Effect of a forced flow on dendritic growth

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The effects of a forced flow on dendritic growth rate are studied theoretically. By using a solvability condition, one determines the eigenvalue $C=\rho^2 V/Dd_0$ as a function of the velocity of the forced flow in the two-dimensional model. Even in the presence of crystalline anisotropy, no solutions for eigenvalues can be found when the flow velocity is larger than a critical velocity. The results are compared critically with recent experiments.

I. INTRODUCTION

A basic problem in dendritic growth is to determine growth rates as a function of various control parameters such as undercooling, concentration of an impurity, or velocity of a controlled external flow.

Growth from a pure undercooled melt is now well understood, at least from a theoretical point of view. Current theory is based on the existence of a one-parameter family of isothermal paraboloids (Ivantsov[1]) growing with constant velocity, with an exact solution of the Stephan problem. For these solutions, only the Péclet number related to the crystal $P_c=\rho V/2D$ is fixed by the undercooling $\Delta=(T_0-T_\infty)c_p/Q$ so that the growth velocity remains arbitrary. Here, ρ is the tip radius of the crystal, V the velocity, D the diffusive coefficient for temperature, T_∞ the temperature of the undercooled liquid, T_0 the crystallization temperature of the pure liquid, c_p the specific heat, and Q the latent heat released per unit volume of solid. When anisotropic surface tension effects are taken into account by the Gibbs-Thomson relation, growth velocity can be determined since another combination between ρ and V, the eigenvalue $C=\rho^2 V/4Dd_0$, where d_0 is the capillary length, is determined by a solvability condition as a function of the anisotropic factor only (Meiron,[2] Barbieri *et al.*,[3] and Ben Amar and Pomeau[4]).

The agreement between theoretical results and experimental data is less clear. On one hand, from a theoretical point of view, growth rates are determined from the needle crystals solutions, i.e., stationary shapes, which is not the case in experiments. This difference appears inessential since numerical simulations of Saito *et al.*,[5] by using the same theoretical model of growth, can produce dendrites, with growth rates in agreement with the previous theoretical results. On the other hand, from the experimental side, if the law $\rho^2 V=C^{te}$ for a given material is well verified, its dependance as a function of the anisotropic factor is less clear. Measurements of anisotropic factors are difficult, essentially when they are small, and there is still some controversy about the experimental results. Thus it appears essential to study the influence of other control parameters on the dendritic growth rates.

When impurity is added to the melt, the Ivantsov paraboloids are still exact solutions of the Stephan problem without surface tension, at once for the thermal and solutal fields. Still here, only the thermal and solutal Péclet numbers related to the crystal are determined by T_∞ and c_∞, the concentration of impurity in the melt, so that the growth velocity is not determined. When anisotropic surface tension effects are taken into account, a solvability condition leads to the determination of the same eigenvalue C as a function of the anisotropic factor and c_∞ only (Ben Amar and Pelcé[6]). For a fixed anisotropic factor, $1/C$ is a linear function of the concentration of impurities, result not in agreement with experiments. Nevertheless both theory and experiments agree that, for a fixed undercooling with respect to the liquidus temperature, the growth velocity has a maximum for some finite value of c_∞ (Lipton *et al.*[7]).

When effect of an axial external flow is considered, experiments show that for a given flow velocity U, $\rho^2 V$ is still a constant, linearly increasing function of the external velocity U (Bouissou *et al.*[8]). In this case, the Ivantsov paraboloids are still solutions of the problem when surface tension effects are neglected, either in the large-Reynolds-number limit [potential flow approximation (Dash and Gill[9] and Ben Amar *et al.*[10])], or in the small-Reynolds-number limit [Oseen approximation (Dash and Gill[9] and Saville and Beaghton[11])]. Only the Péclet number related to the crystal P_c is determined as a function of the undercooling and the Péclet number related to the flow $P_f=\rho U/2D$. Our work will be devoted to the determination of the eigenvalue C as a function of the anisotropic factor and the external flow velocity U by using, as in the previous cases, a solvability condition.

In Sec. II, one introduces the two-dimensional free-boundary problem defined by the equation of diffusion of heat and Navier-Stokes, with thermodynamic boundary conditions applied at the interface. We will consider in the following that the flow is at small Reynolds number $N_{Re}=\rho U/\nu$, which is the case in experiments, so that the flow satisfies the Oseen approximation. In such case, when surface tension effects are neglected, this free

boundary problem admits a one-parameter family of Ivantsov parabolas, that one determines in the third part. Then, we introduce surface tension effects in the problem. Contrary to the previous cases, it is not possible to write an integral equation in which only the shape of the crystal enters. Thus methods based on the resolution of these integral equations (either a whole solution of an analytical continuation of the equation in the complex plane[4] or a solution of a linearization of the equation around the shape without surface tension[3]) cannot be used easily. One will use a solvability condition, in a form previously derived by Pelcé and Bensimon,[12] which is simply the vanishing of an oscillating integral in which shapes without surface tension and dispersion relation for disturbances of a planar interface enter. In Secs. IV A and IV B we present the method and derive the dispersion relation. In Sec. IV C we analyze the effects of a forced flow on velocity selection. We first test our method to study the effects of Péclet number on velocity selection and compare the results to the one obtained by Barbieri and Langer[13] in the two-dimensional case. Then we study the effects of the external forced flow and derive a relation between the eigenvalue C and the external flow velocity. In particular we show that above some critical flow velocity, selection disappears even when anisotropic effects in surface tension are considered, i.e., no steady solution can be found. Then in a final discussion all these theoretical results are compared to experimental data and new experiments are suggested.

II. MODEL OF GROWTH

The liquid-solid interface is a region which is in general very small compared to the macroscopic scale (several Å). During crystal growth, it is there that the latent heat is released, that the temperature field varies rapidly. In order to study the dynamics of growth, one can consider the interface as a discontinuity for the field of temperature and for the velocity field on which boundary conditions are applied.

A. Interface

The interface is assumed to be rough so that the kinetic time of transfer of molecules between solid and liquid is very fast compared to the characteristic diffusion time of heat. In these conditions, a thermodynamic relation, the Gibbs-Thomson relation gives the temperature at the interface, a function of the local curvature $1/R$ of the front. For simplicity, one considers here a two-dimensional model so that the temperature T at the interface is

$$T=T_0-\frac{T_0}{Q}\frac{\sigma}{R} . \tag{1}$$

Here σ is the effective anisotropic liquid-solid surface tension and $1/R$ the curvature of the interface.

B. Temperature field

The temperature field satisfies the diffusion equation

$$\frac{\partial T}{\partial T}=D\Delta T \tag{2}$$

in the solid. In the liquid phase, one must take into account the advection of heat by the fluid velocity field $\mathbf{w}$ and thus the temperature satisfies

$$\frac{\partial T}{\partial t}+(\mathbf{w}\cdot\nabla)T=D\Delta T . \tag{3}$$

At the interface, conservation of energy leads to

$$Q\mathbf{v}\cdot\mathbf{n}=Dc_p(\nabla T_s-\nabla T_1)\cdot\mathbf{n} , \tag{4}$$

where $\mathbf{v}\cdot\mathbf{n}$ is the normal velocity of the interface. Far at infinity, the temperature is uniform and equal to T_∞.

C. Velocity field

The velocity field $\mathbf{w}$ satisfies the Navier-Stokes equation

$$\frac{\partial \mathbf{w}}{\partial t}+(\mathbf{w}\cdot\nabla)\mathbf{w}=-\frac{1}{\rho_1}\nabla p+\nu\Delta\mathbf{w} \tag{5}$$

and the mass conservation relation

$$\nabla\cdot\mathbf{w}=0 . \tag{6}$$

Here ν is the kinetic viscosity of the fluid, p the pressure field, and ρ_1 the density of the liquid. In the following, one assumes that crystal and melt densities are equal so that there is no exchange of mass between the fluid and the crystal as it grows and the normal component of the flow vanishes at the interface. Furthermore, one assumes a no-slip condition at the interface so that the tangential component of the flow vanishes too (see Gliksman *et al.*[14]).

III. CASE WITHOUT SURFACE TENSION

A family of exact solutions can be found when the temperature is constant on the interface $T=T_0$, and when the fluid flow is potential[9,10] or Oseen type.[9,11] In both cases, as for the Ivantsov solutions, the shape of the interface is a parabola in a two-dimensional model. For a given undercooling, the Péclet number related to the velocity of the needle crystal P_c is a function of the Péclet number related to the velocity of the flow P_f.

In the following, one will study the experimentally relevant case of the small Reynolds number limit where the flow is Oseen type. In that approximation, the nonlinear term of the Navier-Stokes equation is replaced by the term $U(\partial\mathbf{w}/\partial z)$, where U is the external flow velocity far ahead of the crystal. Then, the Navier-Stokes equation becomes the so-called Oseen equation:

$$U\frac{\partial\mathbf{w}}{\partial z}=-\frac{1}{\rho_1}\nabla p+\nu\Delta\mathbf{w} . \tag{7}$$

One assumes that the interface is parabolic, of tip radius ρ, and grows at velocity V along the Oz axis. The external flow at infinity is parallel, in the opposite the direction of growth.

In the parabolic coordinates, where ξ and η are defined as

$$x=\rho\sqrt{\xi\eta}\ ,$$
$$z=\frac{\rho}{2}(\eta-\xi)\ , \tag{8}$$

one finds a solution for the velocity field, in the Oseen approximation, whose components are, in a frame where the crystal is at rest

$$u_\eta=-\frac{f(\eta)}{2\sqrt{\eta+\xi}}\ , \tag{9a}$$

$$u_\xi=\frac{\sqrt{\eta\xi}}{\sqrt{\eta+\xi}}\frac{df}{d\eta}\ , \tag{9b}$$

where

$$f(\eta)=2(U+V)\sqrt{\eta}-2Ug(\eta)\ , \tag{10}$$

with

$$g(\eta)=\sqrt{\eta}\frac{\operatorname{erfc}(\sqrt{N_{\mathrm{Re}}\eta/2})}{\operatorname{erfc}(\sqrt{N_{\mathrm{Re}}/2})}+\sqrt{(2/\pi N_{\mathrm{Re}})}\frac{1}{\operatorname{erfc}(\sqrt{N_{\mathrm{Re}}/2})}\times\left\{\exp\left[-\left[\frac{N_{\mathrm{Re}}}{2}\right]\right]-\exp\left[-\left[\frac{N_{\mathrm{Re}}}{2}\eta\right]\right]\right\}. \tag{11}$$

Then, one integrates the stationary equation of heat (3), with the boundary conditions (1) and (4) in which surface-tension effects have been neglected to obtain a temperature profile which depends only on η. Then, relation between Péclet numbers and undercooling is obtained as

$$\Delta=P_c\exp(P_c-P_f)\times\int_1^{+\infty}\frac{d\eta}{\sqrt{\eta}}\exp\left[-P_c\eta+P_f\left[2+\int_1^{\eta}\frac{g(\eta')}{\sqrt{\eta'}}d\eta'-\eta\right]\right]. \tag{12}$$

IV. EFFECT OF SURFACE TENSION IN THE TWO-DIMENSIONAL MODEL

A. Solvability condition

When surface-tension effects are taken into account the Ivantsov parabolas are no longer solutions of the free-boundary problem. Nevertheless, when surface-tension effects are weak, in a sense that will be discussed in detail later, steady solutions can be found close to an Ivantsov parabola if a solvability condition is satisfied. In its most general form, derived previously by Pelcé and Bensimon[12] (see also Pelcé[15,16]), this condition appears as the vanishing of an oscillating integral as

$$\int_{-\infty}^{+\infty}dl\ G[X_0(l)]\exp\left[i\int_0^l k_m(l')dl'\right]=0\ . \tag{13}$$

Here $X_0(l)$ is the Ivantsov parabola function of the curvilinear coordinate l; G, the curvature operator; and k_m, the local nonzero marginal mode of the conjugate dispersion relation, written in a frame moving with constant velocity U in the z direction (frame at rest with respect to the unperturbed solution). The condition of application of this formula is that $k_m(l)$ is large compared to the inverse of the scale of the unperturbed solution [Wentzel-Kramers-Brillouin (WKB) approximation].

B. Stability analysis of a planar interface moving with constant velocity

One determines the growth rate of a perturbation of the planar interface with a wavelength small compared to the scale of the unperturbed solution. As the perturbation disturbs the fluid on a distance of the order of the wavelength, one can expand the stationary velocity field give by Eqs. (9) and (10) around the parabola ($\eta=1$). To the first order in $\eta-1$, one finds

$$u_\eta=-\left[\frac{1}{1+\xi}\right]^{1/2}V\ , \tag{14a}$$

$$u_\xi=\left[\frac{\xi}{1+\xi}\right]^{1/2}[V+Ua(N_{\mathrm{Re}})(\eta-1)]\ , \tag{14b}$$

where

$$a(N_{\mathrm{Re}})=\left[\frac{2N_{\mathrm{Re}}}{\pi}\right]^{1/2}\frac{\exp(-N_{\mathrm{Re}}/2)}{\operatorname{erfc}(\sqrt{(N_{\mathrm{Re}}/2)})}\ . \tag{15}$$

It is more convenient to use a local Cartesian frame (x,y) fixed to the crystal, where x and y are, respectively, the tangent and normal axes to the interface, at a point where the normal to the interface has an angle θ with the growth axis. In these coordinates, the stationary velocity field is

$$\bar{v}=-V\cos\theta\ ,$$
$$\bar{u}=-V\sin\theta-a\frac{U}{\rho}\sin\theta\cos\theta\, y\ , \tag{16}$$

i.e., a shear flow whose magnitude is a function of the angle θ. A similar expansion can be done for the temperature field and one obtains at leading order

$$\bar{T}=T_0-\frac{Q}{c_p}\frac{V}{D}\cos\theta\, y\ . \tag{17}$$

Let u', v', and T' can be the perturbations of the stationary field, ξ' the perturbation of the steady interface, with a wavelength λ assumed very small compared to ρ. In these conditions, the advective term in the Oseen equation can be neglected and the flow perturbation satisfies the Stokes equation, i.e.,

$$\nabla p'=\nu\rho_1\Delta\mathbf{w}'\ , \tag{18}$$

with the mass conservation relation

$$\nabla\cdot\mathbf{w}'=0\ . \tag{19}$$

Similarly, the temperature equation can be written as

$$\frac{\partial T'}{\partial t}+\bar{u}\frac{\partial T'}{\partial x}+\bar{v}\frac{\partial T'}{\partial y}+v'\frac{d\bar{T}}{dy}=D\Delta T' \tag{20}$$

since the stationary temperature field is independant of x. One conserves the advective terms in the temperature equation and not in the flow equation since the Prandtl number $N_{\mathrm{Pr}}=\nu/D$ is assumed small. As it can be deduced from the relations (1) and (4), on the unperturbed interface ($y=0$) the following boundary conditions are satisfied:

$$u'=0, \quad v'=-\frac{\partial \xi}{\partial t} \tag{21}$$

for the velocity field, and

$$T_s'=T_l'-\xi'\frac{V}{D}\cos\theta=\frac{Q}{c_p}d_0\frac{\partial^2\xi}{\partial y^2}\ , \tag{22}$$

and

$$\frac{Q}{c_p}\frac{\partial \xi}{\partial t}=D\left[\frac{\partial T_s'}{\partial y}-\frac{\partial T_l'}{\partial y}-\xi'\frac{Q}{c_p}\left[\frac{V}{D}\cos\theta\right]^2\right] \tag{23}$$

for the temperature field. As can be deduced from relations (18) and (19), $\Delta p'=0$ and thus p' takes the form

$$p'=A\exp(\omega t+ikx-\epsilon ky)$$

where ϵ has the same sign as $\mathrm{Re}(k)$ since p' cannot diverge as y goes to $+\infty$. Then, by using Eqs. (18) and (19) one obtains the perturbed velocity field as

$$u'=(-i\epsilon Ay+B)\exp(\omega t+ikx-\epsilon ky)\ , \tag{24}$$

$$v'=[A(y+\epsilon/k)+iB\epsilon]\exp(\omega t+ikx-\epsilon ky)\ , \tag{25}$$

where B is for the moment an arbitrary constant. One applies the boundary conditions (21) to determine the constants A and B as a function of the amplitude C of the interface displacement:

$$B=C\sin\theta\cos\theta aU/\rho\ , \tag{26}$$

$$A=-C(\omega k\epsilon+i\sin\theta\cos\theta akU/\rho)\ . \tag{27}$$

One determines now the perturbation of the temperature T' on the form, $T'(y)=g(y)\exp(\omega t+ikx)$ and obtains from Eq. (20)

$$D[g''(y)-k^2g(y)]=\bar{v}g'(y)+g(y)(\omega+ik\bar{u})+v'\frac{d\bar{T}}{dy}\ . \tag{28}$$

As g varies on the length scale λ, the left-hand side of Eq. (28) is dominant compared to the right-hand side at large wave number so that one can solve Eq. (28) by expanding the solution at large k. At leading order, one obtains simply $g_0(y)=E_0\exp(-\epsilon ky)$, so that the following order g_1 can be found in the form $g_1(y)=(E_1+f_1y+G_1y^2)\exp(-\epsilon ky)$, where:

$$F_1=\frac{E_0}{4kD}\left[i\frac{U}{\rho}a\sin\theta\cos\theta-2\epsilon(\omega-ikV\sin\theta+\epsilon kV\cos\theta)\right]+\frac{Q}{c_p}\frac{V^2}{4kD}C\left[3\epsilon\omega+ia\sin\theta\cos\theta\frac{U}{\rho}\right]\ . \tag{29}$$

One can apply now the boundary conditions (22) and (23) at each order of the expansion in order to eliminate the unknown constants E_0, E_1, and C and obtain the dispersion relation for the perturbations as

$$\omega=iVk\sin\theta+\epsilon Vk\cos\theta-2d_0D\epsilon k^3+\frac{V^2}{D}\cos\theta[\tfrac{1}{2}\exp(-i\epsilon\theta)-\cos\theta]+iVd_0k^2\epsilon\sin\theta+\omega\left[\frac{5}{4}\epsilon\frac{V}{Dk}-\epsilon d_0k\right]+d_0kia\frac{U}{4\rho}\sin\theta\cos\theta\ . \tag{30}$$

The first line of this relation corresponds to the usual Mullins-Sekerka growth rate. When all the terms of this line balance one obtains the usual order of magnitude $\omega\approx Vk$ and $k\approx(V/Dd_0)^{1/2}$. The second line of the relation corresponds to the effects of Péclet number related to the crystal, i.e., the correction to the Mullins-Sekerka growth rate due to the advective effects of the motion of the crystal. The relative order of magnitude of these terms is V/Dk, or $P_c\lambda/\rho$. Then, the third line of the dispersion relation corresponds to the effect of the external flow, whose order of magnitude, relative to the dominant terms is $aUd_0/V\rho$, or $aP_f(\lambda/\rho)^2$. The corresponding term is complex so that, at this order of the calculation, the effect of the external flow is to modify the advection of the perturbations along the crystal.

C. Velocity selection

We will consider the two following extreme limits of small or large flow velocity, the crossover between the two regimes being for $a(N_{\mathrm{Re}})P_f\approx P_c(\rho/\lambda)$.

1. *Small velocity limit*

As mentioned at the beginning of the section, one computes first the marginal mode of the conjugate of the dispersion relation (30), in which $U=0$. For this, one changes i in $-i$ in relation (30) and as it is the case in previous works,[15,16] one chooses $\epsilon=-1$. As in this limit there is no external flow, the marginal mode k_m satisfies

$$-k_mV\exp(i\theta)+2d(\theta)Dk_m^3=-\frac{V^2}{D}\cos\theta\left[\frac{\exp(-i\theta)}{2}-\cos\theta\right]-id(\theta)k_m^2V\sin\theta\ . \tag{31}$$

One has, at leading order in V/Dk_m,

$$-k_{m_0}V\exp(i\theta)+2d(\theta)Dk_{m_0}^3=0\ , \tag{32}$$

from which follows

$$k_{m_0}=-\left[\frac{V}{2d(\theta)D}\right]^{1/2}\exp(i\theta/2) \tag{33}$$

since $\mathrm{Re}(k)$ has the same sign as ϵ.

At the first order in V/Dk_m, one has

$$k_{m_1}=\frac{P}{2}\exp(-i\theta)\ . \tag{34}$$

Here, one assumes a fourfold symmetry of the crystal so that the capillary length can be written as $d(\theta)=d_0(1-\beta\cos 4\theta)$, where β is the anisotropic factor. Then the solvability condition (13) can be written as

$$\int_{-\infty}^{+\infty} d\eta\, G[(\eta)]\exp\left[\sqrt{C}\,\Psi(\eta)-\frac{P_c}{2}\left[i\eta+\frac{\eta^2}{2}\right]\right]=0\ . \tag{35}$$

Here

$$\Psi(\eta)=\frac{i}{2}\int_0^{\eta}\frac{(1+i\eta')^{7/4}(1-i\eta')^{5/4}}{\sqrt{B(\eta')}}d\eta'\ , \tag{36}$$

where $B(\eta)=(1+\eta^2)^2(1-\beta)+8\beta\eta^2$ and $\eta=\tan\theta$. Here, we find a relation already found by Barbieri and Langer[13] who solved the linearized equation of the complete integral equation. In order to determine this relation, we have neglected the external flow velocity in the dispersion relation, i.e., we assumed that $aP_f \ll P_c(\rho/\lambda)$. As $\lambda=1/k_{m_0}$, this condition is satisfied if $aU \ll VC^{1/2}$.

2. *Large velocity limit*

Consider now the case where the effects related to the Péclet number P_c are negligible, i.e., when $a(N_{\mathrm{Re}})P_f \gg P_c(\rho/\lambda)$, which is generally the case in experiments.[8] Then the equation for the marginal mode is simply [here we still choose $\epsilon=-1$ and i is changed in $-i$ in relation (30)]

$$-k_m V\exp(i\theta)+2d(\theta)Dk_m^3=\frac{i}{4}ak_m\frac{d(\theta)U}{\rho V}\sin\theta\cos\theta\ , \tag{37}$$

from which the marginal mode k_m can be deduced as

$$k_m=-\left[\frac{V}{2d(\theta)D}\right]^{1/2}\times\left[\exp(i\theta)+\frac{i}{4}a\frac{d(\theta)U}{\rho V}\sin\theta\cos\theta\right]^{1/2}\ . \tag{38}$$

As before, we assume a fourfold symmetry of the crystal so that the solvability condition can be written as

$$\int_{-\infty}^{+\infty} d\eta\ G[(\eta)]\exp[\sqrt{C}\,\Psi_\alpha(\eta)]=0\ . \tag{39}$$

Here

$$\Psi_\alpha(\eta)=\frac{i}{2}\int_0^{\eta}d\eta'\frac{[(1+i\eta')(1+\eta'^2)^{5/2}+i\alpha\eta' B(\eta')]^{1/2}}{\sqrt{B(\eta')}} \tag{40}$$

where $\alpha=ad_0U/4\rho V$. Thus, as a first result, one obtains that the eigenvalue C characterizing a selected state is a function of the anisotropic factor β and the dimensionless flow velocity α. At this level the first discrepancy between theory and experiments occurs. In a given experiment, for a given flow velocity of the dendrite decreases on a long-time scale due to the slow change of the supersaturation, but the eigenvalue C remains a constant. Furthermore, C is function of the external velocity U, by the dimensionless number Ud_0/D. Here, theory predicts that the eigenvalue C is function of the external velocity U by the same dimensionless number multiplied by the factor $1/P_c$. As a result for a given experiment, C is not constant since P_c changes due to the slow change of the supersaturation.

To get more quantitative results, one can evaluate the integral (40) in the limit of small anisotropy. The numerator of the integrand vanishes for values of η close to $\eta=i$ (stationary phase points) and the denominator for $\eta=i(1-\sqrt{2\beta})$ (singularity). The dominant contribution to the integral is determined by the neighborhood of $\eta=i$ so that $\Psi_\alpha(\eta)$ can be approximated by

$$\Psi_\tau(\phi)=2^{9/8}\beta^{7/8}\int_{1/\sqrt{2\beta}}^{\phi}d\phi'\frac{[\phi'^{7/2}-\tau(\phi'^2-1)]^{1/2}}{(\phi'^2-1)^{1/2}}\ , \tag{41}$$

where $\eta=i(1-\sqrt{2\beta}\phi)$ and $\tau=\alpha/2^{5/4}\beta^{3/4}$.

First recall what occurs when $\tau=0$ (no external flow velocity).[3] The initial contour of integration (real axis in the η variable or line parallel to the imaginary axis of the real part $1/\sqrt{2\beta}$ in the ϕ variable) must go close to the stationary phase point $\phi=0$, turn around the singularity $\phi=1$, and come back to the stationary point (Fig. 1). The integral (41) is dominated by the contribution of the loop which gives an oscillating factor to the exponentially small value of the integral on the form

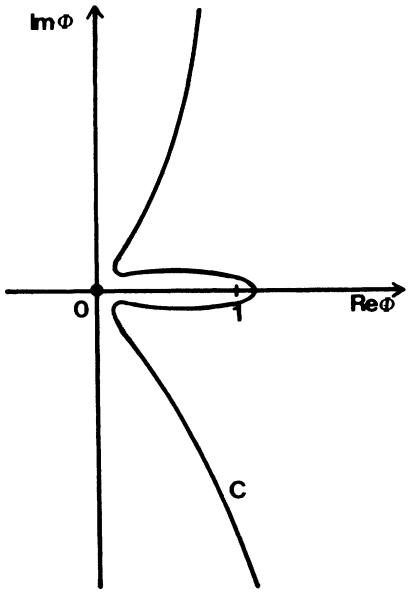

FIG. 1. Contour of integration in the ϕ variable when $\tau=0$.

$$\cos(A\sqrt{C}\,\beta^{7/8}) , \tag{42}$$

where A is a numerical constant.

When $\tau \neq 0$ the stationary phase point $\phi=0$ for $\tau=0$ splits into six displayed symmetrically to the real axis as shown on Fig. 2. The dominant contribution to the integral is given by the two symmetric points with largest real part, i.e., the points with positive real part. The essential feature is that when the parameter τ increases the two points go on the right of the singularity, fuse for some value $\tau_c = \frac{7}{4}(\frac{7}{3})^{3/4}$ on the real axis, and then separate still on the real axis. Thus for small τ the contour goes through the stationary points, turns around the singularity still giving an oscillatory contribution to the integral [Fig. 3(a)]. When $\tau > \tau_c$ all the contour lies on the right of the singularity [Fig. 3(b)]. As a result, the integral is real and the solvability condition cannot be satisfied. Then, even if anisotropic surface tension is taken into account, no stationary solution can be found at sufficiently large external flow velocity. A similar situation was found in the problem of the Saffman-Taylor finger. Solutions for fingers moving at constant velocity can be found only if the relative size of the finger is larger than $\frac{1}{2}$.[17]

For $\tau < \tau_c$, the selected values of C are on the form

$$C = \beta^{-7/4} f\left[\frac{a(N_{\mathrm{Re}})}{\beta^{3/4}}\frac{d_0 U}{\rho V}\right] \tag{43}$$

where f is a function determined numerically.

When τ is small the stationary phase points split from $\phi=0$ at a distance of order $\tau^{2/7}$. We assume that τ is small but sufficiently large in order that the stationary phase points can be treated separately. There are two contributions to the integral: the contribution of the stationary phase points ϕ_- and ϕ_+ and the contribution of the loop. Each stationary phase point contributes by a term $\exp[\sqrt{C}\,\Psi(\phi_\pm)]$ whose oscillating part is

$$\cos[A_1\sqrt{C}\,\beta^{7/8}(1+B_1\tau^{11/14})] ,$$

where A_1 and B_1 are numerical constants. It remains to evaluate the contribution of the loop [Fig. 3(a)] between

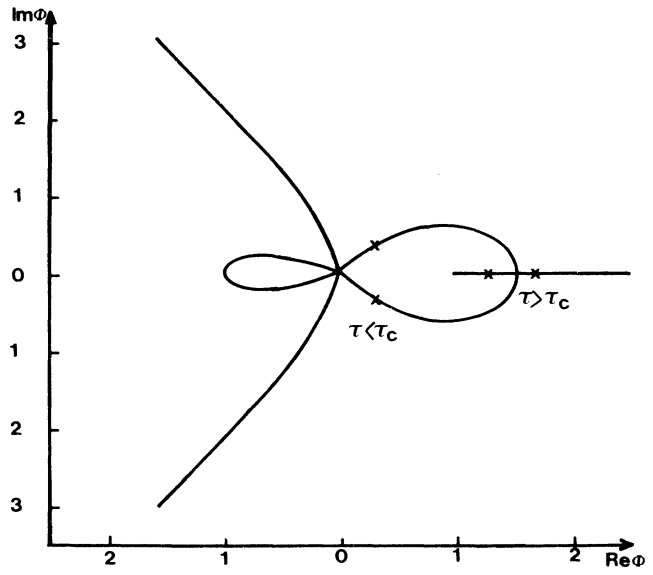

FIG. 2. Stationary phase points.

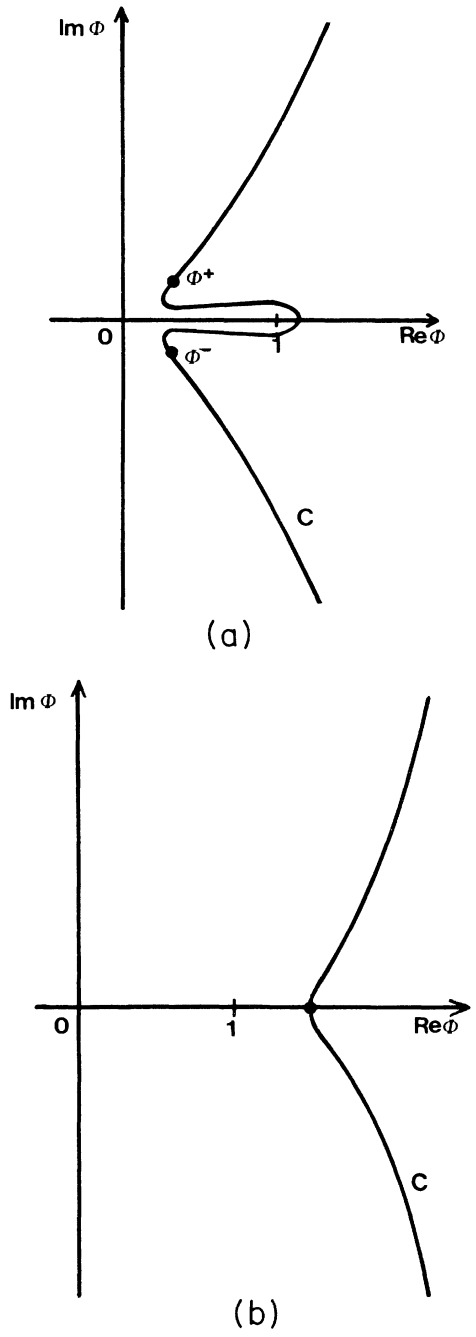

FIG. 3. Contour of integration in the ϕ variable (a) when $\tau < \tau_c$ and (b) when $\tau > \tau_c$.

$\phi_0 \approx \tau^{2/7}$ at the intersection of the steepest descent path and the real axis and $\phi = 1$. It gives an oscillating factor to the exponentially small value of the integral of the form

$$\cos\left[A_2 \sqrt{C}\, \beta^{7/8} \int_{\tau^{2/7}}^{1} d\phi \frac{[\phi^{7/2} - \tau(\phi^2 - 1)]^{1/2}}{(1-\phi^2)^{1/2}} \right], \quad (44)$$

which behaves as $\cos[A_2 \sqrt{C}\, \beta^{7/8}(1 + B_2 \tau^{11/14})]$. The cancellation of the sum of these contributions leads to the following selected values of C:

$$C = \frac{n^2}{\beta^{7/4}} \left[1 + b \left[\frac{a(N_{\mathrm{Re}})}{\beta^{3/4}} \frac{d_0 U}{\rho V} \right]^{11/14} \right], \quad (45)$$

where b is a numerical constant. As mentioned before, this formula holds when $aP_f \gg P_c(\rho/\lambda_m)$, or $aU \gg VC^{1/2}$.

V. DISCUSSION

We determined dendritic growth rates in the presence of an axial external flow in the two-dimensional model. When surface tension effects are neglected, the problem of growth admits a continuum of solutions for needle crystal growing with stationary shape and constant velocity. For a give undercooling, the Péclet number related to the crystal $P_c = \rho V/2D$ is an increasing function of the Péclet number related to the flow $P_f = \rho U/2D$ so that growth rates are not determined at this level. One needs another combination between tip radius and velocity, the eigenvalue $C = \rho^2 V/4Dd_0$ that we determine by taking into account surface-tension effects. This relation can be determined analytically when shapes are close to the shapes without surface tension, as a result of a solvability condition. In the two-dimensional model, the results are the following.

When the external flow velocity is small, $a(N_{\mathrm{Re}})U \ll VC^{1/2}$, C is independent of U. It is a function on the Péclet number P_c by the relation (35). For larger flow velocity $a(N_{\mathrm{Re}})U \gg VC^{1/2}$, C is a function of the dimensionless parameter $\tau = a(N_{\mathrm{Re}})Ud_0/\beta^{3/4}\rho V$, where $a(N_{\mathrm{Re}}) \approx N_{\mathrm{Re}}^{1/2}$ for small Reynolds numbers. When τ is larger than a critical number of order unity no stationary solution can be found. These results are found in the WKB limit, i.e., when C is large, or when the anisotropic factor β is small.

In experiments,[8] $\beta = 0.75$ (pivalic acid), $U/V \approx 10$, $d_0 \approx 20$ Å, and $\rho \approx 10$ μm so that $\tau \approx 10^{-3} a(N_{\mathrm{Re}})$. Furthermore, $N_{\mathrm{Re}} \approx 10^{-6}$ so that $a(N_{\mathrm{Re}}) \approx N_{\mathrm{Re}} \approx 10^{-3}$ in the two-dimensional model. As $\sqrt{C} \approx 5$, $a(N_{\mathrm{Re}})U \ll V\sqrt{C}$ and the effects of the forced flow are dominated by the effects of the Péclet number related to the crystal, i.e., the eigenvalue C is not modified by the effects of the flow. This result is essentially due to the smallness of the factor $a(N_{\mathrm{Re}})$ in the two-dimensional model.

In the three-dimensional model, $a(N_{\mathrm{Re}}) \approx -1/\ln N_{\mathrm{Re}}$ when the Reynolds number is small,[9] so that $a(N_{\mathrm{Re}})U/V\sqrt{C} \approx 1$ and thus one can consider that the external flow may have an observable effect. If one assumes, as it is the case in the absence of forced flow, that the scalings are the same in two-dimensional and three-dimensional case, one will have

$$C = \frac{n^2}{\beta^{7/4}} \left[1 + b \left[\frac{a(N_{\mathrm{Re}})}{\beta^{3/4}} \frac{U}{D/d_0} \frac{1}{P_c} \right]^{11/14} \right]. \quad (46)$$

It is interesting to compare this formula to the experimental results.[8] The linear variation of product $\rho^2 V$ with velocity U must be compared with the theoretical prediction which gives a dependence with $U^{11/14}$. Because of the uncertainty of the data, it is difficult to distinguish a power of U between 1 and 11/14 so that essentially the observed dependance of $\rho^2 V$ with U can be considered in reasonable agreement with our theoretical calculation.

However, experimentally, for a given value of the external flow velocity U, the variation of C as a function of the growth velocity V gives a plateau.[8] If one estimates the constant b in formula (46) so that the value C is equal to the measured one, one finds, for an experimental variation of the growth velocity from 1.5 to 3 μm/s a relative variation for C less than 5% through the term $(1/P_C)^{11/14}$. Thus, under those conditions, one can consider that the dependance of C with V is consistent with experiment.

Furthermore, due to experimental uncertainty, the plot $\rho^2 V$ versus $U_{\parallel}$ [Ref. 8, Fig. 3(a)] is not inconsistent with the existence of the two regimes predicted by the theory: a first one at low flow velocity ($U_{\parallel} < 5$ μm/s), where $\rho^2 V$ does not depend on $U_{\parallel}$ and a second regime corresponding to the effect of the forced flow.

All this indicates that it would be interesting to perform additional experimental work in order to draw out any conclusion concerning the validity of this theory. The main result of the paper is that the eigenvalue C is a function of the dimensionless parameter $\tau = a(N_{\mathrm{Re}})Ud_0/\beta^{3/4}\rho V$ which mixes in a nontrivial way cristalline anisotropy and external flow velocity. Thus, in experiments, it would be particularly interesting to work at larger flow velocity, in a larger range of values of crystal velocity, and use materials of very weak anisotropy like succinonitrile in order to test more completely this scaling law.

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