Interfacial wave theory for dendritic structure of a growing needle crystal. I. Local instability mechanism

Jian-Jun Xu

Department of Mathematics and Statistics, McGill University, 805 Sherbrooke Street W., Montreal, Quebec, Canada H3A 2K6 (Received 18 February 1988; revised manuscript received 2 March 1989)

A global interfacial wave theory is developed to demonstrate the essence and origin of the complicated dendritic structure of a growing needle crystal. In this paper, by using the multiple-variableexpansion method, the local dispersion relation for normal modes is derived in a paraboloidal coordinate system. The local instability mechanism of the interface is explored. Further, in a companion paper [J. J. Xu, following article, Phys. Rev. A 40, 1609 (1989)], a global instability mechanism is established. The existence and significance of a simple turning point in the solidification system is first identified. We obtain uniformly valid solutions in the vicinity of the turning point and investigate the interaction of the interfacial waves in that region. A remarkable mechanism of wave emission and reflection at the turning point is discovered.

I. INTRODUCTION

Dendritic crystal growth from an undercooled melt is a long-standing fundamental subject. This nonequilibrium dynamic process is characterized by propagation of a steady tip and persistent emission of sidebranches. Similar spontaneous pattern-formation phenomena also happen widely in other kinds of nonequilibrium systems in nature. Exploring the essence and origin of such phenomena has great theoretical and practical significance. It is not surprising that many investigators have been attracted to this subject for decades.¹⁻²³

Recently, Barber, Barbieri, and Langer studied the sidebranching problem of 2D needle crystal growth; meanwhile Langer studied the sidebranching problem in the 3D case.²¹⁻²² It is seen that the asymptotic solutions obtained by the above authors are the local solutions, which are not uniformly valid in the tip region and in the vicinity of the turning point. A local, single-branch solution yields no information about the interaction of waves at the turning point and the leading edge of the tip. Hence it cannot be used for the purpose of demonstrating the formation of dendritic structure of a needle crystal. For such a purpose, one must find the global solution to the linear perturbed system and explore the global instability mechanism.

In the present work, we intend to illustrate the stability mechanism of dendritic growth. By using the multiplevariable-expansion method in a paraboloidal coordinate system, we derive the dispersion relationship of local perturbations. These perturbations are all unstable traveling waves, which we call "interfacial waves" or "sidebranching waves." Further, by using asymptotic analysis and matching technique we explore the global instability mechanism and obtain the global mode solutions. Our major results are the following. (1) In the dynamical system of dendritic growth, there exists a simple turning point which plays a crucial role for the formation of the dendritic structure. This turning point reflects the incident wave from the tip region and emits sidebranching waves toward the root of the dendrite. The presence of this wave-emission mechanism is the most important character of the dendritic growth process. (2) At the leading edge of the dendrite tip, there is a signal feedback mechanism. An incident wave from the turning point is transformed into an outgoing wave back to the turning point region. Thus the whole dynamical process of dendritic growth is considered as the wave emission at the turning point and the wave reflections between the turning point and the leading edge of the tip. The global modes and the quantum conditions of eigenvalues for this system are obtained, which will be reported in another paper.

This body of work is divided into two papers. In this paper (paper I) we shall show the local instability theory; in the following paper (paper II)²⁴ we shall demonstrate the wave-emission mechanism at the turning point.

II. MATHEMATICAL FORMULATION OF UNSTEADY DENDRITIC GROWTH

We consider that a dendrite is growing into an undercooled melt in the negative z-axis direction with a constant average speed U (see Fig. 1). As usual, we assume the mass density ρ , the thermal diffusivity κ , and the other thermodynamical constants of the solid state are the



FIG. 1. Cylindrical coordinate system fixed in the needle.

$$\frac{\partial^2 T}{\partial \xi^2} + \frac{\partial^2 T}{\partial \eta^2} + \frac{1}{\xi} \frac{\partial T}{\partial \xi} + \frac{1}{\eta} \frac{\partial T}{\partial \eta}$$
$$= (\xi^2 + \eta^2) \frac{\partial T}{\partial t} + \left[\xi \frac{\partial T}{\partial \xi} - \eta \frac{\partial T}{\partial \eta} \right], \quad (2.1)$$

where we use the thermal diffusion length $l_T = \kappa/U$ as the length scale, thermal diffusion time $t_T = l_T/U = \kappa/U^2$ as the time scale, and $(\Delta H/C_p)$ as the temperature scale $(\Delta H$ is the latent heat per unit volume of solid, C_p is the specific heat).

The relationship between the cylindrical coordinate system and the paraboloidal coordinate system used in Eq. (2.1) is (see Fig. 2)

$$r = \xi \eta, \quad z = \frac{1}{2}(\xi^2 - \eta^2)$$
 (2.2)

or

$$\eta^{2} = -z + (z^{2} + r^{2})^{1/2} ,$$

$$\xi^{2} = z + (z^{2} + r^{2})^{1/2} .$$
(2.3)

The boundary conditions are

as
$$\eta \to \infty$$
, $T \to T_{\infty}$, (2.4)

as
$$\eta \rightarrow 0$$
, T_s regular. (2.5)

At the interface $\eta = \eta_s(\xi, t)$,

$$T = T_s$$
 (thermodynamical equilibrium), (2.6)

$$T_s = -\Gamma K \{\eta_s(\xi, t)\}$$
 (Gibbs-Thomson condition),

where the mean curvature operator

$$\overrightarrow{\mathbf{e}_{1}} = \frac{1}{\sqrt{\tilde{\xi}^{2} + \tilde{\eta}^{2}}} \left(\widetilde{\eta} \cos \theta \, \overrightarrow{\mathbf{i}} + \widetilde{\eta} \sin \theta \, \overrightarrow{\mathbf{j}} + \widetilde{\xi} \, \overrightarrow{\mathbf{k}} \right)$$

$$\overrightarrow{\mathbf{e}_{2}} = \frac{1}{\sqrt{\tilde{\xi}^{2} + \tilde{\eta}^{2}}} \left(\widetilde{\xi} \cos \theta \, \overrightarrow{\mathbf{i}} + \widetilde{\xi} \sin \theta \, \overrightarrow{\mathbf{j}} + \widetilde{\eta} \, \overrightarrow{\mathbf{k}} \right)$$

$$\overrightarrow{\mathbf{e}_{3}} = \left(-\sin \theta \, \overrightarrow{\mathbf{i}} + \cos \theta \, \overrightarrow{\mathbf{j}} \right)$$



FIG. 2. Paraboloidal coordinate system.

$$K\{\eta_{s}\} = -\frac{1}{(\xi^{2} + \eta_{s}^{2})^{1/2}} \times \left[\frac{\eta_{s}^{\prime\prime}}{(1 + \eta_{s}^{\prime 2})^{3/2}} - \frac{1}{\eta_{s}(1 + \eta_{s}^{\prime 2})^{1/2}} + \frac{\eta_{s}^{\prime}(\eta_{s}^{2} + 2\xi^{2}) - \xi\eta_{s}}{\xi(\xi^{2} + \eta_{s}^{2})(1 + \eta_{s}^{\prime 2})^{1/2}}\right].$$
(2.8)

The prime represents the partial derivative with respect to ξ ,

$$\begin{bmatrix} \frac{\partial T}{\partial \eta} - \frac{\partial T_s}{\partial \eta} \end{bmatrix} - \eta'_s \begin{bmatrix} \frac{\partial T}{\partial \xi} - \frac{\partial T_s}{\partial \xi} \end{bmatrix}$$

= $-(\xi \eta_s)' - (\xi^2 + \eta_s^2)^{1/2} \frac{\partial \eta_s}{\partial t}$
(heat-balance condition). (2.9)

In the above, the subscript s refers to the solid state, $\eta_s(\xi, t)$ is the shape function of the interface. The surface tension parameter Γ is defined as

$$\Gamma = \frac{l_c}{l_T} \tag{2.10}$$

and

$$l_c = \frac{\gamma C_p T_{M0}}{(\Delta H)^2} \tag{2.11}$$

is the capillary length, γ is the surface tension, and T_{M0} is the melting temperature of a plane interface. Finally,

$$T_{\infty} = \frac{(T_{\infty})_D - T_{M0}}{\Delta H / C_p}$$

is the dimensionless undercooling, where $(T_{\infty})_D$ is the melt temperature at infinity.

III. BASIC STATES AND LINEARIZED PERTURBED SYSTEM

Our approach, in general, can be applied to the dendrite growth with arbitrary undercooling. In the present paper, however, we only consider the slender dendrite growth with a small undercooling, which is the most practical case. In this case, a slenderness parameter is defined as

$$\delta = \frac{\delta_D}{l_T} \ll 1 \ . \tag{3.1}$$

Hence the interface shape can be expressed in the cylindrical coordinate system as

$$R(z) = \delta R_{\star}(z) . \qquad (3.2)$$

Here we suppose the shape function $R_*(z)$ is known. In the tip region (see Fig. 1), this interface shape of the basic state evidently is approximately a paraboloid different from the Ivantsov paraboloid with the same undercooling. However, in previous works¹²⁻¹⁴ we have shown that when $\delta \ll 1$ in the tip region the isothermal surfaces of temperature field consist of a family of paraboloids up to the order of $O(\delta^4 \ln \delta)$. Namely, we have the following steady temperature distribution and form of shape function:

$$(\eta_s)_B = \eta_0 + \eta_1(\xi)\delta^{\alpha_1} + \cdots ,$$

$$(T)_B = A_1 + A_2 E_1(\eta^2/2) + \delta^4(\ln\delta)T_{B1}(\xi,\eta) + \cdots ,$$

(3.3)

$$(T_s)_B = A_1 + A_2 E_1(\eta_0^2/2) + \delta^4(\ln\delta) T_{sB1}(\xi,\eta) + \cdots,$$

where $E_1(x)$ is the exponential integral function, the index α_1 is a function of the parameters $\{T_{\infty}, \Gamma\}$ defined as

$$\alpha_1 = \frac{4}{1 - \sqrt{1 - x}} - 4 ,$$

$$x = \Gamma / \Gamma_{\text{max}}, \quad \Gamma_{\text{max}} = \frac{T_{\infty}^2}{8 |\ln \delta|} .$$
(3.4a)

As $\Gamma \to 0$, $\alpha_1 \to \infty$. In the realistic cases, $(\Gamma / \Gamma_{\text{max}}) < 0.2$, hence $\alpha_1 > 36$. The coefficients (A_1, A_2, η_0) in (3.3) are

$$A_{1} = T_{\infty} - \frac{\delta^{2}}{4} S'_{*}(0) \ln(\delta^{2}/2) - \frac{\delta^{2}}{4} [S'_{*}(0) + G_{0}],$$

$$A_{2} = \frac{\delta^{2}}{4} S'_{*}(0),$$

$$\eta_{0} = \frac{\delta}{2} \sqrt{S'_{*}(0)},$$

(3.4b)

and

$$S_{*}(z) = R_{*}^{2}(z), \quad S_{*}'(0) = \left[\frac{dS_{*}}{dz}\right]_{z=0},$$

$$G_{0} = \int_{0}^{\infty} [S_{*}''(z) - S_{*}'(z)]e^{-z}\ln(2z)dz \quad .$$
(3.4c)

We use the following tip variables:

$$\widetilde{\xi} = \frac{\xi}{\delta}, \quad \widetilde{\eta} = \frac{\eta}{\delta}, \quad \widetilde{t} = \alpha t, \quad \widetilde{\eta}_s = \frac{\eta_s}{\delta},$$
(3.5)

which implies that the length scale is changed to a quantity proportional to the tip radius of the dendrite. The factor α in (3.5) or the time scale is to be determined. The general unsteady solutions can be separated into two parts—the basic states (3.3) and the infinitesimal perturbations $\{\tilde{\eta}_s, \tilde{T}, \tilde{T}_s\}$. Namely,

$$T(\tilde{\xi},\tilde{\eta},\tilde{t}) = T_B + \tilde{T}(\tilde{\xi},\tilde{\eta},\tilde{t}) ,$$

$$T_s(\tilde{\xi},\tilde{\eta},\tilde{t}) = T_{sB} + \tilde{T}_s(\tilde{\xi},\tilde{\eta},\tilde{t}) ,$$

$$\tilde{\eta}_s = (\tilde{\eta}_s)_B + \frac{1}{\delta^2} \tilde{h}(\tilde{\xi},\tilde{t}) .$$
(3.6)

The linearized perturbed system then is derived by substituting (3.5) and (3.6) into (2.1) and (2.4)–(2.9). The corresponding boundary conditions at the interface are linearized on the paraboloid $\tilde{\eta} = \tilde{\eta}_0$, since $\delta \ll 1$. The procedure is standard. The results are listed as follows:

$$\frac{\partial^{2} \widetilde{T}}{\partial \widetilde{\xi}^{2}} + \frac{\partial^{2} \widetilde{T}}{\partial \widetilde{\eta}^{2}} + \frac{1}{\widetilde{\xi}} \frac{\partial \widetilde{T}}{\partial \widetilde{\xi}} + \frac{1}{\widetilde{\eta}} \frac{\partial \widetilde{T}}{\partial \widetilde{\eta}}$$
$$= \alpha \delta^{4} (\widetilde{\xi}^{2} + \widetilde{\eta}^{2}) \frac{\partial \widetilde{T}}{\partial \widetilde{t}} + \delta^{2} \left[\widetilde{\xi} \frac{\partial \widetilde{T}}{\partial \widetilde{\xi}} - \widetilde{\eta} \frac{\partial \widetilde{T}}{\partial \widetilde{\eta}} \right]. \quad (3.7)$$

The boundary conditions are

as
$$\tilde{\eta} \to \infty$$
, $\tilde{T} \to 0$, (3.8)

as
$$\tilde{\eta} \to 0$$
, \tilde{T}_s regular. (3.9)

At $\tilde{\eta} = \tilde{\eta}_0$,

$$\tilde{T} = \tilde{T}_s + \tilde{\eta}_0 \tilde{h} \quad , \tag{3.10}$$

$$\widetilde{T}_{s} = \frac{\epsilon^{2}}{s(\widetilde{\xi})} \left[\frac{\partial^{2} \widetilde{h}}{\partial \widetilde{\xi}^{2}} + \frac{\widetilde{\eta}_{0}^{2} + 2\widetilde{\xi}^{2}}{\widetilde{\xi}S^{2}(\widetilde{\xi})} \frac{\partial \widetilde{h}}{\partial \widetilde{\xi}} - \frac{\widetilde{h}}{s^{2}(\widetilde{\xi})} \right], \quad (3.11)$$

$$\frac{\partial}{\partial \tilde{\eta}} (\tilde{T} - \tilde{T}_s) + (\alpha \delta) S(\tilde{\xi}) \frac{\partial \tilde{h}}{\partial \tilde{t}} + \tilde{\xi} \frac{\partial \tilde{h}}{\partial \tilde{\xi}} + (2 + \delta^2 \tilde{\eta}_0^2) \tilde{h} = 0 ,$$

where

$$S(\tilde{\xi}) = (\tilde{\xi}^2 + \tilde{\eta}_0^2)^{1/2}, \quad \epsilon = \sqrt{\Gamma} / \delta^2 .$$
(3.13)

The stability parameter ϵ is similar to the parameter σ that Langer used. In the derivation of the above system, the following formulas are used:

$$\begin{cases} \frac{\partial}{\partial \bar{\eta}} (T)_B \\ \frac{\partial}{\partial \bar{\eta}} (T)_B \end{cases}_{\bar{\eta}_0} = -\delta^2 \bar{\eta}_0 + O(\delta^4 \ln \delta) , \qquad (3.14) \\ \begin{cases} \frac{\partial^2}{\partial \bar{\eta}^2} (T)_B \\ \frac{\partial}{\eta_0} = \delta^2 (1 + \delta^2 \bar{\eta}_0^2) + O(\delta^4 \ln \delta) , \\ \end{cases} \\ \begin{cases} \frac{\partial}{\partial \eta} (T_s)_B; \frac{\partial}{\partial \bar{\xi}} (T)_B \\ \frac{\partial}{\partial \bar{\xi}} (T)_B \end{cases} = O(\delta^4 \ln \delta) , \qquad (3.15) \end{cases}$$

and the higher-order terms in δ are omitted without changing the final form of the system described below. From the boundary condition (3.12), it is seen that one must set

$$\alpha = \frac{1}{\delta} \quad . \tag{3.16}$$

Two independent parameters $\{\delta, \Gamma\}$ emerge in the perturbed system (3.7)-(3.12). We set $\delta \rightarrow 0$ and $\Gamma \rightarrow 0$, but fix $\epsilon = \Gamma^{1/2}/\delta^2 = O(1)$. Thus the system (3.7)-(3.12) is reduced to the following:

$$\frac{\partial^2 \widetilde{T}}{\partial \widetilde{\xi}^2} + \frac{\partial \widetilde{T}}{\partial \widetilde{\eta}^2} = -\left[\frac{1}{\widetilde{\xi}}\frac{\partial \widetilde{T}}{\partial \widetilde{\xi}} + \frac{1}{\widetilde{\eta}}\frac{\partial \widetilde{T}}{\partial \widetilde{\eta}}\right].$$
(3.7')

The boundary conditions are

as
$$\tilde{\eta} \rightarrow \infty$$
, $\tilde{T} \rightarrow 0$, (3.8')

as
$$\tilde{\eta} \to 0$$
, $\tilde{T}_s = O(1)$, $\frac{\partial \tilde{T}_s}{\partial \tilde{\eta}} = 0$. (3.9')

At
$$\tilde{\eta} = \tilde{\eta}_0; 0 \leq \tilde{\xi} < \infty$$
,
 $\tilde{T} = \tilde{T}_s + \tilde{\eta}_0 \tilde{h}$, (3.10')

$$\widetilde{T}_{s} = \frac{\epsilon^{2}}{S(\widetilde{\xi})} \left[\frac{\partial^{2}\widetilde{h}}{\partial \widetilde{\xi}^{2}} + \frac{\widetilde{\xi}_{0}^{2} + 2\widetilde{\xi}^{2}}{\widetilde{\xi}S^{2}(\widetilde{\xi})} \frac{\partial\widetilde{h}}{\partial \widetilde{\xi}} - \frac{\widetilde{h}}{S^{2}(\widetilde{\xi})} \right], \quad (3.11')$$

$$\frac{\partial}{\partial \tilde{\eta}} (\tilde{T} - \tilde{T}_s) + S(\tilde{\xi}) \frac{\partial \tilde{h}}{\partial \tilde{t}} + \tilde{\xi} \frac{\partial \tilde{h}}{\partial \tilde{\xi}} + 2\tilde{h} = 0 . \qquad (3.12')$$

The above system (3.7)-(3.12) now only involves one dynamical parameter, ϵ . In the realistic solidification system, the parameter ϵ is actually a very small number. This allows us to seek for the asymptotic expansion solutions for the above system in the limit $\epsilon \rightarrow 0$. In the rest of the present paper, we are going to derive this type of asymptotic solution.

IV. MULTIPLE-VARIABLE-EXPANSION SOLUTIONS

In the system (3.7)-(3.12), the presence of the small parameter ϵ in front of the derivatives in the boundary condition (3.11) suggests the application of the multiple-variable-expansion method for finding asymptotic solutions (refer to Ref. 25). In doing so, we first of all introduce a set of fast variables

$$\widetilde{\xi}_{+} = \frac{\widetilde{\xi}}{\epsilon}, \quad \widetilde{\eta}_{+} = \frac{\widetilde{\eta} - \widetilde{\eta}_{0}}{\epsilon}, \quad \widetilde{t}_{+} = \frac{\widetilde{t}}{\epsilon},$$
(4.1)

and transform the system (3.7)-(3.12) in these fast variables as follows:

$$\left[\frac{\partial^2}{\partial \tilde{\xi}_+^2} + \frac{\partial^2}{\partial \tilde{\eta}_+^2}\right] \tilde{T} = -\epsilon \left[\frac{1}{\tilde{\xi}}\frac{\partial \tilde{T}}{\partial \tilde{\xi}_+} + \frac{1}{\tilde{\eta}}\frac{\partial \tilde{T}}{\partial \tilde{\eta}_+}\right].$$
(4.2)

The boundary conditions are

$$\tilde{\eta}_+ \to \infty, \quad \tilde{T} \to 0 ,$$
 (4.3)

$$\tilde{\eta}_{+} \rightarrow -\infty, \quad \tilde{T}_{s} \rightarrow 0$$
 (4.4)

At $\tilde{\eta}_{+} = 0$ or $\tilde{\eta} = \tilde{\eta}_{0}$,

- -

$$\tilde{T} = \tilde{T}_{s} + \tilde{\eta}_{0}\tilde{h} , \qquad (4.5)$$

$$\tilde{T}_{s} = \frac{1}{S(\tilde{\xi})} \left[\frac{\partial^{2}\tilde{h}}{\partial \tilde{\xi}_{+}^{2}} + \frac{\epsilon(\tilde{\eta}_{0}^{2} + 2\tilde{\xi}^{2})}{\tilde{\xi}S^{2}(\tilde{\xi})} \frac{\partial\tilde{h}}{\partial \tilde{\xi}_{+}} - \frac{\epsilon^{2}}{s^{2}(\tilde{\xi})}\tilde{h} \right] ,$$

$$\frac{\partial}{\partial \tilde{\eta}_{+}} (\tilde{T} - \tilde{T}_{s}) + S(\tilde{\xi}) \frac{\partial h}{\partial \tilde{t}_{+}} + 2\epsilon \tilde{h} = 0 .$$
(4.7)

It should be noted that (1) in the above, we set $\epsilon \rightarrow 0$ for a fixed $\tilde{\xi} > 0$; hence we have

$$\frac{\partial^2 \widetilde{T}}{\partial \widetilde{\xi}^2} \gg \frac{1}{\widetilde{\xi}} \frac{\partial \widetilde{T}}{\partial \widetilde{\xi}} ,$$

so that

$$\frac{\partial^2 \tilde{T}}{\partial \tilde{\xi}^2} + \frac{1}{\tilde{\xi}} \frac{\partial \tilde{T}}{\partial \tilde{\xi}} = \frac{1}{\epsilon^2} \left[\frac{\partial^2 \tilde{T}}{\partial \tilde{\xi}_+^2} + \frac{\epsilon}{\tilde{\xi}} \frac{\partial \tilde{T}}{\partial \tilde{\xi}_+} \right].$$
(4.8)

Obviously, this treatment is invalid in the tip region where $\tilde{\xi} = O(\epsilon)$. As a consequence, it will be seen that the asymptotic solution of the above system will lose the uni-

form validity at $\tilde{\xi} = 0$.

(2) As $\tilde{\eta} \rightarrow 0$, generally, $\tilde{T} = O(1)$; $\partial \tilde{T} / \partial \tilde{\eta} = 0$. Hence, in the vicinity of $\tilde{\eta} = 0$, the following two terms:

$$\left\{\frac{\partial^2 \widetilde{T}}{\partial \widetilde{\eta}^2};\frac{1}{\widetilde{\eta}}\frac{\partial \widetilde{T}}{\partial \widetilde{\eta}}\right\}$$

have the same order. However, we are only interested in the solutions in a region $\tilde{\eta}_* \leq \tilde{\eta} < \infty$, where $\tilde{\eta}_* \in (0, \tilde{\eta}_0)$ is a fixed constant. In this region, we have the inequality

$$\frac{\partial^2 \tilde{T}}{\partial \tilde{\eta}^2} \gg \frac{1}{\tilde{\eta}} \frac{\partial \tilde{T}}{\partial \tilde{\eta}} \quad (\text{as } \epsilon \to 0) \ .$$

Hence, in the derivation of (4.2), we also set

$$\frac{\partial^2 \widetilde{T}}{\partial \widetilde{\eta}^2} + \frac{1}{\widetilde{\eta}} \frac{\partial \widetilde{T}}{\partial \widetilde{\eta}} = \frac{1}{\epsilon^2} \left[\frac{\partial^2 \widetilde{T}}{\partial \widetilde{\eta}_+^2} + \frac{\epsilon}{\widetilde{\eta}} \frac{\partial \widetilde{T}}{\partial \widetilde{\eta}_+} \right].$$
(4.9)

Later we shall see that by making use of (4.9), the form of solutions is simplified but the asymptotic expansions will lose uniform validity at $\tilde{\eta} = 0$.

The system (4.2)-(4.7) contains the fast variables $(\tilde{\xi}_+, \tilde{\eta}_+, \tilde{t}_+)$ and the slow variables $(\tilde{\xi}, \tilde{\eta})$ corresponding to different length scales. All coefficients in this system are the functions of slow variables; whereas all derivatives are with respect to the fast variables. Now, we can make use of the multiple-variable-expansion method for finding the solutions.²⁵ To proceed so, however, in this problem we need to utilize the new fast variables $(\tilde{\xi}_{++}, \tilde{\eta}_{++})$ defined by

$$\frac{d\tilde{\xi}_{++}}{d\tilde{\xi}_{+}} = k(\tilde{\xi}) ,$$

$$\frac{d\tilde{\eta}_{++}}{d\tilde{\eta}_{+}} = k(\tilde{\xi}) ,$$

$$\tilde{t}_{++} = \tilde{t}_{+} ,$$
(4.10)

where the slowly varying function $k(\tilde{\xi})$ of $\tilde{\xi}$ is to be determined. Assuming $\{\tilde{\xi}, \tilde{\eta}, \tilde{\xi}_{++}, \tilde{\eta}_{++}, \tilde{t}_{++}\}$ are independent variables, we must substitute

$$\frac{\partial}{\partial \tilde{\xi}_{+}} = k \frac{\partial}{\partial \tilde{\xi}_{++}} + \epsilon \frac{\partial}{\partial \tilde{\xi}} ,$$

$$\frac{\partial}{\partial \tilde{\eta}_{+}} = k \frac{\partial}{\partial \tilde{\eta}_{++}} + \epsilon \frac{\partial}{\partial \tilde{\eta}} ,$$

$$\frac{\partial^{2}}{\partial \tilde{\xi}_{+}^{2}} = k^{2} \frac{\partial^{2}}{\partial \tilde{\xi}_{++}} + 2\epsilon k \frac{\partial^{2}}{\partial \tilde{\xi} \partial \tilde{\xi}_{++}}$$

$$+ \epsilon \left[\frac{\partial k}{\partial \tilde{\xi}} \right] \frac{\partial}{\partial \tilde{\xi}_{++}} + \epsilon^{2} \frac{\partial^{2}}{\partial \tilde{\xi}^{2}} ,$$

$$\frac{\partial^{2}}{\partial \tilde{\eta}_{+}^{2}} = k^{2} \frac{\partial^{2}}{\partial \tilde{\eta}_{++}} + 2\epsilon k \frac{\partial^{2}}{\partial \tilde{\eta} \partial \tilde{\eta}_{++}} + \epsilon^{2} \frac{\partial^{2}}{\partial \tilde{\eta}^{2}} ,$$
(4.11)

in the system (4.2)–(4.7). The resulting system is given in the Appendix. Now, we let $\epsilon \rightarrow 0$ and make the following multiple-variable expansions:

$$\widetilde{T} = [\widetilde{T}_{0}(\widetilde{\xi}, \widetilde{\eta}, \widetilde{\xi}_{++}, \widetilde{\eta}_{++}) + \cdots]e^{\sigma \widetilde{t}_{++}},$$

$$+ \epsilon \widetilde{T}_{1}(\widetilde{\xi}, \widetilde{\eta}, \widetilde{\xi}_{++}, \widetilde{\eta}_{++}) + \cdots]e^{\sigma \widetilde{t}_{++}},$$

$$\widetilde{T}_{s} = [\widetilde{T}_{s0}(\widetilde{\xi}, \widetilde{\eta}, \widetilde{\xi}_{++}, \widetilde{\eta}_{++}) + \cdots]e^{\sigma \widetilde{t}_{++}},$$

$$\widetilde{h} = [\widetilde{h}_{0}(\widetilde{\xi}, \widetilde{\xi}_{++}) + \epsilon \widetilde{h}_{1}(\widetilde{\xi}, \widetilde{\xi}_{++}) + \cdots]e^{\sigma \widetilde{t}_{++}},$$

$$k = k_{0}(\widetilde{\xi}) + \epsilon k_{1}(\widetilde{\xi}) + \cdots,$$
(4.12)

where the eigenvalue σ is a complex number. By substituting (4.11) and (4.12) into the system (4.2)-(4.7), the successive order approximate solutions can be derived. In the zeroth-order approximation, one can obtain the dispersion relationship formula; whereas in the first-order approximation, one can obtain the amplitude functions of perturbations. The results are shown as follows.

(1) $O(\epsilon^0)$. For the zeroth-order approximation, the following system is obtained:

$$\nabla^{2} \tilde{T}_{0} = \left[\frac{\partial^{2}}{\partial \tilde{\xi}_{++}^{2}} + \frac{\partial^{2}}{\partial \tilde{\eta}_{++}^{2}} \right] \tilde{T}_{0} = 0 , \qquad (4.13)$$

$$\nabla^{2} \tilde{T}_{s0} = \left[\frac{\partial^{2}}{\partial \tilde{\xi}_{++}^{2}} + \frac{\partial^{2}}{\partial \tilde{\eta}_{++}^{2}} \right] \tilde{T}_{s0} = 0 .$$

The boundary conditions are

as
$$\tilde{\eta}_{++} \rightarrow \infty$$
, $\tilde{T}_0 \rightarrow 0$, (4.14)

as
$$\tilde{\eta}_{++} \rightarrow -\infty$$
, $\tilde{T}_s \rightarrow 0$. (4.15)

At $\tilde{\eta}_{++} = 0$ or $\tilde{\eta} = \tilde{\eta}_0$,

$$\tilde{T}_0 = \tilde{T}_{s0} + \tilde{\eta}_0 \tilde{h}_0 , \qquad (4.16)$$

$$\widetilde{T}_{s0} = \frac{k_0^2}{S} \frac{\partial^2 h_0}{\partial \widetilde{\xi}_{++}^2} , \qquad (4.17)$$

$$k_0 \frac{\partial}{\partial \tilde{\eta}_{++}} (\tilde{T}_0 - \tilde{T}_{s0}) + \sigma S \tilde{h}_0 + k_0 \tilde{\xi} \frac{\partial \tilde{h}_0}{\partial \tilde{\xi}_{++}} = 0 . \quad (4.18)$$

We consider the following mode solutions of (4.13):

$$\begin{split} \widetilde{T}_{0} &= A_{0}(\widetilde{\xi}, \widetilde{\eta}) e^{i\widetilde{\xi}_{++} - \widetilde{\eta}_{++}} ,\\ \widetilde{T}_{s0} &= B_{0}(\widetilde{\xi}, \widetilde{\eta}) e^{i\widetilde{\xi}_{++} + \widetilde{\eta}_{++}} ,\\ \widetilde{h}_{0} &= \widehat{D}_{0}(\widetilde{\xi}) e^{i\widetilde{\xi}_{++}} . \end{split}$$

$$(4.19)$$

Denoting

$$A_0(\tilde{\xi}, \tilde{\eta}_0) = \hat{A}_0(\tilde{\xi}), \quad B_0(\tilde{\xi}, \tilde{\eta}_0) = \hat{B}_0(\tilde{\xi}) , \quad (4.20)$$

from the boundary conditions (4.15)-(4.17), we obtain

$$\hat{A}_{0} = \left[\tilde{\eta}_{0} - \frac{k_{0}^{2}}{S} \right] \hat{D}_{0}, \quad \hat{B}_{0} = -\frac{k_{0}^{2}}{S} \hat{D}_{0}$$
(4.21)

and the dispersion relationship formula

$$\sigma = \sigma(k_0) = \frac{k_0}{S} \left[\tilde{\eta}_0 - \frac{2k_0^2}{S} \right] - i\frac{\tilde{\xi}}{S}k_0 . \qquad (4.22)$$

(2) $O(\epsilon^1)$. For the first-order approximation, the governing equation is

$$\nabla^{2} \tilde{T}_{1} = a_{0} e^{i\tilde{\xi}_{++} - \tilde{\eta}_{++}},$$

$$\nabla^{2} \tilde{T}_{s1} = b_{0} e^{i\tilde{\xi}_{++} + \tilde{\eta}_{++}},$$
(4.23)

where

$$a_{0} = -2k_{0}\left[i\frac{\partial A_{0}}{\partial \tilde{\xi}} - \frac{\partial A_{0}}{\partial \tilde{\eta}}\right] - A_{0}\left[i\frac{dk_{0}}{d\tilde{\xi}} + i\frac{k_{0}}{\tilde{\xi}} - \frac{k_{0}}{\tilde{\eta}}\right],$$

$$(4.24a)$$

$$b_{0} = -2k_{0}\left[i\frac{\partial B_{0}}{\partial \tilde{\xi}} + \frac{\partial B_{0}}{\partial \tilde{\eta}}\right] - B_{0}\left[i\frac{dk_{0}}{d\tilde{\xi}} + i\frac{k_{0}}{\tilde{\xi}} + \frac{k_{0}}{\tilde{\eta}}\right].$$

$$(4.24b)$$

To assure the uniform validity of the expansions as $\tilde{\xi}_+ \rightarrow \infty$, we must eliminate the secular term in the righthand side of Eqs. (4.23), or say, set

$$a_0 = b_0 = 0$$
 . (4.25)

From (4.24a) and (4.24b), we derive

$$\left[\frac{\partial}{\partial \tilde{\xi}} + i \frac{\partial}{\partial \tilde{\eta}} \right] \ln(A_0 k_0^{1/2} \tilde{\xi}^{1/2} \tilde{\eta}^{1/2}) = 0 ,$$

$$\left[\frac{\partial}{\partial \tilde{\xi}} - i \frac{\partial}{\partial \tilde{\eta}} \right] \ln(B_0 k_0^{1/2} \tilde{\xi}^{1/2} \tilde{\eta}^{1/2}) = 0 .$$

$$(4.26)$$

At $\tilde{\eta} = \tilde{\eta}_0$, we have

$$\boldsymbol{A}_{0} = \widehat{\boldsymbol{A}}_{0}(\widetilde{\boldsymbol{\xi}}), \quad \boldsymbol{B}_{0} = \widehat{\boldsymbol{B}}_{0}(\widetilde{\boldsymbol{\xi}}) \quad . \tag{4.27}$$

We extend the functions $\{\hat{A}_0(\xi), \hat{B}_0(\xi), k_0(\xi)\}$ analytically to the whole complex ξ plane. Then, the following solutions of (4.26) are obtained:

$$\begin{split} \boldsymbol{A}_{0}(\tilde{\xi},\tilde{\eta}) &= \left[\frac{\tilde{\eta}_{0}}{\tilde{\eta}}\right]^{1/2} \left[\frac{\tilde{\xi}+i\tilde{\eta}_{1}}{\tilde{\xi}}\right]^{1/2} \frac{k_{0}^{1/2}(\tilde{\xi}+i\tilde{\eta}_{1})}{k_{0}^{1/2}(\tilde{\xi})} \\ &\times \hat{\boldsymbol{A}}_{0}(\tilde{\xi}+i\tilde{\eta}_{1}) , \\ \boldsymbol{B}_{0}(\tilde{\xi},\tilde{\eta}) &= \left[\frac{\tilde{\eta}_{0}}{\tilde{\eta}}\right]^{1/2} \left[\frac{\tilde{\xi}-i\tilde{\eta}_{1}}{\tilde{\xi}}\right] \frac{k_{0}^{1/2}(\tilde{\xi}-i\tilde{\eta}_{1})}{k_{0}^{1/2}(\tilde{\xi})} \\ &\times \hat{\boldsymbol{B}}_{0}(\tilde{\xi}-i\tilde{\eta}_{1}) , \end{split}$$
(4.28a)

where

$$\tilde{\eta}_1 = \tilde{\eta} - \tilde{\eta}_0 \ . \tag{4.28b}$$

Due to the condition (4.25), the solutions of Eq. (4.23) are written as

$$\widetilde{T}_{1} = A_{1}(\widetilde{\xi}, \widetilde{\eta}) e^{i\widetilde{\xi}_{++} - \overline{\eta}_{++}} ,$$

$$\widetilde{T}_{s1} = B_{1}(\widetilde{\xi}, \widetilde{\eta}) e^{i\widetilde{\xi}_{++} - \overline{\eta}_{++}} .$$
(4.29)

Setting

and

$$\boldsymbol{A}_{1}(\tilde{\boldsymbol{\xi}},\tilde{\boldsymbol{\eta}}_{0}) = \boldsymbol{\hat{A}}_{1}(\boldsymbol{\tilde{\xi}}), \quad \boldsymbol{B}_{1}(\boldsymbol{\tilde{\xi}},\tilde{\boldsymbol{\eta}}_{0}) = \boldsymbol{\hat{B}}_{1}(\boldsymbol{\tilde{\xi}}) \quad , \tag{4.30}$$

$$\widetilde{h}_1 = \widehat{D}_1(\widetilde{\xi}) e^{i\xi_{++}} , \qquad (4.31)$$

from the boundary conditions on the interface, we derive that at $\tilde{\eta} = \tilde{\eta}_0$,

$$\hat{A}_1 = \hat{B}_1 + \tilde{\eta}_0 \hat{D}_1$$
, (4.32)

$$\hat{B}_{1} = \frac{1}{S} \left[-k_{0}^{2} \hat{D}_{1} - 2k_{0} k_{1} \hat{D}_{0} + 2k_{0} \left[i \frac{\partial \hat{D}_{0}}{\partial \tilde{\xi}} \right] + i \left[\frac{\partial k_{0}}{\partial \tilde{\xi}} \right] \hat{D}_{0} + i \left[\frac{1}{\tilde{\xi}} + \frac{\tilde{\xi}}{S^{2}} \right] k_{0} \hat{D}_{0} \right], \quad (4.33)$$

$$-k_{0}(A_{1}+B_{1})+(\sigma S+i\xi k_{0})D_{1}$$

$$=k_{1}(\hat{A}_{0}+\hat{B}_{0})-\frac{\partial}{\partial\tilde{\eta}}(\tilde{A}_{0}-\tilde{B}_{0})$$

$$-(ik_{1}\tilde{\xi}+2)\hat{D}_{0}-\tilde{\xi}\frac{\partial\hat{D}_{0}}{\partial\tilde{\xi}} \quad (4.34)$$

By using the formula (4.26) and (4.22), one can derive that

$$\hat{A}_{1} = \left[\bar{\eta}_{0} - \frac{k_{0}^{2}}{S} \right] \hat{D}_{1} + I_{1}(\tilde{\xi}) ,$$

$$\hat{B}_{1} = -\frac{k_{0}^{2}}{S} \hat{D}_{1} + I_{1}(\tilde{\xi}) ,$$
(4.35)

where

$$I_{1}(\tilde{\xi}) = \frac{2k_{0}}{S} \left[i \frac{\partial \hat{D}_{0}}{\partial \tilde{\xi}} - k_{1} \hat{D}_{0} \right] + \frac{ik_{0} \hat{D}_{0}}{S} \left[\frac{d \ln k_{0}}{d \tilde{\xi}} + \frac{1}{\tilde{\xi}} + \frac{\tilde{\xi}}{S^{2}} \right]$$
(4.36)

and

$$\left[k_{1} - i\frac{d}{d\tilde{\xi}}\ln\hat{D}_{0}\right] = \frac{W_{1}(\tilde{\xi})}{S(\partial\sigma/\partial k_{0})} , \qquad (4.37)$$

where

$$W_{1}(\tilde{\xi}) = \frac{3}{2} + i \left[\tilde{\eta}_{0} - \frac{14k_{0}^{2}}{S} \right] \frac{d \ln k_{0}^{1/2}}{d\tilde{\xi}} + \frac{i}{2\tilde{\xi}} \left[\tilde{\eta}_{0} - \frac{6k_{0}^{2}}{S} \right],$$
(4.38)

$$\frac{\partial\sigma}{\partial k_0} \left] = \frac{1}{S} (\tilde{\eta}_0 - i\tilde{\xi}) - \frac{6k_0^2}{S^2} \right] .$$
(4.39)

The above procedure can be continued to the furtherorder approximations in the region where $\xi \neq 0$ and $\partial \sigma / \partial k_0 \neq 0$. No difficulty seems to occur. The dispersion relation formula (4.22) and the amplitude formula (4.37) have significant physical implications. In the next section, we shall focus on the discussion of these results.

V. LOCAL INSTABILITY THEORY OF DENDRITIC GROWTH

In the preceding section, as the lowest-order approximation, we derived the dispersion relationship formula (4.22) for the normal mode perturbations of the interface shape:

$$\widetilde{h} \approx d_0 \exp\left[\frac{i}{\epsilon} \int k_0 d\widetilde{\xi} + \frac{\sigma}{\epsilon} \widetilde{t}\right] .$$
(5.1)

The dispersion relationship formula (4.22) can be written into the following form:

$$\sigma_* = k_* (|V_n| - 2k_*^2) - iV_\tau k_* , \qquad (5.2)$$

where

$$V_n \approx \frac{-\tilde{\eta}_0}{S}, \quad V_\tau \approx \frac{\tilde{\xi}}{S}$$
 (5.3)

are the normal component and tangential component of the local growth velocity of interface at $\tilde{\xi}$, respectively, while

$$k_* = \frac{k_0}{S}, \quad \sigma_* = (\sigma_R)_* - i\omega_* = \frac{\sigma}{S} \quad . \tag{5.4}$$

The function $k_*(\tilde{\xi})$ can be regarded as the local wave number at ξ , measured by the local arc length (Δl), of the element $(\Delta \tilde{\xi})$; whereas $(\sigma_R)_*$ and ω_* are the local growth rate of amplitude and frequency of perturbation measured by the local time scale based on the above local arc length. Suppose that we have a local perturbation at $\tilde{\xi}$ with a real wave number k_* , then the first term in the RHS of (5.2) represents the local Müllin-Sekerka morphological instability generated by the normal growth velocity V_n ; whereas the second term in the RHS of (5.2) shows that beside the Müllin-Sekerka morphological instability, the local interface has an oscillation with the frequency $\omega_* = V_\tau k_*$ generated by the tangential component of the local growth velocity of the interface. The local dispersion formula (4.22) or (5.2) has a particular significance for the full understanding of the instability mechanism of general growing interfaces with a nonzero tangential component of growth velocity. As we see that the well-known Müllin-Sekerka dispersion formula is a special case of our results (4.22) for the unidirectional solidifying planar interface, where the purely growing type of modes are the only possible type of unstable modes; the surface tension effect suppresses the short wave perturbations, and the marginal stability wavelength is a constant. However, for the more general dendrite case, the local marginal stability wavelength is a slowly varying function of $\tilde{\xi}$, the energy of perturbations is transferred along the interface with a group velocity V_{τ} . As a consequence, various traveling waves form. The presence of these unstable traveling waves on the interface is one of the most important features in the dendritic crystal growth, which is responsible for the formation of sidebranching structure. We shall explore this in details in the remaining sections of this paper.

VI. DEFECTS OF THE LOCAL NORMAL SOLUTIONS AND A PATTERN FORMATION CONDITION

In the preceding section, we demonstrate the local instability mechanism of the normal modes (5.1) in terms of the dispersion formula (4.22). We verify that the interface of the dendrite permits various local unstable modes. Here, significant questions arise as to whether those local unstable modes can form a coherent structure in the whole region $(0 \le \tilde{\xi} < \infty)$; can one obtain global-mode solutions to the problem with a fixed eigenvalue $\sigma = \sigma_R - i\omega$, which satisfy appropriate boundary conditions? Apparently, for any fixed constant σ , one can solve the wave-number function $k_0(\tilde{\xi})$ from the dispersion formula (4.22), hence determine a normal-mode solution (5.1). This solution, however, is not uniformly valid in the whole region $(0 \le \tilde{\xi} < \infty)$. Therefore it cannot be used as a global-mode solution. In the following, we are going to illustrate this in detail. An alternative form of (4.22) is derived

$$k_0 = M(\tilde{\xi})\cos(z), \quad \sigma = N(\tilde{\xi})\cos(3z) , \quad (6.1)$$

where

$$M(\tilde{\xi}) = \left(\frac{2S(\tilde{\xi})}{3}\right)^{1/2} (\tilde{\eta}_0 - i\tilde{\xi})^{1/2} ,$$

$$N(\tilde{\xi}) = -\frac{M(\tilde{\xi})}{3S(\tilde{\xi})} (\tilde{\eta}_0 - i\tilde{\xi}) .$$
(6.2)

For a given σ , one can obtain three roots from (6.1) as shown in the Fig. 3,

$$k_0^{(1)}(\tilde{\xi}) = M(\tilde{\xi}) \cos\left[\frac{1}{3}\cos^{-1}\left(\frac{\sigma}{N(\tilde{\xi})}\right)\right]$$

(short wave branch),

$$k_{0}^{(2)}(\tilde{\xi}) = M(\tilde{\xi}) \cos\left[\frac{1}{3}\cos^{-1}\left[\frac{\sigma}{N(\tilde{\xi})}\right] + \frac{2\pi}{3}\right], \quad (6.3)$$
$$k_{0}^{(3)}(\tilde{\xi}) = M(\tilde{\xi}) \cos\left[\frac{1}{3}\cos^{-1}\left[\frac{\sigma}{N(\tilde{\xi})}\right] + \frac{4\pi}{3}\right]$$

(long wave branch).

The numerical results show that $\operatorname{Re}\{k_0^{(2)}\} < 0$. In view of the boundary condition (4.14) this root is ruled out. By use of the wave-number function $k_0^{(3)}(\tilde{\xi})$ or $k_0^{(1)}(\tilde{\xi})$, one can write the normal-mode solution (5.1). This solution is not uniformly valid in the region $(0 \leq \tilde{\xi} < \infty)$, since from the formula (4.37) we see that the multiple-variable expansions fail in the tip region $\tilde{\xi} \approx 0$ and in the vicinity of the critical point $\tilde{\xi}_c$, where

$$\left[\frac{\partial\sigma}{\partial k_0}\right]_{\bar{\xi}_c} = \left[\frac{1}{S}(\tilde{\eta}_0 - i\tilde{\xi}) - \frac{6k_0^2}{S^2}\right]_{\bar{\xi}_c} = 0.$$
 (6.4)

One can easily derive that for the given σ , this critical point ξ_c is also the root of the following equation:

$$\sigma = \left(\frac{2}{27}\right)^{1/2} \frac{(\eta_0 - i\tilde{\xi}_c)^{5/4}}{(\eta_0 + i\tilde{\xi}_c)^{1/4}} .$$
(6.5)

JIAN-JUN XU



FIG. 3. Curve of wave-number functions $\{k_0^{(1)}(\xi); k_0^{(2)}(\xi); k_0^{(3)}(\xi)\}$ corresponding to various eigenvalues σ for $\tilde{\eta}_0 = 0.5$.



FIG. 4. Eigenvalues σ of global modes and corresponding critical points ξ_c on the real axis. ($\tilde{\eta}_0 = 0.5$.)

(6.7)

It is seen that

as
$$\sigma = 0$$
, $\tilde{\xi}_c = -i\tilde{\eta}_0$,
as $\sigma = +\infty$, $\tilde{\xi}_c = i\tilde{\eta}_0$. (6.6)

When $\tilde{\xi}_c$ is on the real axis of the complex $\tilde{\xi}$ plane $(0 \leq \tilde{\xi}_c < \infty)$, the corresponding $\sigma = \sigma_c(\tilde{\xi}_c)$, which delineates a curve on the complex σ plane as shown in Fig. 4. Therefore, if the given σ is in the domain (Σ_1) on the complex σ plane, the corresponding critical point $\tilde{\xi}_c$ will be below the real axis on the complex $\tilde{\xi}_c$ plane; if σ is in the domain (Σ_2) , the corresponding $\tilde{\xi}_c$ will be above the real axis.

Figure 5(a) shows that for a eigenvalue σ belonging to the domain (Σ_2) when $\tilde{\xi}$ moves along the real axis and passes by the critical point $\tilde{\xi}_c$ the wave-number functions



FIG. 5. (a) Curves of wave-number function corresponding to an eigenvalue in the domain (Σ_2) ; (b) the curves of wave-number function corresponding to an eigenvalue in the domain (Σ_1) .

 $k_0^{(1)}(\tilde{\xi})$ and $k_0^{(3)}(\tilde{\xi})$ will have a jump. This discontinuity, of course, is not permissible. Thus we deduce that the eigenvalue σ must belong to the domain (Σ_1) on the complex σ plane. In other words, the critical point $\tilde{\xi}_c$ must be located in the lower half of complex $\tilde{\xi}$ plane. We call this necessary condition "the pattern-formation condition," which can be expressed as

$$\operatorname{Im}(\xi_c) \leq 0$$

or

$$\sigma \in (\Sigma_1)$$
.

We also deduce that the glowing global modes can only exist in the low-frequency band $(0 \le \omega < \omega_{max}; as \eta_0=0.5, \omega_{max}=0.27)$. Furthermore, Fig. 5(b) shows that even for σ satisfying the condition (6.7), the normal-mode solutions (5.1) may still not be uniformly valid in the region $(0 < \tilde{\xi} < \infty)$, because as $\tilde{\xi}$ pass by the critical $\tilde{\xi}_c$, the derivatives

$$\frac{d}{d\tilde{\xi}}(k_0)_I \Big/ \frac{d}{d\tilde{\xi}}(k_0)_R = \infty \; .$$

To obtain a uniformly valid global-mode solution, one must study the characteristics of solution near the critical point ξ_c and the tip, respectively. This work will be done in paper II.

VII. CONCLUSIONS

We summarize the above results as follows.

(1) In a dendritic crystal growth process, the local instability mechanism of the interface consists of two interfering parts: (i) a Müllin-Sekerka-type morphological instability generated by the normal component of the local growth velocity of the interface V_n ; (ii) a local oscillation generated by the tangential component of the local growth velocity of the interface V_{τ} . This mechanism results in various local unstable traveling waves, which we call sidebranching waves.

(2) The normal-mode solution (5.1) based on the local dispersion relationship is not uniformly valid in the whole region $(0 \le \tilde{\xi} < \infty)$. It fails validity near the tip and the vicinity of critical point $\tilde{\xi}_c$. In order to obtain a global-mode solution, one must study the characteristics of the solution near the tip and the critical point $\tilde{\xi}_c$, respectively, and propose an appropriate mathematical formulation for the problem. This will be done in the following paper.²⁴

ACKNOWLEDGMENTS

This research was supported by the Microgravity Science and Application Division of the National Aeronautics and Space Administration at Marshall Space Flight Center under Contract No. NAS8-37301.

APPENDIX

The linearized perturbed system written in the multiple variables $\{\tilde{\xi}, \tilde{\eta}, \tilde{\xi}_{++}, \tilde{\eta}_{++}, \tilde{t}_{++}\}$ is listed as follows:

JIAN-JUN XU

$$\left[\frac{\partial^2}{\partial \tilde{\xi}_{++}} + \frac{\partial^2}{\partial \tilde{\eta}_{++}} \right] \tilde{T} = -\frac{2\epsilon}{k} \left[\frac{\partial^2 \tilde{T}}{\partial \tilde{\xi} \partial \tilde{\xi}_{++}} + \frac{\partial^2 \tilde{T}}{\partial \tilde{\eta} \partial \tilde{\eta}_{++}} + \frac{d}{d\tilde{\xi}} \ln(k^{1/2}) \frac{\partial \tilde{T}}{\partial \tilde{\xi}_{++}} \right] - \frac{1}{k^2} \left[\frac{\epsilon}{\tilde{\xi}} \left[k \frac{\partial \tilde{T}}{\partial \tilde{\xi}_{++}} + \epsilon \frac{\partial \tilde{T}}{\partial \tilde{\xi}} \right] + \frac{\epsilon}{\tilde{\eta}} \left[k \frac{\partial \tilde{T}}{\partial \tilde{\eta}_{++}} + \epsilon \frac{\partial \tilde{T}}{\partial \tilde{\eta}} \right] + \epsilon^2 \left[\frac{\partial^2 \tilde{T}}{\partial \tilde{\xi}^2} + \frac{\partial^2 \tilde{T}}{\partial \tilde{\eta}^2} \right] \right].$$
 (A1)

The boundary conditions are

as
$$\tilde{\eta}_{++} \rightarrow \infty$$
, $\tilde{T} \rightarrow 0$, (A2)

as
$$\tilde{\eta}_{++} \rightarrow -\infty$$
, $\tilde{T}_s \rightarrow 0$. (A3)

At
$$\tilde{\eta}_{++} = 0$$
, $\tilde{\eta} = \tilde{\eta}_0$,
 $\tilde{z} = \tilde{z}$, $\tilde{z} = \tilde{z}$

$$\tilde{T} = \tilde{T}_s + \tilde{\eta}_0 \tilde{h} \quad , \tag{A4}$$

$$\widetilde{T}_{s} = \frac{1}{S(\widetilde{\xi})} \left[k^{2} \frac{\partial^{2} \widetilde{h}}{\partial \widetilde{\xi}_{++}^{2}} + 2\epsilon k \frac{\partial^{2} \widetilde{h}}{\partial \widetilde{\xi} \partial \widetilde{\xi}_{++}} + \epsilon \left[\frac{dk}{d\widetilde{\xi}} \right] \frac{\partial \widetilde{h}}{\partial \widetilde{\xi}_{++}} + \epsilon^{2} \frac{\partial^{2} \widetilde{h}}{\partial \widetilde{\xi}^{2}} + \epsilon \frac{\widetilde{\eta}_{0}^{2} + 2\widetilde{\xi}^{2}}{\widetilde{\xi} S^{2}(\widetilde{\xi})} \left[k \frac{\partial \widetilde{h}}{\partial \widetilde{\xi}_{++}} + \epsilon \frac{\partial \widetilde{h}}{\partial \widetilde{\xi}} \right] - \epsilon^{2} \frac{\widetilde{h}}{S^{2}(\widetilde{\xi})} \right], \quad (A5)$$

$$\left[k\frac{\partial}{\partial\tilde{\eta}_{++}} + \epsilon\frac{\partial}{\partial\tilde{\eta}}\right](\tilde{T} - \tilde{T}_{s}) + S(\tilde{\xi})\frac{\partial\tilde{h}}{\partial\tilde{t}_{++}} + \tilde{\xi}\left[k\frac{\partial}{\partial\tilde{\xi}_{++}} + \epsilon\frac{\partial}{\partial\tilde{\xi}}\right]\tilde{h} + 2\epsilon\tilde{h} = 0.$$
(A6)

- ¹G. P. Ivantsov, Dokl. Akad. Nauk SSSR 58, 567 (1947).
- ²G. Horvay and J. W. Cahn, Acta Metall. 9, 695 (1961).
- ³J. S. Langer, Rev. Mod. Phys. **52**, 1 (1980).
- ⁴G. F. Bolling and W. A. J. Tiller, Appl. Phys. **32**, 297 (1967).
- ⁵B. Chalmer, *Principles of Solidification* (Wiley, New York, 1964), p. 105, and reference therein.
- ⁶M. E. Glicksman and R. J. Schaefer, J. Cryst. Growth 1, 297 (1967).
- ⁷M. E. Glicksman and R. J. Schaefer, J. Cryst. Growth 2, 239 (1968).
- ⁸D. E. Temkin, Dokl. Akad. Nauk SSSR 132, 1307 (1960).
- ⁹R. F. Sekerka, J. Phys. Chem. Solids **28**, 983 (1967); in *Crystal Growth*, edited by H. Peiser (Publisher, City, 1967), p. 691.
- ¹⁰R. Trivedi, Acta Metall. **18**, 287 (1970).
- ¹¹G. E. Nash and M. E. Glicksman, Acta Metall. 22, 1283 (1974).
- ¹²J. J. Xu, in Structure and Dynamics of Partially Solidified System, Vol. 125 of NATO Advanced Study Institute, Series E: Subject, edited by D. E. Loper (Plenum, New York, 1987), p. 97.

- ¹³J. J. Xu (unpublished).
- ¹⁴J. J. Xu, Ph.D. thesis, Rensselaer Polytechnic Institute, 1987.
- ¹⁵J. J. Xu, Phys. Rev. A **37**, 3087 (1988).
- ¹⁶D. A. Kessler, J. Koplik, and H. Levine, Phys. Rev. A 30, 2820 (1984), 30, 3161 (1984).
- ¹⁷D. A. Kessler, J. Koplik, and H. Levine, Phys. Rev. A 31, 1712 (1985).
- ¹⁸D. A. Kessler, J. Koplik, and H. Levine, Phys. Rev. A 33, 3352 (1986).
- ¹⁹D. I. Meiron, Phys. Rev. A 33, 2704 (1986).
- ²⁰P. Pelce and Y. Pomeau, Stud. Appl. Math. 74, 245 (1986).
- ²¹M. N. Barber, A. Barbieri, and J. S. Langer, Phys. Rev. **36**, 3340 (1987).
- ²²J. S. Langer, Phys. Rev. 36, 3350 (1987).
- ²³M. B. Amar and Y. Pomeau, Euro Phys. Lett. 2, 307 (1986).
- ²⁴J. J. Xu, following paper, Phys. Rev. A 40, 1609 (1989).
- ²⁵J. Kevorkian and J. D. Cole, in *Perturbation Methods in Applied Mathematics*, Vol. 34 of *Applied Mathematical Sciences*, edited by F. John, J. E. Marsden, and L. Sirovich (Springer-Verlag, New York, 1981).