Numerical Simulation of Three-Dimensional Dendritic Growth

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Dendritic crystal growth in a pure undercooled melt is simulated quantitatively in three dimensions using a phase-field approach. The full non-axisymmetric morphology of the steady-state dendrite tip and $\sigma^*$ are determined as a function of anisotropy for a crystal with a cubic symmetry. Results are compared to experiment and used to critically test solvability theory. (June, 1996).

Dendritic growth has been a central problem in pattern formation \[\text{and} \] metallurgy \[\text{for} \] many years. Considerable theoretical progress has been achieved which has led to the development of solvability theory to determine the steady-state operating state of the dendrite tip (i.e. tip velocity $V$ and tip radius $\rho$) \[\text{and} \] \[\text{theory} \] in 3D, to make comparison with experiment, and applied to critically test the applicability of solvability interface are negligibly small). Our numerical results are

\[\text{modynamic equilibrium (i.e.} \text{where} \text{kinetic effects at the} \text{interface are assumed to relax instantaneously to local thermodynamic equilibrium (i.e. where kinetic effects at the interface are negligibly small).} \]

Paradoxically, our understanding of pattern selection in three dimensions (3D) has remained uncertain, especially with regards to experiment. This is due to the fact that it has so far remained too difficult to simulate reliably the equations of dendritic growth in 3D even on current supercomputers. This, in turn, has prevented: (i) to test whether the global attractor of the growth dynamics is indeed the steady-state needle crystal predicted by solvability theory, and (ii) to test the predictions of this theory which are themselves only approximate in 3D \[\text{and} \] experiment \[\text{are} \] due to the inapplicability of this theory, to the approximate nature of its predictions in 3D, or to some missing physics in the starting equations.

Here we report the results of 3D simulations of dendritic growth in a pure undercooled melt. These simulations are made possible by using a recently developed phase-field approach which renders fully quantitative 3D computations accessible for the first time \[\text{in 3D} \text{and} \text{experiment} \text{are} \text{due} \text{to the} \text{inapplicability} \text{of this} \text{theory, to the approximate nature of its predictions in 3D, or to some missing physics in the starting equations.} \]

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The equations of the 3D free-boundary problem are given by

\[
\begin{align*}
\partial_t T &= D \nabla^2 T \
Lv_n &= c_p D ( \partial_t T |_S - \partial_n T |_L ) \
T_I - T_M &= -\frac{T_M}{L} \sum_{i=1}^2 \left[ \gamma(n) + \frac{\partial^2 \gamma(n)}{\partial \theta_i^2} \right] \frac{1}{R_i} \quad (3)
\end{align*}
\]

where $T$ is the temperature field, $T_I$ is the interface temperature, $T_M$ is the melting temperature, $D$ is the diffusivity, $L$ is the latent heat of melting, $c_p$ is the specific heat, $v_n$ is the normal velocity of the interface, and

\[
\gamma(n) = \gamma_0 (1 - 3 \epsilon_4) \left[ 1 + \frac{4 \epsilon_4}{1 - 3 \epsilon_4} (n_x^4 + n_y^4 + n_z^4) \right] \quad (4)
\]

is the surface energy for a crystal chosen to have a cubic symmetry. In addition, $\theta_i$ are the angles between the normal $n$ and the two local principal directions on the boundary and $R_i$ are the principal radii of curvature. Growth is controlled by the undercooling $\Delta = (T_M - T_\infty)/(L/c_p)$ where $T_\infty$ is the initial melt temperature.

We use three key ingredients to solve the above equations. Firstly, we avoid the usual difficulties of tracking a sharp boundary by using a phase-field approach \[\text{and} \text{experiment} \text{are} \text{due} \text{to the} \text{inapplicability} \text{of this} \text{theory, to the approximate nature of its predictions in 3D, or to some missing physics in the starting equations.} \]

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model parameters to those of the sharp-interface equations. As demonstrated in 2D \cite{12}, this analysis makes it possible: (i) to perform computations with a smaller capillary length, \( d_0 = \gamma_0 T_M c_p / L^2 \), and (ii) to choose \( \lambda \) and the function \( \tau (n) \) in such a way that we obtain a Gibbs-Thomson condition without interface kinetics in the sharp-interface limit \cite{12}. That is we recover exactly Eq. \( \ref{eq:sharp-interface} \) without an additional velocity-dependent term in this limit. Since the computation time scales as \( (d_0/W_0)^5 \) in 3D, this first property dramatically enhances the computational efficiency of the phase-field approach and allows us to model dendritic growth quantitatively, as opposed to non-quantitatively in 3D as previously \cite{14}. Furthermore, it permits us to model an undercooling range where the dendrite tip Péclet number \( P \equiv \rho V / 2 D \) is small enough to compare \( \sigma^* = 2 D d_0 / \rho^2 V \) to its measured small undercooling values \cite{9,10}.

Lastly, we are able to resolve numerically very small anisotropies by incorporating quantitatively the contribution of the grid anisotropy. For a given value of \( \epsilon_4 \) used in \( W(n) \), we compute the equilibrium shape produced by the phase-field model. The grid-corrected anisotropy is then defined as that value of \( \epsilon_4 \) in \( \gamma(n) \) which produces a 3D equilibrium shape that matches exactly that of the phase-field model. A procedure that will be described elsewhere was also developed to obtain a grid-corrected \( \tau(n) \). Numerical tests were performed in 2D to check that this procedure yields accurate values of \( \sigma^* \) over the entire range of \( \epsilon_4 \) investigated here in 3D.

Eqs. \( \ref{eq:sharp-interface} \) were simulated explicitly with a grid spacing ranging between 0.6 and 0.8 with \( W_0 = 1 \), \( D \) ranging between 0.5 and 4, a time step ranging between 0.01 and 0.08, and a constant undercooling \( \Delta = 0.45 \). The anisotropy was varied from \( \epsilon_4 = 0.0066 \) to \( \epsilon_4 = 0.047 \) which spans most of the range of experimental interest. Fig. 1 shows a typical 3D dendrite morphology resulting from the growth of a small spherical seed. Quantitative results which pertain to the steady-state operating state and morphology of the dendrite tip are presented in Table 1 and Figs. 2-3. These were obtained with long simulations that focused on the steady-state growth of a single dendrite tip along the \( z \)-axis. Several runs were performed to check that the values in Table 1 are independent of \( d_0/W_0 \) within an accuracy of 10%.

In order to test solvability theory, we have computed independently the values of \( P \) and \( \sigma^* \) predicted by the numerical solution of the steady-state growth equations using the standard axisymmetric approximation where the surface energy and steady-state shape are assumed to be independent of the polar angle \( \phi \) in the \( x-y \) plane perpendicular to the growth axis \cite{9}. This is the same calculation performed by Kessler and Levine in Ref. \cite{6} and we have checked that our boundary integral code reproduces their results within one percent. For completeness, we also report the predictions of the linear solvability theory of Barbieri and Langer \cite{7} which uses a shape linearized around the Ivantsov paraboloid of revolution \cite{18} and the same axisymmetric approximation.

As shown in Table 1, the numerical solvability theory yields \( \sigma^* \) values which are systematically lower than

![Fig. 1](image-url) Results of 3D phase-field simulation on a 300 x 300 x 300 cubic lattice for \( \epsilon_4 = 0.047 \) which shows dendrite tips growing along the principal \( <100> \) directions. The simulations were performed in the first octant \( (x, y, z \geq 0) \) with a spherical nucleus centered at the origin as initial condition. The solid-liquid boundary shown here corresponds to the \( \psi = 0 \) surface reconstructed by reflection about the \( x = y = z = 0 \) planes. The structure is seen from an angle where all six \( <100> \) directions are visible.

![Fig. 2](image-url) Plot of \( \sigma^* \) vs \( \epsilon_4 \) for \( \Delta = 0.45 \) showing the results of phase-field simulations compared to the approximate predictions of the numerical and linear solvability theories. Also plotted are the small \( \Delta \) experimental values of \( \sigma^* \) for SCN \cite{9} and PVA \cite{10} using the \( \epsilon_4 \) measurements of Ref. [11].
TABLE I. Result of phase-field simulations on a $200 \times 200 \times 200$ cubic lattice compared to the results of the numerical and linear solvability theories for $\Delta = 0.45$. $A$ and $\alpha$ characterize the amplitude of the four-fold symmetry component of the tip morphology in simulations. Typical runs took 60-140 CPU hours on a DEC-ALPHA 3000-700 workstation and shorter times on a CRAY-YMP and a CRAY-T3D.

| $\epsilon_4$ | $P$ | $\sigma^*$ | $\alpha$ | $A$ | $P_{\sigma}$ | $\sigma^*$ | $P_{\sigma}$ | $\sigma^*$ |
|-------------|-----|---------|--------|-----|------------|---------|------------|---------|
| 0.0066      | 0.426 | 0.015 | 1.78 | 13.0 | 0.418 | 0.0129 | 0.471 | 0.0116 |
| 0.0123      | 0.360 | 0.036 | 1.73 | 11.3 | 0.367 | 0.0329 | 0.471 | 0.0250 |
| 0.0171      | 0.312 | 0.063 | 1.68 | 10.1 | 0.324 | 0.0578 | 0.471 | 0.0367 |
| 0.0265      | 0.236 | 0.16  | 1.62 | 7.8  | 0.247 | 0.142 | 0.471 | 0.0602 |
| 0.0369      | 0.150 | 0.47  | 1.57 | 5.7  | 0.172 | 0.365 | 0.471 | 0.0890 |
| 0.0470      | 0.093 | 1.72  | 1.54 | 4.0  | 0.109 | 1.037 | 0.471 | 0.1240 |

The phase-field results, but still reasonably close for small anisotropy. The predicted values of $\sigma^*$, however, start to become significantly inaccurate for anisotropies greater than about 3%, although predictions of the Péclet number remain relatively accurate. Most likely, this does not indicate a breakdown of solvability theory, but of the axisymmetric approximation. This conclusion is supported by the fact that the four-fold deviation from a shape of revolution increases in magnitude with anisotropy (as described below) and, hence, can affect the selection. It is also supported by the fact that we do not observe any sidebranching \cite{1, 9, 19} without adding noise to the phase-field equations. Hence our simulations rule out the possibility of a dynamical attractor other than a steady-state needle crystal. The linear theory is seen to breakdown at much smaller anisotropy. This is because it assumes that the steady-state shape remains close to the Ivantsov paraboloid of revolution. Table 1 shows that the actual Péclet number already starts deviating significantly from its Ivantsov value, $P_{\sigma}$ \cite{19}, at small anisotropy.

The steady-state morphology of the dendrite tip was analyzed using the Fourier decomposition

$$r^2(\phi, z) = \sum_{n} A_n(z) \cos 4n\phi$$  \hspace{1cm} (8)

where $r$ is the radial distance from the $z$-axis. Both $r$ and $z$ are measured in units of $\rho$ with the tip at $z = 0$. This decomposition has the advantage that it is general and does not presupposes a particular analytical form to fit the tip shape. The function $A_0(z)$ is the axisymmetric contribution to this shape. It approaches a paraboloid of revolution, $A_0(z) = -2z$, for small $|z|$ and departs from this shape with increasing $|z|$ as shown in Fig. 3, this departure being more pronounced at larger anisotropy as one would expect. More important is the non-axisymmetric departure from this shape of revolution which is contained in the higher modes, $A_n(z)$ for $n \geq 1$. The amplitude of the first four-fold symmetry mode turns out to be much larger than all the other modes (Fig. 3) and to be well-described by the power law $A_4(z) \sim |z|^\alpha$, which appears as a remarkably straight line on a log-log plot of $A_4(z)$ vs $|z|$ up to a distance of a few $\rho$ behind the tip. Furthermore, the values of $A$ and $\alpha$ in Table 1 clearly show that the amplitude of the four-fold symmetry mode is sensitively dependent on anisotropy which is a qualitatively novel aspect of our results. In contrast, the linear solvability calculation of Ben Amar and Brenner \cite{8} predicts that for small $\epsilon_4$ the tip morphology should be independent of anisotropy, with $A^{-1} = 11$ and $\alpha = 2$ for small $|z|$. The values of $A$ and $\alpha$ listed in Table 1 indicate that this prediction is most likely only valid for values of $\epsilon_4$ which lie outside the range of experimental interest. This is also consistent with the fact that their calculation is only valid in the limit where $\sigma^* \sim \epsilon_4^1$, and that this $7/4$ power law scaling is not yet attained at the smallest computed anisotropy in Fig. 2.

For $\epsilon_4 = 0.0066$, which lies inside the range of uncertainty of the measured value $\epsilon_4 = 0.0055 \pm 0.0015$ for SCN \cite{1}, our simulations yield a value of $\sigma^* \approx 0.015$ with $P = 0.426$. This value is only 20 % smaller than the value $\sigma^* = 0.0192$ measured by Huang and Glicksman \cite{18} for this material. Most of this discrepancy can be accounted for by the finite Péclet number correction which can be estimated, using the numerical solvability code, to decrease $\sigma^*$ by about 15% from its zero Péclet number limiting value. Therefore we obtain a reasonably good agreement with experiment for SCN within the existing uncertainty in the measured value of anisotropy. Pivalic acid \cite{17} (PVA), however, remains problematic as seen in Fig. 2 and a closer examination of kinetic effects for this material could potentially resolve this large discrepancy. Finally, Maurer et al. \cite{20} have found that the tip morphology of NH$_4$Br dendrites is indeed well-described by a single cos 4$\phi$ mode, while LaCombe et al. have found that for SCN dendrites more modes seem necessary to fit the tip morphology \cite{21}. The origin of this difference, which is potentially due to noise amplification, also remains to be understood.

In conclusion, we have demonstrated that quantitative modeling of 3D dendritic growth is possible using a recently developed phase-field methodology \cite{22}. Our results are important in that they leave little doubt about the conceptual validity of solvability theory in 3D and the importance of crystalline anisotropy, and yield results which are consistent with experiment, at least for SCN where kinetic effects are believed to be small. At present, the applicability of solvability theory — in terms of making accurate predictions — remains mainly limited by the axisymmetric approximation. Our results have shown that the three-dimensional morphology of the dendrite tip is dominated by a four-fold component whose amplitude depends sensitively on anisotropy. This prediction should be testable experimentally. A host of other mi-
crostructural pattern formation issues, such as the determination of the steady-state shape further behind the tip and sidebranching, are now amenable to quantitative study using the present computational approach.

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FIG. 3. Steady-state tip morphology for $\epsilon_4 = 0.0123$ shown in: (a) the $\phi = 0^\circ$ (solid line) and $\phi = 45^\circ$ (dash-dotted line) planes, and (b) (100) planes equally spaced along $z$ by one tip radius. Length is measured in units of $\rho$. The Ivantsov parabola $z = -r^2/2$ (dash-line) and a circle of unit radius are superimposed in (a). The Fourier amplitudes at two tip radii behind the tip in (b) are: $A_1(-2) = 0.29$, $A_2(-2) = 0.020$, and $A_3(-2) = 0.0022$, illustrating that the tip morphology is dominated by the four-fold symmetry mode.