Tip Oscillation of Dendritic Patterns in a Phase Field Model

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(Received December 23, 2000)

Dendritic patterns are numerically investigated with a phase field model. Changing the anisotropies of the surface tension and the kinetic coefficient, we find complicated patterns and tip-oscillating patterns in some range of parameter values.

Growing interfaces form various patterns from flat interfaces to complex branched patterns.\(^1\)–\(^3\) Dendrites are intricate patterns found in the crystal growth of most metal alloys and some plastic crystals.\(^4\),\(^5\) There are several types of models to simulate such intricate interface patterns. A nonlocal sharp-interface model is directly related to the basic equations of the diffusion equation with the Gibbs-Thomson condition at the interface.\(^6\),\(^7\) A phase field model has the form of the Ginzburg-Landau equation coupled with the diffusion equation and it is easier to use for numerical simulations.\(^8\)–\(^10\) For melt growth, the Ginzburg-Landau equation describes the dynamics of the phase transition from the liquid phase to the solid phase, and the diffusion process of the latent heat generated at the growing interface is considered to be a limiting factor for the crystal growth. Recently, Karma et al. quantitatively studied the phase field model in two and three dimensions and obtained reasonable results in comparison with the solvability theory.\(^11\),\(^12\)

The standard theory predicts that a needle crystal selected by the solvability condition is stable and that side-branches are produced through the effect of thermal noise.\(^13\),\(^14\) However, there are some experiments suggesting that a kind of side-branches appears as a result of regular tip oscillations.\(^15\),\(^16\) We report some results of the numerical simulation of a phase field model that exhibits tip oscillation.

Our two-dimensional model has the form

\[
\begin{align*}
\tau(\theta) \partial_t p &= \{p - \lambda u(1 - p^2)(1 - p^2) + \partial_x \{W(\theta)^2 \partial_x p - W(\theta)W'(\theta)\partial_y p\} \\
&+ \partial_y \{W(\theta)^2 \partial_y p + W(\theta)W'(\theta)\partial_x p\}\}, \\
\partial_t u &= D\nabla^2 u + \partial_t p/2,
\end{align*}
\]

where \(p(x,y,t)\) is the order parameter, \(\lambda\) is a coupling constant, \(\tau(\theta)\) is the time constant for the order parameter, and \(\theta = \arctan(\partial_y p/\partial_x p)\) is the angle between the normal directions of the contours of constant \(p\) and the \(x\) axis. The solid and liquid phase correspond, respectively, to \(p = 1\) and \(p = -1\). The normalized temperature is denoted by \(u(x,y,t) = (T - T_M)/(L/C_p)\), where \(T(x,y,t), T_M, L\) and \(C_p\) are, respectively, the temperature, the melting temperature, the latent heat and the specific heat. The diffusion constant for \(u\) is denoted by \(D\), and \(W^2(\theta)\) is an anisotropic
diffusion constant for the order parameter.

The generalized Gibbs-Thomson condition (Wilson-Frenkel formula) is assumed for the sharp interface model as

\[ u_i = -d_0(\theta)\kappa - \beta(\theta)v_n, \tag{2} \]

where \( u_i \) is the normalized temperature at the interface, \( d_0(\theta) \), \( \kappa \), \( \beta(\theta) \) and \( v_n \) denote, respectively, the anisotropic capillary length, the interface curvature, the anisotropic kinetic coefficient and the normal interface velocity. The simple growth law of the form \( v\rho^2 = \text{const} \), where \( v \) is the steady growth velocity of the tip and \( \rho \) is the tip radius, is modified by the kinetic effect (nonzero \( \beta \) effect).\(^{17,18}\) Karma and Rappel computed approximate expressions for \( d_0(\theta) \) and \( \beta(\theta) \) in the sharp-interface limit of the phase field model as

\[
d_0(\theta) = \frac{I}{\lambda J} \{ W(\theta) + W''(\theta) \}, \quad \beta(\theta) = \frac{I}{\lambda J} \frac{\tau(\theta)}{W(\theta)} \left[ 1 - \lambda \frac{W^2(\theta)}{2D\tau(\theta)} \frac{K + JF}{I} \right], \tag{3} \]

where \( I = 2\sqrt{2}/3, J = 16/15, K = 0.13604 \) and \( F = \sqrt{2}\ln 2 \). If \( \tau(\theta) = W^2(\theta) \) and the coupling constant \( \lambda \) is assumed to be \( \lambda = (2ID)/(K + JF) \), the anisotropic kinetic coefficient becomes zero, and the Gibbs-Tomson condition is realized. Karma and Rappel compared the numerical simulation of the phase field model with four-fold rotational symmetry for the surface tension and the sharp interface model with \( \beta = 0 \) and obtained good agreement.

We performed numerical simulations of the phase field model with both surface tension anisotropy and kinetic coefficient anisotropy. We first assume four-fold rotational symmetry for the anisotropy given by \( W(\theta) = 1 + e_s \cos(4\theta), \tau(\theta) = W'(\theta)\{1+e_k \cos(4\theta)\} \), where \( e_s \) and \( e_k \) denote the anisotropy parameters of the surface tension and the kinetic coefficient. We have assumed \( \lambda \) to be \( \lambda = 1.8ID/(K + JF) \) with \( D = 2 \). The anisotropy coefficients in (3) are \( d_0(\theta) \propto 1 - 15e_s \cos(4\theta) \) and \( \beta(\theta) \propto 0.1 + (e_k - 0.9e_s) \cos(4\theta) \). The numerical simulation was performed using the finite difference method with \( \Delta x = 0.4 \) and \( \Delta t = 0.015 \). We employed mirror-symmetric boundary conditions at \( x = 0 \) and \( y = 0 \) for the sake of simplicity. The system was initially seeded with a small quarter disk of solid at one corner of the lattice, and a spatially uniform undercooling \( u = -\Delta \) was initially set.

Figure 1 displays the results of numerical simulations for \( \Delta = 0.7 \) and \( e_s = 0.06 \). The anisotropy \( e_k \) is changed as a control parameter. Figure 1(a) displays a snapshot pattern growing in the \( x \) and \( y \) directions (\{10\} and \{01\} directions) at \( e_k = -0.2 \). Since \( \theta = 0 \) and \( \pi/2 \) correspond to the minima of \( \beta(\theta) \), the growth velocity becomes large in these directions if \( u_i + d_0(\theta)\kappa \neq 0 \). On the other hand, the growth velocity is large in the \{11\} direction when \( e_k \) is positive and sufficiently large. Figure 1(d) displays a growth pattern at \( e_k = 0.2 \). Steady growth in the diagonal direction is clearly seen. If \( e_k \) is increased from \(-0.2 \) to \( 0.2 \), the direction of growth changes from the \{10\} direction to the \{11\} direction. In the intermediate parameter region of \( e_k \), complicated patterns were observed. Figure 1(b) displays a pattern growing...
in the (10) and (01) directions for $e_k = 0.065$. The curvature in the tip region is not constant in time, and the interface is deformed. Figure 1(c) displays a tip-oscillating pattern growing in the diagonal direction for $e_k = 0.089$. Side-branches are created mainly in the (10) and (01) directions.

A change in the growth direction can occur more naturally by changing the supercooling $\Delta$ if $e_s e_k > 0$. If the supercooling $\Delta$ is sufficiently small, the growth velocity is very small, and the kinetic effect becomes negligible. If the supercooling $\Delta$ is increased, the kinetic anisotropy becomes important. Especially for $\Delta > 1$, the kinetic effect is necessary for the steady growth of the pattern. Figure 2 shows growth patterns for $e_s = 0.06$ and $e_k = 0.06$. Figure 2(a) displays a pattern growing in the (10) and (01) directions with $\Delta = 0.5$. It is seen that the (10) and (01) directions are preferable, since the surface stiffness is minimum at the orientations $\theta = 0$ and $\pi/2$ for $e_s > 0$. Figure 2(c) displays a pattern growing mainly in the (11) direction with $\Delta = 1.1$. The kinetic effect becomes dominant for $\Delta > 1$. In the range of intermediate values of $\Delta$, the growth direction changes from the (10) direction to the (11) direction, and a complicated pattern appears. Figure 2(b) displays a tip-oscillating pattern with $\Delta = 0.8$. We see that side-branches growing in the (11) direction are created periodically.

A tip-oscillating pattern and a densely-branched pattern have been observed in a real experiment using NH$_4$Cl.\textsuperscript{16} In the experiment, such complicated patterns actually appear in the transition region between ⟨100⟩ growth and ⟨110⟩ growth.

Complicated patterns appear in the parameter region in which the growth velo-

![Fig. 1. Snapshot patterns for the phase field model with four-fold rotational symmetry for $e_s = 0.06, \Delta = 0.7$ and (a) $e_k = -0.2$, (b) 0.065, (c) 0.089 and (d) 0.2.](image1)

![Fig. 2. Snapshot patterns for the phase field model with four-fold rotational symmetry for $e_s = 0.06, e_k = 0.06$ and (a) $\Delta = 0.5$, (b) 0.8, (c) 1.1.](image2)
Fig. 3. Growth patterns for the phase field model with eight-fold rotational symmetry for $e'_s = 0.015, e'_k = -0.01$ and (a) $\Delta = 0.6$ and (b) $\Delta = 0.7$.

Fig. 4. (a) Growth pattern for the phase field model with eight-fold symmetry in a long rectangular box of size $480 \times 80$ for $e'_s = 0.015, e'_k = -0.01$ and $\Delta = 0.7$. The tip oscillation is clearly observed in these figures. The growth velocity is larger, the average tip curvature is larger, and the interval of side-branches is smaller with $\Delta = 0.7$ than with $\Delta = 0.6$. We have checked the tip oscillation in a numerical simulation with the smaller discretization $\Delta x = 0.1$ and $\Delta t = 0.001$. We found that the tip oscillation occurs even when $\tau(\theta) = W^2(\theta)$ and $\lambda = 2ID/(K + JF)$, that is, when the effective value of $\beta(\theta)$ is zero. Surface tension anisotropy with eight-fold symmetry is important for the tip oscillation.

To clarify the regular tip oscillation seen in Fig. 3, we performed a numerical simulation in a narrow rectangular box of size $L_x \times L_y = 480 \times 80$. Mirror-symmetric boundary conditions were again used at $x = 0$, $y = 0$ and $y = L_y$. Figure 4(b) displays the profiles of $p(x, y)$ at $y = 0$. The snapshot profiles of the order parameter $p(x, y)$ and the normalized temperature $u(x, y)$ at $y = 0$. 

ity in the $\langle 10 \rangle$ and $\langle 11 \rangle$ directions is comparable. If eight-fold rotational symmetry is used for the anisotropy, the $\langle 10 \rangle$ and $\langle 11 \rangle$ growth are equivalent due to the symmetry. We have performed a numerical simulation of the model equation with eight-fold symmetry given by $W(\theta) = 1 + e'_s \cos(8\theta), \tau(\theta) = W(\theta)\{1 + e'_k \cos(8\theta)\}$. Figure 3 displays growth patterns for $e'_s = 0.015$ and $e'_k = -0.01$. Since $e'_s$ is positive and $e'_k$ is negative, growth in the $\langle 10 \rangle$ and $\langle 11 \rangle$ directions is preferable. Figure 3(a) displays a growth pattern with $\Delta = 0.6$ and Fig. 3(b) displays a pattern with $\Delta = 0.7$. The tip oscillation is clearly observed in these figures. The growth velocity is larger, the average tip curvature is larger, and the interval of side-branches is smaller with $\Delta = 0.7$ than with $\Delta = 0.6$. We have checked the tip oscillation in a numerical simulation with the smaller discretization $\Delta x = 0.1$ and $\Delta t = 0.001$. We found that the tip oscillation occurs even when $\tau(\theta) = W^2(\theta)$ and $\lambda = 2ID/(K + JF)$, that is, when the effective value of $\beta(\theta)$ is zero. Surface tension anisotropy with eight-fold symmetry is important for the tip oscillation.

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Fig. 5. (a) Time evolution of the normalized temperature $u(x, y)$ on the interface at $y = 0, 12$ and 20. (b) Time evolution of the curvature on the interface at $y = 0, 12$ and 20.

$u(x, 0)$ at $y = 0$. The width of the transition layer of the normalized temperature is sufficiently larger than the width of the transition layer of the order parameter. Figure 5(a) displays the time evolution of the normalized temperature $u(x, y)$ at the interface for $y = 0, 12$ and 20. The interface positions were numerically estimated as positions where $p(x, y) = 0$. Figure 5(b) displays the time evolution of the curvature at the same three interface positions. Regular oscillation is clearly seen both in the time evolution of the normalized temperature and the curvature. The tip velocity oscillates with the same period. The peaks of the temperature profiles are close to the minimum points of the curvature. The peaks of the temperature oscillate amplifies as $y$ is increased. That is, the oscillatory perturbation propagates and is amplified along the interface from the tip region.

In this model $\langle 11 \rangle$ growth and $\langle 10 \rangle$ growth are equally preferable. For this reason, side-branches in the $\langle 11 \rangle$ direction tend to be created. If side-branches are created, more latent heat is released there, and the temperature increases. If the temperature increases, the effective supercooling is decreased, and the side-branching stops. This may be one origin of the tip oscillation.

To study the kinetic effect for the regular tip oscillation, we changed $\tau(\theta)$ to $W(\theta)\{1+e_k \cos(4\theta)\}$ or $W(\theta)\{1+e'_k \cos(8\theta)\}$ with $\Delta = 0.7$, $W(\theta) = 1 + 0.015 \cos(8\theta)$ and $\lambda = 1.8JD/(K+JF)$. As $e_k$ becomes positive and larger with $e'_k = 0$, the $\langle 11 \rangle$ growth becomes more dominant, owing to the four-fold symmetry of $\tau(\theta)$. A tip-oscillating dendritic pattern appears in Fig. 6(a) at $e_k = 0.01$. The tip oscillation becomes weaker as $e_k$ becomes larger, and a steady growth pattern to the $\langle 11 \rangle$ direction appears in Fig. 6(b) for $e_k = 0.035$. Figures 6(c) and (d) display numerical results for $\tau(\theta) = W(\theta)\{1+e'_k \cos(8\theta)\}$. As the kinetic coefficient $e'_k$ becomes negative and smaller, the kinetic anisotropy preferring the $\langle 10 \rangle$ and $\langle 11 \rangle$ growth becomes stronger. Then, the tip oscillation becomes weaker, and a steady growth pattern appears in Fig. 6(c) at $e'_k = -0.12$. The strong kinetic effect seems to suppress the tip oscillation. When $e'_k$ is positive, the kinetic anisotropy prefers the $\pi/8$ and $3\pi/8$ directions, and the kinetic coefficient anisotropy and the surface tension anisotropy compete again. As $e'_k$ becomes larger, the growth direction changes from the $0$ and $\pi/4$ directions to the $\pi/8$ and $3\pi/8$ directions, and complicated patterns appear.
Fig. 6. Growth patterns with the anisotropic kinetic coefficient $\tau(\theta) = W(\theta)\{1 + e_k \cos(4\theta) + e'_k \cos(8\theta)\}$. The other parameters are the same as in the case of Fig. 3(b). (a) $e_k = 0.01, e'_k = 0$, (b) $e_k = 0.035, e'_k = 0$, (c) $e_k = 0, e'_k = -0.12$ and (d) $e_k = 0, e'_k = 0.08$.

in the range of intermediate parameter values. Figure 6(d) displays a complicated dendritic pattern for $e'_k = 0.08$. The pattern grows mainly in the $\pi/8$ and $3\pi/8$ directions for this parameter value.

To summarize, we have performed simulations using a phase field model with anisotropic surface tension and kinetic coefficient of four-fold and eight-fold rotational symmetries. We have found complicated growth patterns in a parameter region where the two anisotropies compete. We have also performed simulations of a phase field model with eight-fold rotational symmetry for the surface tension and found regular tip oscillation. The competitive relation between the $\langle 10 \rangle$ growth and $\langle 11 \rangle$ growth may be one origin of the complicated growth and the tip oscillation in these simulations. However, the detailed mechanism responsible for the oscillation is not yet well understood.