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CRYSTAL GROWTH AND FLUID MECHANICS PROBLEMS IN DIRECTIONAL SOLIDIFICATION

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1. SCIENTIFIC OBJECTIVES:

Broadly speaking, our efforts have been concentrated in two aspects of directional solidification:

- A. A more complete theoretical understanding of convection effects in a Bridgman apparatus.
- B. A clear understanding of scalings of various features of dendritic crystal growth in the sensitive limit of small capillary effects.

For studies that fall within class A, the principal objectives are as follows:

- (A1.) Derive analytical formulas for segregation, interfacial shape and fluid velocities in mathematically amenable asymptotic limits.
- (A2.) Numerically verify and extend asymptotic results to other ranges of parameter space with a view to a broader physical understanding of the general trends.

With respect to studies that fall within class B, the principal objectives include answering the following questions about dendritic crystal growth:

- (B1.) Are there unsteady dendrite solutions in 2-D to the completely nonlinear time evolving equations in the small surface tension limit with only a locally steady tip region with well defined tip radius and velocity? Is anisotropy in surface tension necessary for the existence of such solutions as it is for a true steady state needle crystal? How does the size of such a local region depend on capillary effects, anisotropy and undercooling?
- (B2.) How do the different control parameters affect the nonlinear amplification of tip noise and dendritic side branch coarsening?

2. SCIENTIFIC AND TECHNOLOGICAL RELEVANCE

The vertical Bridgman apparatus (Fig. 1) is used in the directional solidification of a molten binary alloy. The desired crystal for technological applications (as in semi-conductors) is the one with minimal compositional variations (i.e. minimal segregation) and crystalline defects. However, due to significant heat flux through the sides of Bridgman device that is necessary to avoid constitutional supercooling, differences in fluid densities occur that results in convection at any Rayleigh number. Convection without sufficiently vigorous and uniform mixing leads to a significant radial segregation. A clear understanding of the dependence of segregation and interface deformation on the numerous non-dimensional parameters is likely to lead to improved design and control of this industrially relevant process.

Prior theoretical work [see Brown¹ for a review], mostly numerical, has significantly furthered our understanding of the Bridgman problem; nonetheless, without explicit analytical formulas, it is difficult to get a general idea of the trends in various regions of the parameter space. We overcome this problem by deriving explicit formulas in some asymptotic ranges. This is complimented by numerical calculations

to determine the practical range of validity of the asymptotic results as well as to extend the suggested scaling dependences to other ranges of parameter space.

Dendritic crystal growth is an important problem in pattern formation [see References [2-4] for reviews]. In the simplest case, one is interested in the growth of a pure needle crystal in an undercooled melt. It also arises in the context of directional solidification when the drawing speed exceeds some critical value that determines a "cell to dendrite transition". In this case, the growth is controlled by solutal diffusion rather than thermal. Nonetheless, the equations are similar to that in a pure needle crystal growth. Appearance of dendritic structures in directional solidification leads to undesirable striations of solute rich material in the crystal. Further understanding of the basic process of dendritic crystal growth can be both scientifically and technologically rewarding.

By answering question (B1) without the limitations of an assumed steady state, linear stability or a linear WKB wave packet analysis, we address directly the controversy between the premises of "microscopic solvability"^{2,3} and other theories^{5,6} that rely on the equivalent of a "marginal stability" hypothesis made originally by Langer & Muller-Krumhaar. It is to be noted that inclusion of surface tension anisotropy is crucial in the first case but not so for the other. Further, our analytical approach provides us with important scale information in the small capillary limit, where increasingly small scales need to be resolved in any traditional method of numerical computation. An analytical understanding of the parameter dependence of noise amplification and side branch coarsening is also important since it is likely to influence ways of controlling this process.

3. RESEARCH APPROACH

3a. Bridgman Problem

Using a standard quasi-steady assumption, analytical calculations have so far been carried out in the limit when fluid velocities and interfacial deformation are small enough to permit linearization of the Navier-Stokes equation and the interfacial matching condition (about a planar interface). This assumption can formally be justified in the limit of small horizontal heat transfer; nonetheless, our analysis of the equations suggests that the actual expressions for radial segregation at the interface and interface deformation may actually transcend this limitation. Further, with a no-stress boundary condition on the side walls, a convenient radial expansion in terms of a Bessel function representation reduces the problem to an eighth order ordinary differential equation in the melt that is coupled with two second order equations in the crystal through the interfacial conditions. The resulting equations are further analyzed using standard WKB techniques in the two cases of (i) large thermal Rayleigh number R_T in a thermally stable configuration and (ii) large solutal Rayleigh number R_c in a solutally stable configuration.

Further, a numerical code based on a finite difference scheme has been developed to solve the coupled heat-mass transfer and fluid equations with a no-slip as well as no-stress sidewall boundary conditions in order to compare and contrast the two cases and determine the range of validity of the large R_T or $|R_c|$ asymptotics. We also wish to explore the possibility that the complicated dependence of segregation and interfacial deformation on the seventeen nondimensional parameters can be largely understood in terms of only a few lumped parameters that are in turn functions of other parameters, as suggested by the asymptotic results. A systematic bifurcation study of the possible steady states is also planned.

3b. Dendrite Problem

With a one sided diffusion model of the growth of a pure dendrite, but with no assumption about a quasi-stationary field, we have so far looked at two opposite limits: (i) small undercooling when the Peclet

number based on tip radius and velocity of a dendrite is small and (ii) large undercooling when Peclet number based on tip radius and velocity tends to infinity.

i. Small Peclet number limit

In the small Peclet number limit, various regions of an initially near parabolic dendrite are identified where different equations have to be solved to follow the dynamics of disturbances initiated near the tip. Not unexpectedly, there is an $O(1)$ region near the tip where the diffusion problem reduces to a Laplacian.

In this order $O(1)$ region near the tip, the dynamics for $O(1)$ times is studied by using a conformal mapping function $z(\zeta, t)$ that maps the upper half ζ plane to the physical (z) domain. The complex dynamics of the analytically continued $z(\zeta, t)$ is studied in $\text{Im } \zeta < 0$ with a view to understanding the asymptotic behavior of the dynamics for sufficiently small but nonzero capillary effects. This unusual procedure of extending the domain to the complex unphysical ζ plane is a mathematical convenience and allows for a perturbative investigation of small capillary effects on the otherwise ill-posed zero surface tension dynamics. Further, in this formulation, one can mimic small noise by introducing a statistical ensemble of complex singularities that correspond to the same initial interface within some prescribed error. By studying the dynamics of singularities as they come close to the real ζ axis and cause large interfacial distortions, one can understand and identify robust features of this highly sensitive dynamics and determine the dependence on capillary effects, anisotropy and Peclet number.

The analysis of disturbance in the $O(1)$ tip region of the dendrite is complimented by analysis of the solutions in other regions of the semi-infinite dendrite, where a Laplacian approximation for the field is invalid. Study of such solutions is relevant to side branch coarsening when tip noise advects and amplifies along the sides of a dendrite to a sufficiently large distance from the tip.

ii. Large Peclet number limit

In this case, a boundary layer analysis is possible for disturbances of some distinguished length scale and size superposed on a near parabolic initial dendrite. The resulting pair of nonlinear partial differential equations involving one space and time variable is studied.

4. DISCUSSION OF RESULTS

4a. Bridgman Problem

For the Bridgman problem, we have derived explicit analytical formula for radial segregation and interfacial slope in the (i) asymptotic limit of large thermal Rayleigh number⁷ R_T and (ii) large solutal Rayleigh number⁸ $|R_c|$.

Here, we will only present results for case (i). Rather than give the more general formulas, that is a little too complicated to describe in this few pages, we describe the special case obtained under additional assumptions that the vertical height is much larger than the cylinder radius and that the diffusion in the solid is negligible. The expression for radial segregation is of the form

$$\frac{\partial c_s}{\partial r}(r, z_0) \sim \sum_{n=1}^{\infty} A_n J_1(\lambda_n r) \left[1 - \frac{\delta_2}{\lambda_n} \right] e^{-\lambda_n(z_0 - z_I)}. \quad (1)$$

In the above formula, $c_s(r, z)$ is the solute concentration in the solid at a point with cylindrical coordinates (r, z) . Further, λ_n is the n -th zero of the Bessel function $J_1(x)$ not including $x = 0$, A_n and δ_2 are functions of other nondimensional parameters. In deriving formulae (1), it was implicitly assumed that the solutal Peclet number $\ll R_T^{1/6}$ and that the $z_{II} - z_0$ and $z_0 - z_I$ are of the same order. Also, in (1), gravity enters into the formula through an $R_T^{-1/6}$ scaling of A_n . The interfacial slope is also given by an expression

similar to (1). We also have scaling information on the size of fluid velocities in different regions of the Bridgman apparatus.

While Brattkus & Davis⁹ have analyzed a two dimensional version of this problem in the past, their results have been derived in the absence of a thermally insulated so called adiabatic zone, i.e. $z_{II} = z_0 = z_I$ in the notation of Fig. 1. Indeed, the effect of this insulation zone leads us to both important quantitative as well as qualitative differences with their results. In (1), the small scale components radial segregation and interfacial deformation, i.e. coefficients of $J_1(\lambda_n r)$ for large n , is exponentially quenched by factors of $e^{-\lambda_n(z_0 - z_I)}$. This quenching is absent without an insulation zone. Also, if the insulation zone thickness is comparable or much larger than the cylinder radius then an overwhelming part of the contribution in the summation in (1) and similar summation for the slope comes from the first term, implying that each of these quantities will have a $J_1(\lambda_1 r)$ radial dependence. In that case, the Coriell-Serferka hypothesis¹⁰ on the proportionality of radial segregation and interfacial slope is approximately true, though the constant of proportionality is different from what they derived with no fluid motion. Further, in this case, a most interesting aspect of this result is the result that radial segregation and interfacial deformation can each be reduced very significantly by choosing $\delta_2 = \lambda_1 = 3.83$. Physically, the nondimensional parameter δ_2 is the logarithmic derivative of the horizontal heat loss with respect to z , evaluated in the limit of approaching $z = z_I$ from below. Recall that z is the vertical distance from the base of the cylinder scaled by the cylinder radius, while $z = z_I$ is the location of the lower edge of the insulation zone. Thus, to the extent these asymptotic results hold, we have a prediction on optimal growth condition.

We have also carried out some independent numerical verification of our analytic results. There was one point of concern to us – use of an unphysical no-stress boundary conditions at the cylinder side walls, rather than a no slip condition. Fortunately, numerical results¹¹ with no-stress and no-slip side wall conditions, so far obtained for the linearized 2-D problem, show that in the large thermal Rayleigh number regime, there is very little difference between the two with regards to our central results.

Further, the numerical results are found to be consistent with the asymptotic scaling relations, though precise quantitative comparison was not possible due to resolution difficulties at very large Rayleigh number. However, the optimal analytically predicted condition adapted to the 2-D geometry, $\delta_2 = \pi$, appears to hold quite well for moderately large thermal Rayleigh number, well within the limits of accurate computation. As δ_2 increases past this value, the interface shape changes from being concave to convex towards the melt.

b. Dendrite problem

In the large Peclet number limit, a system of nonlinear hyperbolic equations is found to describe¹² the temporal development of disturbances of some distinguished spatial scales and sizes; this develop “shock” in finite time when the lateral diffusion becomes important.

In the small Peclet number limit¹³, the equations in the $O(1)$ region around the tip has been analyzed. Exact solutions describing both tip splitting and side branching have been found when surface tension effects are completely ignored (Figs. 2 and 3). These correspond to moving pole singularities of $z_\zeta(\zeta, t)$ in the unphysical region $Im \zeta < 0$. As these singularities move towards the real ζ axis without actually hitting it in finite time, we obtain the different stages of the evolution shown in the figures. Generally, it has been established that every initial singularity (not just poles) of z_ζ in $Im \zeta < 0$ has a component of motion towards the real domain. This result is analogous to what was obtained for the Hele-Shaw case¹⁴. For a class of initial conditions containing singularities that do not impinge $Im \zeta = 0$ in finite time, asymptotic analysis reveals that for a long time, the zero surface tension dynamics is the leading order behavior of the actual solution for small nonzero surface tension. However, these class of initial conditions

is a small subclass of the general class of initial conditions that will generically contain singularities and zeros of z_ζ that impinge the the real ζ axis in finite time, causing a cusp or a corner or some other singularity in the interfacial shape. Fig. 4 shows the effect of a zero impinging the real ζ axis at $\zeta = 0$. At about the time when singularities would have otherwise appeared on the interface, capillary effects have to be accounted for. The analytical and numerical evidence¹⁵ obtained by performing an inner equation analysis for the related Hele-Shaw problem suggests that singularity formation is impeded by the effect of surface tension; however for isotropic surface tension, narrow structures formed by the approach of a complex singularity cannot settle down to a steady state; instead it appears to fatten out before tip splitting. This process regenerates itself as other singularities, initially further out in the complex plane approach the real ζ axis. However, with a nonconstant surface tension parameter d_0 modelling anisotropy with minimal surface tension axes aligned appropriately, a cusp or corner formation event for zero surface tension presages a rapid evolution over an inner scale for small but nonzero d_0 . The inner solution settles to a local steady state with tip radius and velocity determined from a steady state dendrite theory^{2,3}. The dendrite tip corresponds to only a small region on the real ζ axis which, unlike isotropic surface tension case, does not expand with time. Further approach of complex singularities towards the real ζ axis leads to side branching. Some features of our results have qualitative consistency with direct numerical simulation of anisotropic Hele-Shaw dynamics¹⁷ or that of a dendrite¹⁸. However, our perturbative approach gives essential scaling information that is difficult to establish otherwise.

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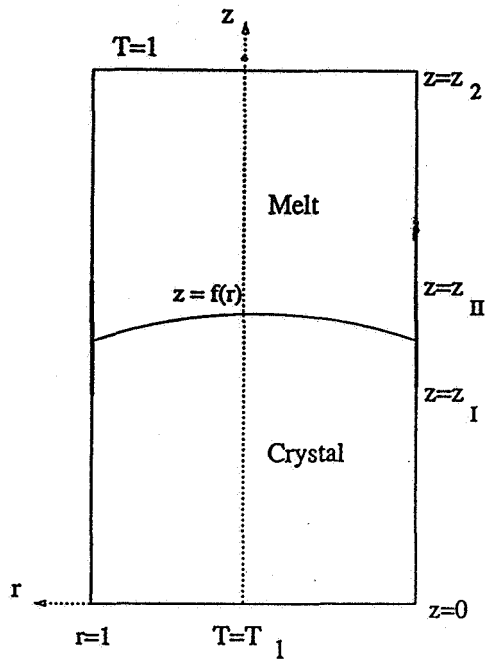


Fig. 1: Sketch of Bridgman device

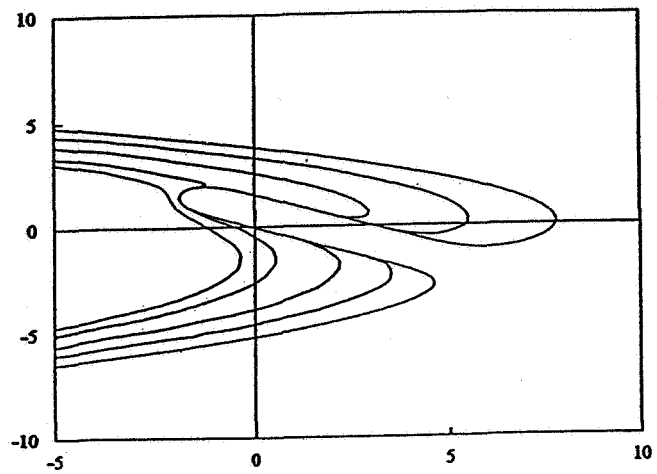


Fig. 2: Evolving tip splitting $d_0 = 0$ needle crystal

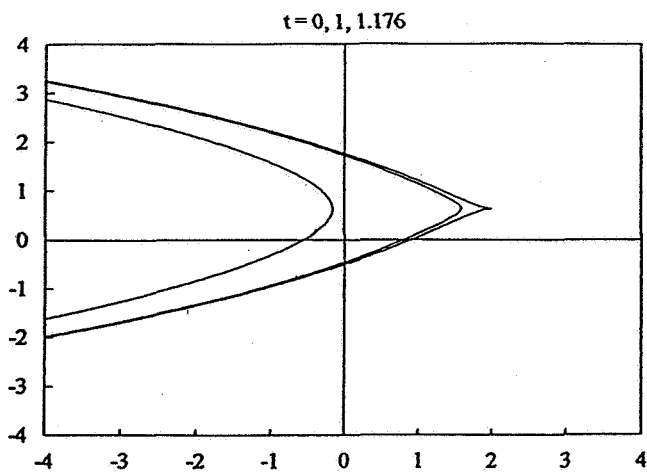


Fig. 4: Cusp formation in $d_0 = 0$ evolution

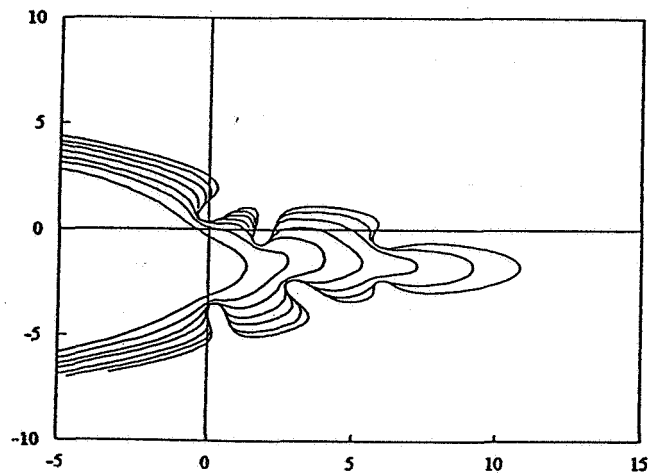


Fig. 3: Evolving side branching $d_0 = 0$ needle crystal