

## CHAPTER 8

# INTERFACIAL INSTABILITIES AND PATTERN FORMATIONS IN SOLIDIFICATION

*[This chapter is based on the lectures of Dr. Jian-Jun Xu of China and Mc Gill University, Montreal, Canada, at the 5th SERC School.]*

## 8.0. Introduction

Pattern formation is a common phenomenon in nature. When a system is driven away from its equilibrium state, a dynamic process will spontaneously take place. During this process, a certain type of pattern is often observed in the system. This pattern is characterized by the distribution of a certain physical quantity, such as streamlines, isothermal lines, iso-concentration lines for species, iso-potential lines, etc.

Stationary, or fractal patterns are often observed at the later stages of a process. The essence and mechanism of fractal, chaotic pattern formation is a wide-open subject in all scientific areas. Our understanding of this complex phenomenon is obviously far from complete and in this talk we will not deal with this topic. From a theoretical point of view, the formation of the stationary patterns is more fundamental, as well as simpler. The essence and mechanism of such formation is, of course, of great interest. Our interest will be restricted in the patterns formed in solidification.

Tree-like interfacial patterns can appear in various growth systems for crystal materials as shown in Figures 8.0.1 and 8.0.2 Kobayashi and Furukawa (1991), Takahashi et al (1995). This kind of patterns is called dendritic pattern. It describes a typical case of dendrite growth. The system is originally

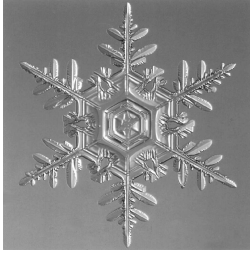


Figure 8.0.1 Photograph of a typical snowflake pattern taken by Furukawa

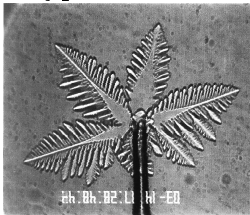


Figure 8.0.2 The photo of an ice crystal taken by Furukuwa et al., which is growing at the undercooling temperature  $\Delta T = 0.37$  K

setup in a metastable state, such as a liquid state with a given uniform undercooling temperature. The dynamical process is initiated by introducing a tiny seed or in other related ways. Once it starts, the crystal growth will proceed spontaneously according to the physical laws.

Another typical example of dendrite growth occurs in unidirectional solidification. In Figures 8.0.3 we show a sketch of a unidirectional solidification device, which consists of a thin sample in a Hele–Shaw cell and two uniform temperature zones at temperatures  $T_1$  and  $T_2$ , which are, respectively, larger and smaller than the melting temperature of the material.

When the sample which is made of a binary mixture is at rest, the interface between solid and liquid is planar. The sample is pulled at a constant imposed velocity  $V$ . When the pulling velocity  $V$  increases to a critical number the flat interface will start a sinusoidal structure, which will gradually evolve into a deeper cellular structure as shown in Figure 8.0.4, with the further increase of the pulling velocity. When the pulling velocity  $V$  exceeds another critical value, the deep cellular structure will transform into arrays of dendrites, as shown in Figure 8.0.5.

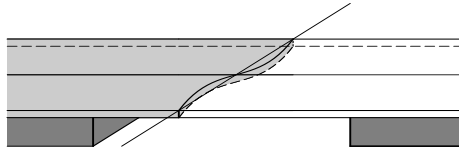


Figure 8.0.3 Sketch of a unidirectional solidification device

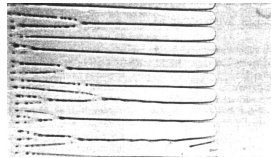


Figure 8.0.4 Deep cellular structure formed during unidirectional solidification

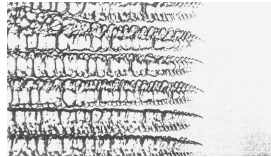
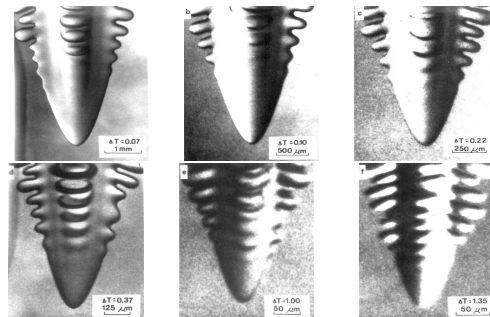


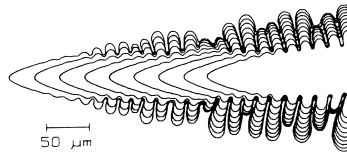
Figure 8.0.5 Dendritic array structure formed during unidirectional solidification

Dendrite growth also frequently appears in many other forms of material processing, such as alloy casting, metal ingot formation, and welding. Dendrite growth is one of the most profound subjects in the area of interfacial pattern formation. A typical single free dendrite growth is shown in Figure 8.0.6a, a photograph of single dendrite growth from a pure organic material melt succinonitrile (SCN) was made by Nash and Glicksman (1974), Huang and Glicksman (1981) while Figure 8.0.6b shows the experimental recording curves for two-dimensional dendrite growth from a supersaturated  $\text{NH}_4\text{Br}$  solution, studied by Dougherty and Gollub (1988).

Experimental observations show that at the later stage of growth a dendrite appears to have a smooth tip moving with a constant velocity. It emits a stationary wave-train, propagating along the interface towards the root.



(a)



(b)

Figure 1.0.6 (a) A photograph of a single free dendrite growth from pure organic material melt succinonitrile (SCN); (b) Experimental curve of two-dimensional dendrite growth from a supersaturated ( $\text{NH}_4\text{Br}$ ) solution

On the basis of this experimental data, it was confirmed that at the later stage of dendrite growth, the tip velocity is a uniquely determined function of the growth condition and the properties of the material.

The basic questions of dendrite growth have been:

1. What mechanism determines the tip growth velocity?
2. What is the origin and essence of the microstructure?

These problems have motivated and inspired a broad range of theoretical and experimental research activities for over a generation. For past several decades Ivantsov (1947), Hurler (1993). It is now recognized that the sinusoidal pattern formed on a initially flat interface is determined by the morphological instability mechanism, that is called the **Mullins-Sekerka Instability**, Mullins and Sekerka (1963, 1964) while the dendritic pattern formed on a curved interface at the later stage of growth is determined by a global wave instability mechanism, that is called the **Global Trapped Wave instability** Xu (1991, 1996, 1997, 2004).

The present talk is going to carry out some mathematical analysis on these interfacial phenomena and demonstrate these two instability mechanisms.

## 8.1. Macroscopic Continuum Model

We use the macroscopic continuum model. This implies that the liquid and solid bulk phases will be treated as continuous media, while the interface is considered as a geometric surface. Let us now imagine that, a front that separates the solid phase and liquid phase, moves into the liquid phase and transforms more and more liquid into the solid phase. Eventually, the whole liquid phase is transformed into the more thermodynamically stable solid phase. During solidification, there may co-exist several interactive macroscopic transport processes in the system. First of all, due to the phase transition, the latent heat is released at the interface. Hence, a solidification system intrinsically has an inhomogeneous temperature field, and heat transfer through conduction is unavoidable. Furthermore, if the system involves two species, for instance, if it contains an impurity, then this binary mixture should be described not only by the temperature field, but also by the impurity concentration field. It is known that for a binary system with co-existing liquid and solid phase, the impurity concentration will have a jump at the interface because of the segregation effect. Therefore, the concentration field in a solidifying binary mixture system is always inhomogeneous. A mass transfer through mass diffusion in the concentration field will be present. Finally, the liquid phase in a solidification system may be in motion induced by various driving forces. For instance, the density change during phase transition will cause convection in the bulk liquid, because the interface acts as a mass sink when the liquid density is smaller than the solid density, or as a mass source when the liquid density is larger than the solid density. The buoyancy effect due to gravity may also cause convection, because the inhomogeneous temperature induces inhomogeneous density. Moreover, other forces on the body and external flow can all cause convection motion in the liquid phase. When convection exists in the liquid phase, there will be a momentum transfer in the system governed by fluid dynamics. If the above three kinds of macroscopic transport processes co-exist, they will obviously interact with one another. For simplicity, in this lecture we shall restrict ourself in the case of solidification of pure melt.

### 8.1.1. Macroscopic transport equations

Consider a front moving from solid phase towards the melt, with a characteristic velocity  $V$  in the reference frame in which the solid phase is at rest. For convenience, it may be called the rest frame. We assume that the liquid phase has mass density  $\rho$ , thermal diffusivity  $\kappa_T$ , specific heat  $c_p$ ; while the corresponding thermal characteristics in the solid phase are denoted by  $\rho_S$ ,  $\kappa_{TS}$  and  $c_{pS}$ , respectively.

Convective motion may occur in the liquid phase for a number of reasons as previously mentioned. We assume that the liquid can be considered as an incompressible Newtonian fluid. The density inhomogeneity caused by the inhomogeneity of temperature is not important except in terms of a buoyancy effect. Thus, the Boussinesq approximation is applicable. The state of the system is described by the following macroscopic fields:

1. the temperature field in the liquid phase,  $T(\mathbf{r}, t)$ ,
2. the temperature field in the solid phase,  $T_S(\mathbf{r}, t)$ ,
3. the absolute velocity field in the liquid phase,  $\mathbf{U}(\mathbf{r}, t)$ .

Here, we use  $\mathbf{r}$  and  $t$  to denote the space vector and time, respectively. The governing equations consist of the heat conduction equation, mass diffusion equation and the Navier–Stokes equations in the Boussinesq model. These equations can be written as follows:

1. The heat conduction equation for the liquid phase. In the rest frame, the heat conduction equation can be written in the form:

$$c_p \rho \left( \frac{\partial T}{\partial t} + \mathbf{U} \cdot \nabla T \right) = k_T \nabla^2 T \quad (8.1.1)$$

or

$$\frac{\partial T}{\partial t} + \mathbf{U} \cdot \nabla T = \kappa_T \nabla^2 T \quad (8.1.2)$$

where  $k_T$  is the heat conduction coefficient, while  $\kappa_T = \frac{k_T}{c_p \rho}$  is the thermal diffusivity in the liquid phase.

2. The heat conduction equation for the solid phase. In the rest frame, we similarly have

$$\frac{\partial T_S}{\partial t} = \kappa_{TS} \nabla^2 T_S \quad (8.1.3)$$

where  $\kappa_{TS}$  is the thermal diffusivity in the solid phase.

3. The continuity equation. For the incompressible liquid mixture, we have

$$\nabla \cdot \mathbf{U} = 0. \quad (8.1.4)$$

4. The momentum equation:

$$\frac{\partial \mathbf{U}}{\partial t} + (\mathbf{U} \cdot \nabla) \mathbf{U} = -\frac{1}{\rho} \nabla P + \nu \nabla^2 \mathbf{U} + \beta(T - T_*) \mathbf{g}, \quad (8.1.5)$$

where  $P$  is the reduced pressure. If we introduce the vorticity  $\boldsymbol{\Omega}$  as

$$\boldsymbol{\Omega} = \nabla \times \mathbf{U}, \quad (8.1.6)$$

the momentum equation can be replaced by the vorticity equation:

$$\frac{\partial \boldsymbol{\Omega}}{\partial t} + \nabla \times (\boldsymbol{\Omega} \times \mathbf{U}) = \nu \nabla^2 \boldsymbol{\Omega} + \nabla \times \left\{ \beta(T - T_*) \mathbf{g} \right\}. \quad (8.1.7)$$

Here, we assume that there is no body force in the system, except for gravity;  $\mathbf{g}$  is the acceleration of gravity;  $\nu$  is the kinematic viscosity;  $\beta$  is the thermal expansion coefficient; and  $T_*$  is a reference temperature.

### 8.1.2. The interface conditions

The full set of boundary conditions must be specified for each specific problem. For instance, the far-field conditions will be different for different growth conditions. However, for the problems under investigation here the same type of interface conditions are, in general, always applied. We denote the interface shape in the rest frame by  $S = S(\mathbf{r}, t) = 0$ . For the certainty, we assume that in the domain of solid phase  $S < 0$ , while in the domain of liquid phase,  $S > 0$ . Hence, the unit normal vector of interface,  $\mathbf{n}$  pointing to the liquid phase can be expressed as

$$\mathbf{n} = \frac{\nabla S}{|\nabla S|}.$$

Note that the interface location during solidification is unknown. The determination of this interface shape function is a part of the solution. These problems, in mathematics, are all called **free boundary problems or moving boundary problems**. To determine the interface conditions, we assume that the system is in a local thermodynamic equilibrium at the interface. With this assumption, we can derive the following interface conditions in the rest frame used:

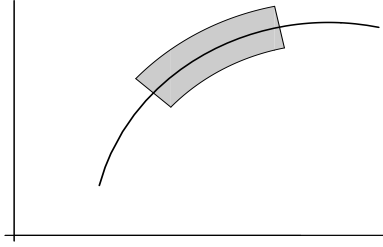


Figure 8.1.1 Sketch of a moving front in an undercooled melt

1. *The thermodynamic equilibrium condition for temperature:*

$$T = T_S . \quad (8.1.8)$$

2. Another thermodynamic equilibrium condition at the interface is the phase equilibrium condition. This implies that the chemical potential of the solid phase must equal the chemical potential of the liquid phase. This condition will lead to the so-called *Gibbs–Thomson condition*, which determines the temperature of the interface.

The Gibbs–Thomson condition states that the solidification temperature at a general curved interface is different from the solidification temperature of the pure melt at a flat interface, say,  $T_{M0}$ . The correction comes from the effect of the curvature of the interface on the surface energy. Assume that the above sub-system is in thermodynamic equilibrium. From the minimum free energy principle at the local equilibrium state one finds that

$$T_S = T_{M0} \left( 1 - \frac{\gamma}{\Delta H} \mathcal{K} \right) , \quad (8.1.9)$$

where  $\gamma$  is the surface tension constant,  $\Delta H$  is the latent heat per unit volume of the solid,  $T_{M0}$  is the melting temperature of a flat interface, and  $\mathcal{K}$  is twice the local mean curvature of the interface.

Under some special situations, for instance, for the system of rapid solidification, one may need to consider non-equilibrium interface. This means that due to the kinetic attachment process, the realistic local temperature at the interface differs from the equilibrium temperature by an amount proportional to the local growth speed of the interface. In this case, the above Gibbs–Thomson condition needs the modification by adding a correction



term as follows:

$$T_S = T_{M0} \left( 1 - \frac{\gamma}{\Delta H} \mathcal{K} \right) - \beta_0 U_I, \quad (8.1.10)$$

where  $\beta_0$  is the coefficient of kinetic attachment.

3. *The energy conservation condition:* The enthalpy is conserved during the phase transition, so, if neglecting the part of variation of interfacial energy, one may write

$$\begin{aligned} \Delta H(U_{S_n} - U_I) &= \mathbf{n} \cdot \left[ (k_T \nabla T)_{\text{liquid}} - (k_T \nabla T)_{\text{solid}} \right] \\ &\quad - \left[ (c_p T)_{\text{liquid}} - (c_p T)_{\text{solid}} \right] \rho_S U_I, \end{aligned} \quad (8.1.11)$$

where

- $U_{S_n}$  is the normal component of the solid phase velocity at the interface, observed in the rest frame. We have assumed that it is zero.
- $U_I$  is the local growth velocity of the interface in the rest frame.
- $\mathbf{n}$  is the normal vector of the interface directing to the liquid phase.

The left-hand side of the formula (8.1.11) is the latent heat release per unit volume of solid per unit time. The first part of the right-hand side of (8.1.11) is the total enthalpy flux away from the interface to both the liquid and solid sides due to heat conduction; while the second part is the total enthalpy flux due to the convective motion.

4. *The total mass conservation condition:*

$$\rho(U_{L_n} - U_I) = \rho_S(U_{S_n} - U_I). \quad (8.1.12)$$

5. *The continuity condition for the tangential component of velocity:*

$$U_{L\tau} = U_{S\tau}. \quad (8.1.13)$$

In the above, the symbols have the meanings:

$\mathbf{e}_\tau$ : the local unit tangent vector at the interface;

$U_{Ln}$ : the normal component of the absolute velocity of a liquid element at the interface;

$U_{L\tau}$ : the tangential component of the absolute velocity of a liquid element at the interface;

$U_{S\tau}$ : the tangential component of the absolute velocity of a solid element at the interface.

Besides the above local thermodynamic equilibrium conditions and kinematic conditions, we also have some mechanical equilibrium conditions at the interface. These are the dynamic interface conditions derived from the momentum balance along the normal and tangential directions at the interface. Since the pattern formation problem is mostly not concerned with the stresses and strains in the solid phase, these dynamic conditions will not be needed. Thus, by assuming that the solid phase is unmoved rigid body, one may set  $U_{S\tau} = U_{Sn} = 0$ , so that the interface conditions Ivantsov (1947), Horvay and Chan (1961), Rutter and Chalmers (1953), may be re-written as 3'.

$$-\Delta H U_I = \mathbf{n} \cdot \left[ (k_T \nabla T)_{\text{liquid}} - (k_T \nabla T)_{\text{solid}} \right] - \left[ (c_p T)_{\text{liquid}} - (c_p T)_{\text{solid}} \right] \rho_S U_I, \quad (8.1.14)$$

4'. *The total mass conservation condition:*

$$\rho(U_{Ln} - U_I) = -\rho_S U_I. \quad (8.1.15)$$

5'. *The continuity condition for the tangential component of velocity:*

$$U_{L\tau} = 0. \quad (8.1.16)$$

One item in the above formulation that needs clarification is how to define and calculate the local growth velocity of the interface,  $U_I$ . For the sake of convenience, we often use a moving frame  $(x_1, x_2, x_3)$  with a velocity  $\mathbf{V}$  as the

reference frame. Assume that the velocity  $\mathbf{V}$  is parallel to the coordinate axes  $x_3$ -axis, whose unit vector is  $\hat{\mathbf{e}}_3$ . So that, we can write

$$\mathbf{U} = \mathbf{u} + V\hat{\mathbf{e}}_3, \quad (8.1.17)$$

where  $\mathbf{u}$  represents the relative velocity field of fluid observed in the moving frame. We shall also denote the relative growth velocity of the interface seen in the moving frame by  $\mathbf{u}_I$ . From the moving frame to the rest frame, the following Galilean transformation holds:

$$\begin{cases} X_1 = x_1 \\ X_2 = x_2 \\ X_3 = x_3 + Vt \\ t = t. \end{cases} \quad (8.1.18)$$

Assume that the interface shape equation in the moving frame and rest frame are

$$s(x_1, x_2, x_3, t) = 0$$

and

$$S(X_1, X_2, X_3, t) = 0,$$

respectively. Then, one may write

$$s(x_1, x_2, x_3, t) = S(x_1, x_2, x_3 + Vt, t) = 0 \quad (8.1.19)$$

and

$$S(X_1, X_2, X_3, t) = s(X_1, X_2, X_3 - Vt, t) = 0. \quad (8.1.20)$$

Thus, we can calculate the growth velocity of the interface in terms of the interface shape equation in the moving frame. Due to

$$\frac{\partial S}{\partial t} + (U_I \mathbf{n}) \cdot \nabla S = 0,$$

we have

$$\begin{aligned} U_I &= -\frac{\frac{\partial S}{\partial t}}{|\nabla S|} = -\frac{\frac{\partial s}{\partial t} - V\frac{\partial s}{\partial x_3}}{|\nabla s|} = -\frac{\frac{\partial s}{\partial t} - V(\hat{\mathbf{e}}_3 \cdot \nabla s)}{|\nabla s|} \\ &= -\frac{\frac{\partial s}{\partial t}}{|\nabla s|} + V \left( \hat{\mathbf{e}}_3 \cdot \frac{\nabla s}{|\nabla s|} \right) \\ &= u_I + V\hat{\mathbf{e}}_3 \cdot \mathbf{n}, \end{aligned} \quad (8.1.21)$$

where

$$u_{\text{I}} = -\frac{\frac{\partial s}{\partial t}}{|\nabla s|} \quad (8.1.22)$$

is the local growth velocity of the interface observed in the moving frame.

### 8.1.3. The scaling and the dimensionless system

As the first step of a mathematical treatment, one needs to make the system dimensionless by choosing a set of proper scales. The proper scales reflect the physical nature of the dynamical system. In many cases, using improper scales will cause difficulties in making either mathematical simplifications or physical interpretations.

For the problems formulated in the last section, we shall choose the characteristic growth velocity of the interface  $V$  as the scale for the velocity. The length scale can be chosen from the intrinsic length scales of the system. There are two intrinsic length scales in the system determined by the macroscopic transport processes: the thermal diffusion length  $\ell_{\text{T}} = \kappa_{\text{T}}/V$ . The physical quantity  $\Delta H/(c_{\text{p}}\rho)$  has the scale of temperature, so it is naturally chosen as the unit of temperature. By using these scales, we define the following dimensionless quantities and variables:

$$\begin{aligned} & (\bar{X}_1, \bar{X}_2, \bar{X}_3, \bar{t}) \\ & = (X_1/\ell_{\text{D}}, X_2/\ell_{\text{D}}, X_3/\ell_{\text{D}}, tV/\ell_{\text{D}}) \\ & (\bar{x}_1, \bar{x}_2, \bar{x}_3) = (x_1/\ell_{\text{D}}, x_2/\ell_{\text{D}}, x_3/\ell_{\text{D}}) \\ & \bar{T} = \frac{T - T_{\text{M}0}}{\Delta H/(c_{\text{p}}\rho)} \\ & \bar{\mathbf{u}} = \mathbf{u}/V \\ & \bar{\omega} = \omega\kappa_{\text{T}}/V^2 \quad (\omega = \nabla \times \mathbf{u}), \end{aligned} \quad (8.1.23)$$

where  $T_{\text{M}0}$  is the melting temperature at a flat interface. From (8.1.23), we have

$$\mathbf{U}/V = \bar{\mathbf{U}} = \bar{\mathbf{u}} + \hat{\mathbf{e}}_3. \quad (8.1.24)$$

Then, the governing equations and related boundary conditions in the moving frame can be derived from those in the rest frame by applying the transformation:

$$\frac{\partial}{\partial t} \rightarrow \frac{\partial}{\partial \bar{t}} - \mathbf{V} \cdot \nabla \quad (8.1.25)$$

or

$$\frac{\partial}{\partial \bar{t}} \rightarrow \frac{\partial}{\partial \bar{t}} - \hat{\mathbf{e}}_3 \cdot \nabla. \quad (8.1.26)$$

The dimensionless governing equations are obtained as follows:

1. Heat conduction equation in the liquid phase:

$$\left( \frac{\partial \bar{T}}{\partial \bar{t}} + \bar{\mathbf{u}} \cdot \nabla \bar{T} \right) = \nabla^2 \bar{T}. \quad (8.1.27)$$

2. Heat conduction equation in the solid phase:

$$\alpha_T \left( \frac{\partial \bar{T}_S}{\partial \bar{t}} + \hat{\mathbf{e}}_3 \cdot \nabla \bar{T}_S \right) = \nabla^2 \bar{T}_S. \quad (8.1.28)$$

3. Continuity equation for the liquid state:

$$\nabla \cdot \bar{\mathbf{u}} = 0. \quad (8.1.29)$$

4. Momentum equation for the liquid state:

$$\frac{\partial \bar{\omega}}{\partial \bar{t}} + (\bar{\mathbf{u}} \cdot \nabla) \bar{\omega} - (\bar{\omega} \cdot \nabla) \bar{\mathbf{u}} = \quad (8.1.30)$$

$$\text{Pr} \nabla^2 \bar{\omega} - \frac{\text{Gr}}{T_\infty} \nabla \times (\bar{T} \mathbf{e}_g). \quad (8.1.31)$$

In the above,  $\mathbf{e}_g$  is the unit vector along the direction of gravity and we introduce several parameters: the Prandtl number;

$$\text{Pr} = \nu / \kappa_T,$$

the undercooling parameter,

$$T_\infty = -[T_{M0} - (T_\infty)_D] c_p \rho / \Delta H = -\text{St},$$

where  $(T_\infty)_D$  denotes the dimensional undercooling temperature; the parameter St is sometimes called the Stefan number; the Grashof number;

$$\text{Gr} = g\beta(T_{M0} - (T_\infty)_D)\kappa_T/V^3$$

and the ratio parameter of thermal diffusivities in the solid and liquid

$$\alpha_T = \kappa_T / \kappa_{TS}.$$

Conventionally, the simplification with  $\alpha_T = 1$  is called the symmetric model; while the simplification with  $\alpha_T = 0$  is called the one-sided model.

The dimensionless conditions at the interface  $\bar{S}(\mathbf{r}, \mathbf{t}) = \mathbf{0}$  are:

(i) *Thermodynamic equilibrium for temperature:*

$$\bar{T} = \bar{T}_S; \quad (8.1.32)$$

(ii) *Gibbs–Thomson condition:*

$$\bar{T}_S = \Gamma \{\bar{\mathcal{K}}\}; \quad (8.1.33)$$

where  $\Gamma$  is the surface tension parameter

$$\Gamma = \frac{\ell_c}{\ell_T} \quad (8.1.34)$$

and

$$\ell_c = \frac{\gamma c_p T_{M0} \rho}{(\Delta H)^2} \quad (8.1.35)$$

is a length scale and usually called the capillary length. Since  $\ell_c$  is determined by the interfacial energy  $\gamma$ , it is sometimes considered as a microscopic length scale. It will be seen later that the parameter  $\Gamma$ , the ratio of the macroscopic length  $\ell_T$  and the microscopic length  $\ell_c$ , is the most important parameter for the stability of the system. Moreover, if the interface is considered as non-equilibrium, so, the effect of the kinetic attachment must be taken into account, then the above Gibbs–Thomson condition needs to be modified into the following form:

$$\bar{T}_S = \Gamma \{\bar{\mathcal{K}}\} - \beta \Gamma (\bar{u}_1 + \hat{\mathbf{e}}_3 \cdot \mathbf{n}). \quad (8.1.36)$$

Here the kinetic parameter  $\beta$  is defined as

$$\beta = \frac{\ell_k}{\ell_c}, \quad (8.1.37)$$

where  $\ell_k$  is the kinetic length, defined as

$$\ell_k = \frac{\beta_0 \kappa_T c_p \rho}{\Delta H}. \quad (8.1.38)$$

(iii) *Enthalpy conservation:*

$$\begin{aligned} & \left[ 1 - (1 - \delta)(1 + \alpha)(\bar{T} + \bar{T}_{M0}) \right] (\bar{u}_I + \hat{\mathbf{e}}_3 \cdot \mathbf{n}) \\ & = [\delta(1 + \alpha)\nabla\bar{T}_S - \nabla\bar{T}] \cdot \mathbf{n}. \end{aligned} \quad (8.1.39)$$

Hereby, we have defined

$$\delta = \frac{c_{pS}}{c_p}, \quad \alpha = \frac{\rho_S}{\rho} - 1, \quad \bar{T}_{M0} = \frac{T_{M0}}{\Delta H/(c_p\rho)}, \quad (8.1.40)$$

which measure the changes of the thermodynamic characteristics in the phase transition.

(iv) *Conservation of total mass:*

$$\bar{\mathbf{u}} \cdot \mathbf{n} + \alpha\bar{u}_I + (1 + \alpha)\hat{\mathbf{e}}_3 \cdot \mathbf{n} = 0; \quad (8.1.41)$$

(v) *Continuity of the tangential component of velocity:*

$$\bar{\mathbf{u}} \cdot \mathbf{e}_\tau = -\hat{\mathbf{e}}_3 \cdot \mathbf{e}_\tau. \quad (8.1.42)$$

Later, for the sake of simplicity, we shall omit the bar over all dimensionless quantities without confusion.

## 8.2. Unidirectional Solidification and the Mullins-Sekerka Instability

Before we begin the study of dendritic growth, it is appropriate to examine a simple case first: the instability of a planar interface in unidirectional solidification. Mullins and Sekerka were the first, in 1963, to perform a systematic analysis of this system. Their linear stability analysis is now called the *Mullins–Sekerka instability*. We assume that the system is two-dimensional, the gravity  $g$  and the ratio of densities  $\alpha$  can be set to zero.

### 8.2.1. Solidification with planner interface from a pure melt

We assume that an originally flat, solidifying front advances into a pure undercooled melt with velocity  $V$ . We utilize moving two-dimensional Cartesian

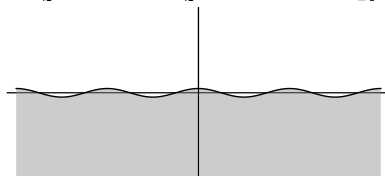


Figure 8.2.1 A simple model for unidirectional solidification

coordinates  $(x, y)$  with the  $x$ -axis fixed on the interface, as shown in Figure 8.2.1. The  $y$ -axis is along the growth direction of the interface. Furthermore, we assume that the mass density  $\rho$ , the thermal diffusivity constant  $\kappa_T$ , and the specific heat per unit volume  $c_p$  are the same for both liquid and solid, namely, we adopt the symmetric model.

To make all physical properties dimensionless, we adopt the scale defined in Section 8.3.3 with the thermal length  $\ell_T$  as the length scale. It should be noted that with the present system, the notion defined in Section 8.1 is translated to  $\mathbf{e}_3 = \mathbf{e}_y$ , and the axis  $x_3$  is directing to the liquid phase. Accordingly, the local growth velocity of the interface observed in the present moving frame can be described as

$$u_I = -\frac{\frac{\partial s}{\partial t}}{|\nabla s|}, \quad (8.2.1)$$

while the the normal vector of the interface can be expressed as

$$\mathbf{n} = \frac{\nabla s}{|\Delta s|}. \quad (8.2.2)$$

The dimensionless system can be derived from (8.1.27) in Section 8.1 by setting the relative velocity field  $\mathbf{u} = -\mathbf{e}_y$  as follows:

$$\nabla^2 \bar{T} = \frac{\partial \bar{T}}{\partial t} - \frac{\partial \bar{T}}{\partial y}, \quad (8.2.3)$$

where  $\bar{T} = \bar{T}(x, y, t)$  is the temperature field in the liquid phase. For the solid phase we replace  $\bar{T}$  by  $\bar{T}_S$ . The dimensionless boundary conditions are the following:



(1) The up-stream far-field condition: as  $\bar{y} \rightarrow \infty$ ,

$$\bar{T} \rightarrow T_\infty = \frac{(T_\infty)_D - T_{M0}}{\Delta H / (c_p \rho)}. \quad (8.2.4)$$

(2) At the interface,  $\bar{y} = \bar{h}(\bar{x}, \bar{t})$ ,

(i): the thermodynamic equilibrium condition:

$$\bar{T} = \bar{T}_S, \quad (8.2.5)$$

(ii): the Gibbs–Thomson condition:

$$\bar{T}_S = \Gamma \frac{\bar{h}_{\bar{x}\bar{x}}}{(1 + \bar{h}_{\bar{x}}^2)^{\frac{3}{2}}} - \beta \Gamma U_I, \quad (8.2.6)$$

(iii): the enthalpy balance:

$$\frac{\partial}{\partial \bar{y}} (\bar{T} - \bar{T}_S) - \bar{h}_{\bar{x}} \frac{\partial}{\partial \bar{x}} (\bar{T} - \bar{T}_S) + \bar{h}_{\bar{t}} + 1 = 0. \quad (8.2.7)$$

For the sake of convenience, we shall hereafter omit the bar over the dimensionless quantities.

### 8.2.2. Basic steady state solution

The above system allows a one-dimensional steady solution with a flat interface. The interface equation is  $y = h_B = 0$ . The exact, one-dimensional steady state solution can be obtained by setting  $\frac{\partial}{\partial x} = 0$  and  $\frac{\partial}{\partial t} = 0$  in the above system. Thus, the basic state of the system becomes

$$T_B = \begin{cases} e^{-y} - 1 - \beta \Gamma, & y \geq 0 \text{ (for the liquid);} \\ -\beta \Gamma, & y \leq 0 \text{ (for the solid).} \end{cases} \quad (8.2.8)$$

From the far field condition, we derive

$$T_\infty = -1 - \beta \Gamma, \quad (8.2.9)$$

or

$$\Gamma = \frac{|T_\infty| - 1}{\beta}. \quad (8.2.10)$$

This determine the speed of the interface of the basic state,

$$U = V = \frac{\kappa}{\ell_c} \left( \frac{|T_\infty| - 1}{\beta} \right). \quad (8.2.11)$$

In what follows, for simplicity, we only discuss the case:  $\beta = 0$  and  $T_\infty = -1$ .

### 8.2.3. Unsteady perturbed solutions and Mullins-Sekerka instability

The unsteady solutions can be expressed in the following form:

$$\begin{aligned} T(x, y, t) &= T_B(y) + \tilde{T}(x, y, t), \\ T_S(x, y, t) &= T_{SB}(y) + \tilde{T}_S(x, y, t), \\ h(x, t) &= h_B + \tilde{h}(x, t), \end{aligned} \quad (8.2.12)$$

where  $\tilde{T}$ ,  $\tilde{T}_S$ , and  $\tilde{h}$  are small perturbations around the steady state. The governing equation for the perturbation is

$$\nabla^2 \tilde{T} = \frac{\partial \tilde{T}}{\partial t} - \frac{\partial \tilde{T}}{\partial y}. \quad (8.2.13)$$

The boundary conditions are the following:

- (1) The up-steam far field condition: as  $y \rightarrow \infty$ ,

$$\tilde{T} \rightarrow 0. \quad (8.2.14)$$

- (2) The interface conditions: assume that the deformation  $\tilde{h}(x, t)$  of the interface, measured using the characteristic amplitude  $\delta \ll 1$ , is very small. One can linearize the original boundary conditions on the interface  $y = \tilde{h}(x, t)$  by expanding them in a Taylor series in  $\delta$  around  $y = h_B = 0$ . As the leading order approximation, we obtain the following linear interface conditions at  $y = 0$

$$\tilde{T} = \tilde{T}_S - (\Delta G_1) \tilde{h}, \quad (8.2.15)$$

$$\tilde{T}_S = \Gamma \tilde{h}_{xx} - G_{1S} \tilde{h}, \quad (8.2.16)$$

$$\frac{\partial (\tilde{T} - \tilde{T}_S)}{\partial y} + (\Delta G_2) \tilde{h} + \frac{\partial \tilde{h}}{\partial t} = 0, \quad (8.2.17)$$

where

$$G_{1L} = \frac{\partial T_B}{\partial y}(0) = -1; \quad (8.2.18)$$

$$G_{1S} = \frac{\partial T_{BS}}{\partial y}(0) = 0, \quad (8.2.19)$$

$$\Delta G_1 = (G_{1L} - G_{1S}) = -1, \quad (8.2.20)$$

$$\Delta G_2 = \left( \frac{\partial^2 T_B}{\partial y^2} \right)_{y=0} - \left( \frac{\partial^2 T_{SB}}{\partial y^2} \right)_{y=0} = 1. \quad (8.2.21)$$

This linear system contains one parameter  $\Gamma$ . In practice,  $\Gamma$  is a very small parameter and it appears in front of the second derivatives  $\tilde{h}_{xx}$  in (8.2.16). Thus, the above system gives rise to a singular perturbation problem. One can let  $\Gamma \rightarrow 0$  and look for an asymptotic expansion solution. In the limiting process  $\Gamma \rightarrow 0$ , all perturbed quantities  $\{\tilde{T}, \tilde{T}_S, \tilde{h}\}$  should have the same order of magnitude. From the interface condition (8.2.16), it is seen that in order for  $\tilde{T}_S$  to have the same order magnitude as  $\tilde{h}$ , one must have  $\tilde{h}_{xx} = O(\frac{\tilde{h}}{\Gamma})$ . This is possible only when  $\tilde{h}$  is also a function of the variable  $\frac{x}{\varepsilon}$ , where

$$\varepsilon = \sqrt{\Gamma}. \quad (8.2.22)$$

It will be seen that the parameter  $\varepsilon$  plays a vital role for interfacial stability. We call this parameter *the interfacial stability parameter*. The above argument provides us with an important hint that the solution must have the structure of multiple length scales; so one may derive an asymptotic expansion form of the solution by using the so-called *multiple variables expansion (MVE) method* (refer to Ke-Cole). The idea of the MVE method is that one may define a set of fast variables such that

$$\begin{aligned} x_+ &= \frac{k(\varepsilon)x}{\varepsilon} \\ y_+ &= \frac{g(\varepsilon)y}{\varepsilon} \\ t_+ &= \frac{\sigma(\varepsilon)t}{\varepsilon}, \end{aligned} \quad (8.2.23)$$

and consider the exact solution as a function of  $(x_+, y_+, t_+, x, y, \varepsilon)$ , where the faster variables  $(x_+, y_+, t_+)$  and the slow variables  $(x, y)$  are formally treated

as the independent variables. Thus, as  $\varepsilon \rightarrow 0$ , the solution  $\tilde{q} \equiv \{\tilde{T}, \tilde{T}_s, \tilde{h}\}$  is expanded in the following MVE form:

$$\begin{aligned}
\tilde{q}(x, y, x_+, y_+, t_+, \varepsilon) &\sim e^{t_+} \{ \tilde{q}_0(x, y, x_+, y_+) + \varepsilon \tilde{q}_1(x, y, x_+, y_+) + \dots \} \\
k(\varepsilon) &\sim k_0 + \varepsilon k_1 + \dots \\
g(\varepsilon) &\sim k_0 + \varepsilon g_1 + \dots \\
g_s(\varepsilon) &\sim k_0 + \varepsilon g_{s1} + \dots \\
\sigma(\varepsilon) &\sim \sigma_0 + \varepsilon \sigma_1 + \dots
\end{aligned} \tag{8.2.24}$$

Note that in the above, we have used  $g_s$  for the solutions in the solid state, which may be different from  $g$  for the solutions in the liquid state. It implies that for the solutions in the solid state, we shall use the fast variable  $y_+ = g_s y / \varepsilon$ . Moreover, to obtain the asymptotic solution, one needs to make expansions in  $\varepsilon$  for the parameter  $\sigma$  and the wave numbers,  $k$  and  $g$ . One can verify that the leading terms of the expansions for  $k(\varepsilon)$ ,  $g(\varepsilon)$ , and  $g_s(\varepsilon)$  can be set the same, but their higher-order terms may be different. Now we need to transform the perturbed system (8.2.13)–(8.2.17) into the form with the above multiple variables. This can be done by replacing all the derivatives in (8.2.13)–(8.2.17) by using the following transformation:

$$\begin{aligned}
\frac{\partial}{\partial x} &\Rightarrow \frac{\partial}{\partial x} + \frac{k}{\varepsilon} \frac{\partial}{\partial x_+}, \\
\frac{\partial}{\partial y} &\Rightarrow \frac{\partial}{\partial y} + \frac{g}{\varepsilon} \frac{\partial}{\partial y_+}, \\
\frac{\partial}{\partial t} &\Rightarrow \frac{\sigma}{\varepsilon} \frac{\partial}{\partial t_+}, \\
\frac{\partial^2}{\partial x^2} &\Rightarrow \frac{\partial^2}{\partial x^2} + \frac{2k}{\varepsilon} \frac{\partial^2}{\partial x \partial x_+} + \frac{k^2}{\varepsilon^2} \frac{\partial^2}{\partial x_+^2}, \\
\frac{\partial^2}{\partial y^2} &\Rightarrow \frac{\partial^2}{\partial y^2} + \frac{2g}{\varepsilon} \frac{\partial^2}{\partial y \partial y_+} + \frac{g^2}{\varepsilon^2} \frac{\partial^2}{\partial y_+^2}.
\end{aligned} \tag{8.2.25}$$

The multiple-variables form of the system is then obtained as follows:

$$\begin{aligned}
k^2 \frac{\partial^2 \tilde{T}}{\partial x_+^2} + g^2 \frac{\partial^2 \tilde{T}}{\partial y_+^2} &= \varepsilon \left( \sigma \tilde{T} - g \frac{\partial \tilde{T}}{\partial y_+} - 2k \frac{\partial^2 \tilde{T}}{\partial x \partial x_+} - 2g \frac{\partial^2 \tilde{T}}{\partial y \partial y_+} \right) \\
&\quad - \varepsilon^2 \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial}{\partial y} \right) \tilde{T};
\end{aligned} \tag{8.2.26}$$

$$\begin{aligned}
k^2 \frac{\partial^2 \tilde{T}_s}{\partial x_+^2} + g_s^2 \frac{\partial^2 \tilde{T}_s}{\partial y_+^2} &= \varepsilon \left( \sigma \tilde{T}_s - g_s \frac{\partial \tilde{T}_s}{\partial y_+} - 2k \frac{\partial^2 \tilde{T}_s}{\partial x \partial x_+} - 2g_s \frac{\partial^2 \tilde{T}_s}{\partial y \partial y_+} \right) \\
&\quad - \varepsilon^2 \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial}{\partial y} \right) \tilde{T}_s, \tag{8.2.27}
\end{aligned}$$

with the boundary conditions:

(1) As  $y_+ \rightarrow \infty$ ,

$$\tilde{T} \rightarrow 0. \tag{8.2.28}$$

(2) As  $y_+ \rightarrow -\infty$ ,

$$\tilde{T}_s \rightarrow 0. \tag{8.2.29}$$

(3) At the interface  $y = y_+ = 0$ ,

(i)

$$\tilde{T} = \tilde{T}_s - (\Delta G_1) \tilde{h}, \tag{8.2.30}$$

(ii)

$$\tilde{T}_s = k^2 \frac{\partial^2 \tilde{h}}{\partial x_+^2} + 2\varepsilon k \frac{\partial^2 \tilde{h}}{\partial x \partial x_+} + \varepsilon^2 \frac{\partial^2 \tilde{h}}{\partial x^2} - G_{1s} \tilde{h}, \tag{8.2.31}$$

(iii)

$$g \frac{\partial \tilde{T}}{\partial y_+} - g_s \frac{\partial \tilde{T}_s}{\partial y_+} + \varepsilon \frac{\partial}{\partial y} (\tilde{T} - \tilde{T}_s) + \sigma \tilde{h} + \varepsilon (\Delta G_2) \tilde{h} = 0. \tag{8.2.32}$$

Hereby, it is seen that as  $\varepsilon \rightarrow 0$ , in the leading order approximation, both the convective and unsteady heat transfer terms in the bulk are negligible, whereas the Gibbs–Thomson effect at the interface is important. By substituting (8.2.24) into the above system (8.4.26)–(8.2.32), one can successively derive each order of approximation.

### Zeroth-order approximation solutions

Assuming  $k_0 \gg \varepsilon$ , in the leading order approximation, we obtain the governing equation

$$k_0^2 \left( \frac{\partial^2}{\partial x_+^2} + \frac{\partial^2}{\partial y_+^2} \right) \tilde{T}_0 = 0, \tag{8.2.33}$$

and the boundary conditions:

(1) As  $y_+ \rightarrow \infty$ ,

$$\tilde{T}_0 \rightarrow 0. \quad (8.2.34)$$

(2) As  $y_+ \rightarrow -\infty$ ,

$$\tilde{T}_{S0} \rightarrow 0. \quad (8.2.35)$$

(3) At the interface,  $y = y_+ = 0$ ,

$$\tilde{T}_0 = \tilde{T}_{S0} - (\Delta G_1) \tilde{h}_0, \quad (8.2.36)$$

$$\tilde{T}_{S0} = k_0^2 \frac{\partial^2 \tilde{h}_0}{\partial x_+^2} - G_{1S} \tilde{h}_0, \quad (8.2.37)$$

$$k_0 \frac{\partial}{\partial y_+} (\tilde{T}_0 - \tilde{T}_{S0}) + \sigma_0 \tilde{h}_0 = 0. \quad (8.2.38)$$

The system (8.2.33)–(8.2.38) allows the mode solutions:

$$\begin{aligned} \tilde{T}_0 &= A_0(x, y) e^{ix_+ - y_+} \\ \tilde{T}_{S0} &= A_{S0}(x, y) e^{ix_+ + y_+} \\ \tilde{h}_0 &= \hat{D}_0 e^{ix_+}. \end{aligned} \quad (8.2.39)$$

With the notation

$$\hat{A}_0 = A_0(x, 0), \quad \hat{A}_{S0} = A_{S0}(x, 0) \quad (8.2.40)$$

and applying the boundary conditions (8.2.36)–(8.2.38) we derive

$$\begin{aligned} \hat{A}_0 &= \hat{A}_{S0} - \Delta G_1 \hat{D}_0, \\ \hat{A}_{S0} &= -k_0^2 \hat{D}_0 - G_{1S} \hat{D}_0, \\ -k_0 (\hat{A}_0 + \hat{A}_{S0}) + \sigma_0 \hat{D}_0 &= 0. \end{aligned} \quad (8.2.41)$$

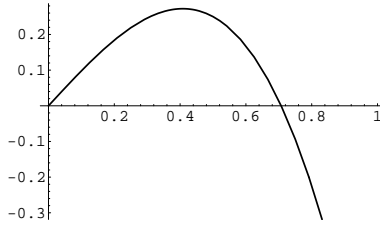


Figure 8.2.2 Mullins-Sekerka dispersion relation for a pure melt system

From this system of homogeneous equations, one deduces that the amplitude functions  $A_0, A_{S0}$  must be independent of  $x$  and  $\hat{A}_0, \hat{A}_{S0}, \hat{D}_0$  must be constants. Moreover, for a nontrivial solution, one must have:

$$\Delta = \det \begin{pmatrix} 1 & -1 & \Delta G_1 \\ 0 & 1 & k_0^2 + G_{S1} \\ -k_0 & -k_0 & \sigma_0 \end{pmatrix} = 0. \quad (8.2.42)$$

Thus the wave number  $k_0$  and the eigenvalue  $\sigma_0$  in the mode solutions cannot be arbitrarily chosen. These two quantities must be subject to the following dispersion relation:

$$\sigma_0 = k_0 \left( -(G_{1L} + G_{1S}) - 2k_0^2 \right). \quad (8.2.43)$$

For the system under consideration, we have

$$\sigma_0 = k_0 (1 - 2k_0^2). \quad (8.2.44)$$

This formula was first obtained by Mullins–Sekerka in 1963. It is now called the Mullins–Sekerka dispersion relation. From this dispersion relation, with a given eigenvalue  $\sigma_0$ , the corresponding wave numbers  $k_0$  are determined (see Figure 8.2.2). Note that if the wave number  $k_0$  has an imaginary part, the perturbation will tend to infinity either as  $x \rightarrow \infty$ , or as  $x \rightarrow -\infty$ .

Furthermore, if the wave number  $k_0$  is a negative real number, the perturbed temperature,  $\tilde{T}_0$ , will tend to infinity as  $y \rightarrow \infty$ . Evidently, these results are physically unacceptable. Thus, the eigenvalue  $\sigma_0$  of the system must be real. There are three cases:

- (i)  $0 < \sigma_0 < \frac{1}{3}\sqrt{\frac{2}{3}}$ . From the local dispersion relation (8.2.44), one solves for the three real roots:  $k_0^{(1)} > 0, k_0^{(2)} < 0, k_0^{(3)} > 0$ . Only the positive roots  $k_0^{(1)}, k_0^{(3)}$  are physically meaningful. The temperature field corresponding to  $k_0^{(2)}$  will increase exponentially as  $y \rightarrow \infty$ , violating the boundary condition (8.2.34).

The general solution of the perturbed states is then

$$\tilde{h} \approx \text{Re} \left\{ \left( D_0^{(1)} e^{\frac{i}{\varepsilon} k_0^{(1)} x} + D_0^{(3)} e^{\frac{i}{\varepsilon} k_0^{(3)} x} \right) e^{\frac{\sigma_0}{\varepsilon} t} \right\}. \quad (8.2.45)$$

These solutions are growing unstable modes. A special case is  $\sigma_0 = 0$ , for which we have  $k_0^{(3)} = 0$  and  $k_0^{(1)} > 0$ . The corresponding solutions are neutrally stable modes.

- (ii) If  $\sigma_0 < 0$ , the local dispersion relation (8.2.44) only allows one real root  $k_0^{(1)}$  and the system has only one corresponding decaying mode.
- (iii) If  $\sigma_0 > \frac{1}{3}\sqrt{\frac{2}{3}}$ , the local dispersion relation (8.2.44) has no real root, so the system has no corresponding mode.

Returning to the original slow variables  $(x, y, t)$ , the normal mode solution is written in the form:

$$\begin{aligned} \tilde{T}_0 e^{\sigma_0 t} &= A_0 e^{\tilde{k}_0(i x - y) + \tilde{\sigma}_0 t} \\ \tilde{T}_{S0} e^{\sigma_0 t} &= A_{S0} e^{\tilde{k}_0(i x + y) + \tilde{\sigma}_0 t} \\ \tilde{h}_0 e^{\sigma_0 t} &= \hat{D}_0 e^{i \tilde{k}_0 x + \tilde{\sigma}_0 t}, \end{aligned} \quad (8.2.46)$$

where

$$\tilde{k}_0 = \frac{k_0}{\varepsilon}; \quad \tilde{\sigma}_0 = \frac{\sigma_0}{\varepsilon}. \quad (8.2.47)$$

Thus, the dispersion relation (8.2.43) is written in the form:

$$\tilde{\sigma}_0 = \tilde{k}_0 \left( - (G_{1L} + G_{1S}) - 2\varepsilon^2 \tilde{k}_0^2 \right). \quad (8.2.48)$$



It is clear from this formula that, when the mean temperature gradient  $\bar{G}_1 = \frac{G_{1L} + G_{1S}}{2} < 0$ , the heat conduction represented by the term  $-2\bar{G}_1\tilde{k}_0$  is an unstable factor; while the surface tension, represented by the term  $\varepsilon^2\tilde{k}_0^3$ , is a stable factor. If the surface tension parameter is zero, the dispersion relation reduces to

$$\tilde{\sigma}_0 = -2\bar{G}_1\tilde{k}_0. \quad (8.2.49)$$

Hence, for any  $\tilde{k}_0 > 0$ , one always has  $\tilde{\sigma}_0 > 0$ . This implies that the system will always be unstable. When the surface tension parameter is nonzero, the surface tension suppresses perturbations with a short wavelength. But perturbations with long wavelengths all grow with time. More precisely, for  $\tilde{k}_0 > \sqrt{-\bar{G}_1/\varepsilon}$ ,  $\tilde{\sigma}_0 < 0$ , and mode solutions are decaying and stable. For  $0 < \tilde{k}_0 < \sqrt{-\bar{G}_1/\varepsilon}$ , the mode solutions are growing and unstable. The critical number  $\tilde{k}_c = \sqrt{-\bar{G}_1/\varepsilon}$  corresponds to the so-called neutral modes.

From the above results, one can conclude that in unidirectional solidification from a pure melt, due to the Mullins–Sekerka instability, any perturbed states are either purely growing or purely decaying. The system allows a unique neutrally stable state, which represents a steady state. It does not allow an oscillatory state. When  $(G_{1L} + G_{1S}) > 0$ , the system will be stable for all  $k_0 > 0$ . This implies that when the solidification interface advances to a high temperature liquid region, the interface will always be smooth. It should be noted that so far these conclusions are drawn from the leading order approximation of the solution in the limit  $\varepsilon \rightarrow 0$ . The dispersion relation (8.2.43) is not exact. For more accurate information, one needs to examine the higher-order approximation solutions.

### First-order approximation solutions

In the first-order approximation, we obtain the solution

$$\begin{aligned} k_0^2 \left\{ \frac{\partial^2}{\partial x_+^2} + \frac{\partial^2}{\partial y_+^2} \right\} \tilde{T}_1 &= \sigma_0 \tilde{T}_0 - k_0 \frac{\partial \tilde{T}_0}{\partial y_+} - 2k_0 \frac{\partial^2 \tilde{T}_0}{\partial x \partial x_+} - 2k_0 \frac{\partial^2 \tilde{T}_0}{\partial y \partial y_+} \\ &\quad - 2k_0 k_1 \frac{\partial^2 \tilde{T}_0}{\partial x_+^2} - 2k_0 g_1 \frac{\partial^2 \tilde{T}_0}{\partial y_+^2}, \end{aligned} \quad (8.2.50)$$

$$\begin{aligned}
k_0^2 \left\{ \frac{\partial^2}{\partial x_+^2} + \frac{\partial^2}{\partial y_+^2} \right\} \tilde{T}_{S1} &= \sigma_0 \tilde{T}_0 - k_0 \frac{\partial \tilde{T}_{S0}}{\partial y_+} - 2k_0 \frac{\partial^2 \tilde{T}_{S0}}{\partial x \partial x_+} - 2k_0 \frac{\partial^2 \tilde{T}_{S0}}{\partial y \partial y_+} \\
&\quad - 2k_0 k_1 \frac{\partial^2 \tilde{T}_{S0}}{\partial x_+^2} - 2k_0 g_{s1} \frac{\partial^2 \tilde{T}_{S0}}{\partial y_+^2}. \tag{8.2.51}
\end{aligned}$$

Consequently,

$$\begin{cases} k_0^2 \left\{ \frac{\partial^2}{\partial x_+^2} + \frac{\partial^2}{\partial y_+^2} \right\} \tilde{T}_1 &= a_0 e^{i(x_+ - y_+)} \\ k_0^2 \left\{ \frac{\partial^2}{\partial x_+^2} + \frac{\partial^2}{\partial y_+^2} \right\} \tilde{T}_{S1} &= b_0 e^{i(x_+ + y_+)}, \end{cases} \tag{8.2.52}$$

where

$$\begin{cases} a_0 &= 2k_0 \left\{ A'_0(y) + \left( \frac{1}{2} + \frac{\sigma_0}{2k_0} + k_1 - g_1 \right) A_0(y) \right\} \\ b_0 &= -2k_0 \left\{ A'_{S0}(y) + \left( \frac{1}{2} - \frac{\sigma_0}{2k_0} - k_1 + g_{s1} \right) A_{S0}(y) \right\}. \end{cases} \tag{8.2.53}$$

In order to obtain a uniformly valid asymptotic solution, one must eliminate the secular terms by setting  $a_0 = b_0 = 0$ . Moreover, without loss of generality, we assume  $A'_0(y) = A'_{S0}(y) = 0$ , so that

$$A_0(y) = \hat{A}_0, \quad A_{S0}(y) = \hat{A}_{S0}. \tag{8.2.54}$$

This leads to

$$\begin{cases} g_1 - k_1 = \frac{1}{2} \left( 1 + \frac{\sigma_0}{k_0} \right) \\ g_{s1} = g_1 - 1. \end{cases} \tag{8.2.55}$$

From  $\nabla^2 \tilde{T}_1 = \nabla^2 \tilde{T}_{S1} = 0$ , we obtain the solutions:

$$\begin{aligned}
\tilde{T}_1 &= A_1(y) e^{i(x_+ - y_+)} \\
\tilde{T}_{S1} &= A_{S1}(y) e^{i(x_+ + y_+)} \\
\tilde{h}_1 &= \hat{D}_1 e^{ix_+}. \end{aligned} \tag{8.2.56}$$

Hereby, one can easily justify that the amplitude functions  $A_1$  and  $A_{S1}$  are only dependent on  $y$ , as the coefficients of the system are constant. Let

$$\hat{A}_1 = A_1(0); \quad \hat{A}_{S1} = A_{S1}(0). \tag{8.2.57}$$

For the first-order approximation, we derive the following boundary conditions at the interface,  $y = y_+ = 0$ :

$$\tilde{T}_1 = \tilde{T}_{S1} - (\Delta G_1)\tilde{h}_1, \quad (8.2.58)$$

$$\tilde{T}_{S1} = k_0^2 \frac{\partial^2 \tilde{h}_1}{\partial x_+^2} + 2k_0 \frac{\partial^2 \tilde{h}_0}{\partial x \partial x_+} + 2k_0 k_1 \frac{\partial^2 \tilde{h}_0}{\partial x_+^2} - G_{1S} \tilde{h}_1, \quad (8.2.59)$$

$$\begin{aligned} k_0 \frac{\partial}{\partial y_+} (\tilde{T}_1 - \tilde{T}_{S1}) + g_1 \frac{\partial \tilde{T}_0}{\partial y_+} - g_{s1} \frac{\partial \tilde{T}_{S0}}{\partial y_+} + \frac{\partial}{\partial y} (\tilde{T}_0 - \tilde{T}_{S0}) \\ + \sigma_0 \tilde{h}_1 + \sigma_1 \tilde{h}_0 + (\Delta G_2)\tilde{h}_0 = 0. \end{aligned} \quad (8.2.60)$$

In order for the mode solution (8.2.57) to satisfy the boundary conditions (8.2.58)–(8.2.60), we must have

$$\hat{A}_1 = \hat{A}_{S1} - (\Delta G_1)\hat{D}_1, \quad (8.2.61)$$

$$\hat{A}_{S1} = -(k_0^2 + G_{S1})\hat{D}_1 + I_2\hat{D}_0, \quad (8.2.62)$$

$$\begin{aligned} -k_0(\hat{A}_1 + \hat{A}_{S1}) - g_1(\hat{A}_0 + \hat{A}_{S0}) + \hat{A}_{S0} + \\ + (\Delta G_2)\hat{D}_0 + \sigma_0\hat{D}_1 + \sigma_1\hat{D}_0 = 0, \end{aligned} \quad (8.2.63)$$

or

$$-k_0(\hat{A}_1 + \hat{A}_{S1}) + \sigma_0\hat{D}_1 = I_3\hat{D}_0. \quad (8.2.64)$$

In the above, we have used the notation

$$\begin{cases} I_2 = -2k_0k_1 \\ I_3 = k_0^2 + G_{1S} + \frac{\sigma_0g_1}{k_0} - \sigma_1 - \Delta G_2. \end{cases} \quad (8.2.65)$$

The determinant of the above inhomogeneous system is  $\Delta = 0$ . Hence, for a nontrivial solution  $\{\hat{A}_1, \hat{A}_{S1}, \hat{D}_1\}$ , the following solvability condition must hold:

$$\det \begin{pmatrix} 1 & -1 & 0 \\ 0 & 1 & I_2 \\ -k_0 & -k_0 & I_3 \end{pmatrix} = 0, \quad (8.2.66)$$

or

$$I_3 + 2k_0I_2 = 0. \quad (8.2.67)$$

From (8.2.67), one obtains

$$\sigma_1 = g_1(1 - 2k_0^2) + k_0^2 - 1 - 4k_0^2k_1. \quad (8.2.68)$$

We set  $k_1 = 0$ . Thus, with  $\Delta G_1 = -1$  and  $\Delta G_2 = 1$ , we get

$$\sigma_1 = g_1(1 - 2k_0^2) + k_0^2 - 1. \quad (8.2.69)$$

It follows from (8.2.55) that

$$g_1 = \frac{1}{2} \left( 1 + \frac{\sigma_0}{k_0} \right). \quad (8.2.70)$$

From the above, we obtain the modified dispersion relation:

$$\begin{aligned} \sigma &= \sigma_0 + \varepsilon\sigma_1 + O(\varepsilon^2) \\ &= k_0(1 - 2k_0^2) - \varepsilon + \varepsilon k_0^2 + \varepsilon g_1(1 - 2k_0^2) + O(\varepsilon^2) \\ &= -\varepsilon + \varepsilon k^2 + \left( k + \frac{\varepsilon}{2} + \frac{\varepsilon\sigma_0}{2k} \right) (1 - 2k^2) + O(\varepsilon^2). \end{aligned} \quad (8.2.71)$$

Note that we have replaced  $k_0$  by  $k$  in the final expression. The above procedure can be continued to even higher-order approximations if necessary. However, as indicated before, the above MVE solution (8.2.24) is not applicable in the long-wavelength range  $k_0 = O(\varepsilon)$ . In fact, as  $k_0 \ll 1$ , the effect of higher-order terms on the right-hand side of (8.2.26), as well as in the boundary condition (8.2.32), becomes important. They must be taken into account in the leading order approximation. Moreover, the MVE solution (8.2.24) is not valid in the extremely short-wavelength range  $k_0 = O(\frac{1}{\varepsilon})$  either. For any given small  $\varepsilon > 0$ , as  $k_0 \gg 1$ , we have  $|\sigma_0| \ll |\varepsilon\sigma_1|$  and  $|g_0| \ll |\varepsilon g_1|$ . Therefore, in order to study the behaviors of solution in the ranges  $k \ll 1$  and  $k \gg 1$ , one needs to make different asymptotic expansions. In what follows, we shall look into these cases separately.

#### 8.2.4. Asymptotic solutions in the long-wavelength regime, $k = O(\varepsilon)$

In the long-wavelength regime,  $\{k = O(\varepsilon); g = O(\varepsilon); \sigma = O(\varepsilon)\}$ , the solution will not have the structure of multiple scales, as both the fast variables  $x_+, y_+, t_+$  and the slow variables  $x, y, t$  are of the same order. We denote

$$\hat{k} = \frac{k}{\varepsilon}; \quad \hat{g} = \frac{g}{\varepsilon}; \quad \hat{\sigma} = \frac{\sigma}{\varepsilon} \quad (8.2.72)$$

and introduce the new slow variables

$$\begin{cases} \hat{x} &= \hat{k}x \\ \hat{y} &= \hat{g}y \\ \hat{t} &= \hat{\sigma}t. \end{cases} \quad (8.2.73)$$

We expand the solution  $\tilde{q} \equiv \{\tilde{T}, \tilde{T}_s, \tilde{h}\}$  in the following asymptotic form:

$$\begin{aligned} \tilde{q}(\hat{x}, \hat{y}, \hat{t}, \varepsilon) &\sim e^{\hat{t}} \left\{ \tilde{q}_0(\hat{x}, \hat{y}) + \varepsilon^2 \tilde{q}_1(\hat{x}, \hat{y}) + \dots \right\} \\ \hat{k}(\varepsilon) &\sim \hat{k}_0 + \varepsilon^2 \hat{k}_1 + \dots \\ \hat{g}(\varepsilon) &\sim \hat{g}_0 + \varepsilon^2 \hat{g}_1 + \dots \\ \hat{g}_s(\varepsilon) &\sim \hat{g}_{s0} + \varepsilon^2 \hat{g}_{s1} + \dots \\ \hat{\sigma}(\varepsilon) &\sim \hat{\sigma}_0 + \varepsilon^2 \hat{\sigma}_1 + \dots \end{aligned} \quad (8.2.74)$$

Note that for solutions of the temperature distribution in the solid phase, a different slow variable

$$\hat{y} = \hat{g}_s y \quad (8.2.75)$$

is used. In terms of the above variables, the linear system (8.2.13)-(8.2.17) is transformed to

$$\left\{ \hat{k}^2 \frac{\partial^2}{\partial \hat{x}^2} + \hat{g}^2 \frac{\partial^2}{\partial \hat{y}^2} \right\} \tilde{T} = \hat{\sigma} \tilde{T} - \hat{g} \frac{\partial \tilde{T}}{\partial \hat{y}}, \quad (8.2.76)$$

with the boundary conditions:

- (1) As  $\hat{y} \rightarrow \infty$ ,  $\tilde{T} \rightarrow 0$ .
- (2) As  $\hat{y} \rightarrow -\infty$ ,  $\tilde{T}_s \rightarrow 0$ .
- (3) At the interface,  $\hat{y} = 0$ ,

(i)

$$\tilde{T} = \tilde{T}_s - (\Delta G_1) \tilde{h},$$

(ii)

$$\tilde{T}_s = \varepsilon^2 \hat{k}^2 \frac{\partial^2 \tilde{h}}{\partial \hat{x}^2} - G_{1s} \tilde{h},$$

(iii)

$$\hat{g} \frac{\partial \tilde{T}}{\partial \hat{y}} - \hat{g}_s \frac{\partial \tilde{T}_s}{\partial \hat{y}} + \hat{\sigma} \tilde{h} + \Delta G_2 \tilde{h} = 0. \quad (8.2.77)$$

From the above, it is seen that as  $\varepsilon \rightarrow 0$ , in the leading order approximation for the solution in the long-wavelength regime, both heat convective and unsteady transfer terms in the bulk are important, but the Gibbs–Thomson effect on the interface is negligible. By substituting (8.2.74) into the above system (8.2.76)–(8.2.77), one can successively derive each order of approximation.

$O(\varepsilon^0)$  : In the leading order approximation, we derive the governing equation

$$\hat{k}_0^2 \frac{\partial^2 \tilde{T}_0}{\partial \hat{x}^2} + \hat{g}_0^2 \frac{\partial^2 \tilde{T}_0}{\partial \hat{y}^2} = \hat{\sigma}_0 \tilde{T}_0 - \hat{g}_0 \frac{\partial \tilde{T}_0}{\partial \hat{y}}. \quad (8.2.78)$$

The above equation yields the solutions

$$\begin{aligned} \tilde{T}_0 &= \hat{A}_0 e^{i\hat{x} - \hat{y}} \\ \tilde{T}_{s0} &= \hat{A}_{s0} e^{i\hat{x} + \hat{y}} \\ \tilde{h}_0 &= \hat{D}_0 e^{i\hat{x}} \end{aligned} \quad (8.2.79)$$

and we derive

$$\hat{g}_0^2 - \hat{g}_0 - (\hat{k}_0^2 + \hat{\sigma}_0) = 0. \quad (8.2.80)$$

For the liquid phase,

$$\hat{g}_0 = \frac{1}{2} + \sqrt{\hat{k}_0^2 + \hat{\sigma}_0 + \frac{1}{4}}, \quad (8.2.81)$$

while for the solid phase,

$$\hat{g}_{s0} = \hat{g}_0 - 1. \quad (8.2.82)$$

With  $\Delta G_1 = -1$ ,  $\Delta G_2 = 1$ , and  $G_{1s} = 0$ , we derive from the boundary conditions (8.2.77),

$$\begin{cases} \hat{A}_0 = \hat{A}_{s0} + \hat{D}_0 \\ \hat{A}_{s0} = 0 \\ -\hat{g}_0 \hat{A}_0 - \hat{g}_{s0} \hat{A}_{s0} + (\hat{\sigma}_0 + 1) \hat{D}_0 = 0. \end{cases} \quad (8.2.83)$$

Thus, it is found that

$$\hat{\sigma}_0 = -1 + \hat{g}_0 = -\frac{1}{2} + \sqrt{\hat{k}_0^2 + \hat{\sigma}_0 + \frac{1}{4}}$$

or

$$\hat{\sigma}_0 = \hat{k}_0. \quad (8.2.84)$$

$O(\varepsilon^2)$ : In the first-order approximation, we derive the governing equations

$$\begin{aligned} & \hat{k}_0^2 \frac{\partial^2 \tilde{T}_1}{\partial \hat{x}^2} + \hat{g}_0^2 \frac{\partial^2 \tilde{T}_1}{\partial \hat{y}^2} - \hat{\sigma}_0 \tilde{T}_1 + \hat{g}_0 \frac{\partial \tilde{T}_1}{\partial \hat{y}} \\ & = (2\hat{k}_0 \hat{k}_1 - 2\hat{g}_0 \hat{g}_1 + \hat{\sigma}_1 + \hat{g}_1) \tilde{T}_0 \end{aligned}$$

and

$$\begin{aligned} & \hat{k}_0^2 \frac{\partial^2 \tilde{T}_{S1}}{\partial \hat{x}^2} + \hat{g}_{s0}^2 \frac{\partial^2 \tilde{T}_{S1}}{\partial \hat{y}^2} - \hat{\sigma}_0 \tilde{T}_{S1} + \hat{g}_{s0} \frac{\partial \tilde{T}_{S1}}{\partial \hat{y}} \\ & = (2\hat{k}_0 \hat{k}_1 - 2\hat{g}_{s0} \hat{g}_{s1} + \hat{\sigma}_1 - \hat{g}_{s1}) \tilde{T}_{S0}. \end{aligned} \quad (8.2.85)$$

To eliminate the secular terms, we must set

$$\begin{cases} 2\hat{k}_0 \hat{k}_1 - 2\hat{g}_0 \hat{g}_1 + \hat{\sigma}_1 + \hat{g}_1 = 0 \\ 2\hat{k}_0 \hat{k}_1 - 2\hat{g}_{s0} \hat{g}_{s1} + \hat{\sigma}_1 - \hat{g}_{s1} = 0. \end{cases}$$

We put  $k_1 = 0$ . It follows that

$$\hat{g}_1 = \hat{g}_{s1} = \frac{\hat{\sigma}_1}{2\hat{g}_0 - 1}. \quad (8.2.86)$$

The first-order approximation yields the solutions

$$\begin{aligned} \tilde{T}_1 &= \hat{A}_1 e^{i\hat{x} - \hat{y}} \\ \tilde{T}_{S1} &= \hat{A}_{S1} e^{i\hat{x} + \hat{y}} \\ \tilde{h}_1 &= \hat{D}_1 e^{i\hat{x}}. \end{aligned}$$

From the boundary conditions (8.2.77), we derive

$$\begin{cases} \hat{A}_1 = \hat{A}_{S1} + \hat{D}_1 \\ \hat{A}_{S1} = -\hat{k}_0^2 \hat{D}_0 \\ -\hat{g}_0 \hat{A}_1 - \hat{g}_{s0} \hat{A}_{S1} + (\hat{\sigma}_0 + 1) \hat{D}_1 = \hat{A}_0 \hat{g}_1 + \hat{A}_{S0} \hat{g}_{s1} - \hat{\sigma}_1 \hat{D}_0. \end{cases}$$

From this system, we obtain

$$\hat{\sigma}_1 = \hat{g}_1 + (1 - 2\hat{g}_0)\hat{k}_0^2$$

or

$$\hat{\sigma}_1 = -\frac{\hat{k}_0}{2}(1 + 2\hat{k}_0)^2. \quad (8.2.87)$$

Combining (8.2.86) with (8.2.87), we derive

$$\hat{g}_1 = -\hat{k}_0^2 \left[ 1 + \frac{1}{2(\hat{g}_0 - 1)} \right]$$

or

$$\hat{g}_1 = -\frac{\hat{k}_0}{2}(1 + 2\hat{k}_0).$$

Thus, we have

$$\begin{aligned} \hat{\sigma} &= \hat{\sigma}_0 + \varepsilon^2 \hat{\sigma}_1 \\ &= (-1 + \hat{g}_0) + \varepsilon^2 \hat{g}_1 + \varepsilon^2 (1 - 2\hat{g}_0) \hat{k}_0^2 \\ &= -1 + \varepsilon^2 \hat{k}^2 + (\hat{g}_0 + \varepsilon^2 \hat{g}_1)(1 - 2\varepsilon^2 \hat{k}^2) + 2\varepsilon^4 \hat{g}_1 \hat{k}^2. \end{aligned}$$

Therefore, expressing the equations in terms of the original parameters  $(k, \sigma)$ , in the long-wavelength regime  $(k \rightarrow 0)$ , we have the dispersion relation:

$$\left\{ \begin{array}{l} \sigma_0 = -\frac{\varepsilon}{2} + \sqrt{k^2 + \varepsilon\sigma_0 + \frac{\varepsilon^2}{4}} \\ g_0 = \frac{\varepsilon}{2} + \sqrt{k^2 + \varepsilon\sigma_0 + \frac{\varepsilon^2}{4}} \\ \varepsilon^2 g_1 = -\varepsilon k^2 \left\{ 1 + \frac{\varepsilon}{2(g_0 - \varepsilon)} \right\} \\ \sigma = -\varepsilon + \varepsilon k^2 + (g_0 + \varepsilon^2 g_1)(1 - 2k^2) + 2\varepsilon^2 g_1 k^2. \end{array} \right. \quad (8.2.88)$$

Note that in this case one can write

$$G_1 = g_0 + \varepsilon^2 g_1 = \frac{\varepsilon}{2} + \sqrt{k^2 + \varepsilon\sigma + \frac{\varepsilon^2}{4}} + O(\varepsilon^3).$$

Comparing (8.2.71) with (8.2.88), it is seen that if one applies the approximate formula  $(k + \frac{\varepsilon\sigma_0}{2k}) \approx \sqrt{k^2 + \varepsilon\sigma_0}$  to the dispersion relation (8.2.71) then the resultant formula

$$\sigma = -\varepsilon + \varepsilon k^2 + \left( \frac{\varepsilon}{2} + \sqrt{k^2 + \varepsilon\sigma_0} \right) (1 - 2k^2) + O(\varepsilon^2) \quad (8.2.89)$$



will be applicable to the whole range  $0 \leq k \ll \frac{1}{\varepsilon}$ .

### 8.2.5. Asymptotic solutions in the extremely short-wavelength regime, $k = O(\frac{1}{\varepsilon})$

In the extremely short-wavelength regime,  $\{k = O(\frac{1}{\varepsilon}); g = O(\frac{1}{\varepsilon})\}$ , one can still find a MVE solution with the fast variable  $x_+, y_+, t_+$  and the slow variables  $x, y, t$ . We write

$$\begin{cases} \tilde{k} = \varepsilon k = O(1) \\ \tilde{g} = \varepsilon g = O(1) \\ \tilde{\sigma} = \varepsilon^\alpha \sigma = O(1), \end{cases}$$

where the exponent  $\alpha$  is to be determined. The MVE system (8.2.26)–(8.2.32) is still applicable to the present case if we change  $k, g$  and  $\sigma$  to  $\tilde{k}, \tilde{g}$  and  $\tilde{\sigma}$ . Thus, we have

$$\begin{aligned} \left( \tilde{k}^2 \frac{\partial^2}{\partial x_+^2} + \tilde{g}^2 \frac{\partial^2}{\partial y_+^2} \right) \tilde{T} &= \varepsilon^{3-\alpha} \tilde{\sigma} \tilde{T} - \varepsilon^2 \tilde{g} \frac{\partial \tilde{T}}{\partial y_+} - 2\varepsilon^2 \tilde{k} \frac{\partial^2 \tilde{T}}{\partial x \partial x_+} \\ &\quad - 2\varepsilon^2 \tilde{g} \frac{\partial^2 \tilde{T}}{\partial y \partial y_+} - \varepsilon^4 \left( \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} - \frac{\partial}{\partial y} \right) \tilde{T}, \end{aligned} \quad (8.2.90)$$

with the boundary conditions:

- (1) As  $y_+ \rightarrow \infty, \tilde{T} \rightarrow 0$ .
- (2) As  $y_+ \rightarrow -\infty, \tilde{T}_S \rightarrow 0$ .
- (3) At the interface,  $y = y_+ = 0$ ,

(i)

$$\tilde{T} = \tilde{T}_S - (\Delta G_1) \tilde{h}, \quad (8.2.91)$$

(ii)

$$\tilde{T}_S = \frac{\tilde{k}^2}{\varepsilon^2} \frac{\partial^2 \tilde{h}}{\partial x_+^2} + 2\tilde{k} \frac{\partial^2 \tilde{h}}{\partial x \partial x_+} + \varepsilon^2 \frac{\partial^2 \tilde{h}}{\partial x^2} - G_{1S} \tilde{h}, \quad (8.2.92)$$

(iii)

$$\begin{aligned} \tilde{g} \frac{\partial}{\partial y_+} (\tilde{T} - \tilde{T}_S) + \varepsilon^2 \frac{\partial}{\partial y} (\tilde{T} - \tilde{T}_S) + \varepsilon^{1-\alpha} \tilde{\sigma} \tilde{h} \\ + \varepsilon^2 (\Delta G_2) \tilde{h} = 0. \end{aligned} \quad (8.2.93)$$

One can see from (8.2.92) that  $\tilde{h} = O(\varepsilon^2 \tilde{T})$ . Furthermore, from (8.2.93) one can prove that the exponent  $\alpha$  must be chosen as  $\alpha = 3$ . Hence the system allows the following asymptotic expansions:

$$\begin{aligned} \tilde{T} &\sim e^{t_+} \left\{ \tilde{T}_0(x, y, x_+, y_+) + \varepsilon^2 \tilde{T}_1(x, y, x_+, y_+) + \dots \right\} \\ \tilde{T}_S &\sim e^{t_+} \left\{ \tilde{T}_{S0}(x, y, x_+, y_+) + \varepsilon^2 \tilde{T}_{S1}(x, y, x_+, y_+) + \dots \right\} \\ \tilde{h} &\sim \varepsilon^2 e^{t_+} \left\{ \tilde{h}_0(x, y, x_+, y_+) + \varepsilon^2 \tilde{h}_1(x, y, x_+, y_+) + \dots \right\} \\ \tilde{k}(\varepsilon) &\sim \tilde{k}_0 + \varepsilon^2 \tilde{k}_1 + \dots \\ \tilde{g}(\varepsilon) &\sim \tilde{g}_0 + \varepsilon^2 \tilde{g}_1 + \dots \\ \tilde{g}_s(\varepsilon) &\sim \tilde{g}_{s0} + \varepsilon^2 \tilde{g}_{s1} + \dots \\ \tilde{\sigma}(\varepsilon) &\sim \tilde{\sigma}_0 + \varepsilon^2 \tilde{\sigma}_1 + \dots \end{aligned} \quad (8.2.94)$$

By substituting (8.2.94) into the system (8.2.90)–(8.2.93), one can successively derive each order of approximation.

$O(\varepsilon^0)$  : In the zeroth-order approximation, we have

$$\tilde{k}_0^2 \frac{\partial^2 \tilde{T}_0}{\partial x_+^2} + \tilde{g}_0^2 \frac{\partial^2 \tilde{T}_0}{\partial y_+^2} = \tilde{\sigma}_0 \tilde{T}_0,$$

which allows the mode solutions:

$$\begin{aligned} \tilde{T}_0 &= A_0(y) e^{ix_+ - y_+} \\ \tilde{T}_{S0} &= A_{S0}(y) e^{ix_+ + y_+} \\ \tilde{h}_0 &= \hat{D}_0 e^{ix_+} \end{aligned}$$

with

$$\tilde{g}_0 = \sqrt{\tilde{\sigma}_0 + \tilde{k}_0^2}.$$

With the notation

$$\hat{A}_0 = A_0(0), \quad \hat{A}_{S0} = A_{S0}(0),$$

from the boundary conditions (8.2.91)–(8.2.93), we derive

$$\begin{cases} \hat{A}_0 = \hat{A}_{S0} \\ \hat{A}_{S0} = -\tilde{k}_0^2 \hat{D}_0 \\ -\tilde{g}_0(\hat{A}_0 + \hat{A}_{S0}) + \tilde{\sigma}_0 \hat{D}_0 = 0. \end{cases}$$

It is seen that in the leading order approximation for the solution in the extremely short-wavelength regime, the unsteady heat transfer term in the bulk is important, but the effect of interface displacement is negligible. We derive the following dispersion relation:

$$\tilde{\sigma}_0 = -2\tilde{g}_0\tilde{k}_0^2.$$

This may be rewritten as

$$\sigma_0 = -2k_0^2 \sqrt{k_0^2 + \varepsilon\sigma_0}.$$

$O(\varepsilon^2)$  : In the first-order approximation, we have

$$\begin{aligned} \tilde{k}_0^2 \frac{\partial^2 \tilde{T}_1}{\partial x_+^2} + \tilde{g}_0^2 \frac{\partial^2 \tilde{T}_1}{\partial y_+^2} - \tilde{\sigma}_0 \tilde{T}_1 &= \tilde{a}_0 e^{ix_+ - y_+} \\ \tilde{k}_0^2 \frac{\partial^2 \tilde{T}_{S1}}{\partial x_+^2} + \tilde{g}_0^2 \frac{\partial^2 \tilde{T}_{S1}}{\partial y_+^2} - \tilde{\sigma}_0 \tilde{T}_{S1} &= \tilde{b}_0 e^{ix_+ + y_+}, \end{aligned}$$

where

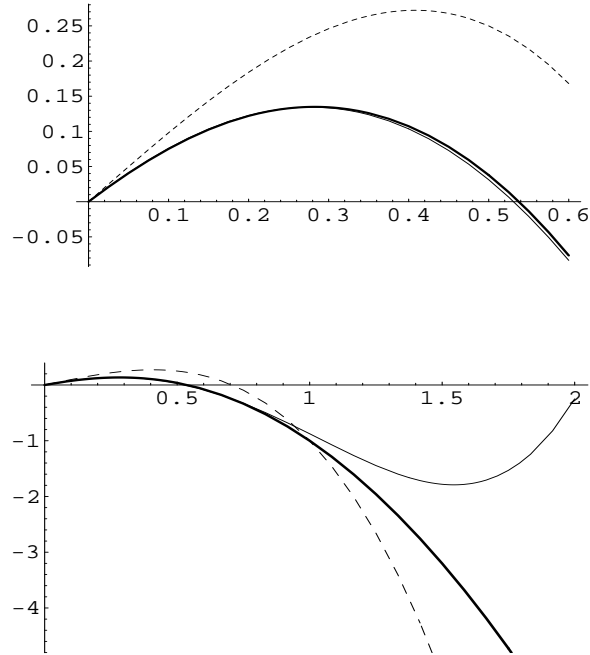
$$\begin{cases} \tilde{a}_0 = 2\tilde{g}_0 A'_0(y) + (\tilde{\sigma}_1 + \tilde{g}_0 + 2\tilde{k}_0\tilde{k}_1 - 2\tilde{g}_0\tilde{g}_1) A_0(y) \\ \tilde{b}_0 = -2\tilde{g}_0 A'_{S0}(y) + (\tilde{\sigma}_1 - \tilde{g}_0 + 2\tilde{k}_0\tilde{k}_1 - 2\tilde{g}_0\tilde{g}_{s1}) A_{S0}(y). \end{cases}$$

To eliminate the secular terms, we set  $\tilde{a}_0 = \tilde{b}_0 = 0$ . Moreover, we assume  $A'_0(y) = A'_{S0}(y) = 0$ , and consequently,

$$A_0 = \hat{A}_0, \quad A_{S0}(y) = \hat{A}_{S0}.$$

Thus, it follows that

$$\begin{cases} \tilde{g}_1 = \frac{1}{2} + \frac{\tilde{\sigma}_1}{2\tilde{g}_0} \\ \tilde{g}_{s1} = \tilde{g}_1 - 1 \end{cases}$$



**Fig. 8.2.3a,b.** The dispersion curves for  $\varepsilon = 0.5$ . The dashed line is the zeroth-order approximation, the thin solid line is the modified first-order approximation, whereas the bold solid line is the exact solution: **(a)** for the range  $0 \leq k \leq 0.6$ ; **(b)** for the range  $0 \leq k \leq 2$

and

$$\tilde{\sigma}_1 = -\tilde{g}_0 - 2\tilde{k}_0\tilde{k}_1 + 2\tilde{g}_0\tilde{g}_1. \quad (8.2.95)$$

As before, we can let  $\tilde{k}_1 = 0$ . Hence, we have

$$\tilde{\sigma}_1 = \tilde{g}_0(2\tilde{g}_1 - 1).$$

The first-order system allows the mode solutions:

$$\begin{aligned} \tilde{T}_1 &= A_1(y)e^{ix_+ - y_+} \\ \tilde{T}_{S1} &= A_{S1}(y)e^{ix_+ + y_+} \\ \tilde{h}_1 &= \hat{D}_1 e^{ix_+}. \end{aligned}$$

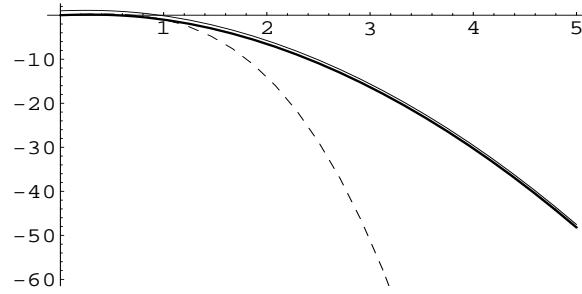


Figure 8.2.4 Dispersion curves: The dashed line is for the zeroth-order approximation, the thin solid line is the extremely short-wavelength approximation, whereas the bold solid line is the exact solution

We write

$$\hat{A}_1 = A_1(0), \quad \hat{A}_{S1} = A_{S1}(0).$$

From the boundary conditions (8.2.91)–(8.2.93), we derive

$$\begin{cases} \hat{A}_1 = \hat{A}_{S1} + \hat{D}_0 \\ \hat{A}_{S1} = -\tilde{k}_0^2 \hat{D}_1 + I_2 \hat{D}_0 \\ -\tilde{g}_0(\hat{A}_1 + \hat{A}_{S1}) + \tilde{\sigma}_0 \hat{D}_1 = I_3 \hat{D}_0, \end{cases}$$

where

$$\begin{cases} I_2 = -2\tilde{k}_0 \tilde{k}_1 \\ I_3 = -2\tilde{k}_0^2 \tilde{g}_1 + \tilde{k}_0^2 - \tilde{\sigma}_1. \end{cases}$$

For a nontrivial solution, the condition

$$I_3 + 2\tilde{g}_0 I_2 + \tilde{g}_0 = 0$$

is necessary. Thus, it follows that

$$\tilde{k}_0^2 - \tilde{\sigma}_1 - 2\tilde{k}_0^2 \tilde{g}_1 - 4\tilde{k}_0 \tilde{g}_0 \tilde{k}_1 + \tilde{g}_0 = 0. \quad (8.2.96)$$

As  $\tilde{k}_1 = 0$ , from (8.2.95)–(8.2.96) we derive

$$\tilde{g}_1 = 1 - \frac{\tilde{k}_0^2}{2(\tilde{g}_0 + \tilde{k}_0^2)}$$

and

$$\begin{aligned}\tilde{\sigma} &= \tilde{\sigma}_0 + \varepsilon^2 \tilde{\sigma}_1 = -2\tilde{g}_0 \tilde{k}_0^2 + \varepsilon^2 (\tilde{k}_0^2 + \tilde{g}_0 - 2\tilde{k}_0^2 \tilde{g}_1) \\ &= \varepsilon^2 \tilde{k}_0^2 + (\tilde{g}_0 + \varepsilon^2 \tilde{g}_1)(\varepsilon^2 - 2\tilde{k}_0^2) - \varepsilon^4 \tilde{g}_1.\end{aligned}$$

Returning to the original parameters  $\{k, \sigma\}$ , we derive the dispersion formula in the extremely short-wavelength regime as

$$\begin{cases} \sigma_0 &= -2(\sqrt{k^2 + \varepsilon\sigma_0})k^2 \\ g_0 &= \sqrt{k^2 + \varepsilon\sigma_0} \\ \varepsilon^2 g_1 &= \varepsilon - \frac{1}{2} \frac{\varepsilon^2 k^2}{g_0 + \varepsilon k^2} \\ \sigma &= \varepsilon k^2 + (g_0 + \varepsilon^2 g_1)(1 - 2k^2) - \varepsilon^2 g_1. \end{cases} \quad (8.2.97)$$

In the present case, one can show that

$$G_1 = g_0 + \varepsilon^2 g_1 = \frac{\varepsilon}{2} + \sqrt{k^2 + \varepsilon\sigma} + O(\varepsilon^3).$$

It should be noted that the system (8.2.13)–(8.2.17) is a linear system with constant coefficients, so that it can be solved exactly. The exact solutions are:

$$\begin{aligned}\tilde{T} &= \hat{A}e^{(\frac{\sigma t}{\varepsilon} + \frac{ikx}{\varepsilon} - \frac{qy}{\varepsilon})} \\ \tilde{T}_s &= \hat{A}_s e^{(\frac{\sigma t}{\varepsilon} + \frac{ikx}{\varepsilon} + \frac{qy}{\varepsilon} - y)} \\ \tilde{h} &= \hat{D}e^{(\frac{\sigma t}{\varepsilon} + \frac{ikx}{\varepsilon})},\end{aligned}$$

where

$$g = \frac{\varepsilon}{2} + \sqrt{k^2 + \varepsilon\sigma + \varepsilon^2/4}.$$

The exact dispersion relation is

$$\sigma = -\varepsilon + \varepsilon k^2 + \left(\frac{\varepsilon}{2} + \sqrt{k^2 + \varepsilon\sigma + \varepsilon^2/4}\right)(1 - 2k^2). \quad (8.2.98)$$

By comparing (8.2.71) with (8.2.98), one sees that the dispersion formula of the MVE solution obtained above, in the regime  $(\varepsilon \ll k \ll \frac{1}{\varepsilon})$ , is the regular perturbation expansion of the exact solution as  $\varepsilon \rightarrow 0$ . In Figure 8.2.3, we have shown all these dispersion curves in the  $(\sigma, k)$  plane. It is seen that up to  $\varepsilon = 0.5$ , the agreement between the exact solution and the first-order MVE solution (8.2.90) is very good in the regime  $0 \leq k < 1$ . However, as  $k > 1.5$ ,

the first-order MVE solution (8.2.71), as expected, is no longer close to the exact solution, even qualitatively. Nevertheless, the asymptotic solution in the extremely short-wavelength regime agrees with the exact solution (8.2.97) very well (see Figure 8.2.4).

## References

- Dougherty, A. and Gollub, J.P. (1988). Steady-State Dendritic Growth of NH<sub>4</sub>Br from Solution, *Phys. Rev. A*, **38**, 3043-3053.
- Horvay, G. and Cahn, J.W. (1961). Dendritic and Spheroidal Growth, *Acta Metall.*, **9**, 695-705.
- Huang, S.C. and Glicksman, M.E. (1981). Fundamentals of Dendritic Solidification -I. Steady-State Tip Growth; II. Development of Sidebranch Structure, *Acta Metall.*, **29**, 701-734.
- Hurle (Ed.), D.T.J. (1993). *Handbook of Crystal Growth, Vol. 1: Fundamentals, Part B: Transport and Stability*, (Elsevier Science, North-Holland, Amsterdam.
- Ivantsov, G.P. (1947). Temperature Field around a Spheroidal, Cylindrical and Acicular Crystal Growing in a Supercooled Melt, *Dokl. Akad. Nauk, SSSR*, **58(4)**, 567-569.
- Kobayashi, T. and Furukawa, Y. (1991). *Snow crystals*, Snow Crystal Museum Asahikawa, Hokkaido.
- Mullins, W.W. and Sekerka, R.F. (1963). Morphological Stability of a Particle Growing by Diffusion or Heat Flow, *J. Appl. Phys.*, **34**, 323-329.
- Mullins, W.W. and Sekerka, R.F. (1964). Stability of a Planar Interface During Solidification of a Dilute Binary Alloy, *J. Appl. Phys.*, **35**, 444-451.

Nash, G.E. and Glicksman, M.E. (1974). Capillarity-limited Steady-State Dendritic Growth I. Theoretical Development, *Acta Metall.*, **22**, 1283-1299.

Rutter, J.W. and Chalmers, B. (1953). A Prismatic Substructure Formed During Solidification of Metals, *Can. J. Phys.*, **31**, 15-39.

Takahashi, K., Furukawa, Y. and Takahashi, Y. (1995). *Story of snow crystals*, Koudansya, Tokyo.

Xu, J.J. (1991). Interfacial Wave Theory of Solidification - Dendritic Pattern Formation and Selection of Tip Velocity, *Phys. Rev.*, **A15 43(2)**, 930-947.

Xu, J.J. (1996). Generalized Needle Solutions, Interfacial Instabilities and Pattern Formations, *Phys. Rev.*, **E 53(5)**, 5051-5062.

Xu, J.J. (1997). *Interfacial Wave Theory of Pattern Formation: Selection of Dendritic Growth and Viscous Fingering in Hele-Shaw Flow*, Springer-Verlag Publisher in the series of Synergetic.

Xu, J.J. (2004). *Dynamical Theory of Dendritic Growth in Convective Flow*, published by Springer Publisher in the series of Advances of Mechanics and Mathematics.