Dendritic growth at very low undercoolings

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Abstract

We have performed numerical simulations of dendritic growth at very low undercoolings in two spatial dimension using a phase-field model. In this regime of growth, the dendrites present sharp corners in the tip region while the trailing region is parabolic, and the corresponding side-branching structures resemble the shape of the tip. The scaling $v\rho^2 \sim \text{constant}$, where $\rho$ is the tip radius of curvature from the fitting to the parabolic trailing region, still holds approximately. We find that the values of $v\rho^2$ are consistent with those given by microscopic solvability theory. The sharpness of the tip region of the dendrite can be characterized in terms of the deviation $\lambda$ with respect to an Ivantsov parabola. We observe that this length scales as $\lambda \sim \rho$, consistent with experimental measurements.
Dendritic crystal growth constitutes one of the most interesting examples of pattern formation phenomena in nonequilibrium dynamical systems \[1,2,3,4,5,6\]. While most studies of dendritic growth have concentrated on relatively high undercoolings, recent experimental measurements in dendritic growth at low undercooling \[7\] have revealed a new feature of this nonequilibrium process, that is, the appearance of a different dendritic morphology with a faceted structure and sharp corners at the dendritic tip. This regime, being close to equilibrium, has been addressed theoretically \[8\] to give an explanation of the self-similarity of the interfacial profile for different undercooling values. There are however still many open questions regarding the dynamical evolution of the pattern, the shapes of the side-branches, the detailed study of the interfacial roughness near the dendritic tip, and, most importantly, whether new elements should be included in the theoretical description, or if such patterns selected by nature are compatible with our current theoretical understanding \[9\].

Motivated by the interesting results of Refs. \[7\] and \[8\], in this paper we use a phase-field model to investigate the small undercooling regime of dendritic growth with an emphasis on steady-state pattern selection. Phase-field models have proven to be a breakthrough for the numerical simulation of unstable interfaces \[10,11,12\], and so far very impressive results have been obtained \[11,13\]. The method is especially useful here since the problem involves high values of interface curvature. Our numerical data show that dendrites with sharp-cornered tips and sharp-shaped side-branchings (see below), are the steady-state pattern at low driving force. With appropriate characterization of the size of the sharp-cornered tip (see below), certain scaling laws hold. It is also found that the relation between growth velocity at the dendrite tip and tip radius at large driving force, still holds here. Finally, our numerical data are consistent with experimental measurements \[7\].

We use the phase-field model proposed for solidification in a pure liquid \[11,12\], which has been shown to provide a reliable means of studying dendritic growth phenomena \[13\]. The model includes two fields. First, a dimensionless temperature field \(u\) is defined as \(u(x, y, t) = (T(x, y, t) - T_S)/(T_S - T_L)\), where \(T_S\) is the melting temperature of the solid phase and \(T_L\) is the temperature of the liquid phase far from the interface. Second, a phase field \(\phi\) is the order parameter of our system; thus \(\phi = 0\) and \(\phi = 1\) represent solid and liquid phases, respectively. The interface locus is determined by positions at which \(\phi = 1/2\). The dynamical equations are the following \[13\]:

\[
\frac{\epsilon^2 \partial \phi}{m \partial t} = \phi(1 - \phi) \left[ \phi - \frac{1}{2} + 30 \epsilon \alpha \Delta u \phi(1 - \phi) \right] + \epsilon^2 L(\eta(\theta)) \phi \tag{1}
\]

and

\[
\frac{\partial u}{\partial t} = -\frac{30}{\Delta} \phi^2 (1 - \phi)^2 \frac{\partial \phi}{\partial t} + \nabla^2 u, \tag{2}
\]

where the generalized Laplacian operator \(L\) is defined as \(\mathcal{L}(\eta(\theta)) = \partial_x (\eta(\theta) \eta'(\theta) \partial_x \cdot) + \partial_y (\eta(\theta) \eta'(\theta) \partial_y \cdot) + \nabla (\eta(\theta)^2 \nabla \cdot)\). The parameters \(\Delta, \alpha\) and \(m\) are defined as functions of the latent heat per unit volume \(L\), the specific heat \(c\), the interfacial energy and mobility \[13\]. In particular the dimensionless undercooling \(\Delta\) is defined as \(\Delta = c(T_S - T_L)/L\). The
interfacial width is given \[14\] by the length scale \(\epsilon\). In the so-called thin interface limit \((\epsilon \to 0)\) \[11\] one indeed obtains the equivalent macroscopic equations \[1\] from the present phase-field model, as shown by McFadden et al. \[14\]. Thus instead of solving macroscopic equations we shall focus on solving the phase-field model directly.

In the above equations the function \(\eta\) accounts for the symmetry of the crystalline structure of the solid, and therefore the anisotropy of the kinetic and surface tension coefficients at the interface. Here, as usual \[4\], we have chosen a fourfold symmetry, \(\eta(\theta) = 1 + \gamma \cos(4\theta)\), where \(\gamma\) is the surface tension anisotropy parameter. We have been motivated to choose this form of anisotropy for two reasons. First, it is the simplest form and is used in the solvability analysis of dendritic growth \[1\], thus comparison to that theory maybe made at least at large driving forces or small \(\gamma\)'s. Second, this form is adequate for the description of the physics of missing orientations \[15,16,17\] in \textit{equilibrium} crystal shapes. It is well known that missing orientations lead to sharp corners in \textit{equilibrium} crystal shapes \[16,19\], hence they should play a role when the growth is very slow. With this form of the anisotropy it is easy to obtain the \textit{equilibrium} missing orientations using the Frank diagram \[18,16\].

We have integrated Eqs. (1) and (2) on a square lattice of width \(w \in [100, 200]\) and height \(h \in [1000, 2000]\) using an Euler algorithm with mesh size \(7.5 \times 10^{-3}\) and time step \(10^{-5}\). The values of the parameters were \(\alpha = 400\), \(m = 0.05\) and \(\epsilon = 5 \times 10^{-3}\) \[13\]. We have investigated the evolution of the system for values of undercooling \(\Delta\) ranging from 0.02 to 0.35. To our knowledge no other numerical simulations at such low undercooling have been carried out so far. A large computational effort is required due to the increasingly demanding time and space factors. For instance the computational time can be estimated to be proportional to a large negative power of \(\Delta\) \[20\]. We have fixed the surface tension anisotropy parameter \(\gamma = 0.1\); in \textit{equilibrium} this would correspond \[19\] to missing orientations of angles \(\theta\) smaller than \(\pm 21.65^\circ\), \textit{i.e.} surfaces with normal angles in this range are thermodynamically unstable and do not appear on the equilibrium crystal shape. Of course we are facing a non-equilibrium situation here, but this construction for \textit{equilibrium} shapes gives us some idea of what might happen. An additive Gaussian white noise of intensity \(10^{-8}\) has been added to Eq. (1) with the purpose only of helping to trigger the growth of side-branches, assuming the role usually played by numerical noise. However noise with such a low intensity cannot affect the tip profile. A detailed description of the stochastic effects on dendritic growth will be presented elsewhere.

Our starting configuration was a small circular solid nucleus in the center at the bottom of the integration grid. We found that dynamical evolution of the unstable interface progresses as follows. First, there was a relatively long transient, in which the temperature evolved from a initial step-like profile to a diffusive one, with a diffusion length comparable to but smaller than the size of the system. The velocity of the interface was constantly adjusting until the temperature flux at the interface acquired a value compatible with the undercooling imposed. Then, the truly dendritic evolution began. We observed that at these very low undercoolings the tip was no longer parabolic, but had a polygonal structure with a sharp corner (Fig. 1). This in some sense resembles the equilibrium crystal shape, namely a “square” with rounded...
edges but still sharp corners for a fourfold anisotropy [16]. In our simulations we constantly monitored the interfacial profile to find the steady-state tip shape, which usually appeared at the precise moment when the first side-branch started to grow. After this moment the sharp-cornered tip structure changed very little with time. We note that the only important difference between this simulation and the previous ones [13] is the very small undercooling, and thus, very small driving force. However notice that the steady-state shape including the shape of side-branches is here quite different from that which appears in the high driving force case. Here growth of side-branches is largely inhibited as their sizes stay more or less constant along the sides of the dendrite. We have checked this side-branching structure by doubling the system size and obtained the same result. The sharp-cornered tip structure can be characterized using a length $\lambda$ which corresponds to the distance between the tip and the topmost point of the fit to a parabolic shape (regions I and II in Fig. 2, respectively). We were motivated by Ref. [7] to use this distance as a useful characterization since it can be measured in the laboratories [7].

To perform such a fit to compute $\lambda$, a problem is that transition between the behaviors of regions I and II is rather smooth, as shown in Fig. 2. The parabolic profile extends down from region I but is cut off because of the appearance of side-branches. To exclude any ambiguity and thus to reduce error, we have found that the quantity $\xi'(x - x_{\text{tip}})/(x - x_{\text{tip}})$, where $\xi_{\text{int}}$ is the interfacial position, provides a valuable criterion to distinguish the sharp-cornered tip from the parabola. In region I, it is essentially hyperbolic; on the other hand, in region II, it reaches a constant value equal to the curvature at the tip of the parabola. Using this the interface belonging to region II could be extracted, and the fit done in the usual way.

In Fig. 3 the curvature as a function of the steady-state velocity is shown. We have obtained the same scaling behavior as that for the purely parabolic dendrites [4], $\rho \sim v^{-1/2}$. In our case the radius $\rho$ is computed using the fitted parabola discussed in the last paragraph. Furthermore, we have found that the values obtained for $\rho^2v$ lie within 7% of the curve corresponding to the prediction of microscopic solvability theory [4] for $\gamma = 0.1$. In this sense the tip structure, although showing clear sharp corners, acts effectively as a parabolic one which fits the trailing region of the dendrite. This fact suggests that the relevant quantity to be taken into account for the sharp-cornered tip shape is $\rho$, rather than the real curvature at the sharp tip, which is 10 to 20 times larger than $1/\rho$ and could be strongly conditioned by the lattice discretization.

The fact that the aforementioned scaling holds, at least for the range of undercooling values considered here, gives a first indication that there is still only one relevant macroscopic length in this growth regime. A further explicit check can be done. In Fig. 4 the measured length $\lambda$, as defined above, as well as the fitted radius of curvature $\rho$ for different values of undercooling is shown. As $\Delta$ decreases (for data points on the right of the figure), the sharp corner at the dendrite tip is easily distinguishable and a scaling $\lambda \sim \rho$ is obtained. This scaling is in agreement with previously reported experimental results [7]. The whole tip structure is therefore self-similar including the tip region of the dendrite. For large values
of undercooling, the fitted $\rho$ is comparable to the radius of curvature at the tip; thus the measure of $\lambda$ is no longer reliable, shown by the saturation of its values on the left part of Fig. [4].

The appearance of sharp-cornered tips as the steady-state velocity is reduced is not abrupt but gradual. This suggests that the transformation from a dendrite with a parabolic tip profile and a strong side-branching process in the higher velocity regime, to a sharp-cornered tip structure as well as “faceted” looking side-branches whose growth is rapidly inhibited (Fig. [1]), is a crossover behavior and not a dynamical transition. This result could be interpreted in terms of a kinetic roughening phenomenon \cite{3,21} where the pattern with sharp-cornered tips at low driving force is kinetically roughened at large driving force, leading to the usual parabolic tips. To explore this idea, we have looked the change in morphology of the dendrite depending on the surface tension anisotropy $\gamma$, at a fixed undercooling value. The inset in Fig. [3] shows $\Theta_{\text{tip}}$ which is the corner angle of the dendrite tip, or more precisely the extrapolation of the two sides around the tip region to $x = x_{\text{tip}}$, as a function of $\gamma$. We observe a gradual increase of $\Theta_{\text{tip}}$ asymptotically to $45^\circ$ which is the angle that, according to the Frank diagram \cite{18} and the Wulff construction \cite{22}, minimizes the equilibrium surface free-energy when $\gamma = 1$. This energetic consideration seems play a role at the high anisotropy, or similarly, lower undercooling regimes. In this regime the nonequilibrium characteristics of the evolution appear to become less important relative to the equilibrium requirement of free energy minimization.

In summary, we have studied the morphology appearing in dendritic growth of a supercooled pure liquid in the low driving force regime. We found that dendrites develop sharp-cornered tips and the growth and coarsening of the side-branches are inhibited at very low undercooling. The scaling behavior for growth at large velocities, $\rho^2 v \sim \text{constant}$, is still valid at low undercoolings if we use the fitted $\rho$ as described above. Furthermore, this value of $\rho^2 v$ is quantitatively consistent with the microscopic solvability theory. We found that there is only one relevant macroscopic length scale in this slow growth regime, such as the fitted $\rho$. The length associated with the size of the sharp corner scales with this fitted $\rho$ as $\lambda \sim \rho$, and this is consistent with experimental measurements. The appearance of the sharp corner is gradual as undercooling decreases or anisotropy increases. Finally we point out that our work focuses on the sharp-cornered tip region of the dendrite, and the steady-state shapes, whereas in the experiment of Ref. [7] true facets were apparent, suggested that it was carried out below the roughening transition of the material. It is not clear how to treat equilibrium roughening transition within a phase-field model where interfaces are diffuse (with thickness $\sim \epsilon$), but this is an interesting problem and should be pursued further in order to quantitatively compare with experiments \cite{7} in the very slow growth regime below the roughening transition. Other interesting directions include detailed calculations of the dynamics of dendritic growth in this low undercooling regime, and the investigation of this regime in three spatial dimensions. We hope to report on these studies in the near future.

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[20] A rough estimate of the computational time in terms of $\Delta$ can be done. To avoid finite size effects the system size must be larger than the diffusion length in the liquid phase. This requirement, together with the scaling of the radius of curvature $\rho \sim \Delta^{-2}$ and velocity $v \sim \Delta^4$, gives the width of the system $w \sim \rho \sim \Delta^{-2}$ as well as the time of evolution $t \sim \rho/v \sim \Delta^{-6}$. Thus the computational time goes as $\Delta^{-8}$. In addition, the time has to be
rescaled by the mesh size, which in turn has to be fine enough to cope with the extremely high curvature.

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FIGURES

FIG. 1. Interfacial profile for $\Delta = 0.25$ at a time $t = 1.00$. The growth of side-branches is inhibited rapidly after their appearance.

FIG. 2. The sharp-cornered tip region of a dendrite, for $\Delta = 0.07$ at a time $t = 3.50$ (squares). Also shown is the definition of $\lambda$, the corresponding parabolic tip (continuous line), as well as the corresponding regions I and II.

FIG. 3. The radius of curvature $\rho$ of the associated parabola of the dendrite as a function of the velocity for different values of $\Delta$ (circles). The continuous line corresponds to a $\rho \sim v^{-1/2}$ fit, while the dashed line is the prediction of microscopic solvability theory for $\gamma = 0.1$. The error bars correspond to the numerical error in the parabolic fit of the interfacial profile.

FIG. 4. The distance $\lambda$ of the sharp-cornered tip to the fitted parabola \( \mathbb{P} \), as a function of the radius of curvature. The linear fit (continuous line) is extrapolated to the origin (dashed line). Inset: the corner angle $\Theta_{\text{tip}}$ of the dendrite tip as a function of the surface tension anisotropy parameter $\gamma$, for a fixed undercooling $\Delta = 0.10$
