Crystal growth in a channel: Numerical study of the one-sided model

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The growth of two-dimensional crystals with and without anisotropy in a channel is analyzed by a Green's-function method. A one-sided diffusional model is treated in quasistationary approximation. Our numerical results on the steady-state growth of symmetrical fingers are in agreement with approximate analytical predictions (E. Brener, M. Geilikman, and D. Temkin, Zh. Eksp. Teor. Fiz. **94**, 241 (1988) [Sov. Phys. JETP **67**, 1002 (1988)]). For fixed supercooling the dependence of growth velocity versus channel width is nonmonotonic, passing a maximum. We did not find stationary solutions for supercooling less than 0.5. When the width of the channel exceeds some critical value we observed that the symmetrical finger becomes unstable against tip splitting. In this case we found stable steady-state growth of nonsymmetrical fingers. We have also found an expected instability of two fingers in the common diffusion field caused by competition.

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I. INTRODUCTION

The growth of a crystal in a channel is expected to show pattern selection similar to the problem of a free dendrite (for a review see [1–3]). For the case of a free dendrite, the anistropy of the surface energy has a strong influence on the types of possible patterns—there are no dendrites without anistropy. For crystal growth in a channel, interaction with the channel walls through the diffusion field allows stable stationary patterns even without anistropy. It has been concluded by Pelce and Pumir [4] that in the limit of small Péclet numbers $p = vw/2D \to 0$ (v is the growth velocity, w is the channel width, D is the diffusion coefficient), formally this problem is equivalent to the Saffman-Taylor problem [5]. The following results have been obtained by Kessler, Koplik, and Levine [6] within the limit $p \to 0$. For isotropic surface energy the stationary growth is only possibly if the dimensionless supercooling $\Delta > \frac{1}{2}$ and the resulting velocity is $v \sim w^{-2}(\Delta - \frac{1}{2})^{-3/2}$ (a similar result was obtained in the Saffman-Taylor problem). This would mean that the growth velocity decreases with increasing supercooling Δ and therefore this solution should be irrelevant to real dendritic growth (but not to the Saffman-Taylor problem). Indeed, it has been shown by Pelce [7] that this solution is unstable. However, Brener, Geilikman, and Temkin [8] have found a second branch of solutions on which the growth velocity is higher and increases with supercooling. When the surface energy becomes anisotropic, the second branch describes a transition to the growth of a free dendrite within the limit $w \to \infty$, and seems to be relevant to the physical process. These results seem to be consistent with a numerical linear stability analysis (Kessler and Levine [9]). The second branch is associated with the fact that the interface profile differs form the Saffman-Taylor profile due to the finite growth velocity (in other words, due to the difference between the Laplace and the diffusion equations).

A prerequisite for an analytical treatment of this problem is knowledge of the interface profile in the limit of zero surface energy. Since the exact solution is not available for arbitrary Péclet number, a model expression was used in Ref. [8] (the exact result was obtained only in the limit $\Delta \to \frac{1}{2}$, where the Péclet number becomes small).

One aim of the present paper is to find dynamically selected stable patterns depending on the growth conditions. Also we have investigated some time-dependent properties of this system like tip splitting of a finger or competition between fingers in the common diffusion field. All these properties presumably are important for understanding of the pattern formation in so-called "dense-branching" morphology. This will be discussed shortly at the end of the paper.

II. BASIC EQUATIONS AND NUMERICAL METHOD

We consider the two-dimensional growth of a crystal in a channel of width w with nonpermeable walls. We assume that crystallization involves one conserved quantity such as crystallizing material or latent heat of freezing which by diffusion has to be transported towards or away from the moving solidification front. We assume that diffusion takes place only outside the solid phase. This leads to the so-called one-sided model. For simplicity of notation here, we speak of *temperatures* as the diffusing quantity, although the one-sided model in principle is more appropriate for chemical diffusion. With that in mind, the dimensionless temperature field u is governed by the thermal-diffusion equation

$$\frac{\partial u}{\partial t} = D\nabla^2 u \ ; \quad u \to 0 \ , \quad y \to \infty \ . \tag{1}$$

Here $u = (T - T_\infty)/(Lc^{-1})$, T is the temperature field, T_∞ is the temperature far away from the crystal, D is the thermal diffusivity, L is the latent heat, c is the specific heat, and y is the axis along the channel. At the walls the boundary condition is

$$\left.\frac{\partial u}{\partial x}\right|_{x=\pm w/2}=0 \ . \tag{2}$$

The latent heat generated during the freezing process has to be carried away through the diffusion field. This requires a continuity condition at the interface,

$$v_n=-D\hat{\mathbf{n}}\cdot\nabla u \ . \tag{3}$$

v_n is the normal velocity, and $\hat{\mathbf{n}}$ is the unit vector normal to the interface. Due to the Gibbs-Thomson effect, the temperature along the interface differs from the melting temperature T_m by a term proportional to the local curvature of the interface. The assumption of fast attachment kinetics at the interface implies local thermodynamic equilibrium and so defines the other boundary conditions:

$$u|_{\text{int}}=\Delta-dk \ . \tag{4}$$

Here $\Delta=(T_m-T_\infty)/(Lc^{-1})$ is dimensionless supercooling, k is the local curvature of the interface $\zeta(x,t)$:

$$k=-\frac{\partial^2\zeta/\partial x^2}{[1+(\partial\zeta/\partial x)^2]^{3/2}} \ , \tag{5}$$

and d is the capillary length. Its anistropy is described in terms of the angle θ between the normal vector $\hat{\mathbf{n}}$ and the y direction:

$$d(\theta)=[\gamma(\theta)+\gamma''(\theta)]T_m cL^{-2} \ , \tag{6}$$

where γ and γ'' are surface energy and its second angular derivative. We assume that the crystal has fourfold symmetry and write the following simple model expression for $\gamma(\theta)$:

$$\gamma(\theta)=\gamma_0[1+\bar{\epsilon}\cos(4\theta)] \ ,$$

which implies for the capillary length

$$d=d_0[1-\epsilon\cos(4\theta)] \ , \quad \epsilon=15\bar{\epsilon} \ . \tag{7}$$

Equations (1)–(7) together with initial conditions constitute a complete mathematical description of the problem.

A numerical approach to this problem in quasistationary approximation was developed in Ref. [10]. This approximation means that the diffusion field follows the interface instantaneously. So, instead of the exact Eq. (1), one solves a stationary diffusion equation in a frame moving at velocity v:

$$\frac{1}{D}\frac{\partial u}{\partial t}\approx 0=\nabla^2 u+\frac{v}{D}\frac{\partial u}{\partial y} \ . \tag{8}$$

This does not mean that all the time dependence is lost, because v_n in (3) is still a time-dependent quantity. In free growth the velocity of the coordinate system v should be chosen to closely approximate that of the actual finger (in order to maintain the quality of quasistationary approximation); therefore an additional equation is introduced, to have it relax to the tip velocity of the finger:

$$\frac{dv}{dt}=-\frac{1}{\tau}[v(t)-v_{\text{tip}}(t)] \ , \tag{9}$$

where the relaxation τ is taken small in comparison with all physical time scales; v_{tip} is the velocity of the most advanced point of the finger. Further details of this numerical approach can be found in Ref. [10].

We conclude this paragraph by introducing the dimensionless velocity V and the dimensionless width of the channel W:

$$V=\frac{vd_0}{D} \ , \quad W=\frac{w}{d_0} \ . \tag{10}$$

For steady-state growth (where quasistationary approximation is exact), the global conservation law requires the asymptotic finger width λ to be equal to Δw. The steady-state velocity V is a function of Δ, W, and ϵ. Even instabilities of stationary structures can be exactly located as long as they are not of Hopf type, so we hope that the procedure can describe a number of time-dependent phenomena at least in a semiquantitative sense.

III. NUMERICAL RESULTS

We first look at the behavior of a crystalline finger which is symmetrical about its axis growing in the channel. For the first set of runs we keep the channel width W fixed and vary the supercooling Δ. For Δ smaller than some critical value Δ^* depending on W we never found a steady-state solution. The finger becomes fatter and fatter thereby slowly filling the channel width. The conservation law accordingly forces the velocity of the finger to decrease in time approximately as $t^{-1/2}$ for long times as expected for a flat interface. For $\Delta>\Delta^*$ we found the steady-state behavior after some transition time. The velocity goes to a finite value which depends on Δ, and the width of the finger also reaches an asymptotic value which satisfies the conservation condition $\lambda=w\Delta$ within 1%. The dependence of the steady-state velocity versus supercooling Δ for fixed channel width $W=100$ is presented in Fig. 1. The critical value $\Delta^*=0.67\pm0.01$ was obtained by fitting the data with a square-root dependence of V versus $(\Delta-\Delta^*)$ (solid line in Fig. 1).

For the second set of runs we kept Δ fixed and varied the channel width W. Again we found the steady-state solution only for W bigger than some critical value,

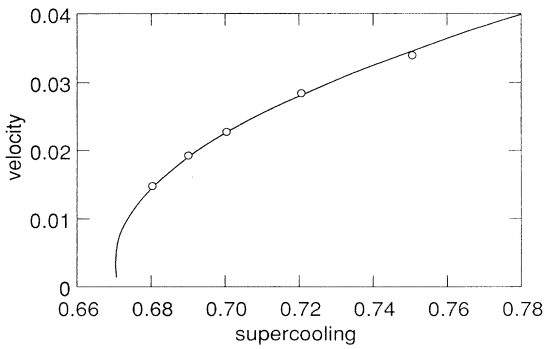

FIG. 1. Growth velocity vs supercooling for fixed (relative) channel width $W=100$. The solid line corresponds to a square-root fit to the data.

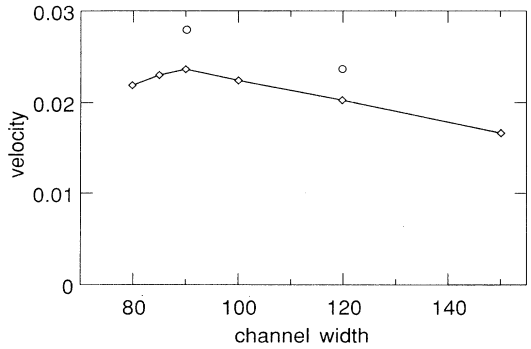

FIG. 2. Growth velocity vs channel width for fixed $\Delta=0.7$. The $\diamond$ symbols correspond to the isotropic case, the open circles to an anisotropy parameter $\epsilon=0.05$ for the capillary length.

which depends on Δ. The dependence of the steady-state velocity V versus channel width W for fixed $\Delta=0.7$ is presented in Fig. 2. This dependence is nonmonotonic, containing a maximum, and in particular the velocity finally decreases with increasing channel width. We also have checked that small anisotropy slightly increases the velocity but does not change the qualitative behavior.

If we increase the channel width up to some new critical value, the symmetrical finger becomes unstable against tip splitting. It is not clear whether this is a *linear* instability. The finger splits into two symmetrical fingers (Fig. 3). Note that there is a mirror symmetry in the middle of the channel along its axis. Therefore, the final configuration here corresponds to a finger in a channel with half the width of the initial one. However, now there is no symmetry implied on the shape of the fingers, its symmetrical shape about the axis being dynamically selected.

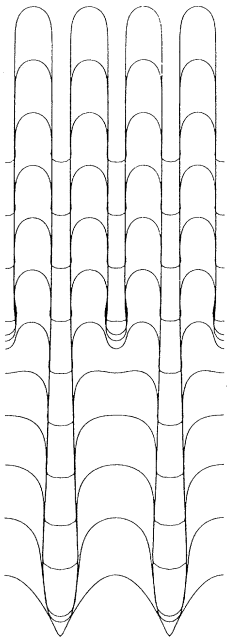

FIG. 3. Splitting of the symmetrical finger: Stroboscopic plot for $\Delta=0.7$, $W=200$.

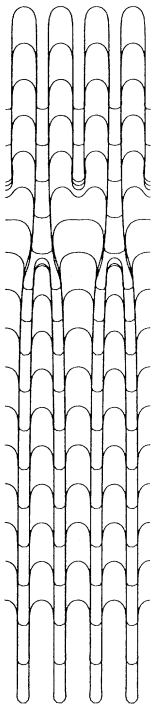

FIG. 4. Two symmetrical fingers competing for the common diffusion field. For details see text.

In the following figures we present some results for the collective behavior of several fingers in a channel. In Fig. 4 there are two symmetrical fingers competing for the common diffusional field (symmetry of both initial fingers implied by the boundary condition). Because of this competition, one of them wins and "kills" the other one. Immediately after this, the winner becomes unstable against tip splitting and arrives at the final situation, as just described (Fig. 3).

Furthermore, we investigated the competition between two fingers in a channel without any additional symmetry conditions. In order to show that competition provides an instability, we started from a symmetrical configuration and looked at the time evolution (Fig. 5). Again, one finger wins in this competition, but now the winner is nonsymmetrical. An attempt to split this finger apparently was not successful. In the final configuration mirror-symmetry is imposed in the troughs corresponding to the channel walls. Clearly, a single nonsymmetric finger (here shown with mirror images) occurs as a stationary solution, which we have checked for stability by a very long run.

Finally we have compared the essentially free dendrite (in a wide channel) with the growth in a channel of a width of about twice the diffusion length. The result is shown in Fig. 6. Parameters were $D=1$, $d_0=0.0003$, $w=0.7$ (0.2), $\Delta=0.58$, $\epsilon=0.1$, leading to $v=22.3$ (18.7) in absolute units. Now the growth speed decreases with decreasing channel width. Note, however, that this is op-

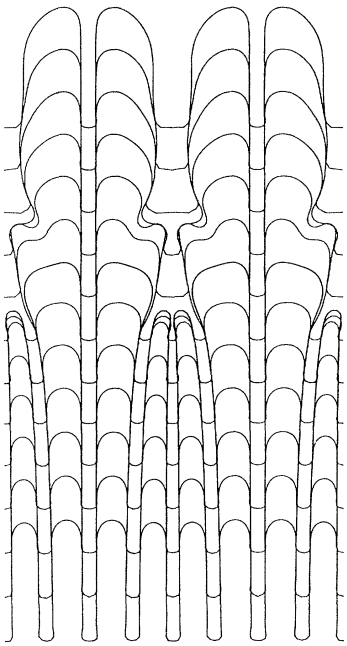

FIG. 5. A single nonsymmetrical finger (here shown with mirror images) as a consequence of a coarsening process. For details see text.

posite to the behavior found for relatively small channel width and for zero or small anisotropy (Fig. 2), as is to be expected.

IV. DISCUSSION

Our numerical results on the steady-state growth of a symmetrical finger are in agreement with analytical predictions of Ref. [8]. For fixed width of the channel, steady-state growth exits only for Δ bigger than some critical value $\Delta^* > \frac{1}{2}$. The growth velocity increases with Δ (Fig. 1) and corresponds to the upper branch of solutions in Ref. [8] in agreement with the arguments given there. We have not found any evidence of another steady-state solution which could correspond to the Saffman-Taylor branch. A recent indication [11] for dendritic solutions without anistropy in a channel seems to be induced by numerical effects.

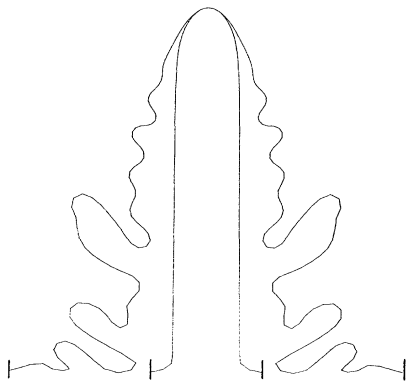

FIG. 6. Comparison of two fingers growing in a wide and in a narrow channel at supercooling $\Delta > \frac{1}{2}$ with capillary anisotropy $\epsilon = 0.1$ (for details see text). The wide-channel dendrite fulfills dendritic scaling relations within 10%. The narrow-channel finger moves about 17% slower due to self-competition for the diffusion field. The diffusion length $2D/v$ in the narrow channel was about half the channel width. All parameters except the channel width were the same.

For fixed Δ the dependence of growth velocity versus channel width is nonmonotonic, passing a maximum (Fig. 2), and again qualitatively agrees with the analytical result of Ref. [8]. Even quantitative agreement exists apart from factors of order unity which presumably come from approximations in Ref. [8]. Inclusion of a small anisotropy does not change the qualitative behavior.

These findings support the analysis given in Ref. [8] stating that there are no stationary solutions for $\Delta < \Delta^*$ and that Δ^* approaches $\frac{1}{2}$ from above as the Péclet number goes to zero.

If the width of the channel exceeds some critical value, we observe that the symmetrical finger becomes unstable against tip splitting (Fig. 3). It is not clear whether this is a linear instability, whether it is induced by external (numerical) noise or whether we did not have appropriate initial conditions when changing the channel width.

When the symmetrical fingers were unstable in that sense, we have found stable steady-state growth of nonsymmetrical fingers (Fig. 5). This is a somewhat unexpected result, since in the theory of Saffman-Taylor fingers there is the statement that only symmetrical finger should be selected [12,13]. On the other hand, in the somewhat related problems of directional solidification and eutectic growth, broken-parity solutions were found [14,15] to appear with increasing wavelength. We have also demonstrated that the competition between two fingers in the common diffusion field leads to the death of one of them. It means that a periodic set of fingers is unstable against coarsening processes because of this competition. On the other hand, tip-splitting processes lead to finer structures.

These two opposite processes finally could be responsible for the formation of so-called "dense-branching" morphology [16], or "seaweed" structure [17,18] in free-growth conditions. Of course, the resulting pattern cannot be expected to be spatially periodic but should be chaotic. Nevertheless, the experiments [18] and computer simulations [19,20] indicate that this structure has a well-defined characteristic length scale and average velocity of the envelope. In our previous paper [17] we assumed the operating point of this structure to be related to the maximum-velocity point (Fig. 2) and consequently to depend only on Δ. Of course, it is still conceivable that the length scale where tip splitting occurs depends on noise, and therefore the length and time scales of the resulting pattern also depend on noise. Even the notion of a specific "phase" (dense branching or compact seaweed) in contrast to "compact dendritic" morphology in this case may become questionable. More work, therefore, is needed to explain the free-growth patterns at low anisotropy in detail.

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