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Abstract

The effect of a temperature dependent diffusion coefficient on the morphological stability of a binary alloy during directional solidification is treated by a linear stability analysis. The Soret effect is also included in the analysis. Specific calculations are carried out for a tin alloy containing silver for which the diffusion coefficient has a linear dependence on temperature. Although the temperature dependence of the diffusion coefficient has little effect on the critical concentration for the onset of morphological stability, it causes a significant change in the wavelength at the onset of instability.

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1 Introduction

Morphological stability during the directional solidification of a binary alloy has been intensively studied [1] since the seminal analysis of Mullins and Sekerka [2]. These analyses reveal the important role played by solute diffusion in determining interface stability during growth. In particular, diffusion determines the solute gradient in the liquid at a given growth rate and, consequently, determines the value of the constitutional supercooling. However, since the liquid temperature gradient is often high, thermodiffusion (the Soret effect) should be considered as well. The possible influence of thermodiffusion on the stability analysis was considered by a number of authors [3–5]. Hurle [3] concluded that thermodiffusion modifies significantly the wavelength and, in a less pronounced way, the critical value of the control parameters.

Hurle’s analysis was carried over with a “normal” Soret effect. In many cases, however, thermodiffusion may have a strong dependence on composition. In particular, for dilute solutions, the thermodiffusion flux is proportional to the mean concentration. This has been confirmed in tin alloys by the measurements in microgravity of Praizey et al. [6, 7]. To account for this, the following form of the solute flux \mathbf{J} can be used:

$$\mathbf{J} = -D \nabla C - A_T(C) \nabla T, \quad (1)$$

where

$$A_T(C) = D_T(\beta_0 + \beta_1 C). \quad (2)$$

Here D is the solute diffusion coefficient, C is the solute concentration, T is the temperature, D_T is the thermodiffusion coefficient, and β_0 and β_1 are constants. The nonlinear dependence of the thermodiffusion flux ($\beta_1 \neq 0$) was considered previously by Coriell et al. [4] in order to extend the analysis performed by Hurle, who studied the influence of thermodiffusion on morphological stability of planar solidification (with $\beta_1 = 0$). Indeed, for dilute solutions and for all cases where β_1/β_0 is large, the transport is of phoretic form and may exhibit unusual dynamics [4, 8]. This dependence on composition is, in the terminology of the thermodynamics of irreversible processes, a nonlinear effect [9]. To be consistent in this

respect, the diffusion coefficient is considered here as a function of the temperature. Recently measurements of the temperature dependence of the diffusivity have been performed in concentrated solutions [10, 11] and in dilute solutions [12, 13]. For dilute solutions a power law dependence is observed, while for concentrated solutions the dependence is more difficult to assess, and is system dependent [14, 15].

In dilute solutions, the dependence of the diffusion coefficient on concentration is not significant and the dependence of the thermodiffusion coefficient on temperature is on the order of $1/T$ in dilute atomic solutions [7]. The dependencies lead to contributions $[\partial D/\partial C](\nabla C)^2$ and $[\partial D_T/\partial T](\nabla T)^2$ in the species balance equation. The first case has been considered by Wollkind [16] who assumed such a dependence to be small, which allowed the use of a perturbative technique. However, exceptions to this are observed in concentrated solutions [17], which introduces additional terms in the perturbation equations. Such a dependence would therefore require a separate analysis that shall not be carried out here. In our approach we do not assume that the other dependences are small. Our interest here shall therefore be to analyze the influence of a dependence of the form

$$\mathbf{J} = -D(T)\nabla C - D_T(\beta_0 + \beta_1 C)\nabla T. \quad (3)$$

on morphological stability. In particular, our analysis addresses how the temperature dependence of the diffusion coefficient contributes to the destabilizing mechanisms.

2 Theory

We consider directional solidification of a binary alloy at constant velocity V in the z -direction and treat the morphological stability of an initially planar crystal-melt interface. We choose an (x, y, z) coordinate system (moving with the macroscopically planar interface) such that the crystal-melt interface is described by $z = h(x, y, t)$, where t is the time. We assume that there is no fluid flow in the melt, that diffusion and thermodiffusion can be neglected in the crystal, and that the thermal diffusivities in crystal and melt are sufficiently large that the thermal field can be approximated by Laplace's equation.

The temperature fields, $T_L(x, y, z)$ and $T_S(x, y, z)$, in the melt and crystal and the solute field in the melt, $C_L(x, y, z, t)$, satisfy the partial differential equations

$$\nabla^2 T_L = 0, \quad (4)$$

$$\nabla^2 T_S = 0, \quad (5)$$

and

$$\frac{\partial C_L}{\partial t} = -\nabla \cdot \mathbf{J} + V \frac{\partial C_L}{\partial z}, \quad (6)$$

We assume that the solute flux \mathbf{J} has the general form given by eq. (3). This form includes Soret diffusion, as treated previously [4, 8], and temperature dependence of the diffusion coefficient. We also assume that the other thermophysical properties are independent of temperature and solute concentration.

The boundary conditions at the crystal-melt interface are

$$(\mathbf{V} \cdot \mathbf{n})(1 - k)C_L = \mathbf{J} \cdot \mathbf{n}, \quad (7)$$

$$L_v(\mathbf{V} \cdot \mathbf{n}) = (k_S \nabla T_S - k_L \nabla T_L) \cdot \mathbf{n}, \quad (8)$$

$$T_S = T_L = T_M + mC_L - T_M \Gamma \mathcal{K}, \quad (9)$$

$$C_S = kC_L, \quad (10)$$

where \mathbf{V} is the solidification velocity vector, \mathbf{n} is the unit normal to the interface, k is the partition coefficient, L_v is the latent heat per unit volume, k_S and k_L are the thermal conductivities of crystal and melt, respectively, T_M is the melting point, m is the liquidus slope, $\Gamma = \gamma/L_v$ is the capillary length, γ is the crystal-melt surface tension, \mathcal{K} is the mean curvature of the crystal-melt interface, and C_S is the concentration in the solid. We fix the far-field boundary conditions by specifying the bulk alloy concentration C_∞ in the liquid far from the interface, and the temperature gradient in the liquid, G_L , at the planar crystal-melt interface. The steady-state solutions of the temperature equations for the planar crystal-melt

interface are

$$T_L^0(z) = T_0 + G_L z, \quad (11)$$

$$T_S^0(z) = T_0 + G_S z, \quad (12)$$

where the planar interface temperature is given by $T_0 = T_M + mC_L$, and the temperature gradients G_S and G_L are related by $L_v V = k_S G_S - k_L G_L$. The differential equation for the steady-state solute field can be integrated to obtain the first order equation,

$$-D(T) \frac{\partial C_L}{\partial z} - D_T (\beta_0 + \beta_1 C_L) \frac{\partial T}{\partial z} = V(1 - k)C_L. \quad (13)$$

We shall assume that the dependence of D on T is linear. For some tin alloys this approximation is valid over a wide range of temperature [18]. Thus we assume that:

$$D(T) = D_I + (\partial D / \partial T)(T - T_0), \quad (14)$$

where D_I the value of the diffusion coefficient at T_0 . The unperturbed solute distribution is then given by

$$C_L - C_\infty = [1/k][\xi + (\mu - k)C_\infty][1 + \alpha V z / D_I]^{-\mu/\alpha}. \quad (15)$$

where

$$\xi = D_T G_L \beta_0 / V \quad (16)$$

$$\mu = 1 + (D_T G_L \beta_1 / V) \quad (17)$$

$$\alpha = (\partial D / \partial T) G_L / V \quad (18)$$

When α tends to zero, eq. (15) tends towards

$$C_L - C_\infty = [1/k][\xi + (\mu - k)C_\infty] \exp[-\mu V z / D_I] \quad (19)$$

which is the expression given previously [4, 8]. The values of α and $\mu - 1$ will usually be of the same order of magnitude. We note that α is always positive, and, for usual values of α , the profiles given by equations (15) and (19) do not differ significantly. The interfacial liquid

solute gradient is given by:

$$G_c = -[V\mu/D_I k][\xi + (\mu - k)C_\infty] \quad (20)$$

and the solute concentration in the solid is given by:

$$C_S = \mu C_\infty + \xi \quad (21)$$

The above quantities do not depend on α . Therefore the base state seems to be affected in a negligible way by the temperature dependence of the diffusion coefficient. For the linear stability analysis of the base state, the variables are written as the superposition of the base state component and a perturbation. The perturbed quantities are Fourier analyzed in the lateral direction and exponential time-dependence is assumed, so that the variables are written as

$$\begin{pmatrix} T_L(x, y, z, t) \\ T_S(x, y, z, t) \\ C_L(x, y, z, t) \\ h(x, y, t) \end{pmatrix} = \begin{pmatrix} T_L^0(z) \\ T_S^0(z) \\ C_L^0(z) \\ 0 \end{pmatrix} + \begin{pmatrix} T_L^1(z) \\ T_S^1(z) \\ C_L^1(z) \\ \hat{h} \end{pmatrix} \exp[\sigma t + i\omega_x x + i\omega_y y], \quad (22)$$

where $\sigma = \sigma_r + i\sigma_i$ governs the time-dependence of the perturbation, ω_x and ω_y are the wavenumbers in the x and y directions, and \hat{h} is the amplitude of the perturbed interface position. The system is unstable if $\sigma_r > 0$ for any wavenumber.

The first order perturbation variables, $T_L^1(z)$, $T_S^1(z)$, and $C_L^1(z)$, satisfy the ordinary differential equations

$$\left[\frac{d^2}{dz^2} - \omega^2 \right] T_L^1 = 0, \quad (23)$$

$$\left[\frac{d^2}{dz^2} - \omega^2 \right] T_S^1 = 0, \quad (24)$$

$$\left[D \frac{d^2}{dz^2} + (V + \frac{\partial D}{\partial T} G_L + D_T \beta_1 G_L) \frac{d}{dz} - D\omega^2 - \sigma \right] C_L^1 +$$

$$\left[\left(\frac{\partial D}{\partial T} + D_T \beta_1 \right) \frac{dC_L^0}{dz} \frac{d}{dz} + \frac{\partial D}{\partial T} \frac{d^2 C_L^0}{dz^2} \right] T_L^1 = 0. \quad (25)$$

where $\omega^2 = \omega_x^2 + \omega_y^2$. The solution for the perturbed temperature field in the solid and liquid which decay at infinity are given by

$$T_S^1(z) = \hat{T}_S \exp(\omega z), \quad (26)$$

$$T_L^1(z) = \hat{T}_L \exp(-\omega z), \quad (27)$$

where \hat{T}_S and \hat{T}_L are constants.

The boundary conditions at $z = 0$ give the following relationships:

$$\hat{T}_L + G_L \hat{h} = \hat{T}_S + G_S \hat{h} = m(C_L^1 + G_c \hat{h}) - T_M \Gamma \omega^2 \hat{h}, \quad (28)$$

$$L_v \sigma \hat{h} = k_S \omega \hat{T}_S + k_L \omega \hat{T}_L, \quad (29)$$

$$\begin{aligned} \sigma(1-k)(C_S/k)\hat{h} + (1-k)V C_L^1 + (1-k)V G_c \hat{h} &= -D \frac{dC_L^1}{dz} - \frac{\partial D}{\partial T} G_c (\hat{T}_L + G_L \hat{h}) \\ -D \frac{d^2 C_L^0}{dz^2} \hat{h} + D_T (\beta_0 + \beta_1 C_S/k) \omega \hat{T}_L - D_T \beta_1 (G_c \hat{h} + C_L^1) G_L, \end{aligned} \quad (30)$$

By eliminating \hat{h} , \hat{T}_S , \hat{T}_L from these equations, we obtain a single boundary condition at $z = 0$ involving the perturbed liquid concentration.

3 Numerical Results

We have solved for the perturbed temperature and concentration fields in the liquid using the computer code SUPORT [25] with the assumption that $\sigma_i = 0$ at the onset of instability ($\sigma_r = 0$). Calculations were carried out for a tin-silver alloy for a range of solidification velocities for a temperature gradient in the liquid of 180 K/cm. The thermophysical properties used are summarized in Table I. The values of k and m are deduced from the phase diagram. The values of the diffusion coefficient and of its variations with composition were obtained by Bruson and Gerl [18] in a shear cell. Although those values may be altered by convection as studied by Garandet et al. [19], the measurements by Bruson and Gerl are

thought to provide good estimates. Thermodiffusion data are known to be very sensitive to gravitational convection [19], and it is presently necessary to rely on measurements performed in microgravity conditions. Such measurements are available only for some tin alloys [12, 13]. Values for pure tin are used for the other thermal quantities, as given in standard thermophysical tables.

The results of the numerical calculations of linear stability are shown in fig. 1 and fig. 2. We plot the critical value of C_∞ above which the interface is unstable as a function of growth velocity V in fig. 1. There are three curves which almost coincide; the solid curve is the usual Mullins and Sekerka theory with the thermodiffusion coefficient $D_T = 0$ and the temperature dependence of the diffusion coefficient $\partial D/\partial T = 0$. The dashed curve is for the actual values of the D_T and $(\partial D/\partial T)$ given in Table I with $\beta_1 = 1$ and $\beta_0 = 0$, while the dashed-dot curve sets the thermodiffusion coefficient to zero, but retains the temperature dependence of the diffusion coefficient. For the lowest velocity used in the calculations ($0.5 \mu\text{m/s}$), the critical concentrations are 0.00321, 0.00308, and 0.00322 atomic fraction silver for the solid, dashed, and dashed-dot curves, respectively. Similar results for the critical wavenumber as a function of velocity are given in fig. 2. At the lower velocities, the dashed and dashed-dot curves almost coincide, but differ significantly from the solid curve. Since the dashed-dot curve neglects the effect of thermodiffusion, we conclude that the temperature dependence of the diffusion coefficient results in the change in critical wavenumber.

4 Approximate Stability Conditions

In order to understand the preceding numerical results, particularly the change in wavenumber that results from the temperature dependence of the diffusion coefficient, we derive an analytical dispersion relation based on approximating the governing equations. The appropriate approximations were found by numerical examination of the governing equations in order to identify the dominant terms.

If we first assume that D_T is negligible, the perturbed solute equation takes the form

$$\left[D \frac{d^2}{dz^2} + \left(V + \frac{\partial D}{\partial T} G_L \right) \frac{d}{dz} - D\omega^2 - \sigma \right] C_L^1 +$$

$$\left[\frac{\partial D}{\partial T} \frac{dC_L^0}{dz} \frac{d}{dz} + \frac{\partial D}{\partial T} \frac{d^2 C_L^0}{dz^2} \right] T_L^1 = 0, \quad (31)$$

and the perturbed solute flux boundary condition takes the form

$$\begin{aligned} \sigma(1-k)(C_S/k)\hat{h} + (1-k)VC_L^1 + (1-k)VG_c\hat{h} = \\ -D \frac{dC_L^1}{dz} - \frac{\partial D}{\partial T} G_c(\hat{T}_L + G_L\hat{h}) - D \frac{d^2 C_L^0}{dz^2} \hat{h} \end{aligned} \quad (32)$$

at $z = 0$. An analytic solution to the perturbed solute equation (31) is difficult because of the spatial variation in the coefficients arising from $D(T)$. The numerical results indicate that the variation is small enough that D in eq. (31) can be assumed to be constant, $D = D_I$; however, the derivative $\partial D/\partial T$ must be retained in these equations. This produces a constant coefficient equation that can be solved analytically. We further find that to a good approximation the base state C_L^0 and its derivatives can be obtained from the solution

$$C_L^0 = C_\infty \left(1 + \frac{(1-k)}{k} \exp[-Vz/D_I] \right), \quad (33)$$

corresponding to a constant diffusion coefficient as well. Eq. (31) then admits an exponential solution

$$C_L^1(z) = A_0 T_L^1 \exp[-rz] + B_0 \exp[-\lambda z], \quad (34)$$

where $r = \omega + V/D_I$,

$$\lambda = \frac{V + G_L \partial D / \partial T}{2D_I} + \sqrt{\left(\frac{V + G_L \partial D / \partial T}{2D_I} \right)^2 + \omega^2 + \frac{\sigma}{D_I}}, \quad (35)$$

and

$$A_0 = \frac{rG_c(\partial D/\partial T)}{\omega(V - G_L \partial D/\partial T) - \sigma - VG_L(\partial D/\partial T)/D_I}. \quad (36)$$

The coefficient B_0 , together with \hat{h} , \hat{T}_L , and \hat{T}_S , is found by applying the boundary condition (32), together with the boundary conditions (28) and (29). In applying (32), we find that the term $d^2 C^0/dz^2$ should be obtained from the exact solution given by eq. (15) (with $\xi = 0$ and $\mu = 1$) rather than eq. (33).

The resulting dispersion relation is given by

$$\begin{aligned} \frac{-\sigma m G_c}{V} + mA_0 g_0 (\lambda - r) + (G_L + g_0) \left[(1 - k) \frac{V}{D_I} - \lambda \right] + \frac{g_0 m G_c \partial D / \partial T}{D_I} \\ + m G_c \left[\lambda - \frac{V}{D_I} \right] - T_M \Gamma \omega^2 \left[\lambda - (1 - k) \frac{V}{D_I} \right] = 0, \end{aligned} \quad (37)$$

where

$$g_0 = \frac{\sigma L_V / \omega + k_S (G_S - G_L)}{k_L + k_S} \quad (38)$$

is a proportionality factor relating the perturbed liquid temperature and the interface perturbation, $\hat{T}_L = g_0 \hat{h}$.

Since the critical wavelength is found to be small compared to the diffusion length D_I/V , we consider the dispersion relation in the limit of large wavenumber. The resulting approximation to the dispersion relation, with $\sigma = 0$, has the form

$$\Omega \left(1 - \frac{\bar{G}}{m G_c} \right) - \frac{\mathcal{A} \Omega^3}{k} - k + \frac{\alpha k_S}{2(k_L + k_S)} \left(\frac{G_S}{G_L} - 1 \right) = 0, \quad (39)$$

where $\Omega = \omega D_I/V$, $\alpha = G_L (\partial D / \partial T) / V$, $\bar{G} = (k_L G_L + k_S G_S) / (k_L + k_S)$, and

$$\mathcal{A} = \frac{k T_M \Gamma V^2}{m G_c D_I^2} \quad (40)$$

is the absolute stability parameter of Mullins and Sekerka [2]. The left hand side of eq. (39) has an extremum when

$$\frac{3\mathcal{A}\Omega^2}{k} = 1 - \frac{\bar{G}}{m G_c}, \quad (41)$$

which when combined with eq. (39) results in the relation

$$\Omega^3 = \frac{k}{2\mathcal{A}} \left[k - \frac{\alpha k_S}{2(k_L + k_S)} \left(\frac{G_S}{G_L} - 1 \right) \right]. \quad (42)$$

Numerically solving eq. (39) for C_∞ , and minimizing C_∞ as a function of ω yields the critical concentration and wavenumber as a function of velocity and liquid temperature gradient. A plot of the critical concentration as a function of velocity based on the approximation (39) is indistinguishable from the dashed-dot curve in fig. 1; the results agree to four digits.

The results for the critical wavenumber as a function of velocity based on the approximate formula (39), and the corresponding exact numerical solution, which corresponds to the dashed-dot curve in fig. 2, is shown in fig. 3.

It is difficult to make further analytical progress in solving the equations (39) and (41) to obtain explicit expressions for the critical wavenumber and concentration (or an alternative control parameter such as V or G_L). The parameter \mathcal{A} depends on both V and c_∞ , and the dependence on the control parameters is nonlinear in these expressions. An estimate for the effect of temperature-dependent diffusivity on the critical wavenumber can be obtained from eq. (42) if we ignore the small effect that α has on \mathcal{A} . For tin-silver, the term in brackets on the right hand side of eq. (42) has the value $[k + 0.052]$ for a velocity $V = 1 \mu\text{m/s}$; since $k = 0.018$ in this case, this results in a significant change in the critical wavenumber.

5 Discussion

For the low solidification velocities considered here, morphological instability is, to a good approximation, given by the modified constitutional supercooling criterion: instability occurs when the constitutional supercooling term mG_c exceeds the thermal-conductivity-weighted temperature gradient, i.e., $\bar{G}/(mG_c) < 1$ in eq. (39). The surface tension term ($\mathcal{A}\Omega^3/k$) is small compared to Ω , but plays an important role in determining the critical wavenumber. Similarly, although the term proportional to α is small as well, it also has a significant effect on the critical wavenumber. Noting that k/\mathcal{A} is independent of k for fixed G_c , we see from eq. (42) that the temperature-dependence of the diffusivity effectively modifies the partition coefficient k . At the onset of instability the term in brackets in eq. (42) can be written as $k - [(\partial D/\partial T)g_0/(2V)]$, where g_0 relates the perturbed temperature field and interface shape, $\hat{T}_L = g_0\hat{h}$. When the diffusivity is temperature-dependent and the thermal conductivity in the solid is larger than in the liquid (as in the tin alloy considered here), this term increases the effective partition coefficient and hence, from eq. (42), it increases the critical wavenumber.

In the frozen temperature approximation [26], in which $G_L = G_S$, the perturbed temperature field vanishes and there is no modification of the critical wavenumber associated

with the temperature-dependent diffusion coefficient, see eq. (42). In this approximation, the hills at the interface maxima are hotter than the valleys at the interface minima, and hence the diffusion coefficient is larger at the hills than in the valleys. However, this effect does not cause a modification of the critical wavenumber. The change in wavenumber apparently arises from the perturbed temperature field, which modifies the perturbed concentration field, see eq. (34). The perturbed temperature field depends on the parameter g_0 given by eq. (38), which is the quantity that actually enters the approximate dispersion relation (39).

With the parameter values we have studied for the tin-silver system, the Soret effect is found to be small. We note, however, that Hurle [3] found a significant shift in the critical wavenumber for moderate Soret coefficients for systems with small partition coefficients, $k \leq 0.01$.

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Table I

Thermophysical Properties of Tin-Silver Alloys

Melting point of tin, T_M	505.1	K	[20]
Liquid thermal conductivity, k_L	0.30	J/(cm s K)	[23]
Solid thermal conductivity, k_S	0.60	J/(cm s K)	[23]
Heat of fusion, L_v	418.0	J/cm ³	[20]
Capillary parameter, $T_M\Gamma$	8.6 (10 ⁻⁶)	Kcm	[21]
Liquid diffusion coefficient, $D(T_M)$	2.07(10 ⁻⁵)	cm ² /s	[18]
Temperature coefficient, $(\partial D/\partial T)$	1.72(10 ⁻⁷)	cm ² /(Ks)	[18]
Liquidus slope, m	-284.	K/at. frac.	[22]
Segregation coefficient, k	0.018		[22]
Thermodiffusion coefficient, D_T	7.2(10 ⁻⁹)	cm ² /(Ks)	[24]

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Sn-Ag Alloys

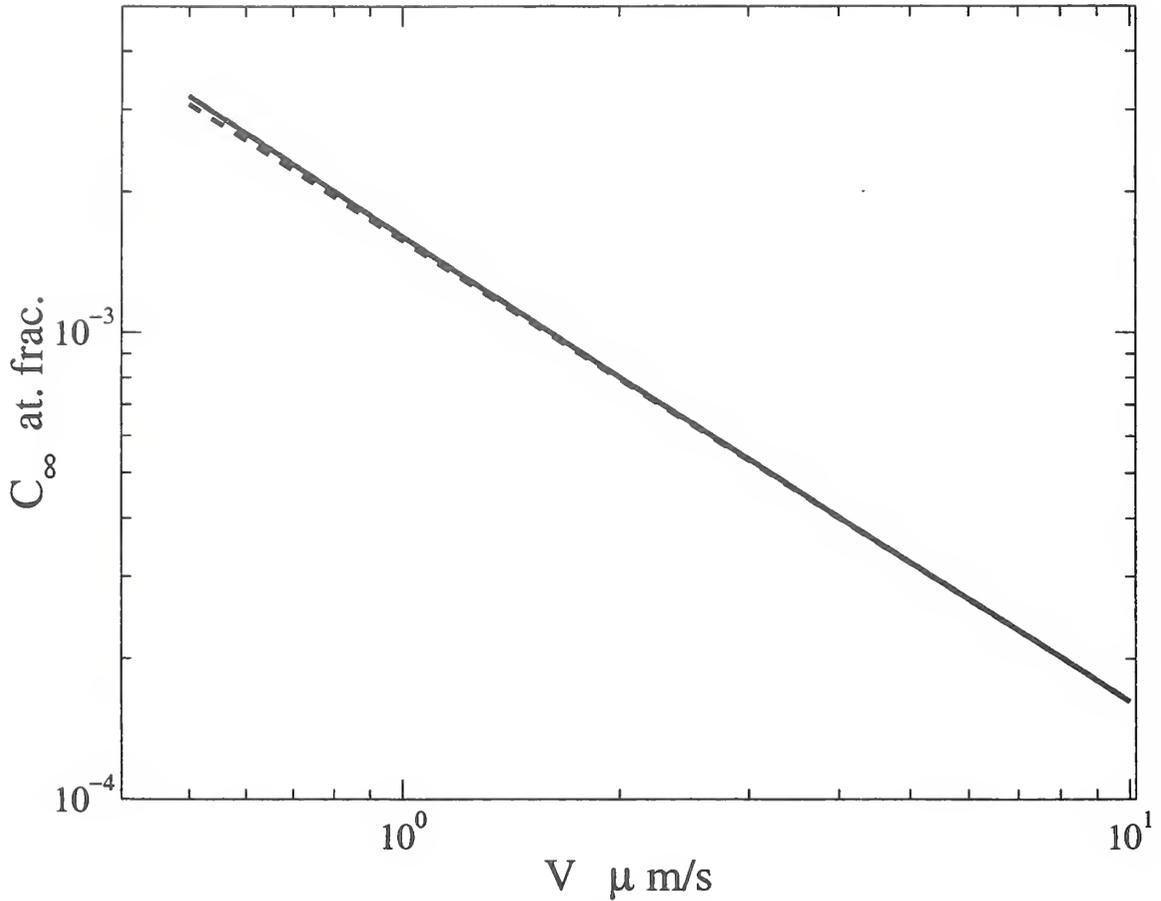


Figure 1: The critical silver concentration above which the interface is morphologically unstable as a function growth velocity for a tin-silver alloy with a liquid temperature gradient of 180 K/cm. The three curves which almost coincide correspond to: solid [$D_T = 0$ and $(\partial D/\partial T) = 0$]; dashed [$D_T = 7.2 \cdot 10^{-9} \text{ cm}^2/(\text{Ks})$ and $(\partial D/\partial T) = 1.72 \cdot 10^{-7} \text{ cm}^2/(\text{Ks})$]; dashed-dot [$D_T = 0$ and $(\partial D/\partial T) = 1.72 \cdot 10^{-7} \text{ cm}^2/(\text{Ks})$].

Sn-Ag Alloys

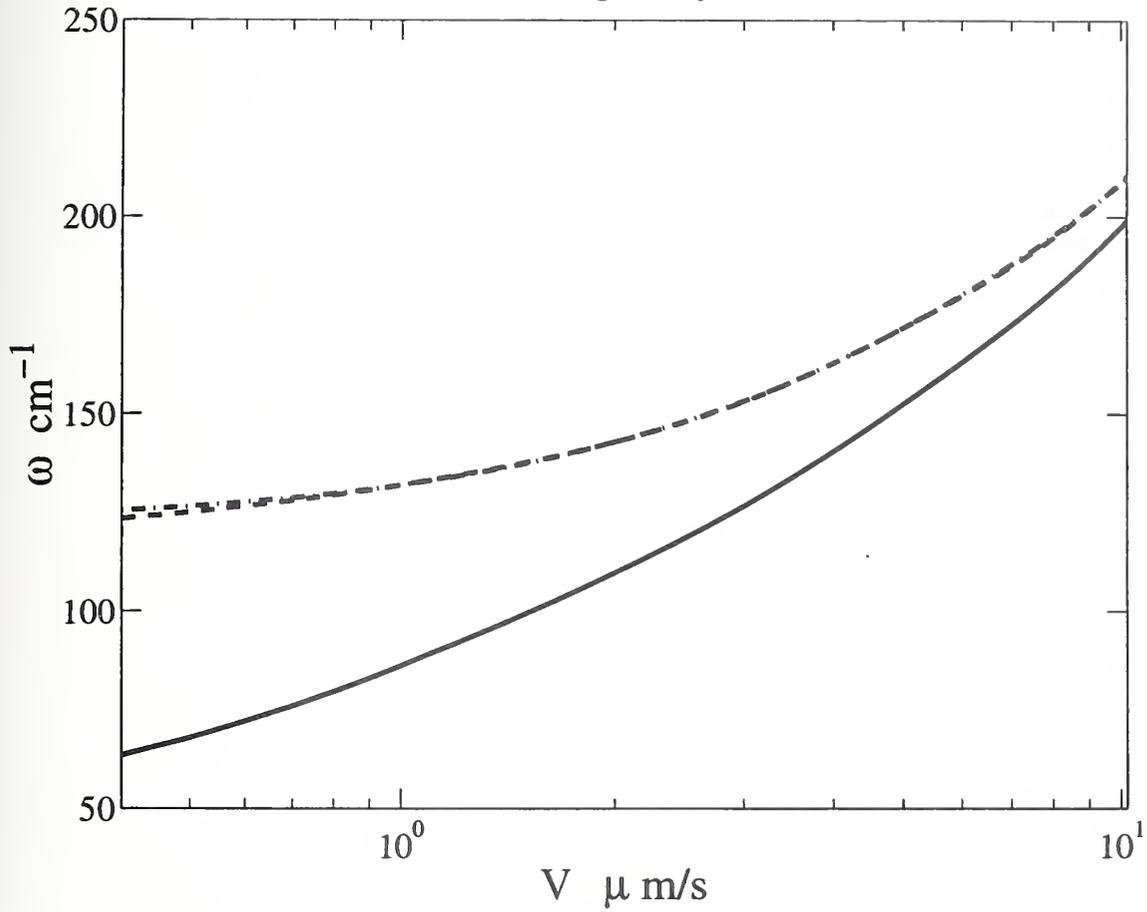


Figure 2: The critical wavenumber as a function growth velocity for a tin-silver alloy with a liquid temperature gradient of 180 K/cm. The solid, dashed, and dashed-dot curves correspond to the same parameters as those in fig. 1; the dashed and dashed-dot curves almost coincide.

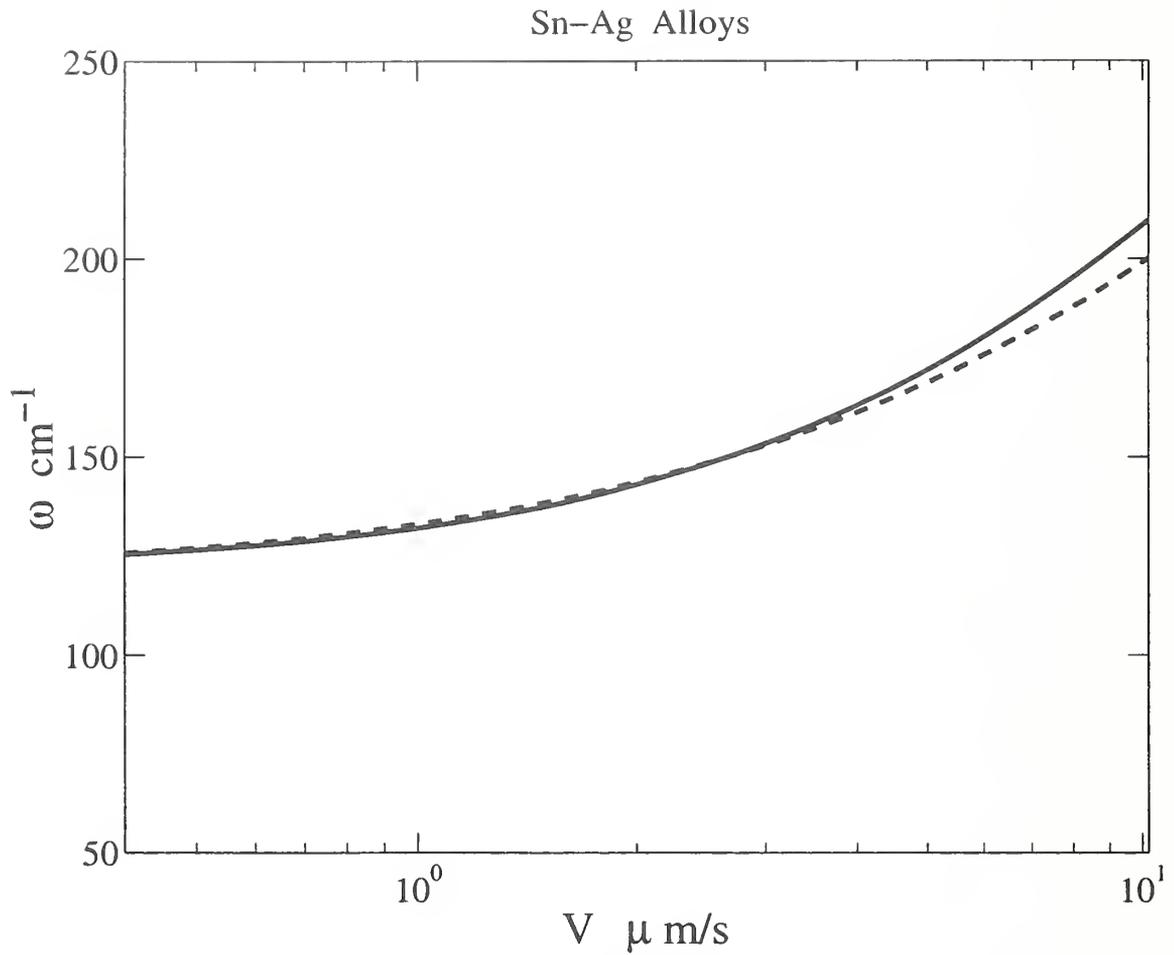


Figure 3: The critical wavenumber as a function growth velocity for a tin-silver alloy with a liquid temperature gradient of 180 K/cm for a temperature-dependent diffusion coefficient. The solid curve is the same as the dashed-dot curve of fig. 2, while the dashed curve is based on numerical solution of the analytic expression given by eq. (39).

