar, Phys. Rev. Lett. **70**, 3083 (1993).

[5] M. Ben Amar, Physica (Amsterdam) **31D**, 409 (1988).

[6] A. Barbieri and J.S. Langer, Phys. Rev. A **39**, 5314 (1989).

[7] D. Kessler and H. Levine, Acta Metall. **36**, 2693 (1988).

[8] M. Ben Amar and E. Brener, Phys. Rev. Lett. **71**, 589 (1993).

[9] R. Almgren, W-S. Dai, and V. Hakim (to be published).

[10] E. Brener and D. Temkin (unpublished).

[11] J.S.Langer, Phys. Rev. A **36**, 3350 (1987).

[12] A. Dougherty and R. Chen, in *Coarsening and the Mean Shape of Three-Dimensional Dendritic Crystals*, Computational Crystal Growers Workshop, edited by J. Taylor (AMS, Providence, RI, 1992), p. 31.