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IN RAPID DIRECTIONAL SOLIDIFICATION

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IMA Preprint Series # 1250
September 1994

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Oscillatory and Cellular Mode Coupling in Rapid Directional Solidification

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August 1994
Abstract

At large rates of solidification, some metallic alloys exhibit periodic microstructures along the growth direction in which layers free of lateral segregation alternate with cellular, dendritic, or eutectic phases. We investigate the formation of novel microstructures such as these bands by studying the nonlinear dynamics of the rapidly solidifying interface for a dilute binary alloy. The model employed in these studies has a velocity-dependent segregation coefficient and liquidus slope, a linear form of attachment kinetics, and the effects of latent heat release and full temperature distribution. Huntley and Davis performed a linear stability analysis on this model which revealed two modes of instability to the planar solid/liquid interface: (i) a steady cellular instability, and (ii) an oscillatory instability driven by disequilibrium effects. In this paper we partially test the conjecture that banded microstructures are due to the nonlinear interaction between these two instabilities by performing a weakly-nonlinear analysis on their coupling. The resulting coupled Landau equations govern the behavior of the disturbance amplitudes. A bifurcation analysis is performed to determine the dynamics present in the system. The results are then applied to several physical systems, and the question of the existence of bands is addressed.
1. Introduction

Rapid directional solidification is an important model technique in materials science for several reasons: (i) the transition region can be fixed in a laboratory reference frame, thereby allowing the detailed control and study of the morphology of the phase change, (ii) geometrical effects are minimized, and (iii) novel microstructures may appear at high rates of solidification when the solid/liquid interface departs from local thermodynamic equilibrium. The physical insights gained by studying this model can be applied to the more complicated rapid-solidification processing techniques (Kurz & Trivedi 1990, Boettinger & Perepezko 1985, Boettinger & Coriell 1986, and Boettinger et al. 1984) used in industry, such as pulsed laser annealing, electron beam treatment, melt atomization/spray casting, spin casting, splat quenching, and chillblock casting.

We wish to investigate the formation of large-transition-rate microstructures by studying the nonlinear dynamics of the rapidly solidifying interface for a dilute binary alloy. The nonequilibrium model we use is one proposed by Baker & Cahn (1971), Boettinger & Perepezko (1985), and Boettinger & Coriell (1986), in which the segregation coefficient and liquidus slope have a velocity dependence represented by the disequilibrium parameter, $\beta_0$, and where a linear law of attachment kinetics measures the ability of solute atoms to attach to the crystalline solid. The parameter $V_0$ is a measure of this kinetic undercooling.

Huntley & Davis (1993a) performed a linear stability analysis on this model; for convenience, they assumed that the thermal conductivities and diffusivities in the solid and liquid are equal. They found that the characteristic equation admits two modes of instability: (i) a steady cellular mode, related to the Mullins & Sekerka (1964) instability, and (ii) an oscillatory mode, related to the oscillatory instability in Coriell & Sekerka (1983) and the pulsatile instabilities in Caroli et al. (1986) and Merchant & Davis (1990). The critical wavenumber of the oscillatory mode can either be zero or nonzero depending on the physical parameters prescribed; the ratio of solutal to thermal diffusivities and the latent heat are the important
parameters for the critical wavenumber. Karma & Sarkissian (1993) also determined some of these results for the oscillatory instability.

There have been several extensions of the linear theory into the nonlinear regime in the absence of latent heat. Braun & Davis (1992) analyzed the weakly-nonlinear behavior of the cellular mode of Merchant & Davis (1990) just past threshold and obtained an amplitude equation that describes the bifurcation structure. Braun & Davis (1991) also analyzed the weakly-nonlinear behavior of the pulsatile mode of Merchant & Davis just past threshold and obtained a complex-coefficient Ginzburg-Landau equation that governs the evolution of the amplitude of the interface. They found that the zero-wavenumber mode is sharply selected as the pulling speed is increased. Merchant et al. (1992) performed a strongly-nonlinear analysis of this zero-wavenumber oscillatory mode and derived amplitude equations which describe relaxation oscillations and the bifurcation structure. Brattkus & Meiron (1992) observed many of these features in their direct numerical simulation of this mode.

Karma & Sarkissian (1993) included the effects of latent heat and the full thermal fields and studied the oscillatory instability numerically. They examined the pulsation of the planar interface, though in their case the zero-wavenumber mode is not preferred, and described strongly nonlinear “relaxation oscillations” of the interface as controlled by the diffusion of latent heat. Huntley & Davis (1993b) extended the analysis of Braun & Davis (1991) by introducing the effects of latent heat and the full thermal fields into the weakly-nonlinear analysis of the pulsatile mode. They found that thermal effects promote supercritical bifurcation and give regions in parameter space where either there is a sharp wavenumber selection of the fundamental mode or there are no stable two-dimensional solutions. Novick-Cohen (1987) analyzed a model closely related to that of Corriell & Sekerka (1983), identified a large-morphological-number limit, and derived a Kuramoto-Sivashinsky equation that governs the evolution of steady cells.

When the pulsatile mode (zero critical wavenumber) is present, the flat interface has a speed that is modulated in time. This would result in what Merchant & Davis (1990)
called solute bands (Fig. 1a), concentration distributions periodic in the pulling direction and free of lateral segregation. If the oscillatory instability has nonzero critical wavenumber but a much larger scale than the cells, there will be weak lateral segregation with corresponding microstructure approximately represented by solute bands. These solute bands are distinct from the bands (Fig. 1b) seen in experiment, structures periodic in the pulling direction in which layers free of lateral segregation alternate with cellular, dendritic, or eutectic phases. Boettinger et al. (1984) observed two-dimensional banded microstructures using Ag-Cu alloys for a range of pulling speeds. At still higher pulling speeds, these structures disappeared. There was also a concentration observed below which no bands appeared for the range of pulling speeds. Gremaud et al. (1990) and Zimmermann et al. (1991) observed similar banded structures in Al-Fe and Al-Cu, respectively, using laser surface-treatment experiments. Hoglund et al. (1991) did not find banded structures in their experiments on Si-Sn. A summary of materials that have exhibited a banding structure at high pulling speeds is given in Kurz & Trivedi (1990).

Several theories have attempted to describe these banded structures. Merchant & Davis (1990) conjectured that the bands observed in the solid are a result of an interaction of the oscillatory and cellular instabilities. Thus, they should occur only where the two instabilities coexist. Braun et al. (1992) looked at the weakly-nonlinear coupling of the pulsatile and cellular modes from Merchant and Davis in the absence of latent heat. Though bands were not found, they did define mathematical conditions under which a banded microstructure could appear. Carrard et al. (1992) have developed a phenomenological theory in the absence of latent heat to describe banding by comparing the undercoolings of dendritic and planar fronts. In their formulation, the solidification front is assumed to undergo jump transitions in speed as a result of quasi-steady changes in undercooling. Certain constants are chosen so as to give the best fit with experimental data on the transition speeds from dendrites to bands and bands to microsegregation-free structure. This theory gives good agreement with certain of the observations but, for example, does not give a cutoff in concentration below
which bands are impossible. Karma & Sarkissian (1993) extended this work by including full thermal effects.

In the present work we use a weakly-nonlinear analysis to couple together the cellular and oscillatory modes described in the linear stability analysis of Huntley & Davis (1993a). In physical scales, this analysis is applicable to the region near the intersection of the cellular and oscillatory branches of the neutral stability curve, i.e. the codimension-2 point. For the physical systems which exhibit bands (Kurz & Trivedi 1990), this intersection region corresponds to a nonzero critical wavenumber for the oscillatory instability. Therefore we shall consider the general case of coupling steady cells to an oscillatory mode with nonzero-wavenumber, and apply the results to several physical systems. The resulting coupled Landau equations (CLE) govern the behavior of the disturbance amplitudes. Note that within this analysis we recover the weakly-nonlinear behavior of the cellular instability (set the amplitude of the oscillatory instability to zero), the weakly-nonlinear behavior of the oscillatory instability for traveling waves and standing waves (set the amplitude of the cellular instability to zero), the weakly-nonlinear behavior of the pulsatile instability (set the amplitude of the cellular instability and the critical wavenumber of the oscillatory instability to zero), and the case of coupling together cells and pulsations (set the critical wavenumber of the oscillatory instability to zero). A bifurcation analysis is applied to determine if the amplitude equations exhibit structure characteristic of banding behavior. Therefore we shall check the possibility that bands are due to the \emph{weakly-nonlinear coupling} of cells and oscillations. The alternating layers of structure present in bands may be suggestive of relaxation-oscillation behavior for the standing-wave instability. We also determine whether bands are due to the weakly-nonlinear behavior of the oscillatory instability (nonzero critical wavenumber) alone.

2. Formulation

We consider a dilute binary alloy of mean concentration $C_\infty$ contained between two closely spaced parallel plates. The temperature is fixed at each end of the apparatus such that a
solid/liquid interface forms in the interior. The apparatus is then pulled at constant speed $V$ through the laboratory-fixed temperature gradient (Fig. 2). In this manner the liquid mixture is continuously solidified at the constant rate $V$. For purposes of simplification, we study the case of uni-directional solidification in which the apparatus is assumed to be infinitely long and the geometry is two-dimensional; many of the results obtained apply to the three-dimensional case as well.

We shall apply the assumption of no flow, and so neglect the effects of gravity and volume change due to solidification. In doing so, the bulk equations are heat and solute diffusion in the liquid and solid. For many metallic systems, the solutal diffusivity in the solid is several orders of magnitude smaller than that in the liquid. Therefore we shall study the one-sided model in which the effect of solute diffusion in the solid is neglected.

We specify conditions on the temperature, temperature gradient, and the solute concentration far away from the solid/liquid interface. Far from the interface in the liquid, the concentration approaches the mean value $C_\infty$ and the temperature approaches its laboratory-fixed value. The far-field condition in the solid specifies the gradient of the temperature. By specifying the temperature at one end of the apparatus and the temperature gradient at the other end (Huntley & Davis 1993b), we obtain a natural specification that uniquely determines the various orders of problems in the weakly-nonlinear analysis.

At the solid/liquid interface, we specify conditions which are thermodynamically consistent with the nonequilibrium effects associated with large solidification rates. A full discussion of this formulation can be found in Huntley & Davis (1993a); we will summarize the results. Firstly, there is continuity of temperature. Secondly, there is a jump in the concentration of the secondary component due to its different solubilities in the solid and liquid phases. This jump is represented by the velocity-dependent segregation coefficient, $k(u^*)$, which is assumed less than unity so that solute is rejected by the solid. We choose the form for the segregation coefficient proposed by Aziz (1982) and Jackson et al. (1980) and experimentally tested by Aziz et al. (1986). The next two conditions are conservation
of heat, which relates the jump in the temperature gradients to the latent heat release, and conservation of solute. Finally, there is a modified Gibbs-Thomson equation which includes a velocity-dependent liquidus slope, \( m(v_n^*) \), and a linear law of attachment kinetics. By using thermodynamic arguments for a planar interface (\( v_n^* = V \)), Boettinger & Perepezko (1985) and Boettinger & Coriell (1986) derived the appropriate form for the temperature at the solid/liquid interface and, hence, the liquidus slope.

Coriell & Sekerka (1983) examined the same physical problem of rapid directional solidification and developed a model that allows for more general forms of disequilibrium effects than does the one we are using. The conditions formulated by Coriell and Sekerka are identical to the ones used in this work when the functionals are restricted appropriately (Huntley & Davis 1993a).

3. Basic State

Nondimensionalize the system using spatial, \( D/V \), and temporal, \( D/V^2 \), scales based on solute diffusion. The scaled equations which result govern the solute concentration fields \( C^L(x, z, t) \) and \( C^S(x, z, t) \), the thermal fields \( T^L(x, z, t) \) and \( T^S(x, z, t) \), and the interface deflection \( h(x, t) \) pulled at constant speed \( V \):

\[
\begin{align*}
\text{Liquid in } z > h(x, t): \\
\nabla^2 C^L + C_z^L - C_t^L &= 0, \\
\nabla^2 T^L + D_L(T_z^L - T_t^L) &= 0,
\end{align*}
\]

(3.1a, 3.1b)

\[
\begin{align*}
\text{Solid in } z < h(x, t): \\
C_z^S - C_t^S &= 0, \\
\nabla^2 T^S + D_S(T_z^S - T_t^S) &= 0,
\end{align*}
\]

(3.1c, 3.1d)

\[
\begin{align*}
\text{Interface at } z = h(x, t): \\
T^S &= T^L.
\end{align*}
\]

(3.1e)
\[ C^S + \frac{k_E}{k_E - 1} = \left( C^L + \frac{1}{k_E - 1} \right) k(v_n), \]  
(3.1f)
\[ \mathcal{L}(1 + h_t) = n(T^S_z - h_x T^S_x) - (T^L_z - h_x T^L_x), \]  
(3.1g)
\[ (1 + h_t) \left[ (k_E - 1) C^L + 1 \right] \left( \frac{k(v_n) - 1}{k_E - 1} \right) = C^L_z - h_x C^L_z, \]  
(3.1h)
\[ M^{-1} T^L = \tilde{m}(v_n) C^L + \frac{\tilde{m}(v_n) - 1}{k_E - 1} - \mu v_n + \frac{\Gamma h_{xx}}{(1 + h_x^2)^{3/2}}, \]  
(3.1i)
and

**Far-field conditions:**

\[ C^L = 1 \quad \text{as} \quad z \to \infty, \]  
(3.1j)
\[ T^L = T_H \quad \text{as} \quad z \to \infty, \]  
(3.1k)
\[ T^S_z = G_S \exp(-D_S z) \quad \text{as} \quad z \to -\infty. \]  
(3.1l)

Here the interfacial velocity \( v_n \) and segregation coefficient \( k(v_n) \) are given by

\[ v_n = \frac{1 + h_t}{\sqrt{1 + h_x^2}}, \quad k(v_n) = \frac{k_E + \beta v_n}{1 + \beta v_n}, \]  
(3.2a)

and the liquidus slope \( \tilde{m}(v_n) \) is given by

\[ \tilde{m}(v_n) = 1 - \frac{1}{(k_E - 1)} \left[ k_E - k(v_n) \right] \left[ 1 - \ln(k(v_n)/k_E) \right]. \]  
(3.2b)

The familiar non-dimensional parameters appearing in system (3.1) are the equilibrium segregation coefficient \( k_E \), the morphological number \( M \), and the surface energy \( \Gamma \),

\[ M = \frac{m_E(k_E - 1)C_{\infty}V}{DGk_E}, \quad \Gamma = \frac{T_M \gamma Vk_E}{L D m_E(k_E - 1)C_{\infty}}. \]  
(3.3)

Two parameters arise from the nonequilibrium formulation of the BAJ-model. They are the attachment-kinetics parameter \( \mu \) and the disequilibrium parameter \( \beta \),

\[ \mu = \frac{Vk_E}{(k_E - 1)^2 C_{\infty}V_0}, \quad \beta = \beta_0 V. \]  
(3.4)

When \( \mu = 0 \), there is infinitely rapid interface kinetics; when \( \beta = 0 \), there is equilibrium segregation. Five parameters arise because of the inclusion of thermal effects. They are the latent heat parameter \( \mathcal{L} \), the ratio of thermal conductivities \( n \), the scaled basic state thermal...
gradient on the liquid side of the planar interface \( G_L \), and the ratios of solutal to thermal diffusivities \( \mathcal{D}_L \) and \( \mathcal{D}_S \):

\[
\mathcal{L} = \frac{LV}{k_L G}, \quad n = \frac{k_S}{k_L}, \quad G_L = \frac{G_L^*}{G}, \quad \mathcal{D}_L = \frac{D}{D_{th}^{(L)}}, \quad \text{and} \quad \mathcal{D}_S = \frac{D}{D_{th}^{(S)}}.
\] (3.5)

The physical constants which appear in the non-dimensional parameters are defined as follows: \( T_M \) is the melting temperature of the pure material, \( \gamma/L \) is the capillarity constant, \( m_E \) is the equilibrium slope of the liquidus, \( L \) is the latent heat release, \( V_0 \) is the upper bound for the rate at which crystallization can occur, \( \beta_0 \) is the measure of departure from thermodynamic equilibrium, \( D \) is the solutal diffusivity in the liquid, \( D_{th}^{(L)} \) (\( D_{th}^{(S)} \)) is the thermal diffusivity of the liquid (solid), \( k_L \) (\( k_S \)) is the thermal conductivity of the liquid (solid), \( C_\infty \) is the mean concentration of the solute, \( V \) is the constant mean pulling speed, \( G_L \) is the thermal gradient on the liquid side of the planar interface, and \( G \) is the averaged thermal gradient at the planar interface.

System (3.1) possesses a steady-state solution for a planar interface, given by

\[
\Phi(x, z, t) = \Phi(z),
\] (3.6a)

where

\[
\Phi(x, z, t) = \begin{bmatrix} C^L(x, z, t) \\ C^S(x, z, t) \\ T^L(x, z, t) \\ T^S(x, z, t) \\ h(x, t) \end{bmatrix}, \quad \Phi(z) = \begin{bmatrix} 1 - \frac{k_E}{(k_E + \beta)} \exp(-z) \\ 0 \\ T_0 + \left( \frac{G_L}{\mathcal{D}_L} \right) [1 - \exp(-\mathcal{D}_L z)] \\ T_0 + \left( \frac{\mathcal{L} + G_L}{n \mathcal{D}_S} \right) [1 - \exp(-\mathcal{D}_S z)] \\ 0 \end{bmatrix}.
\] (3.6b)

Here the planar interfacial temperature \( T_0 \) is given by

\[
T_0 = -M \mu + \frac{M}{(k_L - 1)} \left[ \frac{\bar{m}(1)}{k_1} - 1 \right].
\] (3.6c)

This steady-state solution is used as the basic-state solution in the stability analysis to follow.

At low pulling speeds, \( \beta \ll 1 \), the basic state concentration profile reduces to the exponential form in Mullins & Sekerka (1964), although the temperature at the interface may be quite low due to the presence of attachment kinetics. For high pulling speeds, \( \beta \gg 1 \),
the concentration profile loses its exponential character and approaches a uniform value of unity everywhere, consistent with the absence of segregation. For $\mathcal{D}_S, \mathcal{D}_L \ll 1$ and $\mathcal{L} = 0$, we recover the solution consistent with the linear temperature profile used in Merchant & Davis (1990).

4. Weakly–Nonlinear Analysis

We wish to investigate the onset of the time-oscillatory and the cellular disturbances from the planar basic state. We simplify this problem by assuming equal thermal conductivities, $n = 1$, and equal thermal diffusivities, $\mathcal{D}_S = \mathcal{D}_L = \mathcal{D}$. Then we disturb the basic state $\Phi$,

$$\Phi(x, z, t) = \Phi(z) + \Phi'(x, z, t),$$

and obtain the nonlinear disturbance equations for $\Phi'$.

4.1 Linear Theory

The linearized version of (4.1) has been analyzed in Huntley & Davis (1993a). They wrote the small disturbances to the basic state as

$$\Phi'(x, z, t) = \hat{\Phi}(z) \exp(\sigma t + iax),$$

where $\sigma$ is the complex growth rate and $a$ is the spatial wavenumber. They proceeded much as did Mullins & Sekerka (1964), linearizing the governing system about the basic state, to obtain the following characteristic equation that determines the eigenvalues $\sigma$, viz.

$$(2\mathcal{R}_S + \mathcal{D}) \left\{ k_E \left( \frac{1 + \frac{\sigma}{k_E + \beta}}{1 + \beta} \right) \hat{\mu}(1) + \left( R + \overline{k} - 1 \right) \left[ \frac{G_L}{M} + \Gamma a^2 - \hat{\mu}(1) \frac{k_E}{k_E + \beta} + \frac{\mu}{k_E + \beta} \frac{\sigma \ln(\overline{k}/k_E)}{(k_E + \beta)(k_E - 1)(1 + \beta)} \right] \right\} + \frac{\mathcal{L}}{M} \left( R + \overline{k} - 1 \right) \left( \sigma + \mathcal{R}_S + \mathcal{D} \right) = 0,$$

where

$$\overline{k} = k(1) = \frac{k_E + \beta}{1 + \beta}, \quad \hat{\mu}(1) = 1 - \frac{1}{k_E - 1} \left[ k_E - \overline{k} \left( 1 - \ln(\overline{k}/k_E) \right) \right],$$

$$R = \frac{1}{2}, \quad \mathcal{R}_S = -\frac{1}{2} \mathcal{D} + \frac{1}{2} \sqrt{\mathcal{D}^2 + 4 \sigma \mathcal{D} + 4 a^2}.$$
The planar interface is unstable to infinitesimal disturbances if any \( \text{Re} (\sigma) > 0 \) and stable if every \( \sigma \) satisfies \( \text{Re} (\sigma) < 0 \). Neutral or marginal conditions are determined by setting \( \text{Re} (\sigma) = 0 \). This characteristic equation admits two types of neutrally stable modes: (i) steady cellular disturbances, \( \text{Re} (\sigma) = \text{Im} (\sigma) = 0 \), and (ii) oscillatory disturbances, \( \text{Re} (\sigma) = 0 \) and \( \text{Im} (\sigma) \neq 0 \). Note that using the additional assumption that the latent heat vanishes, \( \mathcal{L} = 0 \), recovers the characteristic equation of Merchant & Davis (1990).

The two neutral stability modes from the linear theory, one cellular and one oscillatory, are shown in Fig. 3. Several results from the linear stability analysis of Huntley & Davis (1993a) are worth noting in order to motivate the subsequent nonlinear analysis. In Fig. 3, the oscillatory and steady cellular curves have absolute minima at \( a_{oc} \) and \( a_c \), respectively. Although not shown in this figure, there are also regions in the physical parameter space where the minima of the curves occur at their point of intersection/termination. We refer to these as endpoint minima; the weakly-nonlinear stability analysis in Section 4.2 cannot be applied to such cases. Note that in addition to the endpoint minima, there are also regions in the physical parameter space where the minimum of the oscillatory curve is at \( a_{oc} = 0 \). Huntley & Davis (1993b) studied the weakly-nonlinear behavior of this pulsatile mode alone. For the present analysis, we shall take the more general case of \( a_{oc} \neq 0 \).

In Fig. 3, the range of unstable wavenumbers for the steady cellular curve is bounded by \( a = 0 \) and the short-wavelength cutoff \( a = a_s \approx 0.25 \). Huntley & Davis (1993a) show that for \( a > a_s \), surface energy stabilizes all disturbances; therefore an absolute-stability boundary exists due to the presence of surface energy. As \( \Gamma \to \Gamma_s^- , a_s \to 0 \) and the minimum of the curve goes to infinity; this is the absolute-stability condition as modified by nonequilibrium effects. From this we see that the surface energy can be used a tuning parameter to adjust the onset of the cellular instability. Choose \( \Gamma = \Gamma_0 \) such that the cellular and oscillatory instabilities simultaneously onset at the value of the bifurcation parameter \( M = M_0 \) (Fig. 3). We shall take advantage of this fact in the subsequent analysis.
4.2 Weakly-Nonlinear Theory

In order to examine the nonlinear selection we wish to analyze the system near the simultaneous onset of the oscillatory and cellular modes, i.e. the codimension-2 point. For generality, we assume that the critical wavenumber of the oscillatory branch is nonzero. We define a small parameter $\epsilon$ by the nearness of $\Gamma$ to $\Gamma_0$,

$$\Gamma = \Gamma_0 + \epsilon^2 \chi,$$  \hspace{.5cm} (4.4)

where $\chi = \pm 1$, depending on whether the initial instability to the planar solid/liquid interface is oscillating or steady cellular. We then take $M$ to be near its critical value,

$$M = M_0 + \epsilon^2 M_2 + \cdots.$$  \hspace{.5cm} (4.5)

All other parameters are taken $O(1)$. A multi-time analysis (Kevorkian & Cole 1981) is employed using the scales suggested by the characteristic equation. We write

$$T = \omega_0 t,$$ \hspace{.5cm} (4.6a)

$$\tau = \epsilon^2 t,$$ \hspace{.5cm} (4.6b)

where $\omega_0$ is the unit-order Hopf frequency, and the following expansions of the solutions:

$$C(x, z, T, \tau; \epsilon) = \epsilon C_1(x, z, T, \tau) + \epsilon^2 C_2(x, z, T, \tau) + \cdots,$$ \hspace{.5cm} (4.7a)

$$T^L(x, z, T, \tau; \epsilon) = \epsilon T^L_1(x, z, T, \tau) + \epsilon^2 T^L_2(x, z, T, \tau) + \cdots,$$ \hspace{.5cm} (4.7b)

$$T^S(x, z, T, \tau; \epsilon) = \epsilon T^S_1(x, z, T, \tau) + \epsilon^2 T^S_2(x, z, T, \tau) + \cdots,$$ \hspace{.5cm} (4.7c)

$$h(x, T, \tau; \epsilon) = \epsilon h_1(x, T, \tau) + \epsilon^2 h_2(x, T, \tau) + \cdots.$$ \hspace{.5cm} (4.7d)

We ignore $C^S$ in this analysis because it is not needed for the weakly-nonlinear description of the interface; we are using the one-sided model approximation. To simplify notation, we drop the superscript for the concentration in the liquid and simply use $C$.

We do not consider the slow-spatial modulation because at this point we are interested in determining the temporal behavior needed to produce the banded microstructures, not
the details of this microstructure. It would be straightforward to include this slow-space behavior later if warranted.

Note that this analysis is valid when the difference between the thresholds of the oscillatory and cellular instabilities is small compared to the difference between their critical wavenumbers. Therefore we cannot apply these results to the region around the point of intersection of these instabilities (Fig. 3, \( a \approx 0.085 \)). Near such a point, we would need to do an analysis of the double eigenvalue to get an amplitude equation which is second order in time; see, for example, Schöpf & Zimmerman (1993).

The two time scales alter the nonlinear disturbance equations in the usual way and, after substituting the expansions, we obtain a sequence of linear problems. At each order \( \varepsilon^j \), the system of linear problems to solve is given by

\[
\mathcal{L} u_j = R_j^u, \quad (4.8a)
\]

\[
B u_j = R_j^b \quad \text{at} \quad z = 0, \quad (4.8b)
\]

\[
C_j \to 0, \quad T_j^L \to 0 \quad \text{as} \quad z \to \infty, \quad (4.8c)
\]

\[
T_{jz}^S \to 0 \quad \text{as} \quad z \to -\infty, \quad (4.8d)
\]

where

\[
u_j \equiv \begin{bmatrix} C_j & T_j^L & T_j^S \\ T_j^S - T_j^L + \mathcal{L} h_j & T_{jz}^L - T_{jz}^S + \mathcal{L} \omega_0 h_{jT} + \mathcal{L} h_j \\ \omega_0 C_j - (\tilde{k} - 1) C_j - \frac{k_E}{1+\beta} \left( h_j + \frac{\omega_0 h_{jT}}{k_E+\beta} \right) & \tilde{m}(1) C_j + \omega_0 h_{jT} \left[ \mu - \frac{\beta k_E \ln(k_E)}{(1+\beta)(k_E-1)(k_E+\beta)} \right] + \frac{\tilde{m}(1) k_E}{k_E+\beta} \right] - \Gamma_0 h_{jxx} \end{bmatrix}
\]

\[
\mathcal{L} u_j = \begin{bmatrix} C_{jzz} + C_{jzx} + C_{jz} - \omega_0 C_{jT} \\ T_{jzz}^L + T_{jxx}^L + \mathcal{D} T_{jz}^L - \mathcal{D} \omega_0 T_{jz}^L \\ T_{jzz}^S + T_{jxx}^S + \mathcal{D} T_{jz}^S - \mathcal{D} \omega_0 T_{jz}^S \end{bmatrix}, \quad (4.8e)
\]

\[
B u_j = \begin{bmatrix} T_{jz}^L - T_{jz}^S + \mathcal{L} \omega_0 h_{jT} + \mathcal{L} h_j \\ \omega_0 C_j - (\tilde{k} - 1) C_j - \frac{k_E}{1+\beta} \left( h_j + \frac{\omega_0 h_{jT}}{k_E+\beta} \right) & \tilde{m}(1) C_j + \omega_0 h_{jT} \left[ \mu - \frac{\beta k_E \ln(k_E)}{(1+\beta)(k_E-1)(k_E+\beta)} \right] + \frac{\tilde{m}(1) k_E}{k_E+\beta} \right] - \Gamma_0 h_{jxx} \end{bmatrix}
\]

\[
(4.8f)
\]
\[
\mathbf{R}_j^u = \begin{bmatrix}
  r_{1j}^u \\
  r_{2j}^u \\
  r_{3j}^u \\
  r_{4j}^u
\end{bmatrix}, \quad \text{and} \quad \mathbf{R}_j^b = \begin{bmatrix}
  r_{1j}^b \\
  r_{2j}^b \\
  r_{3j}^b \\
  r_{4j}^b
\end{bmatrix}.
\] (4.8g)

Here \( \mathbf{L} \mathbf{u}_j \) is the linearized operator with the first two rows operating on \( z > 0 \) and the third row operating on \( z < 0 \).

At \( O(\varepsilon) \), we recover the linear-theory problem under neutral stability conditions,

\[
\mathbf{R}_1^u = 0 \quad \text{and} \quad \mathbf{R}_1^b = 0. \tag{4.9}
\]

Since we are on the neutral curve, the general solution is of the form:

\[
\begin{aligned}
C_1 &= c_{11} B_1^L(\tau) \exp \left[ i \left( T + a_{oc} x \right) + \alpha_1 z \right] + c_{11} B_1^R(\tau) \exp \left[ i \left( T - a_{oc} x \right) + \alpha_1 z \right] + \\
T_1^L &= d_{11} B_1^L(\tau) \exp \left[ i \left( T + a_{oc} x \right) + \gamma_1 z \right] + d_{11} B_1^R(\tau) \exp \left[ i \left( T - a_{oc} x \right) + \gamma_1 z \right] + \\
T_1^S &= e_{11} B_1^L(\tau) \exp \left[ i \left( T + a_{oc} x \right) + \delta_1 z \right] + e_{11} B_1^R(\tau) \exp \left[ i \left( T - a_{oc} x \right) + \delta_1 z \right] + \\
h_1 &= B_1^L(\tau) \exp \left[ i \left( T + a_{oc} x \right) \right] + B_1^R(\tau) \exp \left[ i \left( T - a_{oc} x \right) \right] + \\
&\quad F_1(\tau) \exp \left[ i a_{oc} x \right] + c.c.,
\end{aligned}
\] (4.10)

where \( a_{oc} \) and \( a_c \) are the critical wavenumbers of the oscillatory and cellular branches, respectively. Algebraic expressions for the amplitude coefficients and the exponential coefficients can be found in Huntley (1993). We include right-traveling waves \( (B_1^R \neq 0) \) for this solution because of the reflection symmetry in \( x \) for the characteristic equation. We only plot the neutral stability curves for positive wavenumbers (Fig. 3), although there also is the corresponding curve for negative wavenumbers.

At \( O(\varepsilon^2) \), we obtain

\[
\mathbf{R}_2^u = 0 \quad \text{and} \quad \mathbf{R}_2^b = \begin{bmatrix}
  r_{12} \\
  r_{22} \\
  r_{32} \\
  r_{42}
\end{bmatrix}^T.
\] (4.11)

The forcing terms \( r_{ij} \) are displayed in Appendix A. The solution to this problem consists of twelve different modes and their complex conjugates. We simplify the calculation by making
use of the symmetries present in this system. In particular, we obtain the needed information by considering two simpler problems (as outlined in Schöpf & Zimmerman 1993): (i) the coupled problem between left-traveling waves and cells \((B^L_1 = 0)\), and (ii) the standing wave problem \((F_1 = 0, \quad B^L_1 = B^R_1 \equiv B_1)\).

For case (i), the solution to the \(O(\varepsilon^2)\) problem is of the form

\[
\begin{cases}
C_2 = c_{20}|B_1|^2 \exp(-z) + c_{21}|F_1|^2 \exp(-z) + c_{11} B_1^L(\tau) \exp[i(T + a_{oc}x) + \alpha_1 z] + \\
c_{12} F_1(\tau) \exp[i a_{c} x + \eta_1 z] + c_{22} B_1^L \exp[2i(T + a_{oc}x) + \alpha_2 z] + \\
c_{23} B_1^L \exp[2ia_{c} x + \eta_2 z] + c_{24} B_1^L F_1 \exp[i(T + (a_c + a_{oc}) x) + \phi_4 z] + \\
c_{25} B_1^L F_1^* \exp[i(T - (a_c - a_{oc}) x) + \phi_- z] + c.c.,
\end{cases}
\]

\[
T_2^L = d_{20}|B_1|^2 \exp(-Dz) + d_{21}|F_1|^2 \exp(-Dz) + d_{11} B_1^L(\tau) \exp[i(T + a_{oc}x) + \gamma_1 z] + \\
d_{12} F_1(\tau) \exp[i a_{c} x + \xi_1 z] + d_{22} B_1^L \exp[2i(T + a_{oc}x) + \gamma_2 z] + \\
d_{23} B_1^L \exp[2ia_{c} x + \xi_2 z] + d_{24} B_1^L F_1 \exp[i(T + (a_c + a_{oc}) x) + \Delta_+ z] + \\
d_{25} B_1^L F_1^* \exp[i(T - (a_c - a_{oc}) x) + \Delta_- z] + c.c.,
\]

\[
T_2^S = e_{11} B_1^H(\tau) \exp[i(T + a_{oc}x) + \delta_1 z] + e_{12} F_2(\tau) \exp[i a_{c} x + \zeta_1 z] + \\
e_{22} B_1^H \exp[2i(T + a_{oc}x) + \delta_2 z] + e_{23} F_2 \exp[2ia_{c} x + \zeta_2 z] + \\
e_{24} B_1^H F_1 \exp[i(T + (a_c + a_{oc}) x) + \theta_+ z] + \\
e_{25} B_1^H F_1^* \exp[i(T - (a_c - a_{oc}) x) + \theta_- z] + c.c.,
\]

\[
h_2 = l_{20}|B_1|^2 + l_{21}|F_1|^2 + B_1^L(\tau) \exp[i(T + a_{oc}x)] + F_2(\tau) \exp[i a_{c} x] + \\
l_{22} B_1^L \exp[2i(T + a_{oc}x)] + l_{23} F_2 \exp[2ia_{c} x] + \\
l_{24} B_1^L F_1 \exp[i(T + (a_c + a_{oc}) x)] + l_{25} B_1^L F_1^* \exp[i(T - (a_c - a_{oc}) x)] + c.c.
\]

(4.12)

The algebraic expressions for the amplitude coefficients and exponential coefficients can be found in Huntley (1993). For case (ii), the solution to the \(O(\varepsilon^2)\) problem is of the form

\[
\begin{cases}
C_2 = c_{20}|B_1|^2 \exp(-z) + c_{20}|B_1|^2 \exp[2iT + p_1 z] + c_{22} B_1^2 \exp[2i(T + a_{oc}x) + \alpha_2 z] + \\
c_{27} B_1^2 \exp[2ia_{oc}x + q_1 z] + c.c.,
\end{cases}
\]

\[
T_2^L = d_{20}|B_1|^2 \exp(-Dz) + d_{26}|B_1|^2 \exp[2iT + p_- z] + \\
d_{22} B_1^2 \exp[2i(T + a_{oc}x) + \gamma_2 z] + d_{27} B_1^2 \exp[2ia_{oc}x + r_- z] + c.c.,
\]

\[
T_2^S = e_{26}|B_1|^2 \exp[2iT + p_+ z] + e_{22} B_1^2 \exp[2i(T + a_{oc}x) + \delta_2 z] + \\
e_{27} B_1^2 \exp[2ia_{oc}x + r_+ z] + c.c.,
\]

\[
h_2 = l_{20}|B_1|^2 + l_{26}|B_1|^2 \exp[2iT] + l_{22} B_1^2 \exp[2i(T + a_{oc}x)] + \\
l_{27} B_1^2 \exp[2ia_{oc}x] + c.c.
\]

(4.13)
Again the algebraic expressions for the amplitude coefficients and exponential coefficients can also be found in Huntley (1993). We do not retain terms proportional to the exponential mode \([2i(-T + a_ox)]\) for the standing wave solution because it is not needed to determine the coefficients of the coupled amplitude equations.

At \(O(\varepsilon^3)\), we obtain

\[
\mathbf{R}_3^a = \begin{bmatrix} C_{1\tau} \\ \mathcal{D} T_{L\tau}^L \\ \mathcal{D} T_{S\tau}^L \end{bmatrix} \quad \text{and} \quad \mathbf{R}_3^b = \begin{bmatrix} r_{13} \\ r_{23} \\ r_{33} \\ r_{43} \end{bmatrix}.
\]

(4.14)

The forcing terms \(r_{i3}\) are displayed in Appendix A. We do not need to solve for the particular solution at this order to get a restriction on the fundamental amplitudes \(B_{1L}, B_{1R}, \) and \(F_1\).

The solution to this problem with homogeneous boundary conditions is given by

\[
\begin{align*}
C_3 &= \left( B_{C3}^L + \frac{c_{11} z B_{Lz}^L}{1 + 2\alpha_1} \right) \exp[i(T + a_ox) + \alpha_1 z] + \\
B_{C3}^R + \frac{c_{12} z F_{1z}^L}{1 + 2\eta_1} \exp[ia_c x + \eta_1 z] + c.c.
\end{align*}
\]

\[
T_3^L = \left( B_{L3}^L + \frac{d_{12} z DB_{Lz}^L}{\mathcal{D} + 2\gamma_1} \right) \exp[i(T + a_ox) + \gamma_1 z] + \\
B_{R3}^L + \frac{d_{12} z DB_{Rz}^L}{\mathcal{D} + 2\gamma_1} \exp[i(T - a_ox) + \gamma_1 z] + \\
F_{L3} + \frac{d_{12} z DF_{1z}^L}{\mathcal{D} + 2\eta_1} \exp[ia_c x + \xi_1 z] + c.c.
\]

\[
T_3^S = \left( B_{S3}^L + \frac{e_{11} z DB_{Lz}^L}{\mathcal{D} + 2\delta_1} \right) \exp[i(T + a_ox) + \delta_1 z] + \\
B_{S3}^R + \frac{e_{11} z DB_{Rz}^L}{\mathcal{D} + 2\delta_1} \exp[i(T - a_ox) + \delta_1 z] + \\
F_{S3} + \frac{e_{11} z DF_{1z}^L}{\mathcal{D} + 2\xi_1} \exp[ia_c x + \zeta_1 z] + c.c.
\]

(4.15)

\[
h_3 = B_3^L \exp[i(T + a_ox)] + B_3^R \exp[i(T - a_ox)] + F_3 \exp[ia_c x] + c.c.
\]

The terms above proportional to \(z \exp[i(T + a_ox)], z \exp[i(T - a_ox)],\) and \(z \exp[ia_c x]\) are due to the forcing terms \(\mathbf{R}_3^a\). We substitute (4.15) into the interfacial conditions (4.8b), make use of the linear independence of the modes, and apply the solvability condition in Appendix B (the Fredholm Alternative) to this system of equations to find a restriction on
the slow-time-dependent amplitudes $B^L_1(\tau)$, $B^R_1(\tau)$, and $F_1(\tau)$. They must satisfy coupled Landau equations (CLE).

5. Coupled Amplitude Equations

The CLE govern the evolution of the amplitude of the interface on the slow scales, and can be represented as follows:

\[
\frac{d B^L_1}{d\tau} = \left( \sigma_0 M_2 - \sigma_1 \chi - g_1 |B^L_1|^2 - g_2 |F_1|^2 - g_5 |B^R_1|^2 \right) B^L_1, \tag{5.1a}
\]

\[
\frac{d B^R_1}{d\tau} = \left( \sigma_0 M_2 - \sigma_1 \chi - g_5 |B^L_1|^2 - g_2 |F_1|^2 - g_1 |B^R_1|^2 \right) B^R_1, \tag{5.1b}
\]

\[
\frac{d F_1}{d\tau} = \left( \sigma_2 M_2 - \sigma_3 \chi - g_3 |F_1|^2 - g_4 |B^L_1|^2 - g_4^* |B^R_1|^2 \right) F_1. \tag{5.1c}
\]

The asterisk denotes complex conjugation. The algebraic expressions for the coefficients are given in Huntley (1993). Here $\sigma_0, \sigma_1, g_1, g_2, g_4,$ and $g_5$ are complex constants and $\sigma_2, \sigma_3,$ and $g_3$ are real constants.

We now represent the amplitudes in terms of modulus and phase via

\[
F_1 = |g_3|^{-1/2} r_1(\tau)e^{i\theta(\tau)}, \quad B^L_1 = |g_1|^{-1/2} r_2(\tau)e^{i\theta_2(\tau)}, \quad \text{and} \quad B^R_1 = |g_1|^{-1/2} r_3(\tau)e^{i\theta_3(\tau)} \tag{5.2}
\]

to get the real equations:

\[
\frac{d r_1}{d\tau} = \left[ \gamma_1 - a_{11} r_1^2 - a_{12} (r_2^2 + r_3^2) \right] r_1, \tag{5.3a}
\]

\[
\frac{d r_2}{d\tau} = \left[ \gamma_2 - a_{21} r_1^2 - a_{22} r_2^2 - a_{23} r_3^2 \right] r_2, \tag{5.3b}
\]

\[
\frac{d r_3}{d\tau} = \left[ \gamma_2 - a_{21} r_1^2 - a_{23} r_2^2 - a_{22} r_3^2 \right] r_3, \tag{5.3c}
\]

\[
\frac{d \theta}{d\tau} = \sigma_{01} M_2 - \sigma_1 \chi - g_{11} L^2 - g_{21} C^2 - g_{51} R^2, \tag{5.3d}
\]

\[
\frac{d \phi}{d\tau} = \sigma_{01} M_2 - \sigma_1 \chi - g_{51} L^2 - g_{21} C^2 - g_{11} R^2, \tag{5.3e}
\]

\[
\frac{d \phi}{d\tau} = -g_{41}(L^2 - R^2). \tag{5.3f}
\]

where
\[
C = r_1 |g_3|^{-1/2}, \quad L = r_2 |g_{1R}|^{-1/2}, \quad R = r_3 |g_{1R}|^{-1/2}, \quad (5.3g)
\]
\[
\gamma_1 = \sigma_2 M_2 - \sigma_3 \chi, \quad \gamma_2 = \sigma_0 R M_2 - \sigma_1 R \chi, \quad (5.3h)
\]
\[
a_{11} = \frac{g_3}{|g_3|} = \pm 1, \quad a_{21} = \frac{g_{2R}}{|g_3|}, \quad (5.3i)
\]
\[
a_{12} = \frac{g_{1R}}{|g_{1R}|}, \quad a_{22} = \frac{g_{1R}}{|g_{1R}|} = \pm 1, \quad a_{31} = \frac{g_{5R}}{|g_{1R}|}. \quad (5.3j)
\]

The subscripts \( R \) and \( I \) denote the real and imaginary parts of the complex coefficients, respectively.

We shall concentrate on the behavior of the amplitudes of the oscillatory and cellular disturbances; note \( r_1 \) corresponds to cells, \( r_2 \) corresponds to left-traveling waves, and \( r_3 \) corresponds to right-traveling waves. Such equations have been analyzed by a number of coworkers, so we shall rely on their results whenever possible.

### 5.1 Bifurcation Structure Near the Codimension-2 Point

The steady state solutions of the amplitude equations in (5.3) are as follows:

1.) planar interface: \( r_1 = r_2 = r_3 = 0 \), \( (5.4a) \)

2.) pure cells: \( r_1^2 = \frac{\gamma_1}{a_{11}}, \quad r_2 = r_3 = 0 \), \( (5.4b) \)

3.) pure left-traveling wave: \( r_2^2 = \frac{\gamma_2}{a_{22}}, \quad r_1 = r_3 = 0 \), \( (5.4c) \)

4.) pure right-traveling wave: \( r_3^2 = \frac{\gamma_2}{a_{22}}, \quad r_1 = r_2 = 0 \), \( (5.4d) \)

5.) pure standing wave: \( r_2^2 = r_3^2 = \frac{\gamma_2}{a_{22} + a_{31}}, \quad r_1 = 0 \), \( (5.4e) \)

6.) mixed mode with cells and left-traveling wave:
\[
\begin{align*}
      r_1^2 & = \left( \frac{a_{22} \gamma_1 - a_{12} \gamma_2}{a_{11} a_{22} - a_{12} a_{21}} \right), \quad r_2^2 = \left( \frac{a_{11} \gamma_2 - a_{21} \gamma_1}{a_{11} a_{22} - a_{12} a_{21}} \right), \quad r_3 = 0, \quad (5.4f)
\end{align*}
\]

7.) mixed mode with cells and right-traveling wave:
\[
\begin{align*}
      r_1^2 & = \left( \frac{a_{22} \gamma_1 - a_{12} \gamma_2}{a_{11} a_{22} - a_{12} a_{21}} \right), \quad r_2^2 = \left( \frac{a_{11} \gamma_2 - a_{21} \gamma_1}{a_{11} a_{22} - a_{12} a_{21}} \right), \quad r_2 = 0, \quad (5.4g)
\end{align*}
\]

8.) mixed mode with cells and standing wave:
\[
\begin{align*}
      r_1^2 & = \left( \frac{a_{22} + a_{31}}{a_{22} + a_{31}} \right) \gamma_1 - 2 a_{12} \gamma_2, \quad r_2^2 = r_3^2 = \left( \frac{a_{11} \gamma_2 - a_{21} \gamma_1}{a_{22} + a_{31} a_{11} - 2 a_{12} a_{21}} \right). \quad (5.4h)
\end{align*}
\]

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These solutions exist only when the right-hand sides of the terms are non-negative. We perform a linear stability analysis for each of these steady-state solutions in order to find the potential bifurcation points.

**Case 1**

The planar interface solution always exists and is linearly stable if

\[
\sigma_2 (M_2 - M_{2C}) < 0 \quad \text{and} \quad \sigma_0R (M_2 - M_{2C}) < 0, \quad (5.5a)
\]

where \( M_{2C} = \frac{\sigma_3 \chi}{\sigma_2}, \quad M_{2O} = \frac{\sigma_1 R \chi}{\sigma_0 R}. \quad (5.5b) \)

The values \( M_{2C} \) and \( M_{2O} \) are the bifurcation points from the planar state to the pure-mode cells and oscillations (standing or traveling), respectively.

**Case 2**

The pure-cell mode exists only when \( r_1^2 = \sigma_2(M_2 - M_{2C})/a_{11} > 0 \), and is linearly stable if

\[
a_{11} > 0 \quad \text{and} \quad \left( \sigma_0R - \frac{a_{21}}{a_{11}} \sigma_2 \right) (M_2 - M_{2CM}) < 0, \quad (5.6a)
\]

where \( M_{2CM} = \frac{(a_{21} \sigma_3 - a_{11} \sigma_1 R)}{(a_{21} \sigma_2 - a_{11} \sigma_0 R)} \chi. \quad (5.6b) \)

The value \( M_{2CM} \) is the possible bifurcation point from pure cells to one of the mixed-mode solutions.

**Cases 3 and 4**

The pure left-traveling mode exists only when \( r_2^2 = \sigma_0R(M_2 - M_{2O})/a_{22} > 0 \). For the case of the pure right-traveling mode, we get the same result substituting \( r_3 \) for \( r_2 \). Each of these modes is linearly stable if

\[
a_{22} > 0, \quad a_{31} > a_{22}, \quad \text{and} \quad \left( \sigma_2 - \frac{a_{12}}{a_{22}} \sigma_0R \right) (M_2 - M_{2OM}) < 0, \quad (5.7a)
\]

where \( M_{2OM} = \frac{(a_{22} \sigma_3 - a_{12} \sigma_1 R)}{(a_{22} \sigma_2 - a_{12} \sigma_0 R)} \chi. \quad (5.7b) \)
The value $M_{2OM}$ is the possible bifurcation point from the pure traveling wave to the mixed-mode solution which couples cells and traveling waves.

**Case 5**

The standing wave exists only when $r_2^2 = r_3^2 = \sigma_0 (M_2 - M_{20}) / (a_{22} + a_{31}) > 0$, and is linearly stable if

\[
a_{31} < a_{22}, \quad a_{31} + a_{22} > 0, \quad \text{and} \quad \left( \sigma_2 - \frac{2a_{12}}{a_{22} + a_{31}} \sigma_{0R} \right) (M_2 - M_{2SM}) < 0, \quad (5.8a)
\]

where

\[
M_{2SM} = \left( \frac{(a_{22} + a_{31})\sigma_3 - 2a_{12}\sigma_{1x}}{(a_{22} + a_{31})\sigma_2 - 2a_{12}\sigma_{0R}} \right) \chi. \quad (5.8b)
\]

The value $M_{2SM}$ is the possible bifurcation point from the pure standing wave to the mixed-mode solution which couples cells and standing waves. Note that it is not possible to have both stable standing and traveling waves.

**Cases 6 and 7**

The mixed-mode solution which couples cells and left-traveling waves exists only when

\[
r_1^2 = \left( \frac{a_{22}\sigma_2 - a_{12}\sigma_{0R}}{a_{11}a_{22} - a_{12}a_{21}} \right) (M_2 - M_{2OM}) > 0 \quad \text{and} \quad (5.9a)
\]

\[
r_2^2 = \left( \frac{a_{31}\sigma_{0R} - a_{21}\sigma_1}{a_{11}a_{22} - a_{12}a_{21}} \right) (M_2 - M_{2CM}) > 0. \quad (5.9b)
\]

For the case of the mixed-mode solution which couples cells and right-traveling waves, we get the same result with $r_2$ and $r_3$ switched. Each of these mixed modes is linearly stable if

\[
a_{11}a_{22} - a_{12}a_{21} > 0, \quad (a_{22} - a_{31})(a_{11}\sigma_{0R} - a_{21}\sigma_2)(M_2 - M_{2CM}) < 0 \quad \text{and} \quad (5.10a)
\]

\[
[a_{22}(a_{11} - a_{21})\sigma_2 - a_{11}(a_{12} - a_{22})\sigma_{0R}] (M_2 - M_{2TH}) > 0,
\]

where

\[
M_{2TH} = \left[ \sigma_3 - \frac{a_{11}}{a_{22}} \left( \frac{a_{12} - a_{22}}{a_{11} - a_{21}} \right) \sigma_{1R} \right] \left[ \sigma_2 - \frac{a_{11}}{a_{22}} \left( \frac{a_{12} - a_{22}}{a_{11} - a_{21}} \right) \sigma_{0R} \right]^{-1} \chi. \quad (5.10b)
\]

The value $M_{2MTH}$ is a possible Hopf bifurcation point from the mixed-mode solution between cells and traveling waves. Necessary conditions for this Hopf bifurcation point to exist are
$a_{11}a_{22} - a_{12}a_{21} > 0$ and $a_{12}a_{21} < 0$.

Case 8

The mixed-mode solution which couples cells and standing waves exists only when

\begin{align}
    r_1^2 &= \left( \frac{(a_{22} + a_{31})\sigma_2 - 2a_{12}\sigma_{0R}}{(a_{22} + a_{31})a_{11} - 2a_{12}a_{21}} \right) (M_2 - M_{2SM}) > 0 \quad \text{and} \\
    r_2^2 &= r_3^2 = \left( \frac{a_{11}\sigma_{0R} - a_{21}\sigma_2}{(a_{22} + a_{31})a_{11} - 2a_{12}a_{21}} \right) (M_2 - M_{2CM}) > 0.
\end{align}

(5.11a)  (5.11b)

This mixed-mode solution is stable if

\begin{align}
    a_{31} - a_{22} < 0, \quad a_{11}(a_{22} + a_{31}) - 2a_{12}a_{21} > 0, \quad \text{and} \\
    [(a_{22} + a_{31})(a_{11} - a_{21})\sigma_2 + a_{11}(a_{22} + a_{31} - 2a_{12})\sigma_{0R}] (M_2 - M_{2SH}) > 0,
\end{align}

(5.12a)  (5.12b)

where

\begin{equation}
    M_{2SH} = \left[ \sigma_3 + \frac{a_{11}(a_{22} + a_{31} - 2a_{12})}{(a_{11} - a_{21})(a_{22} + a_{31})}\sigma_{1R} \right] \left[ \sigma_2 + \frac{a_{11}(a_{22} + a_{31} - 2a_{12})}{(a_{11} - a_{21})(a_{22} + a_{31})}\sigma_{0R} \right]^{-1} \chi.
\end{equation}

(5.12c)

The value $M_{2MSH}$ is a possible Hopf bifurcation point from the mixed-mode solution between cells and standing waves. Necessary conditions for this Hopf bifurcation point to exist are $a_{11}(a_{22} + a_{31}) - 2a_{12}a_{21} > 0$ and $a_{12}a_{21} < 0$.

5.2 Bifurcation Structure Away from the Codimension-2 Point

The CLE (5.1) were derived by performing a weakly-nonlinear analysis in the neighborhood of the simultaneous onset of the cellular and oscillatory instabilities (Fig. 3). We also wish to study the bifurcation behavior of the two modes of instability away from this region, i.e. when the two modes of instability are not coupled together. To study the bifurcation behavior of the cellular instability away from this coupled region, we set in the CLE (5.1)

\begin{equation}
    B_1 F_1 = B_1 \dot{F}_1 = 0, \quad \chi = 0, \quad \text{and} \quad M_2 = \pm 1.
\end{equation}

(5.13)

This then gives us the amplitude equation

\begin{equation}
    \frac{dF_1}{d\tau} = (\sigma_2 M_2 - g_3|F_1|^2) F_1.
\end{equation}

(5.14)
The term $\Gamma_0$ still appears in the amplitude-equation coefficients, but now we no longer require that it corresponds to that at the codimension-2 point. The bifurcation structure for this amplitude equation can then be summarized as follows: For $g_3/\sigma_2 > 0$, the bifurcating solution is supercritical ($M_2 = 1$) and stable, so there will be a smooth transition to cells. For $g_3/\sigma_2 < 0$, the bifurcating solution is subcritical ($M_2 = -1$) and unstable, so there will be a jump transition to a (possibly) large amplitude state.

To study the bifurcation behavior of the oscillatory instability away from the coupled region, we set in the CLE (5.1)

$$F_1 = 0, \quad \chi = 0, \quad \text{and} \quad M_2 = \pm 1. \quad (5.15)$$

This then gives us the coupled amplitude equations

$$\begin{align*}
\frac{dB^L_L}{d\tau} &= \left(\sigma_0 M_2 - g_1 |B^L_I|^2 - g_3 |B^R_I|^2\right) B^L_I, \quad (5.16a) \\
\frac{dB^R_R}{d\tau} &= \left(\sigma_0 M_2 - g_3 |B^L_I|^2 - g_1 |B^R_I|^2\right) B^R_I. \quad (5.16b)
\end{align*}$$

Huntley & Davis (1993b) studied the bifurcation behavior of the oscillatory instability with zero critical wavenumber, which reduces this problem to a single amplitude equation describing the pulsations. When we include a nonzero critical wavenumber, we get either standing-wave or traveling-wave bifurcated solutions. To describe the bifurcation-type, it is easier to look at the scaled form of the problem (5.3) with $r_1 = 0$ and $\gamma_2 = \sigma_{0R} M_2$. The bifurcation theory results can then be summarized as follows: For $a_{22}/\sigma_{0R} < 0$, the traveling-wave solution is subcritical ($M_2 = -1$) and unstable. For $(a_{22} + a_{31})/\sigma_{0R} < 0$, the standing-wave solution is subcritical ($M_2 = -1$) and unstable. For $a_{22}/\sigma_{0R} > 0$, the traveling-wave solution is supercritical ($M_2 = 1$) and stable for $a_{22} > 0$ and $a_{31} > a_{22}$. For $(a_{22} + a_{31})/\sigma_{0R} > 0$, the standing-wave solution is supercritical ($M_2 = 1$) and stable for $a_{22} > \pm |a_{31}|$. Thus the traveling wave is stable, or the standing wave is stable, or neither is stable. We cannot have both modes simultaneously stable.

We now apply the results from the bifurcation theory to several physical systems.
<table>
<thead>
<tr>
<th>parameter</th>
<th>units</th>
<th>Ag-Cu</th>
<th>Al-Cu</th>
<th>Al-Fe</th>
<th>Al-Sn</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k_E$</td>
<td></td>
<td>0.44</td>
<td>0.10</td>
<td>0.03</td>
<td>0.0001</td>
</tr>
<tr>
<td>$T_{M\gamma/L} \times 10^7$</td>
<td>(Km)</td>
<td>1.53</td>
<td>1.</td>
<td>1.</td>
<td>2.05</td>
</tr>
<tr>
<td>$D \times 10^9$</td>
<td>(m$^2$/s)</td>
<td>1.</td>
<td>5.0</td>
<td>1.7</td>
<td>7.</td>
</tr>
<tr>
<td>$V_0$</td>
<td>(m/s at frac)</td>
<td>1000.</td>
<td>1000.</td>
<td>2000.</td>
<td>3000.</td>
</tr>
<tr>
<td>$\beta_0$</td>
<td>(m/s)</td>
<td>5.</td>
<td>5.5</td>
<td>1.7</td>
<td>13.</td>
</tr>
<tr>
<td>$m_E$</td>
<td>(K/at %)</td>
<td>-2.8</td>
<td>-5.37</td>
<td>-7.29117</td>
<td>-6.8</td>
</tr>
<tr>
<td>$G \times 10^{-4}$</td>
<td>(K/m)</td>
<td>2.</td>
<td>500.</td>
<td>500.</td>
<td>4.</td>
</tr>
<tr>
<td>$L \times 10^{-8}$</td>
<td>(J/m$^3$)</td>
<td>9.7*</td>
<td>9.5*</td>
<td>9.5*</td>
<td>10.7</td>
</tr>
<tr>
<td>$D_{th}^L \times 10^5$</td>
<td>(m$^2$/s)</td>
<td>13.0*</td>
<td>3.7*</td>
<td>3.7*</td>
<td>3.6</td>
</tr>
<tr>
<td>$D_{th}^S \times 10^5$</td>
<td>(m$^2$/s)</td>
<td>17.4*</td>
<td>7.0*</td>
<td>7.0*</td>
<td>7.1</td>
</tr>
<tr>
<td>$k_L$</td>
<td>(J/sKm)</td>
<td>374.*</td>
<td>95.*</td>
<td>95.*</td>
<td>93.</td>
</tr>
<tr>
<td>$k_S$</td>
<td>(J/sKm)</td>
<td>429.*</td>
<td>210.*</td>
<td>210.*</td>
<td>213.</td>
</tr>
</tbody>
</table>

Table 1: Physical constants for each of the systems considered. The quantities with asterisks attached correspond to values for the pure substrate (no dilute binary component).

6. Physical Systems

In this section, we present our results for the Ag-Cu, Al-Fe, Al-Cu, and Al-Sn systems; these systems are chosen because all have the codimension-2 point corresponding to the intersection of the oscillatory and cellular modes of instability (Figs. 4-7). Note that this intersection point corresponds to a nonzero critical wavenumber for the oscillatory instability. Table 1 contains a listing of the physical constants used for each system.

We first consider the bifurcation-type of these systems away from the neighborhood of the codimension-2 point. The weakly-nonlinear analysis used to obtain the bifurcation results is not valid for the case where the critical value is an endpoint minimum, as discussed in Section 4.1. Therefore the bifurcation analysis only applies to the segments of the neutral
stability curves which correspond to absolute minima.

We plot the results for the Ag-Cu system in Fig. 4. We denote the transition points by the targets. We see that the bifurcation type for the cellular branch is subcritical for lower pulling speeds $V$ and supercritical for higher pulling speeds. The oscillatory branch has a supercritical bifurcation with the traveling-wave solution stable for the lower portion of the curve and a subcritical bifurcation for the upper portion of the curve.

We plot the results for the Al-Cu system in Fig. 5. There are no transition points for this system. The bifurcation-type for the cellular branch is subcritical for all pulling speeds that correspond to an absolute minimum. The oscillatory branch has a supercritical bifurcation with stable traveling waves.

We plot the results for the Al-Fe system in Fig. 6. There are two transition points for this system. The bifurcation-type for the cellular branch is subcritical below the transition point and supercritical above. The bifurcation is supercritical with the traveling-wave solution stable above the transition point and subcritical below this point. Note that the transition point at the nose of the oscillatory branch of the neutral stability curve only applies to the oscillations.

We plot the results for the Al-Sn system in Fig. 7. The cellular instability has a transition point above (below) which the bifurcation is supercritical (subcritical). The oscillatory instability has a supercritical bifurcation to stable traveling-wave solutions for the region which corresponds to an absolute minimum. Huntley & Davis (1993b) performed the analysis for the region with zero critical wavenumber and found that the bifurcation is subcritical.

We now consider the bifurcation structure in the neighborhood of the intersection point. At each of these codimension-2 points, we get a neutral stability curve in the nondimensional scales that has the same general features as Fig. 3. Table 2 contains the linear-theory nondimensional parameters corresponding to the codimension-2 point of each system. With these values for the parameters, we next calculate the coefficients that appear in the CLE (5.3). Table 3 contains the computed values for the linear-theory coefficients given in Table 2.
<table>
<thead>
<tr>
<th></th>
<th>Ag-Cu</th>
<th>Al-Cu</th>
<th>Al-Fe</th>
<th>Al-Sn</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k_E$</td>
<td>0.44</td>
<td>0.1</td>
<td>0.03</td>
<td>$1.0 \times 10^{-4}$</td>
</tr>
<tr>
<td>$\beta$</td>
<td>$1.176 \times 10^{-2}$</td>
<td>0.1025</td>
<td>$8.877 \times 10^{-2}$</td>
<td>$8.21 \times 10^{-2}$</td>
</tr>
<tr>
<td>$\mu$</td>
<td>$6.804 \times 10^{-4}$</td>
<td>$5.892 \times 10^{-2}$</td>
<td>$9.828 \times 10^{-3}$</td>
<td>$1.45 \times 10^{-4}$</td>
</tr>
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<td>$L$</td>
<td>7.111</td>
<td>0.7025</td>
<td>0.1880</td>
<td>187.</td>
</tr>
<tr>
<td>$D$</td>
<td>$6.579 \times 10^{-6}$</td>
<td>$9.346 \times 10^{-5}$</td>
<td>$3.178 \times 10^{-5}$</td>
<td>$1.31 \times 10^{-4}$</td>
</tr>
<tr>
<td>$\Gamma_0$</td>
<td>2.082</td>
<td>1.975</td>
<td>1.538</td>
<td>$1.87 \times 10^{-2}$</td>
</tr>
<tr>
<td>$M_0$</td>
<td>$1.270 \times 10^{4}$</td>
<td>128.8</td>
<td>102.4</td>
<td>$6.37 \times 10^{6}$</td>
</tr>
<tr>
<td>$a_c$</td>
<td>0.0855</td>
<td>0.170</td>
<td>0.162</td>
<td>0.0865</td>
</tr>
<tr>
<td>$a_{oc}$</td>
<td>0.0251</td>
<td>0.044</td>
<td>0.0293</td>
<td>0.0291</td>
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<tr>
<td>$\omega_0$</td>
<td>0.0116</td>
<td>0.0514</td>
<td>0.0584</td>
<td>0.0173</td>
</tr>
</tbody>
</table>

Table 2: Linear-theory parameters that coalesce the onset of the oscillatory and cellular modes for each of the physical systems under consideration.

Then it is simply a matter of applying the bifurcation-theory results from Section 5.1 to each system. We used Doedel’s (1981) bifurcation package AUTO to test our analytical results.

For the weakly-nonlinear analysis, we defined the small parameter $\varepsilon$ by the nearness of $\Gamma$ to $\Gamma_0$, the value of the surface energy parameter which gives the simultaneous onset of the cellular and oscillatory modes of instability. For larger values of $\Gamma$, the minimum of the cellular branch is postponed to larger values of the bifurcation parameter $M$. Thus for each of the bifurcation diagrams, oscillations are the first mode of instability for $\chi = 1$ and cells are the first mode of instability for $\chi = -1$. We then take $M$ to be near its critical value $M_0$, where $M_2$ is the measure of departure from criticality. For $M_2$ small enough, the planar state is stable to infinitesimal perturbations.

Representative bifurcation diagrams are displayed in Figs. 8–10. We first consider the Ag-Cu and Al-Sn systems, both which have the same qualitative bifurcation structure (Fig. 8). For $\chi = 1$, the planar interface loses stability against oscillations first: the traveling-wave mode remains stable in the region of analysis while the standing-wave mode is unstable.
<table>
<thead>
<tr>
<th></th>
<th>Ag-Cu</th>
<th>Al-Cu</th>
<th>Al-Fe</th>
<th>Al-Sn</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{0R}$</td>
<td>$2.26 \times 10^{-7}$</td>
<td>$1.55 \times 10^{-4}$</td>
<td>$2.47 \times 10^{-4}$</td>
<td>$1.47 \times 10^{-9}$</td>
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<tr>
<td>$\sigma_{1R}$</td>
<td>$6.00 \times 10^{-4}$</td>
<td>$2.02 \times 10^{-3}$</td>
<td>$1.61 \times 10^{-3}$</td>
<td>0.161</td>
</tr>
<tr>
<td>$\sigma_{2}$</td>
<td>$5.65 \times 10^{-7}$</td>
<td>$2.61 \times 10^{-4}$</td>
<td>$3.09 \times 10^{-4}$</td>
<td>$1.58 \times 10^{-9}$</td>
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<tr>
<td>$\sigma_{3}$</td>
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<td>0.0925</td>
<td>0.0777</td>
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</tr>
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<td>$a_{11}$</td>
<td>+1</td>
<td>-1</td>
<td>+1</td>
<td>+1</td>
</tr>
<tr>
<td>$a_{21}$</td>
<td>4.104</td>
<td>15.84</td>
<td>13.99</td>
<td>2.347</td>
</tr>
<tr>
<td>$a_{12}$</td>
<td>63.98</td>
<td>66.04</td>
<td>81.46</td>
<td>13.23</td>
</tr>
<tr>
<td>$a_{22}$</td>
<td>+1</td>
<td>+1</td>
<td>-1</td>
<td>+1</td>
</tr>
<tr>
<td>$a_{31}$</td>
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<td>$1.167 \times 10^{3}$</td>
<td>$1.302 \times 10^{3}$</td>
<td>$5.429 \times 10^{2}$</td>
</tr>
</tbody>
</table>

Table 3: Computed values of the coefficients of the amplitude equations for the linear-theory values given in Table 2.

An unstable mixed-mode comprised of cells and traveling waves (MT-mode) branches from and stabilizes the unstable cellular mode for larger values of $M_2$. There is also an unstable mixed-mode comprised of cells and standing waves (MS-mode) which has a range of existence bounded by the cellular mode and the standing-wave mode. Note that there is a region for $M_2$ large where both the pure traveling waves and pure cells are stable. For $\chi = -1$, the planar interface loses stability against cells first. For larger values of $M_2$, the unstable MT-mode branches from and stabilizes the traveling-wave mode. The standing-wave mode remains unstable. Note that there is a region for $M_2$ large where both the pure traveling waves and pure cells are stable.

We next consider the Al-Cu system (Fig. 9). For $\chi = 1$, the planar interface loses stability against oscillations first: the traveling-wave mode remains stable in the region of analysis while the standing-wave mode is unstable. The unstable MT-mode and MS-mode both branch from the unstable cellular mode. The MS-mode terminates upon intersection with the standing-wave mode. For $\chi = -1$, the planar interface loses stability against unstable
cells first. For larger values of $M_2$, the unstable MT-mode branches from and stabilizes the traveling-wave mode. The standing-wave mode remains unstable.

We finally consider the Al-Fe system (Fig. 10). For $\chi = 1$, the planar interface loses stability against oscillations first: both the traveling-wave and standing-wave modes are unstable. The unstable MT-mode and MS-mode both branch from and stabilize the unstable cellular mode. The MS-mode terminates upon intersection with the standing-wave mode. For $\chi = -1$, the planar interface loses stability against stable cells first. For larger values of $M_2$, the unstable MT-mode branches from the unstable traveling-wave mode. The standing-wave mode remains unstable.

For each of the physical systems under consideration, we find that the pure standing-wave solution is unstable. Therefore with the present analysis, one cannot describe the bands observed in Ag-Cu, Al-Cu, and Al-Fe by investigating the nonlinear development of standing waves. In addition, we find that neither of the four systems has the stable mixed-mode solution of cells and traveling waves or cells and standing waves. There are no Hopf bifurcations from the mixed-mode solutions. Such a bifurcation would modulate the amplitude of the cells and oscillations in a way reminiscent of the banded structure observed in materials. Therefore with the present analysis, one cannot describe bands observed in Ag-Cu, Al-Cu, and Al-Fe by coupling together the steady cells and the oscillations.

In Figs. 4–6, we plot the zero-wavenumber oscillatory instability though it is not preferred. Karma & Sarkissian (1993) numerically found that this mode gives relaxation-oscillation behavior. The linear-theory position of this mode as compared to the data locating bands looks very promising. If the oscillatory instability has a supercritical bifurcation at the critical nonzero wavenumber, the zero-wavenumber mode may be the dominant mode driving the banding instability. We initially considered studying the weakly-nonlinear coupling between this zero-wavenumber mode and the cellular instability, but realized that the region of simultaneous onset occurs at an endpoint minimum for the cellular instability for all the systems; the present weakly-nonlinear theory does not apply to this case.
7. Discussion

In this paper we have studied the nonlinear dynamics of the rapidly solidifying interface for a dilute binary alloy. We have performed a weakly-nonlinear analysis near the simultaneous onset of the oscillatory and steady cellular instabilities (Fig. 3). This nonlinear analysis results in coupled Landau equations (5.3) with complex coefficients. The CLE govern the evolution of the amplitude of the fundamental modes of instability to the planar solid/liquid interface. These modes correspond to the cellular instability, left-traveling waves, and right-traveling waves. We investigated the rich variety of nonlinear behaviors of these cellular and oscillatory modes near the onset of instability.

We first considered the bifurcation-type of the oscillatory and cellular modes away from the codimension-2 point (Figs. 4–7). We found that standing waves are never stable; the traveling waves are stable for certain regions of the neutral stability curve. Therefore with the present analysis we cannot describe the bands observed in Ag-Cu, Al-Cu, and Al-Fe by the nonlinear development of standing waves. We also determined that the cellular mode is subcritical for low-enough pulling speeds. We must restrict ourselves to those areas of the neutral stability curve which do not give an endpoint minimum in order for this analysis to be valid.

We next considered the bifurcation-type of the oscillatory and cellular modes near their point of intersection. The type of bifurcation near this codimension-2 point was numerically determined for the Ag-Cu, Al-Cu, Al-Fe, and Al-Sn systems (Figs. 8–10). A variety of structures occurred, corresponding to the competition between the planar state, pure cellular mode, pure traveling-wave solutions, pure standing-wave solutions, mixed-mode solutions with cells and traveling waves, and mixed-mode solutions with cells and standing waves. There were a number of subcritical bifurcations present. Noticeably absent from our analysis are stable mixed-mode solutions. A Hopf bifurcation from such a solution would produce the type of behavior suggestive of the bands observed in various rapidly solidified alloys.
We tried to apply our theory to the nonlinear coupling of cells with the zero-wavenumber mode of the oscillatory instability. The coexistence sector corresponding to these two modes of instability corresponds quite well with the experimental data on bands. Karma & Sarkissian (1993) numerically found that the pulsatile mode gives relaxation-oscillation behavior. If the oscillatory instability has a supercritical bifurcation at the critical nonzero wavenumber, this zero-wavenumber mode may be the dominant mode driving the banding instability. Because the simultaneous onset of cells and pulsations occurs at an endpoint minimum for the cellular instability for all the systems, our weakly-nonlinear results do not apply to this region.

Bands have not been found in the weakly-nonlinear regime near the point of intersection of the neutral curves for Ag-Cu, Al-Cu, Al-Fe, and Al-Sn alloys. There are several possibilities for this result. Firstly, higher-order saturation terms may be needed in the CLE (5.3) to capture the turnaround of the curves because of the large number of subcritical bifurcations in the coupled region. Secondly, bands may not occur in a small neighborhood of the simultaneous onset of the oscillatory and steady cellular instabilities (the region where our weakly-nonlinear analysis is valid). Other regions of interest which are not covered by this work but are amenable to analysis are the intersection point between the cellular and oscillatory instabilities (Fig. 3), the endpoint minima regions, and the high-velocity branch of the oscillatory instability (Fig. 4). Finally, the model employed in this analysis may not contain the necessary physics to describe bands; in particular, buoyancy-driven fluid flow may be important when large amounts of latent heat are released in rapid solidification.

Acknowledgements

The authors would like to acknowledge H. Riecke for his insights on the nonlinear analysis and R.J. Braun and D.M. Anderson for several helpful discussions. This work was supported by grants from the National Aeronautics and Space Administration through the Graduate Student Researchers Program (DAH) and the Program on Microgravity Science and Appli-
cations (SHD), and from the National Science Foundation through the NSF Mathematical Sciences Postdoctoral Research Fellowship (DAH).

References


Appendix A. Forcing Terms

The forcings for the $O(\varepsilon^2)$ system are

\begin{align}
R_{12}^b &= (T_{1z}^L - T_{1z}^S) h_1 + \frac{1}{2} \mathcal{L} D h_1^2, \quad (A1a)
R_{22}^b &= - (T_{1zz}^L - T_{1zz}^S) h_1 + \frac{1}{2} \mathcal{L} D^2 h_1^2 + (T_{1x}^L - T_{1x}^S) h_{1x}, \quad (A1b)
R_{32}^b &= - C_{1zz} h_1 - \frac{k_E}{2(1 + \beta)} h_1^2 + \frac{k_E (k_E - 1) \omega_0}{(k_E + \beta)(1 + \beta)^2} h_{1T}^2 + \frac{\beta \omega_0 \omega_0}{(1 + \beta)^2} h_{1z}^2 + \frac{\beta \omega_0^{2k_E}}{(1 + \beta)^2} h_{1T}^2 + \frac{\beta \omega_0^{2k_E}}{(1 + \beta)^2} h_{1z}^2, \quad (A1c)
R_{42}^b &= \frac{- \beta k_E \omega_0}{(1 + \beta)^2} \frac{\ln(k_E/k_E)}{(k_E + \beta)(1 + \beta)^2} h_{1T}^2 + \frac{\beta \omega_0}{(1 + \beta)^2} \ln(k_E/k_E) C_{1z} h_{1T} + \frac{\beta k_E \omega_0}{(1 + \beta)^2} \frac{\ln(k_E/k_E)}{(k_E + \beta)(1 + \beta)^2} h_{1x}^2 + \frac{k_E \omega_0}{(1 + \beta)^2} \frac{\ln(k_E/k_E)}{(k_E + \beta)(1 + \beta)^2} h_{1T}^2 + \frac{k_E \omega_0}{(1 + \beta)^2} \frac{\ln(k_E/k_E)}{(k_E + \beta)(1 + \beta)^2} h_{1z}^2, \quad (A1d)
\end{align}

The forcings for the $O(\varepsilon^3)$ system are

\begin{align}
R_{13}^b &= (T_{2z}^L - T_{2z}^S) h_1 + \frac{1}{2} \left( T_{1zz}^L - T_{1zz}^S \right) h_1^2 + \left( T_{1x}^L - T_{1x}^S \right) h_{1x} + \mathcal{L} D \left( -\frac{1}{6} D h_1^3 + h_1 h_2 + h_{1T}^2 \right), \quad (A2a)
R_{23}^b &= - \mathcal{L} h_{1T} + \mathcal{L} D^2 \left( -\frac{1}{6} D h_1^2 + h_1 h_2 \right) + (T_{1zz}^L - T_{1zz}^S) h_2 - \frac{1}{2} \left( T_{1zz}^L - T_{1zz}^S \right) h_{1T}^2 - \left( T_{2z}^L - T_{2z}^S \right) h_1 + \left( T_{1x}^L - T_{1x}^S \right) h_{1x}, \quad (A2b)
R_{33}^b &= - \frac{1}{2} C_{1zz} h_1^2 - C_{1zz} h_2 - C_{2zz} h_1 + \left( \frac{k_E - 1}{1 + \beta} \right) \left( \frac{1}{2} C_{1zz} h_1^2 + C_{1zz} h_2 + C_{2zz} h_1 + C_{1zz} h_2 \right) + \frac{k_E}{(1 + \beta)^2} \left( h_{1T}^2 + h_{1T}^2 + \frac{1}{2} h_{1T}^2 \right) + \omega_0 (k_E - 1) \left( C_{1zz} h_1^2 + C_{1zz} h_2 + C_{2zz} h_1 + C_{1zz} h_2 \right) + \frac{k_E \omega_0 (k_E - 1)}{(k_E + \beta)^2} \left( h_{1T}^2 + h_{1T}^2 + \frac{1}{2} h_{1T}^2 \right) + \omega_0 (k_E - 1) \left( C_{1zz} h_1^2 + C_{1zz} h_2 + C_{2zz} h_1 + C_{1zz} h_2 \right) + \frac{k_E \beta^2 \omega_0}{(k_E - 1)} \left( \frac{k_E}{k_E + \beta} h_{1T}^2 + C_{1zz} h_1^2 \right) - \frac{2k_E \beta^2 \omega_0}{(k_E + \beta)(1 + \beta)^2} \left( h_{1T}^2 + h_{1T}^2 + \frac{1}{2} h_{1T}^2 \right) + \frac{k_E \beta^2 \omega_0^3}{(k_E + \beta)(1 + \beta)^2} \left( h_{1T}^2 + h_{1T}^2 + \frac{1}{2} h_{1T}^2 \right) + \frac{k_E \beta^2 \omega_0^3}{(k_E + \beta)(1 + \beta)^2} \left( h_{1T}^2 + h_{1T}^2 + \frac{1}{2} h_{1T}^2 \right) + \frac{\omega_0 k_E \beta}{(k_E + \beta)(1 + \beta)^2} \left( C_1 + \frac{k_E h_1}{k_E + \beta} \right) h_{1x}^2 + C_{2zz} h_{1x} + C_{1zz} h_{1x}, \quad (A2c)
\end{align}
\[ R_{43}^b = -\left( \mu - \frac{k_E \beta \ln(\bar{k}/k_E)}{(1 + \beta)(k_E - 1)(k_E + \beta)} \right) (h_{1r} - h_{1x} h_{2x}) + \tilde{m}(1) \left( \frac{1}{2} C_{1z} h_1^2 + C_{1z} h_2 + C_{2z} h_1 \right) + \]
\[ \frac{k_E \bar{m}(1)}{k_E + \beta} \left( \frac{1}{2} h_1^2 h_{1z} - h_1 h_{2z} \right) + \chi h_{1xx} + \frac{\beta \omega_0 \ln(\bar{k}/k_E)}{(1 + \beta)^2} \left( C_1 h_{2T} + C_2 h_{1T} + C_{1z} h_1 h_{1T} \right) - \]
\[ \frac{k_E \beta \omega_0 \ln(\bar{k}/k_E)}{(k_E + \beta)(1 + \beta)^2} \left( \frac{1}{2} h_1^2 h_{1T} - h_2 h_{1T} - h_1 h_{2T} \right) - \frac{1}{M_0} \left( \frac{1}{2} h_1^2 T_{1z}^L + h_2 T_{1z}^L + h_1 T_{2z}^L \right) - \]
\[ \frac{D G_L}{M_0} \left( \frac{1}{2} \rho h_1^3 - h_1 h_{2z} \right) + \frac{\beta^3 k_E \omega_0^3}{6(k_E - 1)(k_E + \beta)^3} \left[ \frac{6(k_E + \beta)^2 \ln(\bar{k}/k_E)}{(k_E - 1)(k_E + \beta)^3(1 + \beta)^3} \right] h_{1T}^3 - \]
\[ \frac{\omega_0^2 \beta^2}{2(1 + \beta)^3(k_E + \beta)} \left( \frac{k_E h_1}{k_E + \beta} + C_1 \right) \frac{h_{1T}^2}{h_{1T}^2 - \frac{3}{2} \Gamma_0 h_{1xx} h_{2x} + \frac{1}{2} \mu \omega_0 h_{1T} h_{1x}^2 - \]
\[ \frac{k_E \beta^2 \omega_0^2}{(k_E - 1)(1 + \beta)^2(k_E + \beta)^3} h_{1T} h_{2T} - \frac{\beta \ln(\bar{k}/k_E)}{2(1 + \beta)^2} \left( \frac{k_E h_1}{k_E + \beta} + C_1 \right) h_{1x}^2 + \]
\[ \frac{M_2}{M_0} \left( T_{1z}^L + G_L h_1 \right) + \frac{\beta \omega_0 k_E h_{1T} h_{1x}^2}{2(1 + \beta)^2(k_E - 1)(k_E + \beta)} \left[ \beta(k_E - 1) \frac{h_{1T}^2}{k_E + \beta} + (\beta - 1) \ln(\bar{k}/k_E) \right] \right. \]

(A2d)

**Appendix B. Solvability Condition**

In this derivation, we obtain an algebraic form for the solvability condition (the Fredholm Alternative). This is entirely analogous to the usual integral form in which the forcing function must be orthogonal to the null space of the adjoint problem under a suitably defined inner product. We will perform the calculation for the general case of the coexistence of the oscillatory and cellular modes.

We first substitute the general homogeneous solution,

\[
\begin{align*}
C &= B_C^C(\tau) \exp \left[ i(T + a_{oc} x) + \alpha_1 z \right] + F_C^C(\tau) \exp \left[ i a_c x + \eta_1 z \right] + c.c., \\
T^L &= B_L^L(\tau) \exp \left[ i(T + a_{oc} x) + \gamma_1 z \right] + F_L^L(\tau) \exp \left[ i a_c x + \zeta_1 z \right] + c.c., \\
T^S &= B_S^S(\tau) \exp \left[ i(T + a_{oc} x) + \delta_1 z \right] + F_S^S(\tau) \exp \left[ i a_c x + \zeta_1 z \right] + c.c., \\
h &= B(\tau) \exp \left[ i(T + a_{oc} x) \right] + F(\tau) \exp \left[ i a_c x \right] + c.c.,
\end{align*}
\]  

(B1)

into the interfacial boundary conditions \( B u_j \) (4.8f). We decompose the right-hand-side forcing terms of these boundary conditions into exponential modes and make use of their linear independence to get the \( \exp \left[ i(T + a_{oc} x) \right] \) and \( \exp \left[ i a_c x \right] \) systems of algebraic equations,
\[
\begin{align*}
B^S - B^L + \mathcal{L}B &= r_{10} \\
\gamma_i B^L - \delta_i B^S + i\omega_0 \mathcal{L}B + \mathcal{L}DB &= r_{20} \\
\alpha_i B^C - (\bar{k} - 1)B^C - \frac{k_E}{1+\beta} B - \frac{i\omega_0 k_E}{(k_E+\beta)(1+\beta)} B &= r_{30} \\
-\tilde{m}(1)B^C + \left( \frac{G_L}{M_0} - \frac{\tilde{m}(1)k_E}{k_E+\beta} \right) B + i\omega_0 \left( \mu - \frac{\beta k_E \ln(k_E/k_E)}{(1+\beta)(k_E-1)(k_E+\beta)} \right) B + \frac{1}{M_0} B^L + \Gamma_0 a^2_{oc} B &= r_{40},
\end{align*}
\] 

and
\[
\begin{align*}
F^S - F^L + \mathcal{L}F &= r_{11} \\
\xi_i F^L - \zeta_i F^S + \mathcal{L}DF &= r_{21} \\
\eta_i F^C - (\bar{k} - 1)F^C - \frac{k_E}{1+\beta} F &= r_{31} \\
-\tilde{m}(1)F^C + \left( \frac{G_L}{M_0} - \frac{\tilde{m}(1)k_E}{k_E+\beta} \right) F + \frac{1}{M_0} F^L + \Gamma_0 a^2_{oc} F &= r_{41},
\end{align*}
\]

Here \( r_{30} \) (\( r_{11} \)) represents the exponential mode \( \exp [i(T + a_{oc}x)] \) component (\( \exp [ia_{oc}x] \) component) of the forcing term \( r_{30} \) and terms due to inhomogeneities in the governing equations.

These sets of equations can be simplified down to
\[
\begin{align*}
\left\{ -\gamma_i + \delta_i \right\} \left[ \frac{k_E \tilde{m}(1)}{1+\beta} \left( 1 + \frac{i\omega_0}{k_E + \beta} \right) + \left( -\alpha_i + \bar{k} - 1 \right) \left( \frac{G_L}{M_0} + \Gamma_0 a^2_{oc} - \frac{\tilde{m}(1)k_E}{k_E + \beta} \right) \right] \\
i\omega_0 \mu - \frac{i\omega_0 k_E \beta \ln(k_E/k_E)}{(k_E + \beta)(k_E - 1)(1 + \beta)} \right] + \frac{\mathcal{L}}{M_0} \left( -\alpha_i + \bar{k} - 1 \right) (i\omega_0 + \delta_i + \mathcal{D}) \right) \bigg\} \\
\frac{B}{(-\gamma_i + \delta_i)(-\alpha_i + \bar{k} - 1)} &= r_{40} + \frac{\tilde{m}(1) r_{30}}{\alpha_i + 1 - \bar{k}} + \frac{r_{20} + \delta_i r_{10}}{M_0(\delta_i - \gamma_i)} \tag{B4}
\end{align*}
\]

and
\[
\begin{align*}
\left\{ -\xi_i + \zeta_i \right\} \left[ \frac{k_E \tilde{m}(1)}{1+\beta} + \left( -\eta_i + \bar{k} - 1 \right) \left( \frac{G_L}{M_0} + \Gamma_0 a^2_{oc} - \frac{\tilde{m}(1)k_E}{k_E + \beta} \right) \right] + \frac{\mathcal{L}}{M_0} \left( \zeta_i + \mathcal{D} \right) \bigg\}.
\right. \\
\left. \frac{F}{(-\xi_i + \zeta_i)(-\eta_i + \bar{k} - 1)} \right) &= r_{41} + \frac{\tilde{m}(1) r_{31}}{\eta_i + 1 - \bar{k}} + \frac{r_{21} + \zeta_i r_{11}}{M_0(\zeta_i - \xi_i)} \tag{B5}
\end{align*}
\]

The left-hand-sides of equations (B4) and (B5) are zero because the two terms in the curly brackets are simply the linear-theory characteristic equation evaluated at the critical mode for the oscillatory and cellular branch, respectively. Thus for solutions to exist, the following solvability conditions must be satisfied:
\[
\left( \frac{\delta_1}{M_0 (\delta_1 - \gamma_1)} \right) r_{10} + \left( \frac{1}{M_0 (\delta_1 - \gamma_1)} \right) r_{20} + \left( \frac{\tilde{m}(1)}{\alpha_1 + 1 - k} \right) r_{30} + r_{40} = 0, \quad \text{(B6a)}
\]

\[
\left( \frac{\zeta_1}{M_0 (\zeta_1 - \xi_1)} \right) r_{11} + \left( \frac{1}{M_0 (\delta_1 - \gamma_1)} \right) r_{21} + \left( \frac{\tilde{m}(1)}{\alpha_1 + 1 - k} \right) r_{31} + r_{41} = 0. \quad \text{(B6b)}
\]
Figure 1: Graphical representation of (a) solute bands and (b) bands. For solute bands the concentration of solute, represented by the degree of shading, varies periodically in the pulling direction but does not vary in the lateral direction. For bands the concentration of solute varies periodically in the pulling direction as well, but there is also lateral segregation of solute in the dark regions of the layered structure. These dark regions represent the cellular, dendritic, or eutectic structures which alternate with the lighter colored, segregation-free structures.
Figure 2: Schematic representation of the uni-directional solidification geometry. The temperature and concentration profiles $\overline{T}$ and $\overline{C}$ correspond to the steady-state problem, assuming a planar interface and strictly a $z$-dependence for the dependent variables.
Figure 3: Neutral stability curve for the Al-Cu system at the codimension-2 point $C_\infty = 0.11814$ at\% and $V = 0.56385$ m/s. The solid (dashed) curve corresponds to the steady cellular (oscillatory) instability branch. The planar interface is unstable to infinitesimal perturbations above each curve and stable below. The critical wavenumbers for the onset of the oscillatory and steady instabilities are $a_{oc} = 0.044$ and $a_c = 0.170$, respectively. The corresponding frequency for the oscillations is $\omega_0 = 0.0514$ and the codimension-2 point is given by $M_0 = 128.8$. Though not shown in this figure, the oscillatory branch intersects the M-axis at $M = 1040$. 
Figure 4: The oscillatory and steady branches of the neutral stability curve for the Ag-Cu system with the thermal gradient $G = 2 \times 10^4 \, \text{K/m}$. The leftmost (rightmost) solid curve is the steady cellular (oscillatory) branch. The short-dotted portion of these curves correspond to an endpoint minimum (zero frequency for the oscillatory branch) where our bifurcation analysis is not valid, while the long-dashed curve corresponds to the zero-wavenumber oscillatory mode. We denote the transition points for the bifurcation type by the targets. For the cellular instability, the bifurcation type is subcritical below this transition point and supercritical above. For the oscillatory instability, the bifurcation type is supercritical with the traveling-wave solution stable below the target and subcritical above. The data points are taken from Boettinger et al. (1984), with the triangles corresponding to the observed transition from dendrites to bands and the squares corresponding to the observed transition from bands to microsegregation-free structures. The vertical, dashed line denotes the concentration below which no bands were observed.
Figure 5: The oscillatory and steady branches of the neutral stability curve for the Al-Cu system with thermal gradient $G = 5 \times 10^6 K/m$. The leftmost (rightmost) solid curve is the steady cellular (oscillatory) branch. The short-dotted portion of these curves correspond to an endpoint minimum (zero frequency for the oscillatory branch) where our bifurcation analysis is not valid, while the long-dashed curve corresponds to the zero-wavenumber oscillatory mode. There exist no transition points for the bifurcation type in this system. The bifurcation is subcritical for the cellular instability, and the bifurcation type is supercritical with the traveling-wave solution stable for the oscillatory instability. The data points are taken from Zimmermann et al. (1991), with the triangles corresponding to the transition from dendrites to bands. The vertical, dashed line denotes the concentration below which no bands were observed.
Figure 6: The oscillatory and steady branches of the neutral stability curve for the Al-Fe system with thermal gradient $G = 5 \times 10^8 \, K/m$. The leftmost (rightmost) solid curve is the steady cellular (oscillatory) branch. The short-dotted portion of these curves correspond to an endpoint minimum (zero frequency for the oscillatory branch) where our bifurcation analysis is not valid, while the long-dashed curve corresponds to the zero-wavenumber oscillatory mode. There are two transition points for this system. For the cellular instability, the bifurcation is subcritical below the transition point and supercritical above. For the oscillatory instability, the bifurcation type is supercritical with the traveling-wave solution stable above the transition point and subcritical below this point. The data points are taken from Gremaud et al. (1990), with the triangles corresponding to the transition from dendrites to bands and the squares corresponding to the transition from bands to microsegregation-free structures.
Figure 7: The oscillatory and steady branches of the neutral stability curve for the Al-Sn system with thermal gradient $G = 4 \times 10^4 \text{ K/m}$. The leftmost (rightmost) solid curve is the steady cellular (oscillatory) branch. The short-dotted portion of these curves correspond to an endpoint minimum (zero frequency for the oscillatory branch) where our bifurcation analysis is not valid, while the segment denoted by asterisks has zero critical wavenumber. The oscillatory instability has a supercritical bifurcation to stable traveling waves for the absolute-minimum portion of the curve and a subcritical bifurcation for the zero critical wavenumber segment. The cellular instability has a transition point, below which the bifurcation is subcritical and above which the bifurcation is supercritical.
Figure 8: The bifurcation diagram for the Ag-Cu and Al-Sn systems. The dashed curves correspond to unstable modes while the solid curves are the stable modes. The following labels are used in this figure: C for the pure cells, TW for the pure traveling-wave mode, SW for the pure standing-wave mode, MT for the mixed mode between cells and traveling waves, and MS for the mixed mode between cells and standing waves.
Figure 9: The bifurcation diagram for the Al-Cu system. The dashed curves correspond to unstable modes while the solid curves are the stable modes. The following labels are used in this figure: C for the pure cells, TW for the pure traveling-wave mode, SW for the pure standing-wave mode, MT for the mixed mode between cells and traveling waves, and MS for the mixed mode between cells and standing waves.
Figure 10: The bifurcation diagram for the Al-Fe system. The dashed curves correspond to unstable modes while the solid curves are the stable modes. The following labels are used in this figure: C for the pure cells, TW for the pure traveling-wave mode, SW for the pure standing-wave mode, MT for the mixed mode between cells and traveling waves, and MS for the mixed mode between cells and standing waves.
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