Growth of a needle-shaped crystal in a channel

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An analysis is made of the growth of a dendrite in a channel under the influence of an anisotropic surface tension. A preferred growth rate is found by considering the surface energy of a singular perturbation in the equation for the shape of a growing dendrite. It is shown that the dependence of the growth rate v on the supercooling Δ and on the channel width λ is double-valued. In the limit $\lambda \to \infty$ the upper branch of the $v(\Delta)$ dependence tends to a curve representing growth of a free dendrite. It is shown that growth is not possible for all the values of the supercooling, but only beginning from a certain minimum value Δ_m. An analysis is made of the selection of the growth rate of a free dendrite due to an anisotropic kinetics of the crystallization front. If the surface tension is ignored, the growth rate obeys $v \propto \nu^{5/4}\Delta^2$ (ν is the anisotropy parameter of the kinetic growth coefficent; $\nu, \Delta \ll 1$).

1. INTRODUCTION

The problem of selection and formation of a structure in nonlinear systems is currently the subject of intensive investigations. Problems of this kind are encountered, in particular, in studies of crystallization[1] and viscous flow.[2] The most significant progress has been made in solving the Saffman-Taylor problem (for a review see Ref. 2 and the literature cited there) and the problem of growth of a free dendrite.[3–10]

An analysis of heat transport during the growth of a free needle-shaped dendrite in a supercooled melt shows that the surface of a growing needle is parabolic and the axial growth rate v is related to the radius of curvature ρ of the tip and to the dimensionless supercooling Δ by an expression of the type

$$v\rho/2D=f(\Delta),$$

where D is the thermal diffusivity of the melt.[11] This dependence does not allow us to determine separately the values of v and ρ. However, the experimental results[12,13] show that for a given supercooling there are unique values of v and ρ. It has been shown numerically[3,4,6,9] and analytically[5,7,8,10] that the parameters v and ρ (and also the growth direction[10]) are selected allowing for the anisotropic surface tension. The preferred velocity in the two-dimensional case is $v \propto \alpha^{7/4}\Delta^4$ ($\alpha \ll 1$ is the anisotropy parameter and it is assumed that $\Delta \ll 1$).

An analysis of the growth of a needle-shaped crystal in a channel (Fig. 1) is faced with an analogous problem of selection. If the channel walls are thermally insulating, this problem is equivalent to one of growth of a periodic cellular structure. (The simplest experimental realization of such growth in a channel is observed not in the case of crystallization of a melt but during growth from a supersaturated solution in a capillary.) The problem of selection of the preferred growth rate in a channel has been studied less than that of crystallization of a free dendrite. It is shown in Ref. 14 in that the limit corresponding to the Péclet number $p = v\lambda/2D \to 0$ (v is the steady-state growth rate and λ is the channel width) this problem is formally equivalent to the Saffman-Taylor problem. The following results are obtained in Ref. 15 in the limit $p \to 0$. In the presence of an isotropic surface tension the growth of a crystal is possible only when the dimensionless supercooling obeys $\Delta > 1/2$. and we have $v \propto \lambda^{-2}(\Delta - 1/2)^{-3/2}$ (a similar result was obtained in solving the Saffman-Taylor problem in Ref. 16). When an allowance is made for the surface tension anisotropy, a crystal can grow even when $\Delta < 1/2$. Moreover, it is concluded in Ref. 15 that, as in the problem of a free dendrite, there is a discrete spectrum of growth rates. This spectrun is investigated numerically in Ref. 17 for the case when $\Delta = 1$.

Naturally, in the problem of growth in a channel when the channel width approaches $\lambda \to \infty$ there should be a transition to the case of a free dendrite. However, the growth rate $v(\Delta)$ obtained in Ref. 15 decreases on increase in the supercooling Δ (which is unimportant in the case of crystallization processes) and does not reduce to $v(\Delta)$ for a free dendrite. In considering the growth of a dendrite in a channel we shall obtain below the other branch of the solution giving $v(\Delta)$. For this new branch the growth rate is higher and it increases with supercooling Δ. It is this branch that reduces in the limit $\lambda \to \infty$ to the solution corresponding to a free dendrite. We shall show that growth is possible not for all values of the supercooling, but only beginning from a certain minimum Δ_m, which decreases on increase in the channel width (this conclusion is also reached in Ref. 15).

An analytic theory of the spectrum of the growth rates of an isolated dendrite is developed in Refs. 5, 7, and 10. In these theories an allowance is made for the circumstance that the surface tension plays the role of a singular perturbation. The preferred growth rate is selected on the basis of the condition of solvability of the problem in the presence of this singular perturbation. Different solvability conditions are given by different authors. In Ref. 5 this condition is presented in an integral form as the condition of existence of the solution of an inhomogeneous linear integrodifferential equation for the correction to the shape of the interface. In Ref. 7 a nonlinear differential equation for the correction to the shape near a singularity is discussed. The solvability condition used in Ref. 10 is the condition for a finite solution of an inhomogeneous linear differential equation determined in the complex plane near a singularity. All these approaches give literally identical expressions for the growth rate of a free dendrite. The numerical spectral parameter is found in Refs. 7 and 10. Analytic expressions for this parameter in the

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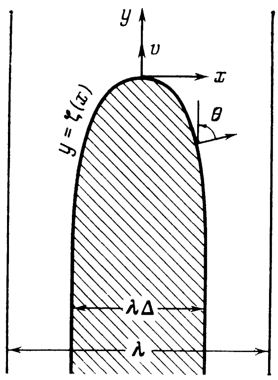

FIG. 1. Schematic representation of the growth of a needle shaped crystal in a channel.

far part of the spectrum, obtained in the WKB approximation, are in fact identical in Refs. 7 and 10.

We shall analyze the growth of a needle-shaped crystal in a two-dimensional channel, adopting the approach described in Ref. 10. Moreover, we shall consider the selection of the growth rate of a free dendrite due to an anisotropic kinetics of the crystallization front. When this effect is allowed for, there is a preferred growth rate even when the surface tension is ignored completely. We shall show that in the two-dimensional case the preferred rate is $v \propto \nu^{5/4}\Delta^2$ if $\Delta \ll 1$ (here, ν is the anisotropy parameter of surface kinetics) and a needle grows in the direction of the maximum kinetic coefficient.

2. EQUATIONS FOR THE GROWTH OF A NEEDLE IN A CHANNEL

We shall consider the two-dimensional problem of the growth of a needle-shaped crystal in a channel (Fig. 1). The steady-state distribution of the temperature in the melt and in the growing crystal is described by the heat conduction equation

$$D\nabla^2 T + v\frac{\partial T}{\partial y} = 0, \tag{2.1}$$

where v is the growth rate along the y axis (Fig. 1). Far from the front in the limit $y \to \infty$ the melt is supercooled and its temperature T_0 is below the melting point T_m. If the channel walls are thermally insulating and are located at $x = \pm\lambda/2$, we have $\partial T/\partial x = 0$. The following heat balance equation is satisfied at the phase boundary $y = \zeta(x)$:

$$c_p D[\mathbf{n}\nabla T_L - \mathbf{n}\nabla T_S] = -Lv_n. \tag{2.2}$$

Here, T_L and T_S are the temperatures of the melt and solid (crystal), respectively; c_p and D are the specific heat and the thermal diffusivity assumed to be the same for both phases; L is the latent heat of crystallization; $\mathbf{n}$ is a unit vector of the outward normal to the phase boundary; v_n is the growth rate along the normal. Allowing for the Gibbs-Thomson effect on the growth kinetics, we find that the temperature at the crystallization front is

$$T(x, \zeta(x)) = T_m + T_m(\tilde{\gamma}(\theta)/L)K(x) - v_n/\beta(\theta). \tag{2.3}$$

Here, $\tilde{\gamma}(\theta) = \gamma(\theta) + d^2\gamma(\theta)/d\theta^2$, where $\gamma(\theta)$ is the anisotropic surface energy; θ is the angle between the normal and the y axis (Fig. 1); $\beta(\theta)$ is an anisotropic kinetic growth coefficient;

$$K(x) = \zeta''/[1+(\zeta')^2]^{3/2}, \quad v_n = v/[1+(\zeta')^2]^{1/2} \tag{2.4}$$

are the curvature of the crystallization front and the normal growth rate.

Using the Green function for Eqs. (2.1) and (2.2), and applying the condition (2.3), we obtain the following integrodifferential equation for the shape of the front:

$$\Delta + d_0(\theta)k(x)/\lambda - v_n/w_k(\theta) = \frac{p}{\pi}\sum_{n=-\infty}^{\infty}\int_{-\Delta/2}^{\Delta/2} dx' \exp\{p[\zeta(x')-\zeta(x)]\} \times K_0\{p[(x-x'+n)^2+(\zeta(x)-\zeta(x'))^2]^{1/2}\}$$
$$= \sum_{n=-\infty}^{\infty}\int_{-\Delta/2}^{\Delta/2} \frac{dx' \cos 2\pi n(x-x')}{[1+(2\pi n/p)^2]^{1/2}} \times \exp\{-p[\zeta(x)-\zeta(x')] - p[1+(2\pi n/p)^2]^{1/2}|\zeta(x)-\zeta(x')|\}. \tag{2.5}$$

Here, $K_0(z)$ is a Macdonald function. In Eq. (2.5) all the lengths (x, x', ζ) are measured in units of the channel width λ;

$$\Delta = (T_m - T_0)c_p/L$$

is the dimensionless value of the supercooling; $p = v\lambda/2D$ is the Péclet number;

$$d_0(\theta) = \tilde{\gamma}(\theta)T_m c_p/L^2$$

is the capillary length; $k(x)$ is the dimensionless curvature; the quantity

$$w_k(\theta) = \beta(\theta)L/c_p$$

has the dimensions of velocity. As in Refs. 3 and 5, the anisotropy of the capillary length is described by

$$d_0(\theta) = \bar{d}_0(1-\alpha\cos 4\theta) \equiv \bar{d}_0 A_\alpha(\theta), \ \mathrm{tg}\,\theta = \zeta'(x) \tag{2.6a}$$

and the anisotropy of the kinetic growth coefficient is

$$1/w_k(\theta) = (1/\bar{w}_k)(1-\nu\cos 4\theta) \equiv (1/\bar{w}_k)A_\nu(\theta). \tag{2.6b}$$

We shall assume that the anisotropy parameters α and ν are small. Integration in the system (2.5) is carried out with respect to the the coordinate x' inside a growing needle. As shown in Ref. 14, the needle width depends on the supercooling and is equal to $\lambda\Delta$ for $\Delta \leqslant 1$ (Fig. 1). In the absence of a surface tension ($d_0 = 0$) and for infinitely fast growth kinetics ($w_k \to \infty$), the surface of the phase boundary is isothermal, whereas the shape of the boundary is $y = \zeta_0(x)$ and this, as shown in Ref. 14, is described in the limit $p \to 0$ by the familiar Saffman-Taylor equation[18]:

$$\zeta_0(x) = \frac{(1-\Delta)}{\pi}\ln\cos\frac{\pi x}{\Delta}. \tag{2.7}$$

In the limit $x \to \Delta/2$, the value of ζ_0 tends to $-\infty$ and Eq. (2.7) yields

$$(\Delta/2-x)\sim\exp[\pi\zeta_0/(1-\Delta)].$$

The exponential form of the asymptote is retained also in the general case:

$$(\Delta/2-x)\sim\exp(s\zeta), \tag{2.8}$$

where the parameter s is governed by the transcendental equation

$$\left(\frac{d_0 s}{\lambda}+\frac{v}{w_k}\right)\left\{\mathrm{tg}\left[\frac{\Delta}{2}(s(s+2p))^{1/2}\right]+\mathrm{tg}\left[\frac{(1-\Delta)}{2}(s(s+2p))^{1/2}\right]\right\}=2p/(s(s+2p))^{1/2}. \tag{2.9}$$

Here, d_0 and w_k are calculated as the values of $d_0(\theta)$ and $w_k(\theta)$ corresponding to $\theta=\pi/2$. Equation (2.9) can be obtained, by analogy with Ref. 15, from Eqs. (2.1)–(2.3) if the asymptote of the temperature field is sought in the exponential form:

$$T_{L(S)}(x,y)=T_m-B_{L(S)}(x)\exp(sy).$$

A convenient model for the description of the shape of the crystallization front is the general expression similar to Eq. (2.7):

$$\zeta_0(x)=\frac{1}{s}\ln\cos\frac{\pi x}{\Delta}, \tag{2.10}$$

where s corresponds to the exact asymptotic equation (2.9). For example, such a shape of the crystallization front was used as the zeroth approximation in the numerical solution reported in Ref. 15. When allowance is made for the effects of the surface energy and kinetics in Eq. (2.5), it is found that the shape of the front differs from $\zeta_0(x)$:

$$\zeta(x)=\zeta_0(x)+\zeta_1(x). \tag{2.11}$$

The linear equation for $\zeta_1(x)$ in the limit of small Péclet numbers, $p\ll 1$, is

$$\frac{[1+(\zeta_0')^2]^{1/2}}{A_\alpha}\int_{-\Delta/2}^{\Delta/2}dx'[\zeta_1(x)-\zeta_1(x')]\times\left\{1+\frac{\mathrm{sh}\,2\pi[\zeta_0(x)-\zeta_0(x')]}{\mathrm{ch}\,2\pi[\zeta_0(x)-\zeta_0(x')]-\cos 2\pi(x-x')}\right\}+\sigma\zeta_1''-3\sigma\frac{\zeta_0''\zeta_0'}{1+(\zeta_0')^2}\zeta_1'+\frac{A_v\zeta_0'}{A_\alpha p_k}\zeta_1'=-\sigma\zeta_0''+\frac{A_v}{A_\alpha p_k}[1+(\zeta_0')^2]. \tag{2.12}$$

Here,

$$\sigma=\bar{d}_0/p\lambda,\quad p_k=\bar{w}_k\lambda/2D,\quad A_\alpha=1-\alpha+8\alpha(\zeta_0')^2/[1+(\zeta_0')^2]^2, \tag{2.13}$$

whereas A_v is given by the same expression as A_α but with α replaced by v.

The regular correction $\zeta_1(x)$ is of the order of σ (or $1/p_k$) and can be found as a solution of Eq. (2.12) ignoring the derivatives of the function ζ_1. It is clear from Eq. (2.12) that this regular correction has a singularity in the complex plane at $\zeta_0'(x)=\pm i$. The derivatives must be allowed for in the vicinity of such singularities. The singular perturbation associated with the derivatives has the effect that Eq. (2.12) is solvable only for certain values of the parameters. The final result is the spectrum of permissible values of the growth rate.

3. EQUATION NEAR A SINGULARITY

We shall consider Eq. (2.12) near the singularity $\zeta_0'=i$, where the contribution of the derivatives is important. Near this singularity Eq. (2.12) becomes purely differential. This is due to the fact that the integral term containing $\zeta_1(x')$ is of the order of σ, i.e., it is of the same order as the values of the function $\zeta_1(x)$ elsewhere outside the singularity. On the other hand, near the singularity we find that $\zeta_1(x)\gg\sigma$, because—as demonstrated by the subsequent analysis—we find that $\zeta_1(x)$ is proportional to a lower power of the small parameter σ. Therefore, the integral term with $\zeta_1(x')$ can be dropped, which makes the equation in question differential. The coefficient in front of $\zeta_1(x)$ in this equation can be obtained by calculating the corresponding integral in Eq. (2.12) for values of x close to a singularity. This calculation can be carried out by a method similar to that described in Refs. 15 and 10. Use is made simply of the analytic properties of $\zeta_0(x)$ and the integral is calculated from the residues at the poles of the integrand. For x in the vicinity of the singularity $\zeta_0'(x)=i$ there are two closely spaced poles and we therefore obtain

$$\int_{-\Delta/2}^{\Delta/2}dx'\left[1+\frac{\mathrm{sh}\,2\pi(\zeta_0(x)-\zeta_0(x'))}{\mathrm{ch}\,2\pi(\zeta_0(x)-\zeta_0(x'))-\cos 2\pi(x-x')}\right]=\frac{1}{2}. \tag{3.1}$$

Therefore, near this singularity Eq. (2.12) becomes

$$\sigma\zeta_1''-\frac{3\sigma\zeta_0''\zeta_0'}{1+(\zeta_0')^2}\zeta_1'+\frac{A_v\zeta_0'}{A_\alpha p_k}\zeta_1'+\frac{[1+(\zeta_0')^2]^{1/2}}{2A_\alpha}\zeta_1=-\sigma\zeta_0''+\frac{A_v}{A_\alpha p_k}[1+(\zeta_0')^2]. \tag{3.2}$$

Changing from a variable x to a new variable t in accordance with the relationship

$$\zeta_0'(x)=i(1-t), \tag{3.3}$$

we find from Eq. (3.2) allowing for the smallness of t that near the singularity we have

$$\frac{d^2\zeta_1}{dt^2}+\left[\frac{d}{dt}\ln\zeta_0''-\frac{3}{2t}+\frac{A_v}{\sigma A_\alpha p_k\zeta_0''}\right]\frac{d\zeta_1}{dt}-\frac{2^{1/2}t^{3/2}}{\sigma(\zeta_0'')^2A_\alpha}\zeta_1=\frac{1}{\zeta_0''}-\frac{2tA_v}{\sigma p_k(\zeta_0'')^2A_\alpha}. \tag{3.4}$$

Here, $A_{\alpha(v)}$ and ζ_0'' are functions of t. According to Eq. (2.13) (when $\alpha,v\ll 1$), we have

$$A_\alpha\approx 1-2\alpha/t^2,\quad A_v\approx 1-2v/t^2. \tag{3.5}$$

It is assumed that the function ζ_0'' has a simple zero near the singularity, i.e., that it can be represented in the form

$$\zeta_0''=-b(t-\varepsilon),\quad \varepsilon\ll 1. \tag{3.6}$$

In the case of the Saffman-Taylor profile of Eq. (2.7) we find that

$$b\approx 4\pi,\quad \varepsilon\approx 4(\Delta-{}^1/_2)\quad \text{for}\quad (\Delta-{}^1/_2)\ll 1. \tag{3.7}$$

If the profile of the crystallization boundary $\zeta_0(x)$ differs from Eq. (2.7) because of the finite growth rate, the values of b and ε depend also on the Péclet number p.

4. PREFERRED GROWTH RATE IN THE CASE OF INFINITELY FAST KINETICS

In the limit $p_k\to\infty$, Eq. (3.4) can be transformed by the substitution

$$\zeta_1(t)=t^{3/4}\psi(t)/b(t-\varepsilon)^{1/2} \tag{4.1}$$

to the following equation which does not contain the first derivative:

$$\psi''(t)+q^2(t)\psi(t)=-t^{-3/4}(t-\varepsilon)^{-1/2},$$

$$q^2(t)=-\frac{2^{1/2}}{\sigma b^2}\frac{t^{7/2}}{(t-\varepsilon)^2(t^2-2\alpha)}+\frac{1}{4(t-\varepsilon)^2}-\frac{21}{16t^2}+\frac{3}{4t(t-\varepsilon)}. \tag{4.2}$$

The system (4.2) is defined in a complex plane t. The function $q^2(t)$ has the following singularities: at $t=0$ there is branching and a second-order pole; at $t=\varepsilon$ there is a second-order pole; at $t=\pm(2\alpha)^{1/2}$ there are simple poles. The behavior of the solution of the system (4.2) depends strongly on the mutual positions of the poles at $t=\varepsilon$ and $t=(2\alpha)^{1/2}$. We shall consider several limiting cases.

a. Weak anisotropy of the surface energy: $\alpha^{1/2}\ll\varepsilon\ll 1$

In this case if $|t|\gg\alpha^{1/2}$, we find from the system (4.2) that

$$\frac{d^2\varphi(\tau)}{d\tau^2}+\left[-\frac{\mu_\varepsilon\tau^{3/2}}{(\tau-1)^2}+\frac{1}{4(\tau-1)^2}-\frac{21}{16\tau^2}+\frac{3}{4\tau(\tau-1)}\right]\varphi(\tau)$$

$$=\tau^{-3/4}(\tau-1)^{-1/2}, \tag{4.3}$$

where $\varphi=-\varepsilon^{-3/4}\psi$. $\tau=t/\varepsilon$, and the small parameters σ and ε are eliminated by introducing

$$\mu_\varepsilon=2^{1/2}\varepsilon^{3/2}/b^2\sigma. \tag{4.4}$$

The solution of Eq. (4.3) in the case when $|\tau|\gg 1$ can be reduced to the regular solution obtained ignoring the derivative and can thus be matched to the solution in the region far from a singularity. However, this is possible only for certain values of the parameter μ_ε, and it is this which determines the spectrum of permissible growth rates. In fact, the asymptote for the general solution of Eq. (4.3) in the case when $|\tau|\gg 1$ is of the form

$$\varphi\propto\tau^{1/8}\exp(\pm{}^4/_3\mu_\varepsilon{}^{1/2}\tau^{3/4}).$$

This solution rises exponentially along a ray $\arg\tau=0$ and it grows in accordance with a power law oscillating along rays $\arg\tau=\pm 2\pi/3$. The required solution becomes a power-law fall $\varphi\propto\tau^{-3/4}$, which follows from Eq. (4.3) when the derivative is ignored, if this behavior along such three rays is suppressed. The relevant three conditions can be satisfied if we select suitably not only the two integration constants, but also the parameter μ_ε. We shall demonstrate this and find the spectrum of μ_ε in the WKB approximation assuming formally that $\mu_\varepsilon\gg 1$.

Instead of the whole plane of the complex variable τ, we shall consider only the region where $\operatorname{Im}\tau\geqslant 0$ and assume that φ is real for real values of $\tau>1$. Then, in view of the symmetry, the finite nature of the solution φ along a ray $\arg\tau=-2\pi/3$ follows asymptotically from the condition that it is finite along a ray $\arg\tau=2\pi/3$.

The spectrum of μ_ε can be described if we, firstly, solve the equation in the range $|\tau|\ll 1$, satisfying then the boundary condition corresponding to the absence of oscillations along a ray $\arg\tau=2\pi/3$; secondly, we solve the equation for $|\tau-1|\ll 1$, satisfying the requirement of finite and real solution if $\tau>1$; thirdly, we match these solutions in the region $0<\tau<1$. Equation (4.3) for the case described by $|\tau|\ll 1$ is

$$\tau^2\varphi''-({}^{21}/_{16}+\mu_\varepsilon\tau^{7/2})\varphi=-i\tau^{5/4}. \tag{4.5}$$

The general solution of this equation can be written in the form of a series

$$\varphi=C_1\tau^{7/4}\sum_{n=0}^{\infty}\frac{({}^4/_{49})^n\mu_\varepsilon{}^n\tau^{7n/2}}{\Gamma(n+1)\Gamma(n+{}^{12}/_7)}$$

$$+C_2\tau^{-3/4}\sum_{n=0}^{\infty}\frac{({}^4/_{49})^n\mu_\varepsilon{}^n\tau^{7n/2}}{\Gamma(n+1)\Gamma(n+{}^2/_7)}$$

$$-i\Gamma({}^6/_7)\Gamma({}^1/_7)\tau^{5/4}\sum_{n=0}^{\infty}\frac{({}^4/_{49})^n\mu_\varepsilon{}^n\tau^{7n/2}}{\Gamma(n+{}^6/_7)\Gamma(n+{}^{11}/_7)}. \tag{4.6}$$

The first two terms representing the solutions of the homogeneous equation can also be expressed in terms of Bessel functions. A ray $\arg\tau=2\pi/3$ in the range $|\tau|\gg 1$ corresponds to a ray $\arg\tau=6\pi/7$ in the range $|\tau|\ll 1$, but we now have $\mu_\varepsilon^{1/2}|\tau|^{7/4}\gg 1$. Therefore, the constants C_1 and C_2 are determined from the requirement of the absence of oscillations along a ray $\arg\tau=6\pi/7$. We can then use the following expression for the asymptote of the series at for $x\gg 1$:

$$\sum_{n=0}^{\infty}\frac{(\mp 1)^n x^n}{\Gamma(n+\alpha)\Gamma(n+\beta)}\approx\frac{x^{3/4-(\alpha+\beta)/2}}{2\pi^{1/2}}\begin{cases}\cos(2x^{1/2}+{}^3/_4-(\alpha+\beta)/2)\\ \exp(2x^{1/2})\end{cases}$$

After determination of the constants C_1 and C_2, the solution of Eq. (4.6) for real values of τ satisfying $\mu_\varepsilon^{1/2}\tau^{7/4}\gg 1$ has the following asymptote:

$$\varphi\propto -ie^{-4\pi i/7}\tau^{-3/8}\exp({}^4/_7\mu_\varepsilon{}^{1/2}\tau^{7/4}). \tag{4.7}$$

A real numerical factor has been omitted.

We shall now consider Eq. (4.3) in the vicinity of $\tau=1$, $|\tau-1|\ll 1$. We then find from Eq. (4.4) that

$$u^2d^2\varphi/du^2-(\mu_\varepsilon-{}^1/_4)\varphi=u^{3/2},\quad u=\tau-1. \tag{4.8}$$

The solution which is real and decreasing in the range $u>0$ has the form

$$\varphi=Cu^{1/2-\mu_\varepsilon{}^{1/2}} \tag{4.9}$$

with a real constant C. Therefore, in bypassing the point $\tau=1$, i.e., in going over from $u<0$ to $u>0$, an additional phase factor $\exp[\pi i(\mu_\varepsilon^{1/2}-\frac{1}{2})]$ is acquired. Allowing for the phase factor in Eq. (4.7), we find that in the range $u>0$ the function φ contains the following phase factor: $\exp[\pi i(\mu_\varepsilon^{1/2}-\frac{4}{7})]$. Since the function φ should be real, we obtain the following spectrum of μ_ε:

$$\mu_{\varepsilon,n}=(n+4/7)^2,\quad n=0,\ 1,\ldots. \tag{4.10}$$

A similar spectrum is reported in Ref. 7.

Equations (4.10) and (4.4) determine the spectrum of the growth rates. However, in order to find the growth rates, we have to specify the parameters ε and b. We recall that these parameters depend on the shape of the phase boundary $\zeta_0(x)$ found ignoring the surface tension and they determine the behavior of ζ_0'' in the vicinity of a singularity [see Eq. (3.6)]. In the case of the Saffman-Taylor profile of Eq. (2.7) the parameters b and ε are given by Eq. (3.7). Combining these relationships with Eqs. (4.4) and (4.10), we obtain the spectrum of growth rates:

$$V=\frac{2^{1/3}\pi^2\mu_{\varepsilon,n}}{\Lambda^2(\Delta-1/2)^{3/2}}, \tag{4.11}$$

where $V=v\bar{d}_0/2D$ and $\Lambda=\lambda/d_0$ represent, respectively, the dimensionless growth rate and the dimensionless channel width. These results are obtained on the assumption that

$$\alpha^{1/2}\ll(\Delta-1/2)\ll1,$$

using the Saffman-Taylor solution which is valid if $p\ll1$. All these conditions can be combined in the following inequalities:

$$\max\{\alpha^{1/2},\ \Lambda^{-2/5}\}\ll(\Delta-1/2)\ll1. \tag{4.12}$$

A result similar to Eq. (4.11) is obtained in Ref. 16 in the specific case of the Saffman-Taylor problem and it is reformulated for the case of crystallization in Ref. 15. It should be noted that, according to Eq. (4.11), the growth rate falls on increase in the supercooling Δ. This is in general unimportant for the crystallization kinetics. However, it was found that in addition to this branch of the solution, there is always a second branch on which the growth rate is higher and increases with the supercooling. The existence of this branch is associated with the fact that the profile $\zeta_0(x)$ differs from the Saffman-Taylor profile because of the finite growth rate. It is this branch that describes the transition to the growth of an isolated dendrite in the limit $\lambda\to\infty$ and seems to us physically realizable, whereas the lower branch is clearly unstable. In an investigation of this new branch and of the qualitative behavior in a wide range of parameters we need to know the needle shape $\zeta_0(x)$ in situations other than that covered by the Saffman-Taylor approximation. Since the exact solution $\zeta_0(x)$ is not available for arbitrary values of the parameters, we shall describe $\zeta_0(x)$ by a model expression of Eq. (2.10), which reduces to the exact expression (2.7) in the limit $p\to0$ and which gives an accurate asymptote for the shape of the boundary at $x\to\pm\Delta/2$. We shall find the parameters ε and b for this model shape:

$$\varepsilon=[1-(\pi/\Delta s)^2]/2,\quad b=2s, \tag{4.13}$$

where

$$s=[p^2+\pi^2/(1-\Delta)^2]^{1/2}-p \tag{4.14}$$

and it satisfies Eq. (2.9) for $d_0=0$ and $w_k=\infty$. Using these relationships and Eq. (4.4), we find the dependence $V(\Delta,\Lambda)$ which can be repesented conveniently in the following parametric form:

$$\Lambda(s)=16\mu_{\varepsilon,n}s^6/[s^2-\pi^2/\Delta^2]^{3/2}[\pi^2/(1-\Delta)^2-s^2], \tag{4.15a}$$

$$V(s)=[\pi^2/(1-\Delta)^2-s^2]/2s\Lambda(s), \tag{4.15b}$$

where

$$\pi/\Delta<s<\pi/(1-\Delta).$$

This last condition may be satisfied only if $\frac{1}{2}<\Delta<1$. The requirement $\alpha^{1/2}\ll\varepsilon\ll1$ leads to an additional restriction on the value of s:

$$\alpha^{1/2}\ll[1-(\pi/\Delta s)^2]\ll1.$$

The $V(\Lambda)$ dependences for a fixed value of Δ and $V(\Delta)$ for a fixed Λ are plotted in Fig. 2 for the case when $\mu_{\varepsilon,0}=\frac{16}{49}$. These dependences are double-valued, i.e., they have two branches. An analytic expression for the lower branch is obtained from Eq. (4.15) in the limit $s\to\pi/(1-\Delta)$: the rate V is described by Eq. (4.11) when the conditions of Eq. (4.12) are satisfied.

An explicit expression for the growth rate corresponding to the upper branch is found from Eq. (4.15) when s is close to π/Δ:

$$V=\frac{\pi(\Delta-1/2)}{\Lambda\Delta(1-\Delta)^2}\left\{1-\frac{(1-2\Delta+2\Delta^2)}{(\Delta-1/2)}\left[\frac{\pi\mu_{\varepsilon,n}(1-\Delta)^2}{\Lambda\Delta(\Delta-1/2)}\right]^{2/3}\right\} \tag{4.16}$$

for

$$\Lambda^{-2/5}\ll(\Delta-1/2)\ll\alpha^{-3/4}\Lambda^{-1}(1-\Delta)^2.$$

For a given channel width Λ, it follows from Eq. (4.15) that the solutions are obtained beginning from a certain minimum supercooling Δ_m (Fig. 2b), which depends on Λ:

$$(\Delta_m-1/2)\propto\Lambda^{-2/5}.$$

In the limit $(\Delta-\frac{1}{2})\ll1$, the Péclet number is, in accordance with Eq. (4.16), also small: $p\equiv V\Lambda\ll1$. In this case the correction to ε associated with the finite value of p can be calculated exactly (see the Appendix). Then according to Eq. (A.7), we have

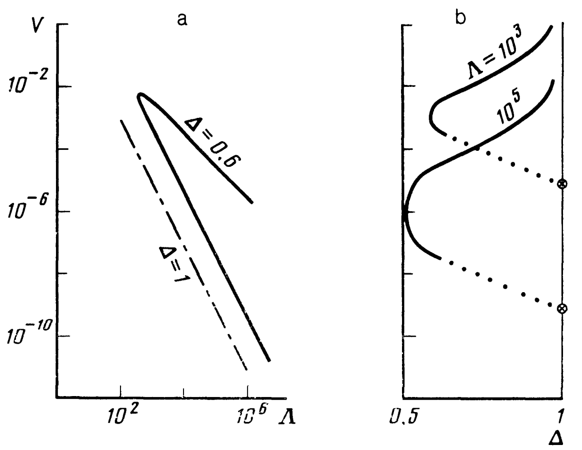

FIG. 2. Dependences of the growth rate V on the channel width Λ (a) and on the supercooling Δ (b) in the absence of the surface tension anisotropy ($\alpha=0$). The continuous curves are plotted using Eq. (4.15) the dash-dot line in Fig. 2a and the crosses enclosed by circles in Fig. 2b are the numerical results from Ref. 17; the dotted curves in Fig. 2b is the proposed interpolation of the curves in the range $\Delta>1/2$, which is outside the framework of the adopted approximations.

$$\varepsilon=4(\Delta-{}^1/_2)-p\pi^{-1}\ln 2.$$

On the other hand, the model equations (4.13) and (4.14) give

$$\varepsilon=4(\Delta-{}^1/_2)-p/2\pi.$$

Therefore, the final answer for the growth rate differs from the results given by Eq. (4.16) in the limit $\Delta\to\frac{1}{2}$ only by numerical factors:

$$V=\frac{1}{\Lambda}\left[\frac{4\pi}{\ln 2}(\Delta-{}^1/_2)-\frac{2\pi^{5/3}}{(\ln 2)^{1/3}}\left(\frac{\mu_{\varepsilon,n}}{\Lambda(\Delta-{}^1/_2)}\right)^{2/3}\right]. \tag{4.17}$$

This expression is valid under the same conditions as Eq. (4.16) and in the limit we have $(\Delta-\frac{1}{2})\ll 1$.

b. Selection of the growth rate on the basis of the surface energy anisotropy.

If we allow for α, we can simplify Eq. (4.2) if $\varepsilon<0$, but $|\varepsilon|\gg\alpha^{1/2}$, when we can ignore t compared with ε. In this case it follows from Eq. (4.2) that if we make the substitutions

$$\tau=t\alpha^{-1/2}\text{and}\varphi=\psi\varepsilon^{1/2}\alpha^{-5/4}, \tag{4.18}$$

we obtain

$$\frac{d^2\varphi}{d\tau^2}-\left[\frac{2^{1/2}\mu_\alpha\tau^{7/2}}{\tau^2-2}+\frac{21}{16\tau^2}\right]\varphi=-\tau^{-3/4}. \tag{4.19}$$

Here,

$$\mu_\alpha=\alpha^{7/4}/\sigma b^2\varepsilon^2. \tag{4.20}$$

Equation (4.19) is considered in Ref. 10 in the specific case of crystallization of an isolated dendrite. The spectrum values of $\mu_{\alpha,n}$ is calculated there and this spectrum determines the spectrum of the growth rates ($\mu_{\alpha,0}\approx 0.48$). The relationships for the growth rate are obtained by solving simultaneously Eqs. (4.20), (4.13), and (4.14). The required $V(\Delta,\Lambda)$ dependence can again be readily represented in the parametric form:

$$\Lambda(s)=2\mu_{\alpha,n}\alpha^{-7/4}[\pi^2/\Delta^2-s^2]^2/s[\pi^2/(1-\Delta)^2-s^2], \tag{4.21a}$$

$$V(s)=[\pi^2/(1-\Delta)^2-s^2]/2s\Lambda(s), \tag{4.21b}$$

where $0<s<\min\{\pi/\Delta,\pi/(1-\Delta)\}$. Moreover, we have an additional restriction on the parameter s which follows from the requirement $|\varepsilon|\gg\alpha^{1/2}$:

$$[(\pi/s\Delta)^2-1]\gg\alpha^{1/2}.$$

The $V(\Lambda)$ and $V(\Delta)$ dependences are plotted in Fig. 3. The $V(\Lambda)$ curves either have two branches described by the system (4.21) or one branch. If $\Delta<1/2$ there are always two branches which go on to the region $\Lambda\to\infty$. The lower branch corresponds to continuation of the Saffman-Taylor solution in the range $\Delta<1/2$, associated with the surface tension anisotropy. This branch is discussed in Ref. 15. The asymptote for this solution is obtained from Eq. (4.21) assuming that $s\to\pi/(1-\Delta)$:

$$V\approx\frac{4\pi^2\mu_{\alpha,n}}{\alpha^{7/4}\Lambda^2}\frac{({}^1/_2-\Delta)^2}{\Delta^4(1-\Delta)^2} \tag{4.22}$$

if $\Delta^4\alpha^{7/4}\Lambda\gg 1$ and $\alpha^{1/2}\ll(\frac{1}{2}-\Delta)\ll\Lambda\alpha^{7/4}$. The growth rate on the upper branch tends to a finite limit on increase in Λ and this limit can be found from Eq. (4.21) by substituting $s\to 0$:

$$V=\frac{\alpha^{7/4}}{4\mu_{\alpha,n}}\left(\frac{\Delta}{1-\Delta}\right)^4\quad\text{for}\quad\Delta^4\alpha^{7/4}\Lambda\gg 1. \tag{4.23}$$

In this limit ($\Lambda\to\infty$) we are dealing in fact with the growth of a dendrite. The relationship (4.23) for the case $\Delta\ll 1$ differs only by a numerical factor from the exact expression applicable to a free dendrite.[5,10] This difference is due to the following circumstance. In the case of a free dendrite characterized by a parabolic shape in the case when $\bar{d}_0=0$ and $w_k\to\infty$, we have[11]

$$2P_0^{1/2}e^{P_0}\int_{P_0^{1/2}}^{\infty}e^{-x^2}dx=\Delta, \tag{4.24}$$

where $P_0=vp/2D$ is the Peclet number and ρ is the radius of curvature of the dendrite tip. The model Eq. (2.10) derived for the tip also describes a parabolic shape with a radius

$$\rho/\lambda=-1/\zeta_0''(0)=\Delta^2s/\pi^2.$$

It follows from Eq. (4.14) in the limit $p\to\infty$ (i.e., when $\lambda\to\infty$) that $s=\pi^2/2p(1-\Delta)^2$, which yields

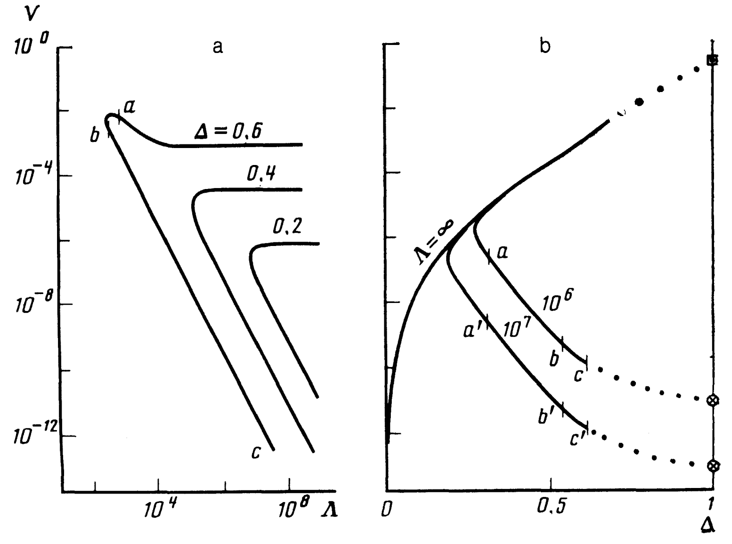

FIG. 3. Dependences of the growth rate V on the channel width Λ (a) and on the supercooling Δ (b) for $\alpha=0.01$. The continuous curves are the results obtained in the present study, the points corresponding to Δ are the results from Ref. 17 (⊗) and from Ref. 19 (□). The dotted curves represent interpolation. Outside the range a–c the continuous curves are plotted using Eq. (4.21), whereas in the range b–c they are plotted using Eq. (4.11); in the range a the curves correspond to the intermediate case characterized by $\varepsilon<\alpha^{1/2}$.

$$P_0=\Delta^2/2(1-\Delta)^2.$$

If $\Delta \ll 1$, it follows from this expression that $P_0 = \Delta^2/2$, so that the only difference from the exact relationship $P_0 = \Delta^2/\pi$ is a numerical factor. We must stress however that in the other limit when $(1-\Delta) \ll 1$, the dependence $V \propto (1-\Delta)^{-4}$ deduced from Eq. (4.23) is invalid. This is because the model expression gives

$$P_0=(^1/_2)(1-\Delta)^{-2}$$

instead of the exact expression

$$P_0=(^1/_2)(1-\Delta)^{-1}.$$

On the other hand, Eq. (4.23) represented as the dependence of v on P_0,

$$v \approx \frac{2D}{\bar{d}_0}\frac{\alpha^{7/4}P_0^2}{\mu_{\alpha,n}}, \tag{4.25}$$

is identical with the exact expression which, as shown in Refs. 8 and 10, is valid for values of P_0 which need not be small. The range of validity of Eq. (4.25) is given in Ref. 10:

$$P_0\alpha^{1/2} \ll 1.$$

It should be pointed out that, according to Eq. (4.25), the growth rate V diverges in the limit $\Delta \to 1$. A model of a boundary layer is used in Ref. 19 to show that the growth rate remains finite in the limit $\Delta \to 1$:

$$v = \frac{2D}{\bar{d}_0}\frac{7}{8}\left(\frac{56\alpha}{3}\right)^{3/4}. \tag{4.26}$$

In the limit of the range of validity of Eq. (4.25), i.e., when $P_0\alpha^{1/2} \sim 1$, Eqs. (4.25) and (4.26) give values of the same order of magnitude.

It is clear from Fig. 3b that the growth becomes possible beginning from the minimum supercooling Δ_m, which in the $\Lambda\alpha^{7/4} \gg 1$ case is

$$\Delta_m=(3^{3/2}\pi\mu_{\alpha,0}/\Lambda\alpha^{7/4})^{1/4} \ll 1,$$

and for this supercooling the growth rate is $V_m = \pi/3^{1/2}\Lambda$. In the limit $\Lambda \to \infty$ the values of Δ_m and V_m tend to zero. On the upper branch the rate $V(\Delta)$ is described by a curve derived for a free dendrite, whereas for the lower curve we have $V \to 0$.

5. SELECTION OF THE GROWTH RATE ON THE BASIS OF THE ANISOTROPY OF THE KINETIC GROWTH COEFFICIENT

For the sake of simplicity, we shall consider the kinetic effects in the case of a free dendrite ($\lambda \to \infty$) with a negligibly small surface energy ($\bar{d}_0 = 0$). The anisotropy of the kinetic growth coefficient $w_k(\theta)$ is described by Eq. (2.6b). In the limit of infinitely fast kinetics, $w_k \to \infty$, the crystallization front is parabolic:

$$\zeta_0(x)=-x^2/2\rho,$$

where ρ is the radius of curvature of the tip of the needle shaped dendrite, and the Péclet number

$$P_0=v\rho/2D$$

is related to the supercooling Δ by Eq. (4.24).

Allowing for the finite nature of the kinetic growth coefficient, we find that Eq. (3.2) for the correction to the shape $\zeta_1(x)$ corresponding to $\sigma = 0$ is

$$x\frac{d\zeta_1}{dx} - \frac{p_k(1+x^2)^{3/2}}{2A_\nu}\zeta_1=-(1+x^2), \tag{5.1}$$

where

$$A_\nu(x) \approx 1+8\nu x^2/(1+x^2)^2.$$

Here, all the lengths are measured in units of ρ; $p_k = \bar{w}_k\rho/2D$; ν is the anisotropy parameter assumed to be small so that $\nu \ll 1$.

Near a singularity $x = i$ on the assumption that $|x-i| \ll 1$ and after the substitution of

$$x=i(1-\tau\nu^{1/2}), \quad \zeta_1=2\nu\varphi,$$

we obtain the following equation for φ in the case when $\tau\nu^{1/2} \ll 1$:

$$\frac{d\varphi}{d\tau}+\frac{2^{1/2}\mu_\nu\tau^{7/2}}{(\tau^2-2)}\varphi=\tau, \tag{5.2}$$

where

$$\mu_\nu=p_k\nu^{5/4}. \tag{5.3}$$

The selection of the growth rate and of the parameter ρ is due to the fact that Eq. (5.2) has the required solution when the numerical parameter μ_ν assumes discrete values $\mu_{\nu,n}$. It then follows from Eq. (5.3) that

$$\rho=\mu_{\nu,n}\frac{2D}{\bar{w}_k}\nu^{-5/4} \tag{5.4}$$

and

$$v=\bar{w}_k\nu^{5/4}P_0(\Delta)/\mu_{\nu,n}, \tag{5.5}$$

where $P_0(\Delta)$ is the solution of the transcendental equation (4.24). In the two limiting cases of $\Delta \ll 1$ and $(1-\Delta) \ll 1$, we have, respectively, $P_0 = \Delta^2/\pi$ and $P_0 = (\frac{1}{2})(1-\Delta)^{-1}$. The direction of the dendrite growth is the same as the direction along which the kinetic growth coefficient is maximal. In the case of a free dendrite the transition from the selection on the basis of the surface energy described by Eq. (4.25) to the selection on the basis of the kinetics defined by Eq. (5.5) occurs at a certain supercooling when both rates become of the same order of magnitude. At this supercooling we have

$$P_0(\Delta) \propto \frac{\bar{w}_k\bar{d}_0}{D}\frac{\nu^{5/4}}{\alpha^{7/4}}. \tag{5.6}$$

The selection on the basis of the kinetics occurs in the case of stronger supercooling. Therefore, for Δ close to 1, where the parameter P_0 is sufficiently large, the selection is always on the basis of the kinetics. However, it then follows from Eq. (5.5) that the rate v rises without limit when $\Delta \to 1$. This divergence is nonphysical and it is obviously related to the following. It is said in Ref. 10 that Eq. (4.25) is valid also for P_0 which is not small, but still obeys $P_0\alpha^{1/2} \ll 1$. Similarly, Eq. (5.5) is valid when $P_0\nu^{1/2} \ll 1$. There is as yet no analytic theory for the case when $\Delta \to 1$ and $P_0\nu^{1/2} \gg 1$. On the other hand, it follows from physical considerations that in the limit $\Delta \to 1$ the growth rate remains finite and this confirms the results of a numerical modeling reported in Ref. 20.

We shall conclude this section by calculating the spectrum of values of μ_ν on the assumption that $\mu_\nu \gg 1$. Such a calculation is similar to the calculation of the spectrum of μ_ε made in the preceding section.

If $\tau \ll 1$, the general solution of Eq. (5.2) is

$$\varphi(\tau)=\exp(2^{1/2}\mu_\nu\tau^{9/2}/9)\left[C+\int_0^\tau t\exp(-2^{1/2}\mu_\nu t^{9/2}/9)\,dt\right]. \quad (5.7)$$

The integration constant C is found on the assumption that the solution is finite along a ray $\arg\tau = 4\pi/9$. [When we go over from the values of $|\tau| \ll 1$ to the range $|\tau| \gg 1$, we find that $\arg\tau = 4\pi/9$ corresponds to $\arg\tau = 2\pi/5$ for which the general solution of Eq. (5.2)

$$\varphi \propto \exp(-2^{3/2}\mu_\nu\tau^{5/2}/5)$$

rises exponentially.] After determination of this constant for real values of τ such that $\mu_\nu^{-9/2} \ll \tau \ll 1$, we find that

$$\varphi\sim\exp(17\pi i/18)\exp(2^{1/2}\mu_\nu\tau^{9/2}/9). \quad (5.8)$$

A real numerical factor is omitted from the above expression.

Near the point $\tau = 2^{1/2}$ the solution of Eq. (5.2) is

$$\varphi \propto (\tau - 2^{1/2})^{-2^{3/4}\mu_\nu}. \quad (5.9)$$

It follows from the above expression that on going from $\tau < 2^{1/2}$ to $\tau > 2^{1/2}$ an additional phase factor $\exp(2^{3/4}\mu_\nu \pi i)$ appears in the solution. Using the phase factor in Eq. (5.8) we find that in the $\tau > 2^{1/2}$ case we finally obtain the phase factor of the function $\varphi(\tau)$:

$$\exp\left[\pi i(2^{3/4}\mu_\nu + {}^{17}/_{18})\right].$$

The spectrum of μ_ν is obtained by requiring that φ be real in the range $\tau > 2^{1/2}$:

$$\mu_{\nu,n}=2^{-3/4}(n+{}^1/_{18}),\quad n=0,\ 1,\ 2,\dots . \quad (5.10)$$

We shall conclude by noting that in the case of a free dendrite we find from the numerical results[4] that the only stable solution corresponds to the maximum growth rate in the discrete spectrum found above. The other solution are unstable against splitting of the tip of a growing needle-shaped crystal.

APPENDIX

Calculation of the correction to ε

In the limit $(\Delta - \frac{1}{2}) \ll 1$ the value of ε is given by Eq. (3.7). A correction linear in p to this equation can be found if we consider a linear (with respect to p) correction to the shape $\zeta_0(x)$. We shall describe the shape of the front in the form

$$\zeta_0(x)=\zeta_{ST}(x)+p\eta(x), \quad (A.1)$$

where ζ_{ST} is the Saffman-Taylor solution given by Eq. (2.7). It follows from the definition of Eq. (3.6) that ε is that value of t in Eq. (3.3) for which we have $\zeta_0''(x) = 0$. Therefore, we can determine ε from two relationships:

$$\zeta_0''(x)=0,\quad \zeta_0'(x)=i(1-\varepsilon). \quad (A.2)$$

When the shape of the front is described by the Saffman-Taylor expression $\zeta_{ST}(x)$, it follows from Eq. (A.2) that

$$\varepsilon_{ST}=4(\Delta-{}^1/_2)\ll 1,\quad x_{ST}=-i\infty. \quad (A.3)$$

In the case when the shape is described by Eq. (A.1) and we have $p \ll 1$, the correction to ε_{ST} is proportional to p, which is small. After linearization of the system (A.2), we obtain

$$\varepsilon=\varepsilon_{ST}-ip\eta'(x_{ST}). \quad (A.4)$$

Therefore, we can find ε from the correction to the shape near a singularity.

The linear equation for $\eta(x)$ is

$$\int_{-1/4}^{1/4} dx'[\eta(x)-\eta(x')]\left\{1+\frac{\cos^2 2\pi x-\cos^2 2\pi x'}{2\sin^2 2\pi(x-x')}\right\}$$

$$=\frac{1}{4\pi^2}\int_{-1/4}^{1/4} dx'\ln\frac{\cos 2\pi x}{\cos 2\pi x'}\ln\frac{|\sin 2\pi(x-x')\sin 2\pi(x+x')|}{\cos^2 2\pi x'}. \quad (A.5)$$

This equation is derived from Eq. (2.5) on the assumption that $d_0 = 0$ and $w_k \to \infty$ in the limit $\Delta \to \frac{1}{2}$. An allowance is made for the fact that the Saffman-Taylor profile $\zeta_{ST}(x)$ satisfies Eq. (2.5) linearized with respect to p. Therefore, Eq. (A.5) corresponds to the expansion of Eq. (2.5) up to terms quadratic in p. The values of x near x_{ST} contain large imaginary contributions. For this reason the integral on the right-hand side of Eq. (2.5) is calculated and the kernel of the integral on the left-hand side simplifies and ceases to depend on x. We thus obtain

$$\int_{-1/4}^{1/4} dx'[\eta(x)-\eta(x')](1-\cos 4\pi x')\approx-\frac{i\ln 2}{2\pi}x. \quad (A.6)$$

An allowance is made here for the fact that

$$\int_{-1/4}^{1/4} dx'\ln\frac{|\sin 2\pi(x-x')\sin 2\pi(x+x')|}{\cos^2 2\pi x}=0.$$

Since $|x| \gg 1$, it follows from Eq. (A.6) that

$$\eta(x)\approx-\frac{i\ln 2}{\pi}x,$$

so that Eq. (A.4) yields

$$\varepsilon\approx 4(\Delta-{}^1/_2)-p\pi^{-1}\ln 2. \quad (A.7)$$

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