Spectrum of growth rates of an isolated dendrite

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An analysis is made of a mechanism which determines the velocity and direction of the growth of a two-dimensional dendrite and which involves an anisotropy of the surface tension. The growth rate is found in the limit of a slight anisotropy, and it is shown that the dendrite grows in the direction of the lowest surface tension.

The crystallization of a supercooled melt may be regarded as a typical problem of structure formation. Steady-state solutions for an isolated two-dimensional dendrite, for the problem incorporating thermal conductivity and heat evolution at the interface, are described by the family of parabolas $y = -x^2/2\rho$, and the growth rate satisfies[1] $v \propto 1/\rho$. The dendrite shape observed experimentally is indeed very nearly a parabola,[2] but the parabola parameter ρ and the velocity v are determined unambiguously by the growth conditions. In an attempt to find a mechanism which selects solutions with a definite velocity it turned out that if there is a nonzero isotropic surface tension at the interface, there are no steady-state solutions at all. A solution of the problem has been found by allowing an anisotropy of the surface tension. The solution consists of the assertion (made on the basis of numerical calculations[3,4]) that in this case there is a discrete spectrum of rates, and the only solution which is stable is that which corresponds to the maximum rate.

A qualitative analysis of the growth equations has shown that at small values of the anisotropy parameter α and of the supercooling parameter Δ the growth rate has the behavior[5] $v \propto \Delta^4 \alpha^{7/4}$. In the present letter we derive an analytic theory for the spectrum of velocities of the crystallization front.

The shape of an isolated dendrite in steady-state growth is described by the equation

$$\Delta + (d_0/\rho) k[y(x)] = (p/\pi) \int_{-\infty}^{\infty} \exp[p[y(x') - y(x)]\} K_0 \times \{p\sqrt{(x - x')^2 + [y(x) - y(x')]^2}\} dx' , \quad (1)$$

where K_0 is the modified Bessel function, all lengths are expressed in units of ρ, $\Delta = (T_m - T_0)c_p L^{-1}$ is the dimensionless supercooling (T_m is the melting point, T_0 is the temperature of the melt, c_p is the specific heat, and L is the heat of fusion), $d_0 = \gamma T_m c_p L^{-2}$ is the capillary length (γ is the surface tension), $k[y(x)] = y''/(1 + y'^2)^{3/2}$ is the curvature of the front, p is the Peclet number, $p = v\rho/2D$, and D is the thermal diffusivity (the thermal characteristics of the phases are identical). In the absence of a surface tension ($d_0 = 0$) a solution of (1) is the parabola[1] $y(x) = -x^2/2$ and $\Delta = 2\sqrt{p}e^{p}\int_{\sqrt{p}}^{\infty} e^{-y^2}dy$. As in Ref. 5, we introduce a nonzero, slightly anisotropic surface tension $d_0 = \bar{d}_0(1-\alpha\cos 4\theta)$, where $\tan\theta = dy/dx$ and $\alpha \ll 1$. The shape of the front is distorted, $y(x) = y_0(x) + \zeta(x)$, and the linear equation for $\zeta(x)$ takes the following form[5] in the limit $\sigma = \bar{d}_0/p\rho \ll 1$, $\Delta \ll 1$:

$$\sigma\zeta'' - \frac{3\sigma\zeta'}{(1+x^2)} - \frac{(1+x^2)^{3/2}}{2\pi A(x)}\int_{-\infty}^{\infty}\frac{x+x'}{x-x'}\,\frac{\zeta(x)-\zeta(x')}{1+(x+x')^2/4}dx' = \sigma\ , \tag{2}$$

where $A(x) \simeq 1 + 8\alpha x^2/(1+x^2)^2$. The integral in (2) is taken over the residues if the function $\zeta(x)$ is split up into the terms $\zeta_+(x)$ and $\zeta_-(x)$, which are analytic in the upper and lower half-planes of the complex variable x. Ignoring the derivatives, we find a Wiener-Hopf equation, $(x+i)\times[\zeta_+(x) - \zeta_+(-x+2i)] - (x-i)[\zeta_-(x) - \zeta_-(-x-2i)] = i\sigma A(x)(1+x^2)^{-1/2}$. Its solution satisfies $\zeta(x)$ and has singularities at the points $x = \pm i$. The terms with derivatives in (2) are important in the region $|x\pm i| \sim \alpha^{1/2}$ and play the role of a singular perturbation.

Let us examine the neighborhood of the point $x = i$ in more detail. Near it, it is sufficient to consider only $\zeta_-(x)$; after making the replacements $x = i(1-\alpha^{1/2}t)$, $\psi(t) = \alpha^{-1}t^{-3/4}\zeta_-[x(t)]$, we find the following equation for ψ:

$$d^2\psi/dt^2 + P^2(t)\psi = -1/(4t^{3/4}), \tag{3}$$

where

$$P^2(t) = -[2^{1/2}\lambda t^{7/2}/(t^2-2) + 21/(16t^2)]. \tag{4}$$

The small parameters σ and α are eliminated by introducing $\lambda = \alpha^{7/4}/\sigma$. Equation (3) is an inhomogeneous Schrödinger equation, defined in the plane of the complex variable t with a cut along the semiaxis $(-\infty, \sqrt{2})$. Its solution for $1 \ll |t| \ll \alpha^{-1/2}$ should have the asymptotic behavior $\psi = t^{-9/4}/(2^{5/2}\lambda)$, which is found by ignoring the derivative in (3) and which insures a joining with the solution of the Weiner-Hopf equation in the region $1 \gg |x-i| \gg \alpha^{1/2}$.

At $|t| \gg 1$ the semiclassical solutions of the homogeneous version of Eq. (3) are of the form $\psi_{1,2} \propto t^{-3/8}\exp[\pm(4+7)2^{1/4}\lambda^{1/2}t^{7/4}]$. The boundary conditions formulated above are therefore equivalent to the condition that the solution of (3) is finite on the rays $\arg t = 0, \pm 4\pi/7$. These conditions can be satisfied at only certain values of the parameter λ, and these particular values determine the spectrum of velocities of an isolated dendrite. Let us calculate the λ spectrum in the semiclassical approximation, making the formal assumption $\lambda \gg 1$. For this purpose, it is sufficient to solve Eq. (3) (first) in that neighborhood of the point $t = 0$ in which the branch point and the second-order pole at $t = 0$ and the turning point at $t_1 \simeq 2^{-7/11}(21/\lambda)^{2/11} \ll 1$ lie and (second) near the turning point $t_2 = \sqrt{2}$, where $P^2(t)$ has a pole. Finally, we join these solutions, making use of the semiclassical asymptotic results which hold between turning points t_1 and $t_2 = \sqrt{2}$.

Near the point $t=0$, a particular solution of inhomogeneous equation (3) can be written in the form $\psi_{in} \propto t^{5/4}\varphi(\lambda t^{11/2})$, where $\varphi(z)$ is a power series in z. Solutions of the homogeneous equation are given by the functions $t^{1/2}J_{\pm 5/11}(2^{7/4}\lambda^{1/2}t^{11/4}/11)$, where J_ν is a Bessel function. The constants in the homogeneous solutions are determined by the condition that the total solution must decay on the rays $\arg t = 2\pi/11$, $6\pi/11$ (as we go from $|t| \ll 1$ to $|t| \gg 1$, these rays correspond to the rays $\arg t = 0$ and $\arg t = 4\pi/7$). On the upper bank of the (t_1, t_2) cut we then have

$$\psi \propto P^{-1/2}(t)\exp\left\{i\left[\int_{t_1}^{t} P(t')dt' - 5\pi/44\right]\right\} \tag{5}$$

Near the turning point, $t_2 = \sqrt{2}$, a solution of (3), which is real and which decays as $t \to \infty$, is found by using the modified Bessel function $K_1(z)$. On the upper bank of the cut this solution is described by

$$\psi \propto P^{-1/2}(t)\exp\left\{-i\left[\int_{t}^{\sqrt{2}} P(t')dt' + 3\pi/4\right]\right\}. \tag{6}$$

A solution for $\psi(t)$ in the lower t half-plane is found by taking the complex conjugate of this solution. Joining solutions (5) and (6), we find a condition on the semiclassical λ spectrum:

$$\int_{t_1}^{\sqrt{2}} P(t)dt = \pi(n + 4/11); \qquad n = 0, 1, 2 \ldots , \tag{7}$$

where t_1 is the root of the equation $P(t) = 0$. In the case $n \gg 1$ we find from (7) $\lambda_n \simeq 3.0n^2$.

The growth velocity at $\Delta \ll 1$ is given by

$$v_n = 2D\Delta^4 \alpha^{7/4}/(\pi^2 \lambda_n d_0) . \tag{8}$$

The spectrum λ_n was also found through a numerical integration of Eq. (3); the results are $\lambda_0 \approx 0.48$ (0.79), $\lambda_1 \approx 5.8$ (7.3), $\lambda_2 \approx 17.5$ (19.7) and $\lambda_3 \approx 34.4$ (38.5), where the numbers in parentheses are the values found from (7). According to the numerical results,[4] only the solution with λ_0 should be stable and therefore experimentally observable.

Equation (2) has been written under the assumptions[5] $p, \Delta \ll 1$. Our analysis shows that, because of the small size of the singular regions near $x = \pm i$, the linear equation for the singular part of the function $\zeta(x)$ will be applicable as long as the condition $p \ll \alpha^{-1/2}$ holds, i.e., even at $p \sim 1$. In this case, we cannot find a solution for the regular correction to the shape of the front, but it is sufficient to use the exact relationship between p and Δ to find the dependence of the front velocity on the supercooling. It follows that an anisotropy of the surface tension, α, is the only small parameter which can be used in this problem.

We calculated the growth velocity of a dendrite oriented in the direction of the lowest surface tension. If these directions differ by an angle φ, we need to replace (t^2-2) by $[t^2-2\exp(-4i\varphi)]$ in expression (4) for $P^2(t)$. It turns out that a solution with the given boundary conditions is possible only in the case $\varphi=0$. A dendrite can therefore grow only in the direction of the lowest surface tension. This conclusion is of a general nature and should not be directly related to the circumstance that the parameter α is small (we have made use of this circumstance in the present calculations). An experimental verification of this conclusion would be strong evidence that the mechanism discussed is responsible for determining the velocities and directions of the growth of a two-dimensional dendrite.

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