Phase field investigation on the initial planar instability with surface tension anisotropy during directional solidification of binary alloys*

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Phase field investigation reveals that the stability of the planar interface is related to the anisotropic intensity of surface tension and the misorientation of preferred crystallographic orientation with respect to the heat flow direction. The large anisotropic intensity may compete to determine the stability of the planar interface. The destabilizing effect or the stabilizing effect depends on the misorientation. Moreover, the interface morphology of initial instability is also affected by the surface tension anisotropy.

Keywords: surface tension anisotropy, directional solidification, interfacial stability
PACC: 7350G, 8130F, 4630L

1. Introduction

Solidification involves a complex interplay of interface dynamics and transport phenomena, which may launch complex interface morphology. During solidification the interface morphology dominates the final material microstructures which affect the mechanical and the electronic properties of materials. Therefore problems relevant to microstructure evolution are of interest to many physicists and metallurgists. For most metallic alloys, the solidification process is dominated by solutal diffusion and/or thermal diffusion. The physical mechanism of interface instability is rooted in the competition between the destabilizing effects of the solutal diffusion and the stabilizing effects of the temperature gradient and of the surface tension.[1] The long-standing investigations on solidification pattern formation have revealed that surface tension anisotropy, although small, plays an important role in the microstructure evolution. Surface tension anisotropy may significantly influence the mechanism of stability and the selection condition of free crystal growth.[2−4] During directional solidification the surface tension anisotropy, an essential factor for stationary cellular arrays, tilted cellular/dendritic arrays and seaweed patterns, may also compete to determine the interfacial stability near some critical points.[5−17]

Most of the previous studies concern the effect of surface tension anisotropy on finial steady state pattern selection. However, the microstructure evolution during directional solidification is usually a history-dependent dynamic process. During directional solidification the cellular/dendritic array originates from the instability of the planar front. The initial instability occurs in the initial transient stage where the incubation time of initial microstructure and the interface morphology are of technical interest.[18,19] Although surface tension anisotropy plays an important role in microstructure evolution, the initial planar instability with surfaced tension anisotropy is seldom considered. In the present paper, a quantitative phase field model is employed to investigate the initial planar instabilities with different anisotropic intensities of the surface tension and different misalignment angles of preferred crystallographic orientation with respect to the heat flow direction. The interface morphology evolutions from the planar interface to the observation instability with different surface tension anisotropies are investigated in order to reveal the effect of surface tension anisotropy on the initial instability of the planar interface.

2. Model description and simulation parameters

The quasi-two-dimensional system of a Hele–Shaw cell is considered here. The cell is pulled at a

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constant velocity V from the hot end to the cold end. The hot end and the cold end present an external imposed temperature gradient G, corresponding to the heat flow. In the 'frozen temperature approximation', the temperature field in the cell is $T = T_{\rm M} + Gz$ in the laboratory frame of reference, where $T_{\rm M}$ is the melting temperature of pure solvent and z the heat flow direction. The solute concentration is denoted by far field concentration c_∞. A typical representation of surface tension anisotropy with four-fold symmetry is $\gamma = \gamma_0(1 + \gamma_4 \cos(4\theta + 4\theta_0))$ in two dimensions, where γ_0 is the isotropic part of the surface tension, γ_4 the anisotropic intensity of the surface tension, θ_0 the misorientation of preferred crystallographic orientation with respect to the heat flow direction, and θ the angle between the normal vector of the interface and the heat flow direction. In the one-sided model, local equilibrium at the interface gives the free boundary conditions as

$$c_{\rm S} = kc_{\rm L}, \tag{1}$$

$$V_{\rm n}(c_{\rm L} - c_{\rm S}) = -D\frac{\partial c_{\rm L}}{\partial z}, \tag{2}$$

$$T_{\rm I} = T_{\rm M} + mc_{\rm L} - \kappa\Gamma[1 - 15\gamma_4 \cos(4\theta + 4\theta_0)], \tag{3}$$

where $c_{\rm S}$ and $c_{\rm L}$ are the concentrations in solid and liquid respectively, k the partition coefficient, D the chemical diffusion coefficient, $T_{\rm I}$ the temperature at the interface, m the slope of liquidus line, κ the curvature, $\Gamma = \gamma_0 T_{\rm M}/L$ and L the latent heat.

Recently the development of the phase field method with diffuse interface makes it more convenient to investigate the free boundary problems.[20−22] The phase field model for directional solidification of binary alloys adopted here was presented by Echebarria *et al.*[22] This quantitative model has been benchmarked by detailed numerical tests in two dimensions. The model with surface tension anisotropy is as follows:

$$\begin{aligned}
&\tau_0\left(1 - (1-k)\frac{z + (mc_\infty/k)/G}{l_T}\right)(\partial_t\phi - V\partial_z\phi) \\
&= \nabla(W(\theta)^2\nabla\phi) - \partial_x[W(\theta)W'(\theta)\partial_y\phi] + \partial_y[W(\theta)W'(\theta)\partial_x\phi] + \phi - \phi^3 + \lambda(1-\phi^2)^2 \\
&\quad \times\left(\frac{2mc}{1 + k - (1-k)\phi} - \frac{z + (mc_\infty/k)/G}{l_T}\right),
\end{aligned} \tag{4}$$

$$\partial_t c - V\partial_z c = \nabla\cdot\left[D_L\frac{1-\phi}{1+k-(1-k)\phi}\nabla c + \left(D_L\frac{1-\phi}{1+k-(1-k)\phi} + \frac{W_0}{\sqrt{2}}\frac{(\partial_t\phi - V\partial_z\phi)}{|\nabla\phi|}\right)\frac{c(1-k)}{1+k-(1-k)\phi}\nabla\phi\right], \tag{5}$$

where ϕ is the phase field variable, c the bulk concentration, $W(\theta) = W_0(1 + \gamma_4 \cos(4\theta + 4\theta_0))$, and $l_T = |m|\, c_\infty(1-k)/(kG)$. The parameters of the interface thickness W_0, the relaxation time for phase field model τ_0 and the coupling constant λ are linked to physical quantities by the relationships of $d_0 = a_1 W_0/\lambda$ and $\tau_0 = a_2\lambda W_0^2/D$. Here $d_0 = \Gamma/(mc_\infty(1-k)/k)$, $a_1 = 0.8839$, and $a_2 = 0.6267$.

Among the organic crystals used in the previous directional solidification experiments, the most widely investigated material is succinonitrile (SCN), of which the important material properties have been accurately investigated.[23] The dilute solute is usually acetone (ACE). Here SCN-ACE 0.05 mol% is selected for the numerical calculation, with the chemical diffusion coefficient in liquid phase $D \approx 1.27 \times 10^{-9}$ m^2/s, partition coefficient $k = 0.1$, Gibbs–Thomson coefficient $\Gamma = 6.48 \times 10^{-8}$ K/m and the slope of liquidus line $m = -222$ K/mol. The simulation is performed in two dimensions with explicit time stepping. The diffuse interface thickness is $W = 1$ μm and the grid size is d$x = 0.4$ W. The calculation domain is 300 μm×1000 μm. In the calculation, an appropriate stochastic noise is imposed on the phase field across the diffuse interface to simulate the effect of fluctuations on the liquid/solid interface.

3. Results and discussion

Due to the long transient behaviour at low pulling velocity, it is difficult to directly evaluate the effect of the surface tension anisotropy on the critical pulling velocity of planar instability by experiments or numerical simulations. Therefore, first of all, the criteria to evaluate the effect of surface tension anisotropy need to be confirmed. Obviously, stabilizing effects can ex-

tend the stage of planar interface while destabilizing effects can shorten the stage of planar interface in the transient process of directional solidification. So, the dynamic evolution of the planar interface instability in transient process can also reflect the effect of surface tension anisotropy. Herein, the temperature gradient is chosen as 100 K·cm^{-1}, and the pulling velocity is $V = 50$ μm/s, with which the planar interface is unstable after a period of time. The interface morphology evolution varies with surface tension anisotropy, which can reveal effects of surface tension anisotropy on the initial planar interface instability.

3.1. Effect of anisotropic intensity

Although the previous experimental measurement gave different values of surface tension anisotropy for a specific material, the property of surface tension anisotropy is almost constant. In this investigation, to obtain the general behaviour about the effect of surface tension anisotropy, the anisotropic intensity parameter γ_4 is intentionally changed. The heat diffusion and the solute diffusion are the leading processes during directional solidification and the capillary effect of surface tension is only a correction quantity. The capillary effect in Eq. (3) with a small surface tension anisotropy ($\gamma_4 = 0.01$) has only 15% deviation from the isotropic case. For all that, the effect of surface tension anisotropy cannot be neglected because the dynamic evolution of planar instability is an accumulating process.[18] This small effect of surface tension anisotropy may compete to determine some characteristics of the interface instability.

The dynamic evolutions of planar interface instability with different values of γ_4 for $\theta_0 = 0°$ and 45° are simulated as shown in Figs. 1 and 2 from 5 s to 6.5 s with 0.5 s interval. Significantly, for $\theta_0 = 0°$ the interface instability with $\gamma_4 = 0.04$ occurs at about 5 s while the interface instability with $\gamma_4 = 0$ occurs at about 6 s. Moreover, the wave number with $\gamma_4 = 0.04$ is three but more than that with $\gamma_4 = 0$. It reveals that for $\theta_0 = 0°$ with a larger anisotropic intensity the planar interface becomes unstable at an earlier time and the initial average wavelength is smaller. However, in Fig. 2, for the case of $\theta_0 = 45°$, the surface tension anisotropy has a reverse behaviour: the planar interface becomes unstable later and the initial average wavelength becomes larger with anisotropic intensity increasing. These results demonstrate that the effect of the surface tension anisotropy may compete to determine interfacial stability near some critical conditions in directional solidification, which is qualitatively in agreement with the previous analysis results.[8] The difference between $\theta_0 = 0°$ and $\theta_0 = 45°$ also indicates that the effect of surface tension anisotropy strongly depends on the misaligned angle, which will be investigated in detail later.

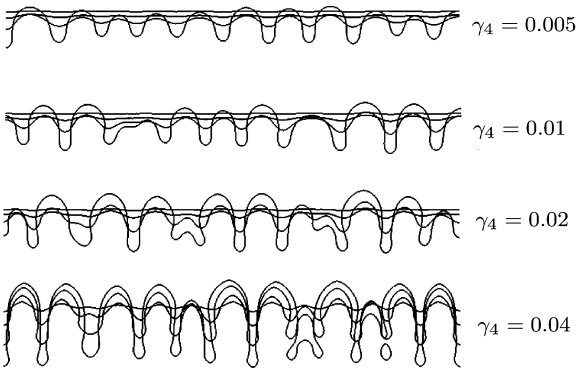


Fig. 1. Dynamic evolutions of planar interface instability with different anisotropic intensities of surface tension when $\theta_0 = 0°$ from 5 s to 6.5 s.

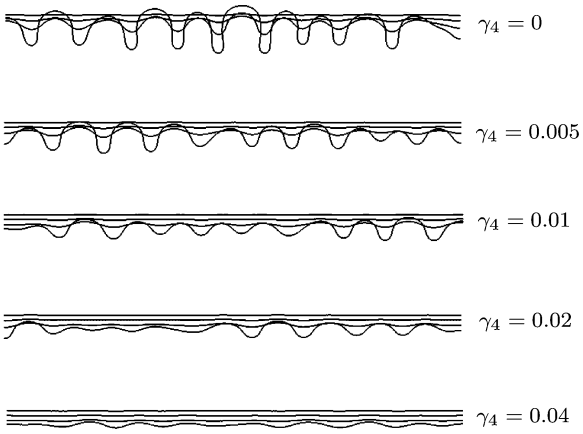


Fig. 2. Dynamic evolutions of planar interface instability with different anisotropic intensities of surface tension when $\theta_0 = 45°$ from 5 s to 6.5 s.

3.2. Effect of misalignment angle

In the linear stability analysis, the anisotropic term of the capillary effect in Eq. (3) can be simplified into

$$1 - 15\gamma_4 \cos(4\theta + 4\theta_0) \approx 1 - 15\gamma_4 \cos(4\theta_0). \quad (6)$$

The results of linear stability analysis reveal that the surface tension anisotropy destabilizes the planar interface when $\cos(4\theta_0) > 0$ but stabilizes the planar

interface when $\cos(4\theta_0) < 0$. Given an anisotropic intensity, the planar interface is more stable with the decrease of $\cos(4\theta_0)$. The simulation results are shown in Fig. 3 when θ_0 varies in $[0, \pi/4]$. As θ_0 increases from 0 to $\pi/4$ the planar interface lasts over a longer period of time, which indicates that the planar interface is more stable with the decrease of $\cos(4\theta_0)$. The planar interface becomes unstable early with wave number being large compared with that in the isotropic case when $\theta_0 = 0°$ and $\theta_0 = 15°$ ($\cos(4\theta_0) > 0$), while the planar interface is destabilized late with wave number being small compared with that in the isotropic case when $\theta_0 = 30°$ and $\theta_0 = 45°$ ($\cos(4\theta_0) < 0$).

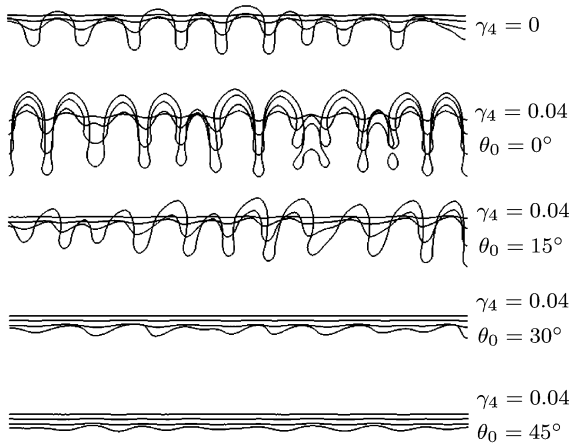


Fig. 3. Effects of misaligned angle between the preferred anisotropic orientation and the heat flow direction on the dynamic evolution of planar interface instability from 5 s to 6.5 s.

The simulation results accord well with the linear stability analysis about the surface tension anisotropy effects except for the tilting modulation with $\theta_0 = 15°$ and $\theta_0 = 30°$. In the linear stability analysis, the surface tension anisotropy is shown to be irrelevant to the tilting growth but the anisotropic kinetics is related to that in Refs. [5]–[7]. However, simulation results and experimental investigations indicate that the anisotropic surface tension may be relevant to the tilting growth in the fully nonlinear regime.[9−17] Close-ups of magnified small interface modulation at an early time are shown in Fig. 4, where the dynamic evolution of interface can be considered in the linear regime. It can be seen that the interface modulation is still well-balanced without leaning to the right side. The wave number is almost the same as the tilting modulation with one second later than that for the corresponding interface morphologies in Fig. 3. Here, the investigations reveal that in the linear growth regime, the anisotropic surface tension does not lead to tilting growth, while in the nonlinear growth regime, the misaligned orientation of surface tension anisotropy does induce travelling wave modulation after the planar interface instability.

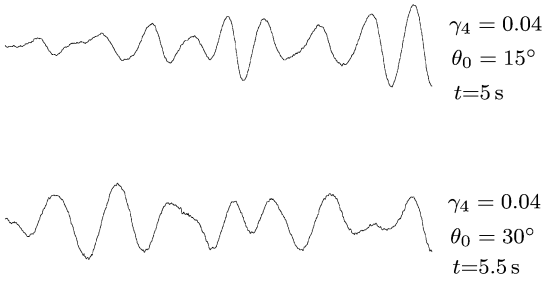


Fig. 4. Close-ups of interface modulation at the early stage with misaligned angle.

4. Conclusion

The effect of the surface tension anisotropy on the onset of the initial planar interface instability during directional solidification is investigated by a quantitative phase field model. The surface tension anisotropy affects the incubation stage of planar instability and the interface morphology of initial instability compared with those in the isotropic case. The misorientation of preferred crystallographic orientation with respect to the heat flow direction determines the stabilizing effect or destabilizing effect. The linear effect of surface tension anisotropy does not lead to leaning growth, but the nonlinear effect with misaligned orientation induces tilting growth.

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