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SHEAR STABILIZATION OF A SOLIDIFYING FRONT

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ABSTRACT

The manufacturing of multi-component single crystals with uniform material properties is frequently hampered by the presence of morphological instabilities during the solidification. We discuss the influence of shear flows on the linear and nonlinear stability of the solid/liquid interface during the directional solidification of binary alloys. The flows are generated by nonplanar harmonic translations of the crystal parallel to the mean interface position. Oscillations with physically realizable amplitudes and frequencies are found to be useful for stabilization purposes.

INTRODUCTION

Directional solidification is a processing configuration which lends itself to experimental observation and mathematical analysis. Here a liquid may be confined to a Hele-Shaw cell or to a fully three-dimensional region (inside a cylindrical tube, for example). The confined material, along with its container, is then pulled through a temperature gradient, so that the fluid solidifies as it moves from a heated region into a cooler one. If the material is pulled at constant speed, the solid-liquid interface will establish itself at a fixed position relative to the heat source and sink.

The instability of the planar front was first explained by Tiller et al [1] and a full linear-stability analysis, including the effects of surface energy, was first done by Mullins and Sekerka [2]. They found that the interface is stable for sufficiently low solute concentrations. For higher concentrations, the interface is unstable for a finite range of pulling speeds. Near the critical values of the pulling speed, the instability results in nearly sinusoidal cells.

When the instability is present, variations in the concentration along the interface are frozen into the solid, resulting in stripes of elevated solute concentration. A number of authors have attempted to use a forced fluid flow to extend the range of pulling speeds for which the interface remains flat (see Davis [3] for a summary of these). Schulze and Davis [4] investigated the effects of translating the crystal in elliptical orbits parallel to the interface, following the example of Kelly and Hu [5], who has found that the same flow has a stabilizing influence on Bénard convection. This nonplanar forcing will generate a three-dimensional version of a Stokes boundary layer in the fluid above the interface. Because the boundary layer will be compressed due to the flow normal to the interface generated by the pulling speed, we refer to this flow configuration as a three-dimensional Compressed Stokes Layer (3D CSL), or CSL for short.

Schulze and Davis [4] found that this flow can, on a linear theory basis, stabilize the interface, provided the pulling speed is sufficiently high. The success of this method requires that the frequency of the flow lie within a calculated range. The outermost curve in Figure 1 is a typical neutral curve for the no-flow system [2], showing the critical pulling speed as a function of the alloy concentration for a fixed temperature gradient. Also shown in this figure are neutral curves for the system when it is forced through the boundary to generate a CSL. As the amplitude of the forcing is increased, the upper branch of the neutral curve is lowered considerably and the nose of the curve moves to the right. For a pulling speed near the nose of the neutral curve, typical values for the frequency and amplitude of the lateral velocity oscillations would be 10^4 hertz and 10^{-3} centimeters respectively. Such scales would lend themselves well to an acoustic forcing. The precise value of these parameters can vary over many orders of magnitude,

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but, as a rule, vary with the solute-diffusion length and time scales and are physically realizable for a sizable range of operating conditions.

In this paper the effect of CSL on the bifurcation structure for morphological instability will be investigated. Our principal aim is to discover under what conditions the flow may provide effective control of interfacial instability. Of particular concern is the possibility that subcritical instabilities could reduce, or even eliminate, the stabilization indicated by the linear theory. When the interface is unstable, there are a variety of potential patterns for the instability to take, including square, hexagonal and two-dimensional (roll) cells, and we shall discuss the use of this flow as a pattern-selection mechanism.

Our focus will be on the upper branch of the curve shown in Figure 1, for it is in this region that the CSL has a strong influence on the interfacial morphology. Along this portion of the neutral curve the critical wavelength of interfacial disturbances is long compared to the solutal boundary-layer thickness, and an analysis which exploits this fact was accomplished by Brattkus and Davis [6]. Our aim is to generalize this work to one in which the CSL is present.

THE EVOLUTION EQUATION

The system is stable for all nondimensional surface tensions $\Gamma > \Gamma_s = 1/k$. We introduce the small parameter ϵ as a measure of closeness to this point, which is known as the absolute stability limit:

$$\epsilon^4 = 1/k - \Gamma . \quad (0.1)$$

In this limit, the system becomes linearly unstable to long-wave disturbances when the morphological number becomes very large. In terms of physical variables, this limit can be thought of as large pulling speed.

The linear stability theory suggests the following slow space and time scales:

$$X = \epsilon^2 x , Y = \epsilon^2 y , T = \epsilon^4 t . \quad (0.2)$$

Near the absolute stability limit, the morphological number M scales like

$$\bar{M} \sim \epsilon^8 M , \quad (0.3)$$

with $M \sim O(1)$. With these scales one obtains the sought after generalization to the equation of Brattkus and Davis [6] valid for the case of the imposed CSL flow.

This equation has the form [7]

$$L[h_0] + R^2 f_0 \nabla^2 |\mathbf{r} \cdot \nabla|^2 F^{-1} \left[\frac{\hat{h}_0}{|k|} \right] + k \bar{M}^{-1} h_0 = N[h_0] , \quad (0.4)$$

where the operators L and N are defined by the Brattkus-Davis [6] equation $L[h_0] = N[h_0]$, F^{-1} is the inverse Fourier transform, R is a scaled Reynolds number, and f_0 is constant. Note that the correction is linear.

RESULTS AND SUMMARY

We have derived [7] a strongly nonlinear evolution equation for the shape of a directionally solidifying interface in the presence of a three-dimensional compressed Stokes layer. We used a one-sided model, where diffusion of solute in the solid is neglected, and we invoked the frozen temperature approximation. The equation is valid when: the surface energy parameter Γ is near the absolute stability boundary $\Gamma_s = 1/k$; and the imposed flow, as measured by R , is weak. In this limit, the critical disturbance

wavelength is long compared to the diffusion length scale D/V and the critical morphological number is large.

Linear theory shows that increasing the flow amplitude R could stabilize the interface, provided the frequency of the flow oscillations is within a calculated range. The stabilization is particularly effective in the long-wave regime. The two-dimensional bifurcation analysis [7] of the derived long-wave evolution equation shows that increasing the amplitude of the CSL will eventually change bifurcations from super- to subcritical when the flow is in the stabilizing parameter regime. Thus, at least some of the stabilization gained according to the linear theory is lost to subcritical instabilities if the flow is made too large; yet it is still possible to stabilize within a range of R values for which the bifurcation is supercritical and, presumably, one could increase R at least somewhat beyond this range before the subcritical instability would cancel the stabilization indicated by the linear theory. It appears that effective stabilization of the two-dimensional system using the CSL will require careful control of both the amplitude and frequency of the flow.

For the three-dimensional system we have examined [7] bifurcations to steady roll, square and hexagonal solutions. When the motion of the crystal is a noncircular elliptical pattern, rolls are the only bifurcating solution in the weakly nonlinear limit. This may be the best scenario for stabilizing the three-dimensional system, as there are fewer types of bifurcating solutions to suppress, at least near onset. However, for systems which are strongly supercritical or for systems forced by a nearly circular motion of the crystal, secondary bifurcations to rectangles and asymmetric hexagons are likely to occur.

When the motion of the crystal is in a circular pattern, the system is isotropic in the plane of the interface, and superposition states involving more than one set of obliquely positioned rolls are possible. This flow may be useful for selecting a preferred pattern among these states, allowing a crystal grower more control over crystal microstructure.

We have examined [7] the competition between roll and square solutions, and found that increasing the flow amplitude tends to reduce, and eventually eliminate, the range of segregation coefficients for which stable square solutions are possible. Increasing the flow amplitude favors subcritical instabilities for both squares and rolls. Unlike the no-flow case, unstable, supercritically bifurcating square solutions are possible for a small range of R values.

We have examined [7] the competition between roll and hexagonal solutions. For values of k away from unity, unstable hexagons bifurcate transcritically and are the only bifurcating solution in the weakly nonlinear limit. When k is sufficiently close to unity, the cubic and quadratic nonlinearities in the evolution equation are balanced, and a complicated bifurcation structure emerges. As with the square and roll solutions, bifurcations to rolls are supercritical when R is sufficiently small, but switch to subcritical as R is increased. Hexagons bifurcate transcritically, but with very shallow subcritical turning points for small R . When R is increased, the bifurcation for the hexagons changes direction, and they become essentially subcritical. So, in general, increasing the flow amplitude favors subcritical instability for all types of solutions examined.

In general, we have found that the flow has little impact on the system when the frequency and Schmidt number are in the destabilizing range. Clearly, this is an effect of the weak-flow limit, as microstructure will be significantly altered by a stronger version of this flow any time the interface remains unstable in its presence. The steady rolls, squares and hexagons indicated by the analysis of this chapter are leading-order approximations for the interface shape in a specific limit. Higher-order corrections to the evolution equation would indicate time-periodic variations of these patterns as one moves vertically through the crystal, and a large amplitude flow would likely render the patterns unrecognizable.

When the flow parameters are in the stabilizing range, we have found that the bifurcation structure for all types of solutions (rolls, squares, hexagons and mixed modes) changes only in scale until a critical value of R is surpassed. Beyond these transition points, which are distinct for each solution type, bifurcations

switch from super- to subcritical. In general, lowering the frequency Ω lowers these transitions, and increasing the frequency raises them.

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REFERENCES

- [1] Tiller, W. A, Jackson, K. A., Rutter, J. W. and Chalmers, B.: The Redistribution of Solute Atoms during the Solidification of Metals, *Acta Metall.* Vol. 1, 1953, 428.
- [2] Mullins, W. W. and Sekerka, R. F.: Stability of a Planar Interface during Solidification of a Dilute Binary Alloy, *J. Appl. Phys.*, Vol. 35, 1964, 444.
- [3] Davis, S. H.: Effects of Flow on Morphological Stability, in "Handbook of Crystal Growth" (Ed. D.T.J. Hurle) North-Holland Press. Amsterdam (1993), 859.
- [4] Schulze, T. P. and Davis, S. H.: Shear Stabilization of Morphological Instability during Directional Solidification, *J. Cryst. Growth*, Vol. 149, 1995, 253.
- [5] Kelly, R. E. and Hu, H.-C.: The Onset of Rayleigh-Bénard Convection in Non-planar Oscillatory Flows, *J. Fluid Mech.*, Vol. 249, 1993, 373.
- [6] Brattkus, K. and Davis, S. H.: Cellular Growth near Absolute Stability, *Phys. Rev.* B38, 1988, 11452.
- [7] Schulze, T. P. and Davis, S. H.: Shear Stabilization of a Solidifying Front: Weakly Nonlinear Analysis in the Long-Wave Limit, *Phys. Fluids*, 1996, in press.

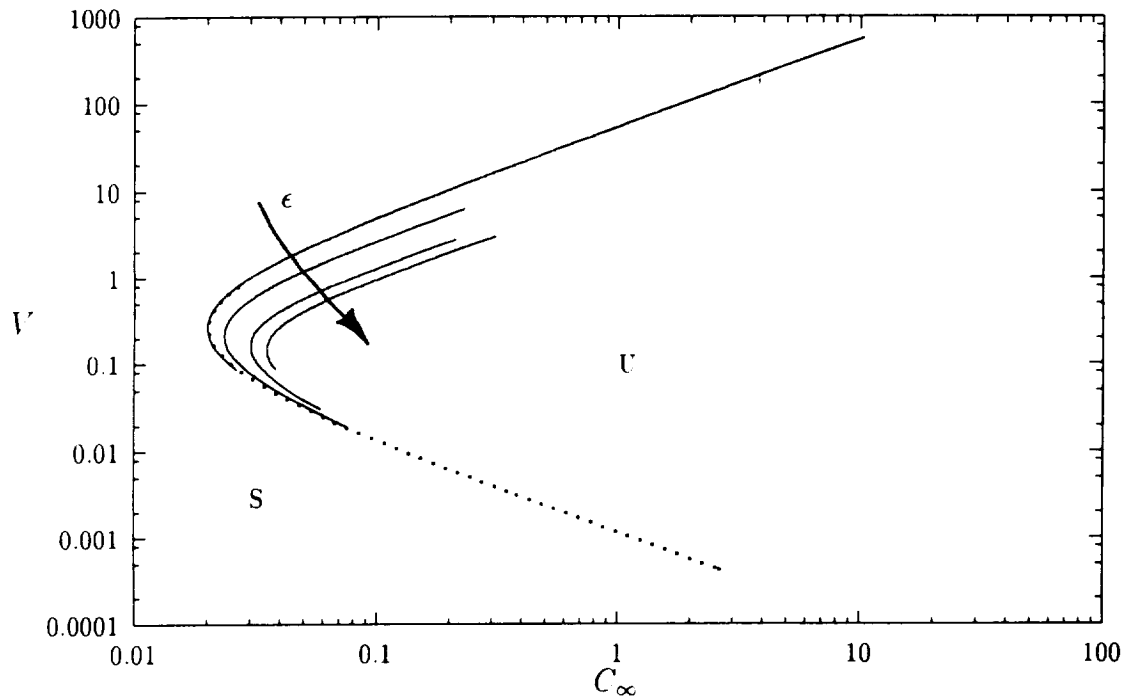


Figure 1: Plot of a typical neutral curve in dimensional form— V versus C_∞ for with a fixed temperature gradient. All of the curves extend infinitely along tangents to the portions shown. The interface is linearly stable (S) when the far-field concentration is to the left of the neutral curve. The no-flow (outermost) curve is divided into a portion corresponding to subcritical instability (dashed portion) and a portion corresponding to supercritical instability (solid portion).

