Side branching in the three-dimensional dendritic growth

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The time-dependent behavior of side branching deformations is considered with allowance for the actual nonaxisymmetric shape of the needle crystal. It is found that the amplitude of the deformation increases faster than that for the axisymmetric paraboloid shape of the needle. It is argued that this effect can resolve the puzzle that the experimentally observed side branches have much larger amplitudes than can be explained by the thermal noise in the framework of the axisymmetric approach. The coarsening behavior of side branches in the nonlinear regime is discussed.

We have studied the problem of a free dendrite growing in a one-component undercooled melt.[1] The control parameter is the dimensionless undercooling $\Delta=(T_M-T_\infty)c_p/L$, where T_M is the melting temperature, L is the latent heat, and c_p is the specific heat. The temperature profile satisfies the diffusion equation with the interface moving at a normal velocity v_n and acting as a source of magnitude v_nL/c_p. Together with the Gibbs–Thomson condition at the interface, it leads to a rather complicated integrodifferential evolution equation.

The steady-state version of this problem was discussed in Refs. 2 and 3. The dendritic tip with the radius of curvature ρ moves at a constant velocity v. The Peclet number $P=\rho v/2D$ (D is the thermal diffusivity) is related to the undercooling Δ by the 3D Ivantsov formula,[4] which for a small Δ is $P(\Delta)=-\Delta/\ln\Delta$. The stability parameter is $\sigma\equiv d_0/(P\rho)=\sigma^*(\alpha)$, where $d_0=\gamma T_M\, c_p/L^2$ is the capillary length proportional to the isotropic part of the surface energy γ, and α is the strength of the crystalline anisotropy. The function $\sigma^*(\alpha)$ is given by the 3D selection theory[2] and $\sigma^*(\alpha)\propto\alpha^{7/4}$ for a small α. These two relations for P and σ determine both v and ρ. The interface shape near the tip is close to the Ivantsov paraboloid and can be described by the equation $z(r,\phi)=-r^2/2+\Sigma A_m\, r^m\cos(m\phi)$, where the amplitudes A_m are given by the 3D selection theory[2] (we measure all lengths in units of ρ and time in units of ρ/v). In the tail region the interface shape deviates from the Ivantsov paraboloid: Four well-developed arms (for cubic symmetry) are formed in the cross section. For a small Δ, not too far from the tip, this shape can be described as[3]

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

where the tip position of the arm is $x_{\rm tip}(z)=(5|z|/3)^{3/5}(\sigma_2^*/\sigma^*)^{1/5}$. The function $\sigma_2^*(\alpha)$ is given by the 2D selection theory and the ratio $\sigma_2^*(\alpha)/\sigma^*(\alpha)$ is independent of α in the limit of small α.

The description of the side branches requires the solution of a time-dependent problem for the perturbation around this missile-shaped, steady-state crystal $z=\zeta_0(x,y)$. Langer *et al.*[5–7] suggested that dendritic side branches might be generated by a selective amplification of a very small, noisy perturbation near the tip of a growing needle crystal. It appears that realistic side branching behavior might be produced by purely thermal fluctuations in the solidifying material. The side branching deformation is described in Ref. 7 as a small (linear) perturbation moving on a cylindrically symmetric needle crystal (the Ivantsov paraboloid[4]). The noise-induced wave packets generated near the tip grow in amplitude, spread, and stretch as they move down the sides of the dendrite, producing a train of side branches. In the linear approximation, the amplitude grows exponentially and the exponent is proportional to $|z|^{1/4}$. These results are in approximate qualitative agreement with the available experimental observations,[8,9] but experimentally observed side branches have much larger amplitudes than can be explained by thermal noise in the framework of the axisymmetric approach.[7] This means that either the thermal fluctuation strength is adequate to produce visible side branching deformations or agreement with experiment would require at least one more order-of-magnitude exponential amplification factor.

The main goal of this paper is to describe the side branching problem, taking into account the actual nonaxisymmetry shape of the needle crystal, defined by Eq. (1). We will show that for this nonaxisymmetric shape the perturbations increase faster than for the axisymmetric shape. This effect allows us to remove the above-mentioned discrepancy between theory and experiment.

As in the Ref. 7, we assume that the perturbation is small and consider its evolution in the linear approximation. Therefore, the first step in this analysis is to linearize the evolution equation about the steady-state solution. For the investigation of the behavior of a noise-induced wave packet as it moves along the dendrite it is important to know the Green's function of our linear problem. According to Ref. 10, the Green's function is given by a path integral

$$G(X,Y,t,X',Y',t')=\int \exp\left[\int_{t'}^{t} \Omega(x,y,k_x,k_y)d\tau - i\int_{X'}^{X} k_x dx - i\int_{Y'}^{Y} k_y dy\right] \times D\{x(\tau)\}D\{y(\tau)\}D\{k_x(\tau)\}D\{k_y(\tau)\}. \tag{2}$$

Here the functional integration is performed along all the trajectories $x(\tau)$, $y(\tau)$, $k_x(\tau)$, and $k_y(\tau)$ which start at the point $x=X'$, $y=Y'$ at $\tau=t'$ and go to the point $x=X$, $y=Y$ at $\tau=t$.

The expression for the Green's function is of the Feynman type, but with the action

$$S=\int_{t'}^{t}\Omega(x,y,k_x,k_y)d\tau - i\int_{X'}^{X}k_x dx - i\int_{Y'}^{Y}k_y dy \tag{3}$$

written in the Hamiltonian form rather than in the Lagrangian form. In this representation all important information about the problem is contained in the local dispersion relation $\Omega(x,y,k_x,k_y)$ of the linear operator. In the WKB approximation the functional integral can be calculated by the method of the steepest descent, where the Green's function behavior is determined by the extremal trajectory which is governed by the Hamilton equations

$$\frac{dx}{d\tau}=-i\frac{\partial\Omega}{\partial k_x},\quad \frac{dy}{d\tau}=-i\frac{\partial\Omega}{\partial k_y},\quad \frac{dk_x}{d\tau}=i\frac{\partial\Omega}{\partial x},\quad \frac{dk_y}{d\tau}=i\frac{\partial\Omega}{\partial y}. \tag{4}$$

Thus, the Green's function is $G\sim\exp(S_{\text{ext}})$ and the problem reduces to the solution of the Hamilton equations for the given Hamiltonian function $\Omega(x,y,k_x,k_y)$.

The important point is that the local dispersion relation for this solidification problem is the well-known local Mullins–Sekerka spectrum. Let us replace the interface of the needle crystal in the vicinity of the arbitrary point x, y, $z=\zeta_0(x,y)$ by a piece of the tangential plane. For the short-wavelength perturbation of the form $\delta n\sim\exp(\Omega t-ik_s s-ik_u u)$, the local Mullins–Sekerka spectrum is

$$\Omega=\sqrt{k_s^2+k_u^2}\,[\cos\Theta-\sigma(k_s^2+k_u^2)]+ik_s\sin\Theta. \tag{5}$$

Here Θ is the angle between the z axis and the local normal $\hat{n}$, k_s, and k_u are the components of the wave vector along $\hat{s}$ and $\hat{u}$, and $\hat{s}$ and $\hat{u}$ are the unit orthogonal vectors in the tangential plane. The unit vector $\hat{s}$ lies in the tangential plane and in the normal n, z plane.

The spectrum (5) is presented in the local orthogonal frame of reference n, s, u. It is convenient to rewrite it in the fixed Cartesian coordinates and to obtain the spectrum in the form $\Omega(x,y,k_x k_y)$.

The main restriction of our calculation comes from the fact that any further analytical progress can be reached only for small values of y, i.e., near the tip of the main arm in the cross section. In this region the unperturbed interface of the needle crystal, which is given by Eq. (1), can be written as follows:

$$(\tfrac{5}{3}|z|)^{3/5}-\frac{y^2}{(\tfrac{5}{3}|z|)^{1/5}}=x,\quad \frac{y}{|z|^{2/5}}\ll 1. \tag{6}$$

Here we omitted the factor $[\sigma_2^*(\alpha)/\sigma^*(\alpha)]^{1/5}$ in (1) which is very close to 1.

The actual values of $k_y(\tau)\sim y(\tau)$ are small in the region of small y. For small y and k_y we can expand the function Ω to the second-order terms:

$$\Omega(x,y,k_x,k_y)=\Omega_0(x,k_x)+\tfrac{1}{2}Ay^2+Byk_y+\tfrac{1}{2}Ck_y^2. \tag{7}$$

Here Ω_0, A, B, and C are the functions of x and k_x only. Straightforward but tedious calculations give for $x\gg 1$ (or $|z|\gg 1$) the equations

$$\Omega_0=\frac{-ik_x}{p_o}\left[1+i\frac{\sigma k_x^2}{p_o^2}+\frac{i}{p_o}\right], \tag{8a}$$

$$A=\frac{b^2k_x}{p_o^2}\left[1+\frac{3\sigma k_x^2}{p_o}+\frac{2i}{p_o}\right], \tag{8b}$$

$$B=-\frac{b}{p_o}\left[1+\frac{3\sigma k_x^2}{p_o}+\frac{i}{p_o}\right], \tag{8c}$$

$$C=\frac{1}{k_x}\left[1+\frac{3\sigma k_x^2}{p_o}\right], \tag{8d}$$

where $p_o=(\partial\zeta_o/\partial x)_{y=0}$, and $b=(\partial^2\zeta_o/\partial y^2)_{y=0}$. These equations are derived for an arbitrary profile with an extremum at $y=0$ and they are valid for $|p_o|\gg 1$. For our profile [Eq. (6)] we have $p_o=-x^{2/3}$ and $b=-2x^{1/3}$.

We would like to find the optimal trajectory, i.e., the four unknown functions $x(\tau)$, $y(\tau)$, $k_x(\tau)$, and $k_y(\tau)$ which are governed by Eqs. (4), (7), and (8) and by four boundary conditions: $x(0)=X'=0$, $y(0)=Y'=0$, $x(t)=X$, and $y(t)=Y$. To accomplish this goal, we use the following iterative strategy:

i) The first step is to solve these equations for the case $y(\tau)=0$ and $k_y(\tau)=0$. This gives the trajectory $x(\tau)$, $k_x(\tau)$ along the ridge of the side arm.

ii) The second step is to find $y(\tau)$, $k_y(\tau)$ for the fixed functions $x(\tau)$ and $k_x(\tau)$ given by the first step.

iii) Finally, we find the corrections to $x(\tau)$, $k_x(\tau)$ due to the functions $y(\tau)$, $k_y(\tau)$ given by the second step.

After a lengthy calculation, which will be published elsewhere,[11] we find the action for the optimal trajectory

$$S(Z,Y,t)=\frac{2(5/3)^{(9/10)}}{3\sqrt{3\sigma}}|Z|^{2/5}\left\{1+i\,\frac{3}{2}\left(\frac{3}{5}\right)^{3/5}\frac{(t-|Z|)}{|Z|^{3/5}}-\frac{3}{8}\left(\frac{3}{5}\right)^{6/5}\frac{(t-|Z|)^2}{|Z|^{6/5}}\right.$$
$$\left.-\frac{9}{4}\left(\frac{3}{5}\right)^{4/5}(\sqrt{1-i/9}-1)\frac{Y^2}{|Z|^{4/5}}-\frac{3\alpha}{2}\left(\frac{3}{5}\right)^{7/5}\frac{Y^2(t-|Z|)}{|Z|^{7/5}}\right\}, \tag{9}$$

where $\alpha\simeq 0.039$.

After the calculation of the action S at the optimal trajectory we can write the Green's function as $G\sim\exp(S)$, where the prefactor comes from the functional integration over the space near the optimal trajectory. The noise-induced correction $\xi_1(Z,Y,t)$ to the interface shape [the profile is described by the relation $X=X_0(Z,Y)+\xi_1(Z,Y,t)$] is given by the general relation

$$\xi_1(Z,Y,t)=\int dZ'dY'\int_{-\infty}^{t}dt'G(Z,Y,t,Z',Y',t')\,\eta(Z',Y',t'), \tag{10}$$

where η is a stochastic field of the noise at the interface. Formally, η is the inhomogeneous term in the linear equation $L\xi_1=\eta$, where L is a linear operator which has the local spectrum (5) and the Green's function G.

The appropriate procedure for introducing thermal noise was described in detail by Langer.[7] Following this procedure, we find the root-mean-square amplitude for the side branches generated by thermal fluctuations

$$\langle \xi_1^2(Z,Y)\rangle^{1/2} \sim \bar{Q}\ \exp\left\{\frac{2(5/3)^{9/10}}{3\sqrt{3\sigma}}|Z|^{2/5}\left[1-\frac{9}{4}\left(\frac{3}{5}\right)^{4/5}(\sqrt{1-i/9}-1)\ \frac{Y^2}{|Z|^{4/5}}\right]\right\}, \quad (11)$$

where the fluctuation strength $\bar{Q}$ is given in Ref. 7, and $\bar{Q}^2=2k_BT^2c_pD/(L^2 v\rho^4)$. An estimate for the double-point correlation function at the points $(Z_1,Y=0)$, $(Z_2,Y=0)$ gives for $Z_1 \simeq Z_2 \simeq Z$

$$\langle \xi_1(Z_1,0)\xi_1(Z_2,0)\rangle$$

$$=\langle \xi_1^2(Z_1,0)\rangle^{1/2}\langle \xi_1^2(Z_2,0)\rangle^{1/2}\cdot\cos\left[\frac{2\pi(Z_1-Z_2)}{\lambda}\right]\exp\left[-\frac{(Z_1-Z_2)^2}{2l_c^2}\right], \quad (12)$$

where

$$l_c^2=4\left(\frac{5}{3}\right)^{3/10}\sqrt{3\sigma}|Z|^{4/5}, \quad \lambda=2\pi\left(\frac{3}{5}\right)^{3/10}\sqrt{3\sigma}|Z|^{1/5}. \quad (13)$$

Equation (11) describes an increase in the amplitude with increasing distance from the tip $|Z|$. This amplitude increases exponentially as a function of $(|Z|^{2/5}/\sigma^{1/2})$. At a fixed distance, $|Z|=\text{const}$, the amplitude decays slightly and oscillates with Y. The important result is that the amplitude of the side branches for the anisotropic needle increases faster than for the axisymmetric paraboloid shape. In the latter case it increases exponentially as a function of[7] $(|Z|^{1/4}/\sigma^{1/2})$. We think that this effect can resolve the puzzle that experimentally observed side branches have much larger amplitudes than can be explained by thermal noise in the framework of the axisymmetric approach.[7] Agreement with experiment would require at least one more order of magnitude in the exponential amplification factor. Indeed, we find that for experimental values of $\sigma=0.02$ and $|Z|$, where the first clear side branches can be seen,[8] the ratio of the amplification factors for the actual anisotropic shape and the parabolic shape is

$$\exp(S_{\text{anis.}})/\exp(S_{\text{parab.}}) \simeq \begin{cases} 7 & \text{for} \quad |Z|=7 \\ 11 & \text{for} \quad |Z|=9. \end{cases}$$

The correlation length (or the width of the wave packet) l_c and the side branch spacing λ predicted by (13) depend on the distance from the tip $|Z|$. These dependences are slightly different from those predicted by the axisymmetric method,[7] but the difference is not as crucial as the difference between the amplitudes which increase exponentially with $|Z|$. For example, at the experimentally relevant distances $|Z|=(7-9)$, where the first clear side branches can be seen, the spacing predicted by (13) is $\lambda \simeq 2.0$, which is in approximate agreement with the experimental observations and with the spacing predicted by the axisymmetric method.[7]

Far from the tip the side branching deformations grow out of the linear regime and eventually start to behave like dendrites themselves. It is clear that the branches begin to grow as free, steady-state dendrites only at a distance from the tip on the order of the diffusion length, which in turn is much larger than the tip radius ρ in the limit of small P. This means that there exists a large range of Z, $1 \ll |Z| \ll 1/P$, where the side branches grow in a strongly nonlinear regime, but do not yet behave as free dendrites. We can think of some fractal object in which the length and thickness of the dendrites and the distance between them increase according to certain power laws with the distance from the tip $|Z|$. The dendrites in this object interact due to the competition in the common diffusion field. Some of them die and some continue to grow in the direction prescribed by the anisotropy. This competition leads to a coarsening of the structure in such a way that the distance between the survived dendrites, $\lambda(Z)$, is adjusted to be of the same order of magnitude as the length of the dendrites, $l(Z)$. The scaling arguments similar to those of Ref. 12 give $\lambda(Z) \sim l(Z) \sim |Z|$. The whole dendritic structure with side branches looks like a fractal object on the scale smaller than the diffusion length and as a compact object on the scale larger than the diffusion length.[13] The mean density of a solid phase in the compact structure is equal to the undercooling Δ.

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