

Phase-field model for dendritic growth in a channel

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We present a phase-field description of dendritic growth in a channel. We observe that both the anisotropic solid-liquid interfacial tension and the geometrical constraint imposed by the channel concur in determining the growth of dendrites: even without interfacial anisotropy there exists a certain critical value of the supercooling Δ above which the governing equations admit steady state solutions and the dendrites advance with constant velocity. In the range considered, for fixed supercooling the growth velocity is a decreasing function of the channel width. When the anisotropy parameter γ is not too low, the computed dendrite tip radius ρ and growth velocity v are consistent with the dependence $\rho^2 v \propto \gamma^{-7/4}$, valid for a free dendrite. On the other hand, for vanishing anisotropy the channel constraint is sufficient to determine a steady growth regime. The present results, taking into account the kinetic undercooling effect and the fully unsteady dynamics of the process, represent an improvement over existing studies based on approximate free boundary models.

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I. INTRODUCTION

The growth of a needle-shaped crystal from an undercooled melt has been addressed in several studies; extensive reviews are given in [1,2]. Experiments conducted on free growth processes [3] show that the dendrite tip is characterized by a radius of curvature ρ and a velocity v that are reproducible functions of the dimensionless supercooling $\Delta = c(T_m - T_0)/L$, where T_0 and T_m represent the initial temperature of the melt and the coexistence temperature of the two phases respectively; c and L are the specific and latent heat per unit volume. Ivantsov's theory [4], neglecting capillarity effects, can only determine the product ρv as a function of Δ . The degeneracy of this solution corresponds to a scale invariance, and is due to the lack of the necessary length scale for the pattern description. Additional physical information is needed to set the scale and to allow for the solution of both the velocity and radius of curvature. This deficiency can be removed taking into account the effects of a finite surface tension σ , that introduces in the model the capillarity length $d_0 = c\sigma T_m/L^2$. In this perspective, the microscopic solvability theory (see [2] for a review) indicates that for a stable and steady tip propagation surface energy is not sufficient, and anisotropy of the surface energy is required. When the dendrite grows in a channel, a preferred direction along the channel's axis is introduced, and stationary patterns are expected even with isotropic surface tension. In the limit of a narrow channel, when the Péclet number $Pe = \lambda v/2D$ is vanishingly small (λ is the channel width and D the thermal diffusivity), Kessler, Koplik, and Levine [5] found, for the isotropic case, that a stationary growth is only possible when $\Delta \geq 0.5$, and the resulting velocity scales as $v \propto \lambda^{-2}(\Delta - 1/2)^{-3/2}$. With anisotropic surface energy the crystal growth is allowed even for $\Delta < 0.5$. According to these results the tip velocity is a decreasing function of the supercooling Δ ; it is an unintuitive behavior and, indeed, these solutions were shown to be unstable [6].

A second branch of solutions was found by Brener, Geilikman, and Temkin with an approximate analytical method [7]. For this new branch the growth rate is an increasing function of Δ and reduces, for $\lambda \rightarrow \infty$, to the solution found for a free dendrite. A successive numerical study [8], based on the quasistationary limit of the free-boundary equations, provided results in qualitative agreement with this model. However, until now no simulations have been performed, based on a fully unsteady formulation of the problem, incorporating the kinetic undercooling effect and allowing for heat diffusion even in the solid phase.

In this paper the phase-field model [9,10] is employed for the numerical simulation of two-dimensional crystal growth in a channel. This approach removes the necessity of tracking the interface position that is found as a part of the numerical solution, and allows a simple treatment even of complicated and interconnected structures. Moreover, it takes into account in a very natural fashion the effects of the interface kinetics that are expected to strongly influence the solution when the supercooling Δ is not too small. On the other hand, the classical free-boundary formulation is recovered asymptotically when the solid-liquid interface is sufficiently sharp [11].

The phase-field model gives a diffuse interface picture of the solidification process, and introduces a length scale, the interface thickness δ . Realistic values of this parameter fall in the range of several atomic dimensions [12]. In two-dimensional numerical calculations the cost of the solution increases as δ^{-4} and, due to the limitations of computing resources, at present even simple problems would result untractable. In this study, along the lines suggested by Wheeler, Boettinger, and McFadden [13], a value is selected for the interface thickness that is small compared to the lowest geometric scale that characterizes the process, namely, the radius of curvature of the dendrite tips, and, nevertheless, more than ten times greater than realistic values.

The numerical results agree with the most significant predictions of the approximate model of Brener, Geilikman, and Temkin [7]. Below a critical value Δ^* of the supercooling Δ no steady solutions are allowed. For $\Delta > \Delta^*$ the tip velocity

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settles to a steady value that increases with Δ . For fixed supercooling, the tip velocity is a decreasing function of the channel width.

The anisotropy of the surface energy plays an important role, extending the range of steady growth to lower values of Δ . When the anisotropy parameter γ is not too low, the dendrite tip radius ρ and the growth velocity v obey the dependence $\rho^2 v \propto \gamma^{-7/4}$ that was proved for a free dendrite [14,15].

II. THE GOVERNING EQUATIONS AND NUMERICAL METHOD

The model describes the solidification of a pure substance in terms of two fields: the scalar phase field ϕ and a dimensionless temperature field $u = c(T - T_m)/L$. The field ϕ is an ordering parameter assuming the values $\phi=0$ in the solid and $\phi=1$ in the liquid; intermediate values correspond to the interface between the two phases. The model is developed along the lines suggested by Penrose and Fife [16,17] and successively followed by Wang *et al.* [18] and Wheeler, Murray, and Schaefer [19]. These authors obtained the dynamic equations by simultaneous variations of an entropy functional with respect to the two fields. The model results in the following dimensionless equations for the phase field and temperature:

$$\frac{\varepsilon^2}{m} \frac{\partial \phi}{\partial t} = \varepsilon^2 \nabla^2 \phi + \phi(1-\phi) \left(\phi - \frac{1}{2} \right) + \varepsilon \alpha \frac{\partial P(\phi)}{\partial \phi} u, \quad (1)$$

$$\frac{\partial u}{\partial t} = \nabla^2 u - \frac{\partial P(\phi)}{\partial \phi} \frac{\partial \phi}{\partial t}. \quad (2)$$

Here and henceforth, lengths are measured in units of some reference length l representative, for example, of the domain sizes, whereas time is measured in units of l^2/D . The function $P(\phi) = \phi^3(10 - 15\phi + 6\phi^2)$, introduced by Wang *et al.* [18], enforces the condition that bulk solid and liquid are described by $\phi=0$ and $\phi=1$, respectively, for every value of u . The solution of these equations guarantees that the total dimensionless entropy of the system, given by

$$\mathcal{S} = \int_{\Omega} \left[s(\phi, u) - \frac{1}{2} \varepsilon^2 (\nabla \phi)^2 \right] d\Omega, \quad (3)$$

increases monotonically in time. In Eq. (3) integration is performed over the computational domain; the last term in the integrand represents a gradient correction to the thermodynamic entropy density, given by

$$s(\phi, u) = \int_0^{\phi} \left[\zeta(1-\zeta) \left(\zeta - \frac{1}{2} \right) + \varepsilon \alpha \frac{\partial P(\zeta)}{\partial \zeta} u \right] d\zeta. \quad (4)$$

Anisotropy of the surface energy can be accounted for allowing the parameter ε , in Eq. (3), to depend on the angle θ , defined as the angle between the normal to the interface and a fixed direction—the channel axis in our calculations. Assuming a dependence $\varepsilon(\theta) = \bar{\varepsilon} \eta(\theta)$, where $\eta(\theta)$ will be defined later, Eq. (1) is modified as [20]

$$\begin{aligned} \frac{\varepsilon^{-2}}{m} \frac{\partial \phi}{\partial t} &= \phi(1-\phi) \left(\phi - \frac{1}{2} \right) + \bar{\varepsilon} \alpha \frac{\partial P(\phi)}{\partial \phi} u \\ &- \bar{\varepsilon}^2 \frac{\partial}{\partial x} \left(\eta(\theta) \eta'(\theta) \frac{\partial \phi}{\partial y} \right) \\ &+ \bar{\varepsilon}^2 \frac{\partial}{\partial y} \left(\eta(\theta) \eta'(\theta) \frac{\partial \phi}{\partial x} \right) \\ &+ \bar{\varepsilon}^2 \nabla \cdot [\eta^2(\theta) \nabla \phi]. \end{aligned} \quad (5)$$

The parameters $\bar{\varepsilon}$, α , and m are related to the physical properties of the substance through the following relations [19]:

$$\alpha = \frac{\sqrt{2} l L^2}{12 c \sigma T_m} = \frac{\sqrt{2} l}{12 d_0}, \quad (6)$$

$$m = \frac{\mu \sigma T_m}{DL}, \quad (7)$$

$$\bar{\varepsilon} = \frac{\delta}{l}, \quad (8)$$

where μ is the interface kinetic coefficient. In the following numerical simulations we put $\alpha=400$, $m=0.05$, and $\bar{\varepsilon}=0.005$; with $l=2.1 \times 10^{-4}$ cm these values mimic as close as possible the thermophysical properties of nickel near its melting temperature. Moreover, we assume

$$\varepsilon(\theta) = \bar{\varepsilon}(1 + \gamma \cos k \theta), \quad (9)$$

where γ specifies the intensity of the anisotropy effects; choosing $k=4$ we enforce fourfold anisotropy.

Equations (2) and (5) have been solved on a two-dimensional domain $0 \leq x \leq x_m$, $0 \leq y \leq \lambda$, characterized by a large value of the aspect ratio x_m/λ , that is varied in the range $18 \leq x_m/\lambda \leq 33$. The length of the channel is fixed at $x_m=6$. Adiabatic conditions were imposed at the channel's boundaries. Initially, in the undercooled melt ($u=-\Delta, \phi=1$) a solid germ ($u=0, \phi=0$) is prepared in the region $x \leq x_0$; the germ's surface is perturbed by a random corrugation, putting $x_0(y) = 0.12(1 + A\chi)$, where $A=0.005$ and χ is a random number uniformly distributed in the range $[-0.5, +0.5]$. The high frequency components of the corrugation spectrum coalesce at the early stage of the growth and the pattern selected by the system is a single finger that propagates into the channel.

To discretize the governing equations, a uniform spatial grid is utilized, with $\Delta x = \Delta y = 0.005$; the Laplace operator is approximated through a five-point formula and an explicit Euler integration scheme is employed to advance forward in time. The ratio of the diffusivities of the two fields $D_u/D_\phi = 1/m$ is very high; as a consequence the time step required to achieve stability is very different for Eqs. (2) and (5). To circumvent this problem and to improve the efficiency of the numerical scheme, the ϕ equation (5) is firstly advanced in time with a step $\Delta t_1 = 0.4 \times 10^{-4}$, then the heat equation (2) is iterated 10 times with a time step $\Delta t_2 = 0.1 \times \Delta t_1$.

III. NUMERICAL RESULTS

The constraint imposed by the side walls is effective when the channel's width λ is not too large with respect to the thermal diffusion length $2D/v$: for $Pe \gg 1$, the free den-

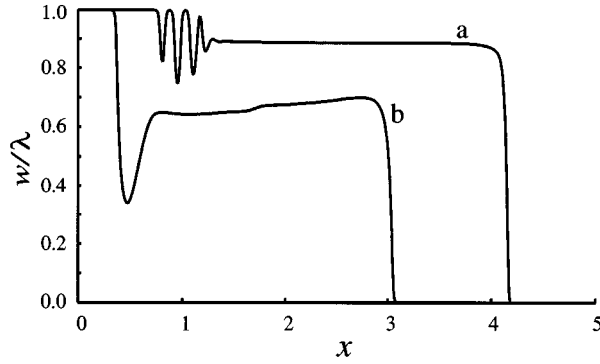


FIG. 1. Width of dendrite, scaled to λ , along the growth direction x . Curve a : at time $t=0.3$, with 0.9 ; curve b : at time $t=2.4$, with $\Delta=0.5$. The channel width is $\lambda=0.18$, and $\gamma=0$ (isotropic surface energy).

solution should be expected. Accordingly, the numerical computations are mainly conducted to focus the growth behavior when the Péclet number is of order one. The numerical results will be presented in terms of nondimensional physical quantities, scaling all the lengths to l , time to l^2/D and velocities to D/l .

The effect of the initial supercooling is investigated first. No anisotropy of the surface energy is assumed, setting $\gamma=0$; the channel width is fixed at $\lambda=0.18$. In these conditions the process selects two different regimes as Δ is varied. For $\Delta \leq 0.63$ stationary growth is not allowed. The width w of the dendrite increases with time and, due to energy conservation, the tip velocity decreases; the expected $t^{-1/2}$ behavior is approached asymptotically. For $\Delta > 0.63$, after a short transient the tip velocity reaches a steady value, and the width stabilizes at the value $w=\lambda\Delta$ required by the energy constraint. These considerations are clarified in Fig. 1, where in the same graph the ratio w/λ is shown along the x direction for two different dendrites. Curve a describes, at time $t=0.3$, a dendrite growing with a supercooling $\Delta=0.9$; the ratio w/λ is settled to a steady value that coincides with Δ . For curve b ($t=2.4$) the supercooling is $\Delta=0.5$; here the finger width is considerably larger than $\lambda\Delta$ and increases with time.

In Fig. 2 a log-log plot represents the growth rate Q versus time and for different values of Δ ; Q is defined as

$$Q = \frac{d}{dt} \int_{\Omega} [1 - \phi(x,y)] dx dy. \quad (10)$$

Notice that for steady growth Q and v are related by $Q = \lambda\Delta v$.

For $\Delta \leq 0.63$ Q is monotonically decreasing with time, while for $\Delta > 0.63$ the curves tend to a saturation level that increases with Δ . As previously stated, the Péclet number is always confined in the range $0.5 < Pe < 1.5$, so that the numerical solution is truly reflecting the effects of the wall's constraint.

The main features of the process are still retained when the effects of anisotropy are accounted for, i.e., when $\gamma \neq 0$, but the range of steady growth is extended towards lower values of Δ . Figure 3 shows the steady tip velocity versus Δ

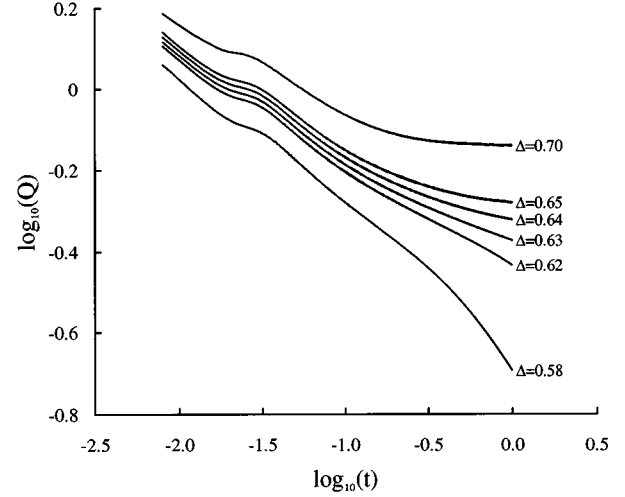


FIG. 2. Log-log plot of the growth rate \dot{Q} vs time, at different values of the supercooling Δ ; the channel width is $\lambda=0.18$ and $\gamma=0$ (isotropic surface energy).

for $\gamma=0$ and 0.02 . Anisotropy results in higher values of v ; with $\gamma=0.02$ the threshold value for steady growth is $\Delta=0.60$.

Figure 4 shows the tip steady velocity versus the channel width λ . Here the supercooling is fixed at $\Delta=0.8$ and we consider the isotropic case $\gamma=0$. In the explored range, v is a monotonically decreasing function of λ .

All the previous results are in qualitative agreement with the predictions of the approximate analytical model of Brener, Geilikman, and Temkin [7]. However, the tip velocities shown in Fig. 3 are considerably smaller than the ones obtained in the successive numerical study [8]; on the contrary our results are comparable with the experimental data of Willnecker, Herlach, and Feuerbacher [21]. This discrepancy had to be expected: the data reported in [8] were obtained neglecting the kinetic undercooling. As pointed out by Pomeau and Ben Amar [2], such an approximation yields a strong overestimate of the tip velocity at large values of Δ .

We now discuss the effects of the anisotropy parameter γ on the computed results. The notion that capillarity is at the origin of the selection mechanism that resolves the ambigu-

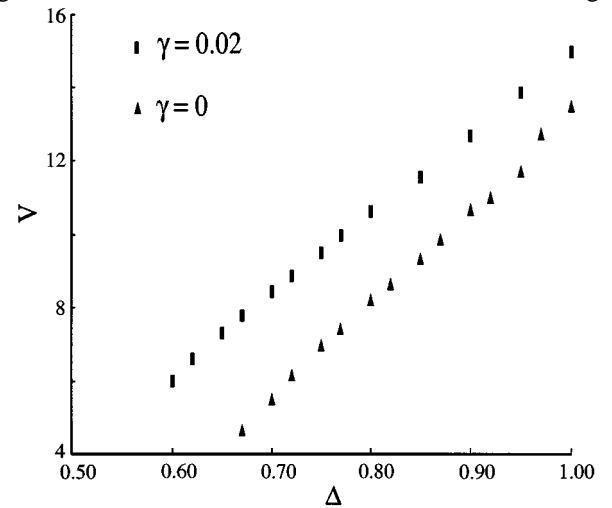


FIG. 3. Steady velocity of the dendrite tip, vs the supercooling Δ ; the channel width is $\lambda=0.18$.

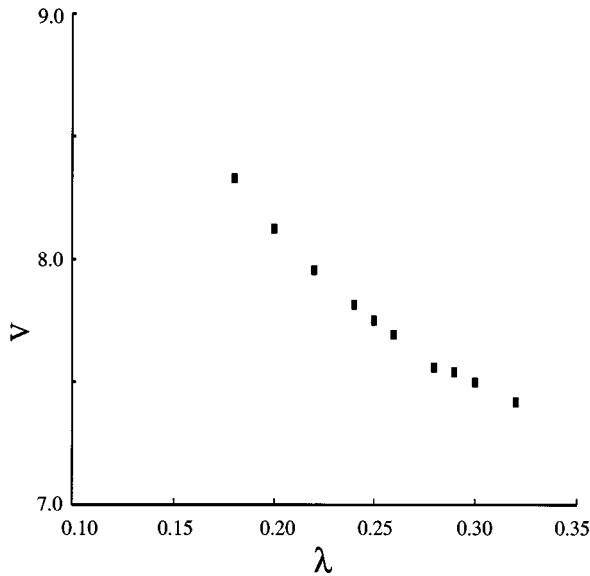


FIG. 4. Steady velocity of the dendrite tip, vs the channel width λ ; $\gamma=0$ (isotropic surface energy), and $\Delta=0.8$.

ity of Ivantsov's solution is now well established. Within this approach, the microscopic solvability theory predicts, for the free growth case, the result $\sigma^* = 2Dd_0/(\rho^2 v) \propto \gamma^{7/4}$. This dependence was confirmed in the numerical study conducted by Wheeler, Murray, and Schaefer [19] with the phase-field model. Brener, Geilikman, and Temkin [7] suggested the same behavior for the growth in a channel, in the limit $Pe \gg 1$. It is not clear whether the same result should apply when the Péclet number is of order one.

The radius of curvature of the dendrite tip was calculated evaluating the expression $\rho = \phi_x / \phi_{yy}$ on the dendrite axis, at the solid-liquid interface defined as the locus $\phi(x, y, t) = 0.5$.

Figure 5 shows $\log_{10}(\rho^2 v)$ versus $\log_{10}(\gamma)$ for $\lambda=0.18$ and $\Delta=0.8$; the Péclet number here is confined in the range $0.8 < Pe < 1.2$. The numerical data fit well the $\gamma^{-7/4}$ power law (the superimposed straight line) when γ is not too low; on the other hand, as $\gamma \rightarrow 0$ the data tend to a finite limit, indicating that the presence of the channel's walls provide the necessary anisotropy to determine a steady growth even without interfacial anisotropy.

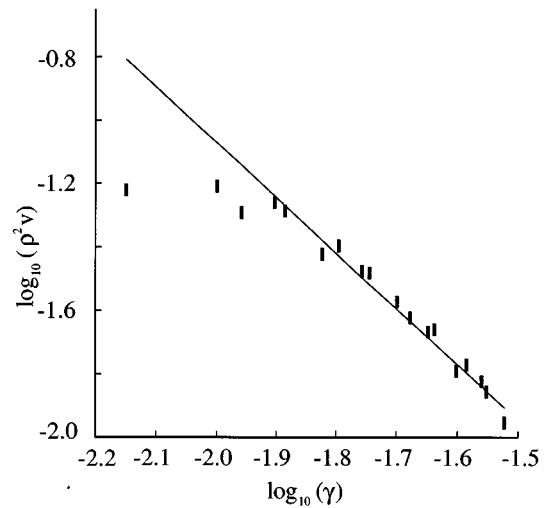


FIG. 5. Log-log plot of the product $\rho^2 v$ vs γ . The superimposed straight line corresponds to the power law $\rho^2 v \propto \gamma^{-7/4}$.

IV. CONCLUSIONS

A numerical study has been conducted on the growth of dendrites in a channel, when the Péclet number is of order one. The fully time dependent equations of the phase-field model were utilized. The results bridge the gap between the numerical study of Wheeler, Murray, and Schaefer [19] on the free dendrite growth, and the one of Brener *et al.* [8] where the growth in a channel is simulated through the free boundary equations, in a one-sided and quasistationary approximation.

Steady solutions for the tip propagation are allowed even with isotropic surface tension, when the supercooling Δ is larger than a threshold value. Anisotropy of the surface tension extends the range of steady solutions towards lower Δ values, and results in larger growth rates. For fixed Δ the tip velocity is a decreasing function of the channel width λ . As the anisotropy parameter γ is varied, the product $\rho^2 v$ shows a saturating behavior at low γ values; as γ increases the numerical results are consistent with the power law dependence $\rho^2 v \propto \gamma^{-7/4}$, predicted by the microscopic solvability theory for a free dendrite.

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