# What happens to the initial planar instability when the thermal gradient is increased during directional solidification?*

Wang Zhi-Jun(王志军), Wang Jin-Cheng(王锦程)†, Li Jun-Jie(李俊杰), Yang Gen-Cang(杨根仓), and Zhou Yao-He(周尧和)

*State Key Laboratory of Solidification Processing, Northwestern Polytechnical University, Xi'an* 710072, *China*

(Received 20 January 2011; revised manuscript received 15 June 2011)

The positive thermal gradient is one of the most important parameters during directional solidification. The increase of the thermal gradient usually stabilizes the planar interface in the steady state analysis. However, in the initial transient range of planar instability, the thermal gradient presents complicated effects. Time-dependent analysis shows that the increase of the thermal gradient can enhance both the stabilizing effects and the destabilizing effects on a planar interface. The incubation time first decreases and then increases with the increase of the thermal gradient. Moreover, the initial average wavelength always increases with the thermal gradient increasing, contrary to the effect of the thermal gradient on the steady cellular/dendritic spacing. This reveals the types of spacing adjustment after planar instability.

**Keywords:** directional solidification, thermal gradient, transient behaviour, initial planar instability

**PACS:** 81.30.fb, 47.20.Ib, 47.20.Ma **DOI:** 10.1088/1674-1056/20/10/108104

## 1. Introduction

Directional solidification of binary alloys has received considerable attention over the past several decades as an example of a non-equilibrium pattern formation system that has technological significance. These patterns are of paramount importance in determining the properties of materials, as well as reflecting the intrinsic features of a nonlinear system.[1−5] Thermal gradient is one of the most important parameters for determining pattern formation during directional solidification. From planar instability to the formation of cellular/dendritic arrays, the positive thermal gradient plays an important role. Abundant theoretical analyses and experimental investigations have revealed the effects of the thermal gradient on microstructural evolution. Linear stability analysis indicates that a positive thermal gradient can stabilize the planar interface during directional solidification. However, in the cell-dendrite transition region, the thermal gradient displays a destabilizing nature with regard to the side-branching instability, which is contrary to the effects in planar instability.[6−8] Experimental and simulation results have also presented the effects of the thermal gradient on the final steady morphology, such as the tip radius, the primary spacing, and so on.[3] Although the effect of the thermal gradient on planar instability has been well described, efforts have been devoted only to the criterion of planar instability rather than to the instability process. The effects of the thermal gradient on directional solidification have not been completely understood yet.

Planar instability is a dynamical process. At the very beginning, the planar interface moves backward with respect to the thermal gradient. The solutal boundary layer is gradually established ahead of the front. The planar instability occurs during the backward movement of the interface. After planar instability, the displaced interface position accelerates. The incubation time of planar instability is the period from the very beginning to the onset of planar instability. In directional solidification, investigations about the effects of the thermal gradient on microstructure selection in steady state were fruitful, while the study

*Project supported by the National Natural Science Foundation of China (Grant No. 51071128), the Program for New Century Excellent Talents in University (Grant No. NCET-09-0683), the Fund of State Key Laboratory of Solidification Processing in Northwestern Polytechnical University, China (Grant Nos. 17-TZ-2007, 03-TP-2008, and 24-TZ-2009), the National Basic Research Program of China (Grant No. 2011CB610401), and the China Postdoctoral Science Foundation (Grant No. 20110491689).

†Corresponding author. E-mail: jchwang@nwpu.edu.cn

© 2011 Chinese Physical Society and IOP Publishing Ltd

of the transient stage was scarce. Investigating the transient behaviour of planar instability is one of the most important steps to understand the details of pattern formation during directional solidification, where the incubation time and the initial wavelength are the focus points.[9−13] The incubation time is related to the instantaneous interface velocity and solute boundary layer at the onset of the initial planar instability. The initial wavelength is the starting point of the adjustment of primary cellular/dendritic spacing, which influences the final steady state microstructure.

According to the linear stability analysis of the steady state, the positive thermal gradient plays a stabilizing role in planar instability.[14−17] However, the steady state analysis cannot give enough information about the planar instability process. With regard to the onset of initial planar instability, the effects of the thermal gradient in the transient stage are seldom considered. The history-dependent characteristic of directional solidification calls for descriptions of dynamic evolution from the initial planar instability to the final steady state, which means that the transient stage is the foundation of subsequent microstructure evolution. Therefore, the effects of the thermal gradient on the initial planar instability need to be clarified for understanding the microstructure evolution and optimal design in manufacture processes. In this paper, the nontrivial phenomenon is revealed for the influence of the thermal gradient on planar instability in the transient stage.

Planar instability in the transient stage during directional solidification was first analysed by Warren and Langer.[9] Information about the initial planar instability can be extracted very well from their analysis. Recently, a simple model was proposed to describe the onset of initial planar instability by combining the Fourier synthesis method and time-dependent linear stability analysis.[13] According to the model, the displaced interface microstructure and the incubation time onset of planar instability can be well predicted. In previous investigations, effects of pulling velocity and surface tension anisotropy have been considered.[13,18] The results are significantly different from the steady state analyses. Therefore, it is necessary to re-examine and quantitatively predict the initial planar instability with different thermal gradients with the consideration of both scientific and technical interest. Herein, the model[13] is employed to study the effects of the thermal gradient on the initial planar instability quantitatively according to the incubation time and initial average wavelength.

## 2. Model description

The directional solidification analysed here is a two-dimensional case with isotropic surface tension, moving in the $z$ direction at a constant pulling velocity $V$ in an externally imposed thermal gradient $G$. The temperature is defined by a frozen field as $T = T_{\rm M} + Gz$, where $T_{\rm M}$ is the melting temperature of pure solvent. During the transient stage, the concentration boundary layer can be approximately described as[9]

$$C(z,t) = C_\infty + [C(z_{\rm I},t) - C_\infty]\exp[-2(z-z_{\rm I})/l], \quad (1)$$

where $C(z,t)$ is the time-dependent concentration field ahead of the planar front, $z_{\rm I}$ the interface position, $l$ diffusion length and $C_\infty$ the concentration far ahead of the solidification front. Here $z_{\rm I}$ and $l$ are time-dependent, and satisfy two coupled differential equations,

$$V_{\rm I} = \frac{\partial z_{\rm I}}{\partial t} = \frac{2D(z_{\rm I}-z_\infty)}{l(1-k)z_{\rm I}} - V, \quad (2)$$

$$\frac{\partial l}{\partial t} = \frac{4D(z_\infty - kz_{\rm I})}{l(1-k)z_{\rm I}} - \frac{l}{z_{\rm I}-z_\infty}\frac{\partial z_{\rm I}}{\partial t}, \quad (3)$$

where $V_{\rm I}$ is the instantaneous interface speed, $D$ the chemical diffusion coefficient in liquid, $k$ the partition coefficient and $z_\infty = mC_\infty/G$, $m$ the liquidus slope. According to Eqs. (2) and (3), the establishment of the solutal boundary layer is affected by the thermal gradient through parameter $z_\infty$.

The dynamics of the initial planar instability can be described by the time-dependent linear stability analysis as[9]

$$\begin{aligned}&\omega^*\left[1+\frac{2(z_{\rm I}-z_\infty)}{l}+\frac{\Gamma\omega^2}{G}\right]\\ &= \frac{V_{\rm I}}{D}+\frac{2(z_{\rm I}-z_\infty)}{l}\\ &\quad\times\left[\frac{V_{\rm I}+V}{D}+\frac{\sigma_\omega(t)}{V_{\rm I}+V}+\frac{1}{z_{\rm I}}+\frac{\Gamma\omega^2}{Gz_{\rm I}}\right]. \end{aligned} \quad (4)$$

Here $\sigma_\omega(t) = \partial_t A_\omega(t)/A_\omega(t)$ is the amplification rate of perturbation with space frequency $\omega$ and amplitude $A_\omega(t)$, $\omega^* = (V_{\rm I}+V)/2D + \left[\omega^2 + ((V_{\rm I}+V)/2D)^2\right]^{1/2}$. $\Gamma = \gamma T_{\rm M}/L$ is the Gibbs–Thomson coefficient, $\gamma$ the isotropic surface tension energy and $L$ the latent heat. According to the Fourier synthesis method[13]

$$z_{\rm I}(x,t) - z_0(t) = \sum A_\omega(t)\cos(\omega x + \phi_\omega(t)), \quad (5)$$

where $z_0(t)$ is the basic interface position, computed approximately according to Eqs. (2) and (3). The

amplitude $A_\omega(t) = A_\omega(0)\exp\left(\int_{t_0}^{t}\sigma_\omega(t)\mathrm{d}t\right)$ and the phase $\phi_\omega(t)$ are assumed to be stochastic in $[0, 2\pi]$ with mean distribution. $t_0$ is the critical time when $\sigma_\omega(t)$ changes from negative to positive and $A_\omega(0) = d_0$, where $d_0 = \Gamma/(L/c_\mathrm{p})$ is the thermal capillary length and $c_\mathrm{p}$ the specific heat at constant pressure. The corresponding phase of the modulation $\phi_\omega(t) = \phi_\omega(t_{0-})$ after $t_0$. This simple model can quantitatively predict the incubation time and the initial average wavelength at the onset of planar instability. The effects of the thermal gradient on the initial planar instability are evaluated according to the variation of the incubation time and the initial average wavelength.

## 3. Results and discussion

With a constant thermal gradient, both the incubation time from the very beginning to the initial visible interface modulation and the initial average wavelength decrease with the increase of pulling velocity in the transient stage of planar instability, which indirectly verifies that the pulling velocity is a destabilizing factor.[13] However, the effects of the thermal gradient on the initial planar instability during transient stage are quite different. In the transient stage, the instantaneous interface velocity and the diffusion length in the time-dependent linear stability analysis are also related to the thermal gradient as shown in Eqs. (2) and (3). These nonlinear coupled relations make the effects of the thermal gradient on the initial planar instability unexpected. In the time-dependent linear stability analysis of Eq. (4), the amplification of the fluctuation depends on the thermal gradient and the time-dependent interface velocity and diffusion length. Therefore, it is uncertain that increasing the thermal gradient means the increase of the incubation time, which is quite different from the steady analysis. Furthermore, the variation of initial average wavelength with the thermal gradient is unclear.

The SCN with the dye coumarin 152 with composition $C_\infty = 0.43$ wt% is employed for analysis. This transparent organic alloy has been used to testify the establishment of solutal boundary layer experimentally.[12] The relevant physical properties of SCN-C152 are $k = 0.05$, $D = 4.5 \times 10^{-10}$ m$^2$/s, and $m = 542$ K/mol. For $V = 50$ μm/s, the establishment of solute boundary layer with different thermal gradients is revealed by the time-dependent interface velocity and diffusion length as shown in Fig. 1 under the assumption of a planar interface. It should be noted that the thermal gradient is considered here only for the theoretical investigation without the consideration of the experimental implementation. It shows that the evolutions of interface velocity and diffusion length greatly depend on the thermal gradient in the transient stage, which affects the establishment process of the solutal boundary layer. For a small thermal gradient, the instantaneous interface velocity increases slowly and the large diffusion length corresponds to thick solutal boundary layer. But for a large thermal gradient, the interface velocity increases rapidly to the pulling velocity with a small diffusion length. Therefore the increase of the thermal gradient will reduce the initial transient stage without considering planar instability. Accordingly, equations (3) and (4) can provide the quantitative information about the design of crystal growth during the initial transient stage.

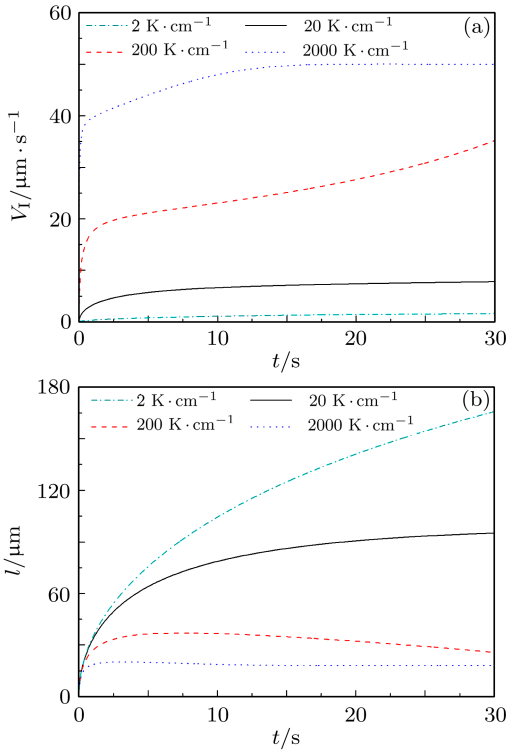



**Fig. 1.** (colour online) Time-dependent interface velocities (a) and time-dependent diffusion lengths (b) for $V = 50$ μm/s.

With regard to the initial planar instability during the transient stage, the thermal gradient plays a more complicated role compared with the steady state analysis. Although the positive thermal gradient directly stabilizes the planar interface in the linear stability analysis, the thermal-gradient-dependent interface velocity and diffusion length indirectly reflect the destabilizing effects of the thermal gradient on the planar interface in the time-dependent analysis. As the

thermal gradient increases, both the destabilizing and the stabilizing effects increase. So, it is unclear that which one will play a predominant role. In the part of model description, the simple model provides the specific information considering the onset of the initial planar instability. Figure 2 shows the variations of the incubation time and the initial wavelength with the thermal gradient at several constant pulling velocities. As shown in Fig. 2(a), the incubation time first decreases and then increases as the thermal gradient increases. For a small thermal gradient, the interface velocity increases slowly, leading to the faint destabilizing effect. For a large thermal gradient, stabilizing effects of the thermal gradient can compensate the strong destabilizing effects introduced by rapidly increasing interface velocity. However, for a medium thermal gradient, the destabilizing effect plays a predominant role. Therefore, the planar interface can last for long period for both relatively small and relatively large thermal gradients. For the monotonically decreasing part of the incubation time curve, the destabilizing effects of the thermal gradient increases faster, while for the increased part the stabilizing effect of the thermal gradient increases faster. This interesting phenomenon has not been reported previously. Results from different pulling velocities present a similar variation regularity of the curve of incubation time versus the thermal gradient and also indicate that the crossover thermal gradient in the incubation time curves increases with the increase of pulling velocity. Moreover, the solidified length onset of the initial planar instability is one of the characteristics concerned for crystal growth design. When $V = 50$ μm/s, planar instability occurs at a time of about 10 s for $G = 20$ K/cm and it appears at a time of 5 s for $G = 200$ K/cm. As shown in Fig. 1(a), before the initial planar instability, the interface velocity varies significantly with the thermal gradient. The crystal growth length from the beginning to the onset of the initial planar instability can be obtained from Fig. 1(a) by the integral value of $\int_{t=0}^{t=t_0} V(t)\mathrm{d}t$. The crystal growth lengths at which planar instability occurs are 53.1 μm, 90.7 μm, 216.6 μm for 20 K/cm, 200 K/cm, and 2000 K/cm, respectively, always increasing with the increase of the thermal gradient.

The planar instability morphology is also obviously influenced by the thermal gradient. As shown in Fig. 2(b), the initial average wavelength increases with the increase of the thermal gradient. The initial planar instability morphology can be obtained by computing Eq. (5). The initial average length is directly obtained from the interface morphology.[13] The results show that the relationship between initial average length and the thermal gradient approximately satisfies the power law of $\lambda_0 \propto G^{0.13}$. The selection of initial average length is determined by the amplification process of background fluctuation. The small gradient and the instantaneous interface velocity codetermine the initial average wavelength. The instantaneous interface velocity increases as the thermal gradient increases as shown in Fig. 1. In the linear stability analysis, the critical instability wavelength decreases as the interface velocity increases while increases as the thermal gradient increases. Accordingly, the thermal gradient plays a dominant role in determining the initial average wavelength compared with the instantaneous interface velocity.

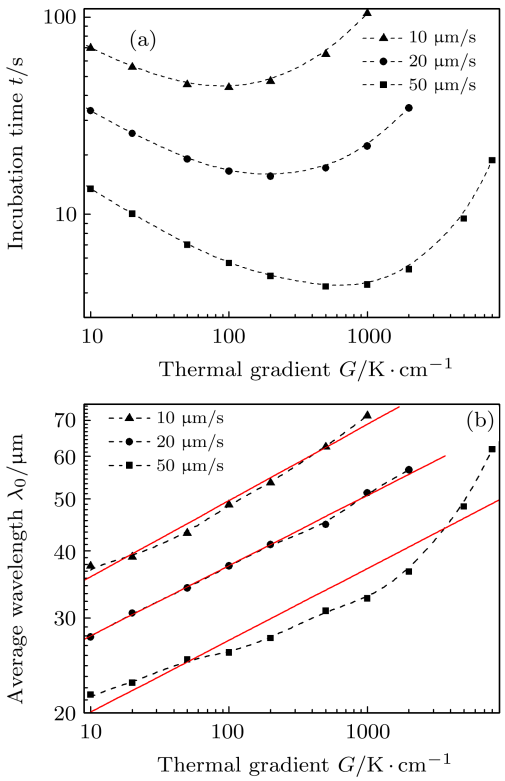



**Fig. 2.** (colour online) Effects of the thermal gradient on the initial instability with different pulling velocities. Panel (a) shows the incubation times versus the thermal gradient and panel (b) exhibits average wavelengths versus the thermal gradient.

In directional solidification, the initial planar instability is the origin of the cellular/dendritic evolution. Usually the overgrowth of several preferred cellular rudiments will suppress the other cells after planar instability, when the cell/dendrite spacing is much larger than the initial average wavelength.

However, the primary cellular/dendritic spacing in a steady state decreases with the thermal gradient as $\lambda_1 \propto G^{-1/2}$.[2] A comparison between the power law of initial average wavelength $\lambda_0 \propto G^{0.13}$ and the steady primary spacing law $\lambda_1 \propto G^{-1/2}$ shows that the overgrowth phenomenon is more likely to happen with a small thermal gradient, while the tip splitting phenomenon is more likely to occur at a large thermal gradient for spacing adjustment after planar instability. This reveals the reason why there are different adjustment manners for the primary spacing after the initial planar instability.[19]

## 4. Conclusion

Increase of the thermal gradient does not always enhance the initial planar stability during the transient stage. Planar instability is determined by the competition between a stabilized factor from the thermal gradient and an unstabilized factor from the solutal boundary layer ahead of the interface during directional solidification. Although the thermal gradient itself stabilizes the planar interface in the linear stability analysis, its effect on the solutal boundary layer shows a reverse action during the transient stage. Their combination presents a crossover phenomenon for the incubation time of planar instability with different thermal gradients. The initial average length always increases with the thermal gradient as $\lambda_0 \propto G^{0.13}$. The detection of these unexpected phenomena can provide a clue for the further optimization of the thermal gradient in the design of crystal growth.

## References


[1] Langer J S 1980 *Rev. Mod. Phys.* **52** 1

[2] Somboonsuk K, Mason J T and Trivedi R 1984 *Metall. Trans.* A **15** 967

[3] Billia B and Trivedi R 1993 *Handbook of Crystal Growth* (Amsterdam: Elsevier) Vol. 1 p. 899

[4] Boettinger W J, Coriell1 S R, Greer A L, Karma A, Kurz W, Rappaz M and Trivedi R 2000 *Acta Mater.* **48** 43

[5] Wang Z J, Wang J C and Yang G C 2010 *Chin. Phys.* B **19** 017305

[6] Georgelin M and Pocheau A 1998 *Phys. Rev.* E **57** 3189

[7] Georgelin M and Pocheau A 1998 *Eur. Phys. J.* B **4** 169

[8] Teng J, Liu S and Trivedi R 2009 *Acta Mater.* **57** 3497

[9] Warren J A and Langer J S 1993 *Phys. Rev.* E **47** 2702

[10] Huang W, Geng X and Zhou Y 1993 *J. Cryst. Growth* **134** 105

[11] Ma D and Jie W 1996 *Acta Phys. Sin.* (Overseas Edition) **5** 306

[12] Losert W, Shi B Q and Cummins H Z 1998 *Proc. Natl. Acad. Sci.* **95** 431

[13] Wang Z J, Wang J C and Yang G C 2009 *Appl. Phys. Lett.* **94** 061920

[14] Mullins W W and Sekerka R F 1964 *J. Appl. Phys.* **35** 444

[15] Wang Z J, Wang J C and Yang G C 2008 *Acta Phys. Sin.* **57** 1246 (in Chinese)

[16] Wang Z J, Wang J C and Yang G C 2009 *Cryst. Res. Technol.* **44** 43

[17] Chen M W, Lan M, Yuan L, Wang Y Y, Wang Z D and Xu J J 2009 *Chin. Phys.* B **18** 1691

[18] Wang Z J, Wang J C and Yang G C 2009 *Phys. Rev.* E **80** 052603

[19] Ding G L 1997 *Morphological Characteristics and Transitions During the Solidification of Binary Alloys* (Xi'an: Xi'an Northwestern Polytechnical University Press) (in Chinese)