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SIDEBRANCHING IN THE THREE DIMENSIONAL DENDRITIC GROWTH

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We consider the time-dependent behavior of sidebranching deformations taking into account the actual nonaxisymmetric shape of the needle crystal. It is found that the amplitude of the deformation grows faster than for the axisymmetric paraboloid shape of the needle. We argue that this effect can resolve the puzzle that experimentally observed sidebranches have much larger amplitudes than can be explained by thermal noise in the framework of the axisymmetric approach. The coarsening behavior of sidebranches in the nonlinear regime is shortly discussed.

We study the problem of a free dendrite growing in 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 the latent heat and c_p the specific heat. The temperature field satisfies the diffusion equation with the interface moving with normal velocity v_n and acting as a source of magnitude $v_n L/c_p$. Together with the Gibbs–Thomson condition at the interface, it leads to a rather complicated integral-differential evolution equation.

The steady-state version of this problem has been discussed in Refs.[2, 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 reads for small Δ: $P(\Delta) = -\Delta/\ln\Delta$. The stability parameter $\sigma \equiv d_0/(P\rho) = \sigma^*(\alpha)$, 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 small α. These two relations for P and σ determine both v and ρ. The interface shape in the tip region is close to the Ivantsov paraboloid and can be described by equation $z(r,\phi) = -r^2/2 + \sum A_m r^m \cos(m\phi)$, with the amplitudes A_m given by the $3D$ selection theory [2] (we measure the 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 small Δ, not too far from the tip, this shape can be described as [3]

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

where the tip position of the arm $x_{tip}(z) = (5|z|/3)^{3/5}(\sigma_2^*/\sigma^*)^{1/5}$. 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 sidebranches necessitates the solution of a time dependent problem for the perturbation about this missile-shaped steady-state crystal

$z = \zeta_o(x,y)$. Langer and coworkers [5, 6, 7] suggested that dendritic sidebranches might be generated by selective amplification of very small, noisy perturbation near the tip of a growing needle crystal. It appeared that realistic sidebranching behavior might be produced by purely thermal fluctuations in the solidifying material. The sidebranching deformation is described in [7] as a small (linear) perturbation moving on a cylindrically symmetric needle crystal (Ivantsov paraboloid [4]). The noise-induced wave packets generated in the tip region grow in amplitude, spread and stretch as they move down the sides of the dendrite producing a train of sidebranches. 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 available experimental observations [8, 9], but experimentally observed sidebranches have much larger amplitudes than explicable by thermal noise in the framework of the axisymmetric approach [7]. It means that either the thermal fluctuation strength turns out not to be quite adequate to produce visible sidebranching deformations, or agreement with experiment would require at least one more order of magnitude in the exponential amplification factor.

The main aim of this paper is to describe the sidebranching problem taking into account the actual nonaxisymmetric shape of the needle crystal, defined by eq.(1). We will show that, for this nonaxisymmetric shape, the perturbations grow faster than for the axisymmetric one. This effect allows to remove the mentioned discrepancy between the theory and experiment.

As in the Ref.[7], we assume the perturbation to be 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

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

Here the functional integration is performed over all the trajectories $x(\tau)$, $y(\tau)$, $k_x(\tau)$, $k_y(\tau)$ which start at the point $x = X', y = Y'$ at $\tau = t'$ and come 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 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 steepest descent method, where the Green's function behavior is determined by the extremal trajectory 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 just $G \sim \exp(S_{ext})$ and the problem is reduced to the solution of the Hamilton equations for the given Hamilton's function $\Omega(x, y, k_x, k_y)$.

The important point is that the local dispersion relation for this solidification problem is just the well-known local Mullins–Sekerka spectrum. Let us replace the interface of the needle crystal in the vicinity of its arbitrary point $x, y, z = \zeta_o(x, y)$ by a piece of its 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

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

Here Θ is the angle between the z–axis and the local normal $\hat{n}$; k_s and k_u are components of a wavevector along $\hat{s}$ and $\hat{u}$; $\hat{s}$ 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 plane (n, z).

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. close to 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

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

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 also small in the region of small y. For small y and k_y, we can expand the function Ω to the second-order terms:

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

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 $\mid z \mid \gg 1$) the following equations

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

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

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

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

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

We would like to find the optimal trajectory, that is four unknown functions $x(\tau)$, $y(\tau)$, $k_x(\tau)$, $k_y(\tau)$, which are governed by eqs.(4), (7), (8) and by four boundary conditions: $x(0) = X' = 0$, $y(0) = Y' = 0$, $x(t) = X$, $y(t) = Y$. In order to do this 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

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

$$\left. - \frac{9}{4} \left(\frac{3}{5}\right)^{4/5} \left(\sqrt{1 - i/9} - 1\right) \frac{Y^2}{\mid Z \mid^{4/5}} - \frac{3\alpha}{2} \left(\frac{3}{5}\right)^{7/5} \frac{Y^2(t - \mid Z \mid)}{\mid Z \mid^{7/5}} \right\}, \tag{9}$$

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 a prefactor comes from the functional integration over the space close to 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 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 is described in detail by Langer [7]. Following this procedure, we find the root–mean–squared amplitude for the sidebranches generated by thermal fluctuations

$$< \xi_1^2(Z,Y) >^{1/2} \sim \bar{Q} \exp \left\{ \frac{2(5/3)^{9/10}}{3\sqrt{3\sigma}} \mid Z \mid^{2/5} \left[1 - \right. \right.$$

$$\left. \left. - \frac{9}{4} \left(\frac{3}{5}\right)^{4/5} (\sqrt{1 - i/9} - 1) \frac{Y^2}{\mid Z \mid^{4/5}} \right] \right\}, \tag{11}$$

where the fluctuation strength $\bar{Q}$ is given in ref.[7], $\bar{Q}^2 = 2k_B T^2 c_p D/(L^2 v \rho^4)$. The estimation for the double–point correlation function at the points $(Z_1, Y = 0), (Z_2, Y = 0)$ gives for $Z_1 \simeq Z_2 \simeq Z$

$$< \xi_1(Z_1,0)\, \xi_1(Z_2,0) >= \tag{12}$$

$$=< \xi_1^2(Z_1,0) >^{1/2} < \xi_1^2(Z_2,0) >^{1/2} \cdot \cos \left[\frac{2\pi(Z_1 - Z_2)}{\lambda} \right] \exp \left[- \frac{(Z_1 - Z_2)^2}{2\ell_c^2} \right],$$

where

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

Eq.(11) describes an increase in the amplitude with the growth of the distance from the tip $|Z|$. This amplitude grows exponentially as a function of $(|Z|^{2/5}/\sigma^{1/2})$. At a fixed distance, $|Z| = \text{const.}$, the amplitude slightly decays and oscillates with Y. The important result is that the amplitude of the sidebranches for the anisotropic needle grows faster than for the axisymmetric paraboloid shape. In the latter case it grows exponentially as a function of $(|Z|^{1/4}/\sigma^{1/2})$ [7]. We think that this effect can resolve the puzzle that experimentally observed sidebranches 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 sidebranches can be seen [8], the ratio between the amplification factors for the actual anisotropic shape and the parabolic shape is

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

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

Far down from the tip the sidebranching deformations grow out of the linear regime and eventually start to behave like dendrites themselves. It is clear that the branches start to grow as free steady-state dendrites only at the distances from the tip which is of the order of the diffusion length which, in turn, is much larger than the tip radius ρ in the limit of small P. It means that there exists the large range of Z, $1 \ll |Z| \ll 1/P$, where the sidebranches grow already in the strongly nonlinear regime, but they do not behave as free dendrites yet. We can think of some fractal object where the length and thickness of the dendrites and the distance between them increase according to some 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 the 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 sidebranches 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 undercooling Δ.

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