

# Nonaxisymmetric patterns in the Saffman-Taylor problem and in three-dimensional directional solidification at low velocity

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The theory of nonsymmetric patterns in the Saffman-Taylor problem and directional solidification in two and three dimensions is presented. We consider various effects that influence these patterns: the transverse gravitational force in the Saffman-Taylor problem, or transverse temperature gradient and anisotropy of surface energy in three-dimensional directional solidification. In the latter case, we show that the nonaxisymmetric shape correction, generated in the tip region, can be matched smoothly to the asymptotic shape in the tail region. Here a three-dimensional problem can be entirely solved analytically, including the tail region and without invoking rotational symmetry. The implications of our results for the description of the tail of three-dimensional dendrite are discussed.

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## I. INTRODUCTION

The Saffman-Taylor (ST) finger problem [1] is one of the simplest examples of pattern formation [2,3]. The selection of a symmetric finger in two dimensions is the most studied and best understood one. This finger is seen in a Hele-Shaw cell (a narrow gap between two parallel plates) when a viscous fluid is displaced by a less viscous one. The flat interface is unstable and either stable finger patterns or unstable fractal structures can be formed. For the stable finger, there is a well-developed theory explaining the selection of the pattern via a solvability mechanism arising at any nonzero value of the surface tension. In the high-velocity limit, where interfacial tension effects have a small magnitude, it amounts to the computation of transcendently small corrections appearing beyond all orders of the regular perturbation expansion. This type of exponentially small term was shown to be present in the ST system via numerical calculations [4]. Finally, matched-asymptotics-behavior methods which carefully took into account the surface-tension-induced singularity in the complex plane did indeed explain why the relative finger width  $\lambda$  approaches 1/2 when its velocity increases [5]. All selected fingers are reflection symmetric, i.e., the extra nonsymmetric degree of freedom found in the original (zero-surface-tension) finger problem [6] is not utilized by the system [7,8].

Brener, Levine, and Tu [9] have generalized the family of solutions, as well as the selection mechanism, via inclusion of nonsymmetric forcing, assuming that there exists a gravitational force which acts on the fluid of density  $\rho$ . If the cell is rotated away from horizontal by the angle  $\omega$ , this will induce a transverse potential across the finger. In the high-velocity limit, where surface-tension effects are small, they have analyzed a local equation around

a specific singular point in the complex plane and have found the scaling relations for the finger width and offset as functions of the surface tension and the rotation angle. They have found that for small enough angle the finger slightly deforms, with width staying at about 1/2, but slightly decreasing. At some intermediate angle (but still small at small surface-tension effects) the rate of deformation and width shrinkage accelerates and finally, the width becomes small and the finger hugs the top sidewall. These results are in a good agreement with experimental study [10]. The closely related problem of the selection of nonreflection symmetric dendrites in the presence of anisotropies which do not have a microscopic mirror symmetry has been discussed in Ref. [11].

The theory of a symmetric ST finger in the opposite limit, where the finger moves at low velocity and fills the channel almost completely, has been developed by Dombre and Hakim [12]. In this limit, where surface-tension effects play a dominant role and cannot be treated as a perturbation, their treatment is analogous to the analysis of coating films done by Landau and Levich [13]. The idea is that the tip region (outer region) and the trailing part (inner region) of the finger can be treated separately and matched afterwards.

The main aim of our paper is a theoretical description of the non-symmetric patterns in two and three dimensions. The approach of Dombre and Hakim is very useful because it allows one to solve this problem analytically. In Sec. II we recall the basic formulation and our recent results [10] for the ST problem in the presence of nonsymmetric gravitational force. We find that for a given rotation angle the finger sticks at the top side wall for some critical minimal velocity (as for a symmetric finger) but does not fill the whole channel. The critical velocity increases and the finger width decreases if the rotational angle increases.

In Sec. III we extend the theory to the three dimensions. Though we can easily define the problem mathematically, the formalism no longer describes the physical problem of a multiphase flow in a simple cell. Neverthe-

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less, as was argued in [14], the ST problem in the tube can yield information about ensemble-averaged diffusion-limited aggregation in the same geometry. Finally, the three-dimensional ST problem can serve as a testing ground for the selection theory that has been successful for the two-dimensional problem.

Another physical problem, related to this ST problem, is a directional solidification in a tube at small Peclet numbers. In this problem we have a concentration field instead of the pressure field and a gradient of temperature instead of the gravitational force. In Sec. IV we consider this problem for the nonaxisymmetric patterns caused by the presence of the anisotropy of surface energy. We show that the nonaxisymmetric shape correction generated in the tip region can be matched smoothly to the asymptotic shape in the tail region. For the closely related problem of free dendritic growth the mentioned matching procedure gives rise to serious difficulties because the deviation from the basic isotropic solution becomes large in the asymptotic region even for small anisotropy [15]. In our case this effect does not appear because of the presence of the tube and asymptotic shape in the tail region can be found. So this is the first example where a three-dimensional problem can be entirely solved analytically including the tail region and without invoking rotational symmetry.

## II. SAFFMAN-TAYLOR FINGERS IN THE PRESENCE OF TRANSVERSE GRAVITATIONAL FORCE

Let us recall the basic formulation of the ST problem in the presence of nonsymmetric gravitational force. A viscous fluid obeying Darcy's law  $\mathbf{v} = -(q/\mu)\nabla p$  is displaced by a second fluid of negligible viscosity. Here  $\mathbf{v}$  is two-dimensional velocity,  $p$  is the pressure field,  $\mu$  the viscosity,  $q = b^2/12$  is the medium permeability, and  $b$  is the plate spacing. Assuming incompressibility, the velocity potential  $\Phi = -(q/\mu)p$  satisfies Laplace's equation

$$\nabla^2 \Phi = 0. \quad (1)$$

For a steady-state finger advancing at constant velocity  $U$  along the  $z$  direction the boundary conditions are

$$\frac{\partial \Phi}{\partial n} = U \cos \theta, \quad (2)$$

$$\Phi = (q/\mu)(TK - \rho g x \sin \omega), \quad (3)$$

$$\left. \frac{\partial \Phi}{\partial x} \right|_{x=\pm a} = 0. \quad (4)$$

Here  $\theta$  is the angle between the local normal vector  $\mathbf{n}$  on the interface and the  $z$  direction,  $T$  is the surface tension,  $K$  the local curvature,  $\rho$  the fluid density,  $g$  the gravitational acceleration,  $\omega$  the tilt angle, and  $a$  is the actual half-width of the channel. We are interested in the limit of low velocity, where the finger fills the channel almost completely. In the tip region the fluid is weakly perturbed by the existence of the thin layers near the side

walls, the flow is uniform [12]:

$$\Phi = Uz, \quad (5)$$

and the interface profile obeys Eq. (3),

$$\gamma K = z_{\text{int}} + \sigma x_{\text{int}}, \quad (6)$$

where all lengths are reduced by the half-width of the channel  $a$ ;  $\gamma = qT/(\mu U a^2)$  and  $\sigma = q\rho g \sin \omega/(\mu U)$  are two dimensionless control parameters. Using the following parametrization of the shape:

$$z(\theta) = - \int d\theta (\sin \theta) / K, \quad (7)$$

$$x(\theta) = \int d\theta (\cos \theta) / K, \quad (8)$$

and differentiating Eq. (6) with respect to the angle  $\theta$ , we find

$$\gamma K \frac{dK}{d\theta} = -\sin \theta + \sigma \cos \theta \quad (9)$$

and finally

$$K = [(2/\gamma)(\cos \theta + \sigma \sin \theta + C)]^{1/2}. \quad (10)$$

This expression for  $K$  contains a nonsymmetric term proportional to  $\sigma$  and corresponds to Eq. (9) of Ref. [12] for  $\sigma = 0$ . The integration constant  $C$  can be found by matching the solution (10) to the solution in the inner region. Note that  $C$  as well as a parameter  $\sigma$  should be small since small curvature and small contact angle are required for the matching to the inner region.

In the narrow gaps between the finger and the side walls the potential  $\Phi$  can be taken as a constant across the thickness of the layer and equal to its value (3) on the interface. This approximation allows us to derive an ordinary differential equation for the shape of the gap. Following directly the derivation of Ref. [12], we get

$$h_i(z) \left( \gamma \frac{d^3 h_i}{dz^3} \pm \sigma \frac{dh_i}{dz} - 1 \right) = -\delta_i. \quad (11)$$

Here  $i = t, b$  and the indexes  $t$  and  $b$  denote the top and bottom gaps, respectively;  $h_i(z)$  is the local thickness of the layer:  $h_t(z) = 1 - x_t(z)$  and  $h_b(z) = -1 + x_b(z)$ ;  $\delta_i \ll 1$  is the asymptotic value of the thickness, which is different now for the top and bottom gaps. Using the rescaling  $z = (\delta_i \gamma)^{1/3} \tilde{z}$  and  $h_i = \delta_i \tilde{h}_i$  we arrive at the parameterless equation

$$\tilde{h} \frac{d^3 \tilde{h}}{d\tilde{z}^3} = \tilde{h} - 1, \quad (12)$$

which is identical to the equation given in Ref. [12]. In this rescaling equation we have neglected the term  $\sigma \delta^{2/3} \gamma^{-1/3} d\tilde{h}/d\tilde{z}$  since  $\delta \ll 1$ .

As shown in Ref. [12], Eq. (12) has the following asymptotic behavior: for  $\tilde{z} \rightarrow -\infty$

$$\tilde{h} = 1 + \alpha \exp(\tilde{z}), \quad (13)$$

and for  $\tilde{z} \rightarrow \infty$

$$\tilde{h} = \frac{\tilde{z}^3}{6} + \frac{\beta\tilde{z}^2}{2} + \eta\tilde{z} + \tau - 3\ln(\tilde{z}), \quad (14)$$

where  $\alpha$ ,  $\beta$ ,  $\eta$ , and  $\tau$  depend on the origin of  $\tilde{x}$ , but the quantity

$$\eta^* \equiv \eta - \beta^2/2 \approx 3.18 \quad (15)$$

is translationally invariant [12]. The matching procedure is very close to the procedure in Ref. [12], the only difference is that we have two matching conditions for the bottom and top gaps:

$$\theta_{mb} = -\pi/2 + \gamma^{-1/3}\delta_b^{2/3}\eta, \quad (16)$$

$$\theta_{mt} = \pi/2 - \gamma^{-1/3}\delta_t^{2/3}\eta, \quad (17)$$

$$K_{mi}^2 = \gamma^{-4/3}\delta_i^{2/3}\beta^2. \quad (18)$$

The index  $m$  denotes the matching point. Combining Eqs. (10) and (15)–(18), we get

$$\delta_b^{2/3} = \gamma^{1/3}(|C| + \sigma)/\eta^*, \quad (19)$$

$$\delta_t^{2/3} = \gamma^{1/3}(|C| - \sigma)/\eta^*. \quad (20)$$

Finally, the condition which allows us to find the integration constant  $C$  is that the meniscus fills the channel almost completely. From Eqs. (8) and (10) we get

$$\left(\frac{\gamma}{2}\right)^{1/2} \int_{\theta_{mb}}^{\theta_{mt}} \frac{d\theta \cos \theta}{(\cos \theta + \sigma \sin \theta + C)^{1/2}} = 2. \quad (21)$$

We can expect that  $C$  and  $\sigma$  will be of the order of  $\delta^{2/3}$  [see Eqs. (19) and (20)]. It means that in Eq. (21) we can replace the limits of integration by  $-\pi/2$  and  $\pi/2$  and neglect the nonsymmetric term  $\sigma \sin \theta$  in the denominator (due to the symmetry reason the integration gives only a quadratic dependence of  $\sigma$ ). By these simplifications we get an expression for  $C$ , which is identical to the expression given in Ref. [12]:

$$C = -A(\gamma^* - \gamma), \quad (22)$$

$$\gamma^* = \frac{2}{[\int_0^{\pi/2} (\cos \theta)^{1/2} d\theta]^2} \approx 1.4, \quad (23)$$

$$A = \frac{\int_0^{\pi/2} (\cos \theta)^{1/2} d\theta}{\gamma^* \int_0^{\pi/2} (\cos \theta)^{-1/2} d\theta} \approx 0.325. \quad (24)$$

Combining Eqs. (19), (20), and (22), we get the final result for the thickness of the gaps as a function of the control parameters  $\gamma$  and  $\sigma$ ,

$$\delta_b^{2/3} = (\gamma^*)^{1/3}[A(\gamma^* - \gamma) + \sigma]/\eta^*, \quad (25)$$

$$\delta_t^{2/3} = (\gamma^*)^{1/3}[A(\gamma^* - \gamma) - \sigma]/\eta^*. \quad (26)$$

These predictions generalize Eq. (25) of [12]. The numerical value of  $\eta^*$  has been corrected according to Ref. [11] of Ref. [16]. Note that all constants in Eqs. (25) and (26) come from the theory of the symmetrical finger. The dependence on the asymmetry parameter  $\sigma$  is remarkably simple. The main effect of gravity is just a rotation of the outer meniscus by a small angle  $\sigma$  [see Eq. (10):  $\cos \theta + \sigma \sin \theta \approx \cos(\theta - \sigma)$  for small  $\sigma$ ]. For more

details and experimental study of the effect of gravity on ST fingers see [10].

### III. SAFFMAN-TAYLOR PROBLEM IN THREE DIMENSIONS

In this section we discuss the ST problem in a three-dimensional geometry, i.e., the ST problem in a tube. As we mentioned in the Introduction, though the problem can be easily defined mathematically, the formalism no longer describes the physical problem of a multiphase flow in a simple cell. Nevertheless, as was argued in [14], the ST problem in the tube can yield information about ensemble-averaged diffusion-limited aggregation in the same geometry and can serve as a testing ground for selection theory.

Let us first discuss the axisymmetric case,  $\sigma = 0$ . We note that the inner equation (12) and the matching conditions (16)–(18) remain unchanged because of the small scale ( $\delta \ll 1$ ) and the three-dimensional effects are unimportant. On the other hand, the outer equation (6) contains three-dimensional curvature and cannot be solved analytically. Let us present Eq. (6) for the meniscus shape  $z(r)$  in the form

$$\frac{z''}{(1+z'^2)^{3/2}} + \frac{z'}{r(1+z'^2)^{1/2}} + \sin \theta_m = -z/\gamma. \quad (27)$$

We chose the origin of  $z$  at the matching point  $r = 1$  and set the first term in the curvature to zero at this point. It means that we set  $\beta = 0$  and  $\eta = \eta^*$  in the matching conditions (16)–(18). At the matching point  $r = 1$  we have

$$-z' = \tan \theta_m \gg 1. \quad (28)$$

For arbitrary values of the parameter  $\gamma$ , the solution of Eq. (27) with these boundary conditions behaves like  $z = \pm\gamma/r$  for small  $r$ . A solution that is smooth at the origin exists only for some special value of  $\gamma$ , which depends on  $\theta_m$ . For  $\theta_m$  close to  $\pi/2$  by numerical integration of Eq. (27) we found

$$\theta_m = \frac{\pi}{2} - A_3(\gamma_3^* - \gamma), \quad (29)$$

with  $\gamma_3^* \approx 1.19$  and  $A_3 \approx 0.36$ . The subscript 3 denotes that these numbers correspond to the three-dimensional case. Combining Eqs. (17) and (29), we find the dependence  $\delta$  on  $\gamma$  for the three-dimensional case:

$$\delta^{2/3} = (\gamma_3^*)^{1/3} A_3(\gamma_3^* - \gamma)/\eta^*. \quad (30)$$

It was noted in Ref. [12] that in the two-dimensional case there is a countable set of solutions characterized by the appearance of  $n$  bumps and  $n - 1$  troughs. Indeed, Eq. (9) for  $\sigma = 0$  can be presented as a pendulum equation, and these solutions where the pendulum performs an odd number of oscillations are admissible. It means that for one of the oscillations of the  $n$ th branch the effective channel width equals  $2/(2n - 1)$ . Therefore the upper bounds  $\gamma_n^*$  for these successive branches behave like  $\gamma_n^*/(2n - 1)^2$ . For the three-dimensional case we also have a discrete set of solutions and for large  $n$

we can use the result of the two-dimensional theory, because three-dimensional effects become important only in the small region near the origin ( $r \sim 1/n$ ). Levine and Tu [14] performed the numerical simulation for the three-dimensional problem and found the set of solutions. However, the solution branches merge and disappear in pairs as  $\gamma$  becomes small enough. Our theory is valid in the limit of  $\gamma$  close to  $\gamma_3^*$  where the system does not exhibit branch merger.

The main effect of the transverse gravitational force (for example, applied along the  $x$  direction) is just a rotation of the outer meniscus by a small angle  $\sigma$  around the  $y$  axis. It gives the correction  $\sigma \cos \phi$  to the matching angle  $\theta_m$  in Eq. (29) ( $\phi$  is the azimuthal angle) and for  $\delta$  instead of Eq. (30) we get

$$\delta^{2/3}(\phi) = (\gamma_3^*)^{1/3} [A_3(\gamma_3^* - \gamma) + \sigma \cos \phi] / \eta^*. \quad (31)$$

This result generalizes Eqs. (25) and (26) to the three-dimensional case. Another possibility to get a nonaxisymmetric shape is to introduce the anisotropy of surface energy. In order to discuss this possibility we turn now to an apparently different problem, namely, the directional solidification in a capillary tube, where the anisotropy of surface energy at the solid-liquid interface is more realistic.

#### IV. THREE-DIMENSIONAL NONAXISYMMETRIC PATTERNS IN DIRECTIONAL SOLIDIFICATION

In most common directional solidification experiments, one is drawing at constant velocity a dilute binary mixture across a linear temperature gradient. Let us discuss the three-dimensional case where the external geometry is given by a small cylindrical tube with the radius  $a$ . We consider here the one-sided model of directional solidification (diffusion only in the liquid part) with locally parallel liquidus and solidus lines and with a constant miscibility gap  $\Delta C$  (for details see [2]). If  $u = (C - C_\infty) / \Delta C$  ( $C$  is the impurity concentration in the liquid,  $C_\infty$  its value far ahead of the solidification front) designates the dimensionless concentration, the mass diffusion equation reads

$$\nabla^2 u + P \frac{\partial u}{\partial z} = 0, \quad (32)$$

where lengths are reduced by the radius of tube  $a$  and Peclet number is  $P = Ua/D$ , where  $U$  is the pulling velocity and  $D$  is the diffusion coefficient. At the liquid-solid interface the conservation law says

$$P \cos \theta = - \frac{\partial u}{\partial n}. \quad (33)$$

Here  $\theta$  is the angle between the local normal vector  $\mathbf{n}$  on the interface and the  $z$  direction. At the tube's boundary there is no flux and

$$\left. \frac{\partial u}{\partial r} \right|_{r=1} = 0. \quad (34)$$

Far ahead of the front  $u(z = \infty) = 0$ . Finally, using the fact that the actual temperature is shifted from the

melting temperature due to the impurities and due to the Gibbs-Thomson effect we obtain the last condition at the interface,

$$u = 1 - z_{\text{int}}/l_T - d_0 \Upsilon. \quad (35)$$

Here  $l_T = m_l \Delta C / (aG)$  is the dimensionless thermal length with  $G$  the applied thermal gradient and  $m_l$  the liquidus slope;  $d_0 = \nu_0 T_m / (aLm_l \Delta C)$  is the dimensionless capillary length proportional to the isotropic part of the surface energy  $\nu_0$  ( $T_m$  is the melting temperature of the pure substance and  $L$  the latent heat);  $\Upsilon$  is the well known [2] Gibbs-Thomson shift in the equilibrium melting point

$$\Upsilon = \frac{1}{R_1} \left( \nu + \frac{\partial^2 \nu}{\partial \Theta_1^2} \right) + \frac{1}{R_2} \left( \nu + \frac{\partial^2 \nu}{\partial \Theta_2^2} \right), \quad (36)$$

where  $R_1, R_2$  are the local principal radii of curvature of the surface,  $\Theta_1, \Theta_2$  are the angles between the normal  $\mathbf{n}$  and the local principal directions on the surface, and  $\nu(n)$  is the dimensionless anisotropic surface energy which is just 1 in the isotropic case.

It is convenient to introduce a new field  $w = [2\chi / (2\chi - 1)](1 - u/P - z/2\chi)$  (here  $2\chi = l_T P$ ) and to rewrite Eqs. (32)–(35) in terms of  $w$ ,

$$\nabla^2 w + P \frac{\partial w}{\partial z} = - \frac{P}{(2\chi - 1)}, \quad (37)$$

$$\cos \theta = \frac{\partial w}{\partial n}, \quad (38)$$

$$w = \gamma[\Upsilon], \quad (39)$$

where we used the same notation  $\gamma$  for the parameter  $\gamma = (d_0/P)2\chi/(2\chi - 1)$  in order to emphasize the relation to the Saffman-Taylor problem.

For the isotropic case the analysis is basically the same as in Ref. [12] for the two-dimensional case. Let us recall the main points. In the limit of small Peclet numbers three regions can be distinguished.

(i) Far ahead of the interface the field  $u$  decays exponentially along the  $z$  direction.

(ii) In the tip region the terms linear in  $P$  are negligible compared to the Laplacian and the problem is identical to the three-dimensional Saffman-Taylor problem, which we discussed in the preceding section. The gap  $\delta_0$  depends on  $\gamma$  and is described by Eq. (30) for  $\gamma$  close to  $\gamma^*$ .

(iii) Far behind the tip ( $z \rightarrow -\infty$ ) due to the presence of a temperature gradient the gap becomes exponentially small,

$$1 - r(z) = \delta(z) = \delta_0 \exp[Pz/(2\chi - 1)]. \quad (40)$$

We will return to this point later.

By invoking the global conservation law it is possible to select the position of the tip region in the temperature field as a function of  $\delta_0$  [12],

$$\frac{z_{\text{tip}}}{l_T} = \frac{2\chi - 1}{2\chi} 2\delta_0 - 2P\gamma^*b. \quad (41)$$

The last term is small and describes the capillary correction. The numerical factor  $b$  equals 1 for the two-dimensional case and can be found numerically for the three-dimensional case.

We turn now to the anisotropic case. Our main aim here is to show that the nonaxisymmetric shape correction generated in the tip region can be matched smoothly to the asymptotic shape in the tail region. For the closely related problem of free dendritic growth the mentioned matching procedure gives rise to serious difficulties because the deviation from the basic isotropic solution becomes large in the asymptotic region even for small anisotropy [15]. In our case this effect does not appear because of the presence of the tube, and the asymptotic shape in the tail region can be easily found.

Let us begin from the tip region. In this region the main effect is just a correction to the matching angle [Eq. (29)] for the outer meniscus. The equation for the outer meniscus reads

$$\gamma\Upsilon = z + \text{const}, \quad (42)$$

$$\left( \frac{f_n''}{(1+z_0'^2)^{3/2}} + \frac{f_n'}{r(1+z_0'^2)^{3/2}} - \frac{3z_0''z_0'f_n'}{(1+z_0'^2)^{5/2}} - \frac{n^2 f_n}{r^2(1+z_0'^2)^{1/2}} \right) + f_n/\gamma$$

$$= - \left( \nu_n + \frac{\partial^2 \nu_n}{\partial \theta^2} \right) \frac{z_0''}{(1+z_0'^2)^{3/2}} - \left( \nu_n + \cot \theta \frac{\partial \nu_n}{\partial \theta} \right) \frac{z_0'}{r(1+z_0'^2)^{1/2}} + \nu_n \frac{n^2(1+z_0'^2)^{1/2}}{r z_0'}, \quad (45)$$

where  $\tan \theta = -z_0'$ . According to our choice  $\beta = 0$  (see the preceding section), at the matching point  $r = 1$  we have  $z_0'' = f_n'' = 0$  and  $\theta_m \approx \pi/2$ . Thus we can deduce from Eq. (45) that at the matching point  $r = 1$

$$f_n' - n^2 f_n z_0'^2 = |z_0'|^3 (1 - n^2 - f_n/\gamma) + O(|z_0'|) \quad (46)$$

for  $|z_0'| \gg 1$  [it is always possible to set  $\nu_n(\pi/2) = 1$  by a proper choice of  $\epsilon_n$ ]. One can expect that the solution  $f_n(r)$  of Eq. (45) which is smooth at the tip is a function of order 1. Since we assume that, we can also expect from Eq. (46) that at the matching point  $r = 1$  for  $|z_0'| \gg 1$

$$f_n(r=1) = \gamma(1 - n^2) + O(1/z_0'^2), \quad (47)$$

$$f_n'(r=1) = \gamma n^2 (1 - n^2) z_0'^2 + O(1).$$

We can now find the nonaxisymmetric correction to the matching angle ( $\tan \theta_m = -z_0' - \epsilon_n f_n' \cos n\phi$ )

$$\theta_m = \frac{\pi}{2} - A_3(\gamma_3^* - \gamma) + \epsilon_n \gamma n^2 (n^2 - 1) \cos n\phi \quad (48)$$

and, using Eq. (17), the correction to the gap

$$\delta(\phi) = \delta_0 + \delta_n \cos n\phi,$$

$$\delta_n = -\frac{3}{2\eta^*} \delta_0^{1/3} \epsilon_n (\gamma_3^*)^{4/3} n^2 (n^2 - 1), \quad (49)$$

where  $\delta_0$  is given by Eq. (30) (we assumed that  $\epsilon_n \ll \delta_0^{2/3}$ ). This shape of the gap cannot be preserved and will

where  $\Upsilon$  is given by Eq. (36). For the isotropic case ( $\nu = 1$ ) it reduces to Eq. (27) with solution  $z_0(r)$ . Let us present the anisotropic surface energy in the form

$$\nu(\theta, \phi) = 1 + \epsilon_0 \nu_0(\theta) + \epsilon_n \nu_n(\theta) \cos n\phi, \quad (43)$$

where  $\phi$  is an azimuthal angle,  $n$  is an integer which depends on the symmetry of a growing crystal, and  $\epsilon_n$  describes the strength of the anisotropy. For simplicity we restrict ourselves just to one azimuthal harmonic. In the linear approximation with respect to small  $\epsilon$  we can look for a solution of Eq. (42) in the form

$$z(r, \phi) = z_0(r) + \epsilon_0 f_0(r) + \epsilon_n f_n(r) \cos n\phi. \quad (44)$$

The axisymmetric shape correction  $f_0(r)$  gives, finally, a small (proportional to  $\epsilon_0$ ) correction to the matching angle, which means just a small renormalization of  $\gamma^*$  in Eq. (30) for the axisymmetric part of the gap. We are mainly interested in the nonaxisymmetric correction to the gap. Linearization of Eq. (42) with respect to  $\epsilon_n$  gives the following linear equation for  $f_n(r)$ :

change in the tail region due to two reasons: (i) the wrinkled interface tends to the equilibrium shape according to the Wolf construction, which corresponds to  $\delta_n \approx -\epsilon_n$ ; (ii) both  $\delta_0$  and  $\delta_n$  will decay because of the presence of a temperature gradient. In order to describe these effects in the tail region let us go back to the original Eqs. (37)–(39). In this region the shape of the interface can be written as

$$r(z, \phi) = 1 - \delta_0(z) - \delta_n(z) \cos n\phi, \quad (50)$$

where  $\delta(z)$  is assumed to be small. We expect that the characteristic scale along the  $z$  direction will be much larger than 1 and we neglect all derivatives with respect to  $z$  in Eqs. (37) and (39). It means that the Gibbs-Thomson shift  $\Upsilon$  [Eq. (36)] is just

$$\Upsilon = 1 - (n^2 - 1)[\delta_n(z) + \epsilon_n] \cos n\phi \quad (51)$$

and the field  $w$ , which satisfies diffusion equation (37) and boundary conditions (34), (39), is

$$w = \gamma - \frac{P}{2(2\chi - 1)} \left( \frac{r^2}{2} - \ln r \right) - \left( \frac{\gamma}{2} \right) (n^2 - 1)$$

$$\times [\delta_n(z) + \epsilon_n] (r^n + r^{-n}) \cos n\phi. \quad (52)$$

Using Eq. (38), we get the following equations for  $\delta_0(z)$  and  $\delta_n(z)$ :

$$\delta_0'(z) = \frac{P}{2\chi - 1} \delta_0(z), \quad (53)$$

$$\delta'_n(z) = \frac{P}{2\chi - 1} \delta_n(z) + \gamma(n^2 - 1)[\delta_n(z) + \epsilon_n]n^2\delta_0(z). \quad (54)$$

The solution of Eq. (53),

$$\delta_0(z) = \delta_0 \exp[Pz/(2\chi - 1)], \quad (55)$$

describes an exponential decay of  $\delta_0(z)$  and recovers the already mentioned result (40). The solution of linear Eq. (54) can also be presented in the closed form, but let us describe several limiting cases. For

$$\delta_0 n^2 (n^2 - 1) \ll P/(2\chi - 1) \quad (56)$$

we can neglect the term proportional to  $\delta_0(z)\delta_n(z)$  in the whole region of  $z$ . By this simplification the solution of Eq. (54) is

$$\delta_n(z) = [\delta_n + \epsilon_n \gamma \delta_0 n^2 (n^2 - 1)z] \exp[Pz/(2\chi - 1)], \quad (57)$$

where  $\delta_n$  is given by Eq. (49). The final asymptotic behavior ( $P|z|/(2\chi - 1) \gg 1$ ) is always described by Eq. (57) (without the term  $\delta_n$ ) because in this region we can always drop the mentioned term. In the case opposite to (56) we have another intermediate asymptotic behavior for  $P|z|/(2\chi - 1) \ll 1$ . In this region we can replace  $\delta_0(z)$  by  $\delta_0$  and drop the first term on the right-hand side of Eq. (54). It gives

$$\delta_n(z) = -\epsilon_n + (\delta_n + \epsilon_n) \exp[\gamma \delta_0 n^2 (n^2 - 1)z]. \quad (58)$$

This expression describes the relaxation of the wrinkled interface with “initial” amplitude (49) to the “equilibrium” shape  $\delta_n = -\epsilon_n$ , which corresponds to the Wolf construction, and holds in the intermediate region of  $z$ ,

$$(2\chi - 1)/P > |z| \gg [\gamma n^2 (n^2 - 1)\delta_0]. \quad (59)$$

For larger  $|z|$ ,  $\delta_n(z)$  decays according to Eq. (57).

In the presence of a transverse temperature gradient  $G_t$  there is a correction to the gap which is proportional to  $\cos \phi$ . In the tip region it is described by Eq. (31) with  $\sigma = (G_t/G)/(2\chi - 1)$ . In the tail region this harmonic decays according to Eq. (57) with  $n = 1$  and  $\sigma$  instead of  $\gamma \epsilon_n n^2 (n^2 - 1)$ .

We note that due to the additional power of  $z$  in Eq. (57) the non-axisymmetric correction  $\delta_n(z)$  becomes larger than the axisymmetric one for any small  $\epsilon_n$  or  $\sigma$ . It means that a crystal sticks to the wall at some finite distance  $z$ , where  $\delta_0(z) = |\delta_n(z)|$ .

## V. CONCLUSION

In this paper we have presented a theoretical description of nonsymmetric pattern formation caused by various effects: transverse gravitational force in the Saffman-Taylor problem, or transverse temperature gradient and anisotropy of surface energy in three-dimensional directional solidification. In the problem of three-dimensional directional solidification we have shown that the nonax-

isymmetric shape correction, generated in the tip region, can be matched smoothly to the asymptotic shape in the tail region. The most important conclusion to be drawn from this result is a new interpretation of dendritic growth. We can think of the selection as happening only at the tip of the dendrite while the matching of the tail is always possible, since the mathematical structure of the relevant equations (53) and (54) is that of an initial value problem. This statement will be made more precise below. In the case of growth in a cylindrical tube, this leads to a fully analytic solution of both the selection and shape problems including the tail region. No assumptions about rotational symmetry have to be invoked.

In addition, our result has very important consequences for the other growth situations, especially for free dendritic growth. It frees us from the idea that selection somehow requires matching the tail region to the tip. Instead the tail shape follows from that of the tip after solution of the selection problem for the tip where the needle crystal is still close to the Ivantsov solution to render linearization possible. For the three-dimensional case, this selection problem has been solved in Ref. [15]. Using the same ideas as for the growth in a tube, we now have the key for an understanding of three-dimensional free dendritic growth. It will not be possible to make the same amount of analytic progress as in growth in a tube, but our approach essentially solves the problem with details to be worked out numerically. The crucial point is that the nonaxisymmetric shape correction, generated in the tip region, can be used as an “initial” condition for the “time”-dependent two-dimensional (cross-section) problem of the interface evolution. For the steady-state growth in the  $z$  direction  $z$  plays the role of time for this two-dimensional problem. For example, Eqs. (53) and (54) are evolution equations and the values of  $\delta_0$ ,  $\delta_n$  in the tip region are the initial conditions for these equations. The important difference between free dendritic growth and growth in the tube is that for free growth the deviation from the basic isotropic Ivantsov solution remains small only in the initial period of the evolution. This initial deviation is given by selection theory [15]. As “time” goes on, the deviation increases due to the Mullins-Sekerka instability and a nonlinear theory must be applied. The two-dimensional time-dependent problem of the interface evolution in the presence of the anisotropic surface tension has been considered in [17] in the framework of a Laplacian approximation. After some transition time the system shows an asymptotic behavior which is independent of the initial conditions. Four well-developed arms are formed (for fourfold symmetry). The length of the arms increases in time as  $t^{3/5}$  and the width of the arms increases as  $t^{2/5}$  (see figures in [17]). This scaling law and the shape of the arms have been found in [17] both numerically and analytically. In our case it means that the length and width of the arms increases as  $|z|^{3/5}$  and  $|z|^{2/5}$ , respectively, where  $|z|$  is the distance from the dendritic tip. This shape, which is very different from the Ivantsov paraboloid, should hold as long as the two-dimensional diffusion length is larger than the size of the arms, before subsequent crossover to the classical constant velocity regime of the two-dimensional dendrite.

In this regime the length of the arms increases as  $|z|$ . A more detailed description is given in [18].

Another interesting and analytically unsolved problem is a broken-parity finger discovered via numerical simulations of a crystal growing in a channel without any external asymmetrical force [19, 20]. This problem resists even a qualitative analytical explanation because of the lack of a zeroth-order solution playing the role of an exact solution [21] for the ST problem. The model [22],

which has been successful for symmetrical fingers growing in a channel, cannot explain this result because of the same reasons that there are no asymmetrical fingers in the ST problem without external asymmetrical forces.

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