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CRYSTAL GROWTH AND FLUID MECHANICS PROBLEMS IN DIRECTIONAL SOLIDIFICATION

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1 Scientific objectives

The following is an outline of the directions of our efforts;

A. A more complete theoretical understanding of convection effects in a vertical Bridgman apparatus, looking toward doing the following:

A1 Construct analytical solutions for interfacial shape and fluid velocity in mathematically sensible and physically meaningful limit cases. It appears that Biot number, Prandtl number and the Rayleigh numbers are crucial. Certain canonical nonlinear problems may be treated as well.

A2 Numerically verify and extend asymptotic results from A1 to other ranges of parameter space with a view to a broader physical understanding of the general trends.

A3 Numerically and analytically investigate the nature of the initial-value problem for the melt/solid combination, paying special attention to the end conditions. Are the often tacitly-assumed quasi-steady states in fact realizable?

B. A clear understanding of scalings of various features of dendritic crystal growth in the case that both the surface energy and undercooling are small. We need to answer the following questions:

B1 Are there unsteady dendritic solutions in two dimensions to the fully nonlinear, time-evolving dynamical equations in the small-surface-energy limit (with small undercooling as well), for which there is a steady tip region with well defined tip radius and velocity, but with persistent unsteadiness in the side-branching? Most importantly, is anisotropic surface energy necessary for the existence of such solutions, as it is for a true steady-state needle crystal? How does the size of such a tip region depend on capillary effects, anisotropy and the undercooling?

B2 What is the source of noise in the dendrite problem? How does the linear theory of amplification of noise differ from a theory that includes nonlinear effects? How do nonlinear effects determine dendrite coarsening ?

2 Scientific and technological relevance

The vertical Bridgman apparatus is useful for the directional solidification of certain molten binary alloys. Such alloys, which are often components in semi-conductor devices, require minimal material segregation and minimal crystalline defects to be useful. The problem with the device is that the significant heat loss through the side walls of the material ampoule, necessary to avoid constitutional supercooling, produces significant convection in the melt and its concomitant variation in solutal concentration along the crystal-melt interface, and hence frozen in to the solid product. A clear understanding of the dependence of segregation and interfacial deformation on the numerous non-dimensional parameters of the device/melt combination is likely to lead to improved design and control of this industrially relevant process. Brown[1] has reviewed much of the important prior theoretical/numerical work, and since we have further discussed it in our 1994 report, we do not repeat it here.

Dendritic crystal growth is an important problem in pattern formation, (See References [2-4] for reviews.) but the connection with problems in directional solidification is that dendrites may arise in the context of solidification when the drawing speed in a Bridgman device, for example, exceeds some critical value that determines a "cell-to-dendrite transition". In this case, the dendrite growth is controlled by solutal diffusion rather than thermal. Nonetheless, the equations are similar to those for 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, comparing and contrasting with existing theories [2,3,5,6], can be both scientifically and technologically rewarding.

3 Research approach

3.1 Bridgman problem

3.1.1 Analytical approaches

One method of approach is to explore various asymptotic solutions of the Bridgman equations for extreme values of particular parameters. We utilize a number of differing methods to uncover something of the dependence of the exact solutions on these parameters, including the WKBJ technique and matched asymptotic expansions. The advantage of the former technique is that the scales of velocities, temperature, *et al*, come out naturally from the analysis–without any *ad hoc* prescriptions of the gauge functions. Scaling guidance from such a procedure allows matched asymptotics methods to be used. The advantage of the latter technique is that certain canonical, parameter-free, weakly nonlinear problems may be formulated and solved–gaining an entrance into the nonlinear regime without resort to full numerical computations. A number of significant results are reported in the next section.

The efficacy of analytic approaches has already been demonstrated[7,8] in simple optimization conditions for minimization of interfacial deflection, and hence material segregation. It appears now that in a wide range of Rayleigh numbers such optimization is possible [9].

3.1.2 Numerical approaches

Our approach to seeking optimal operating conditions for a Bridgman device is divided into two Parts. First, we seek steady-states[10] in which the crystal grows steadily inside the insulation zone. The solution is a quasi-stationary state in that the top and bottom of the ampoule are assumed to be sufficiently far away as to have no effect on the local behavior near the interface and insulation zone. We utilize Newton's method to solve the discrete system. Secondly, we investigate the transient development of the flow in the ampoule, paying special attention to the end boundary conditions, and differences between behavior in finite and infinitely-long ampoules. We use an ADI technique to advance the temperature and a semi-implicit method to advance the interface in time.

3.2 The dendrite problem at small Peclét number

We consider the time evolving aspects of a one-sided model for two-dimensional dendritic growth in a weakly undercooled melt that corresponding to small Peclet number. Further, our focus is almost exclusively to the case when the surface energy effects are appropriately small. It is to be noted that for a dendrite that is approximately parabolic, surface energy effects are reduced further away from the tip due to decreasing curvature and this makes the small surface energy limit of obvious relevance. We combine analytical and numerical methods to shed light on this limiting dynamics.

The essence of the approach rests on that of Tanveer[11] in solving the problem of the evolution of a Hele-Shaw finger, *viz.*, extending the domain into a complex plane, thereby making well-posed a mathematical problem for the interfacial evolution that is ill-posed in the physical plane as surface energy goes to zero. The sensitivity to noise in the laboratory setting is then seen as having its origin in the motion of a distribution of singularities in the complex plane. A variety of such initial distributions correspond to essentially identical laboratory initial conditions–but lead to quite different behaviors in time.

4 Discussion of results

4.1 Bridgman problem

We have carefully studied the nature of the asymptotic solutions for flow/solidification in the Bridgman device, under a 'quasi-steady' approximation, in the case that the Biot number is small and one or both of the inverse Rayleigh numbers (for temperature or solutal concentration) are small[7,8,9]. Matched asymptotic expansion techniques have provided particular information about the flow: The boundary- and shear-layer regions are shown schematically as Figure 1, where the dashed lines indicate the boundaries of the insulation zone that brackets the crystal/melt interface. The flow in the side-wall boundary layer drives the motion, with details of the material segregation being rather different depending on the relative sizes of the two Rayleigh numbers. Our most recent results[9] depend to some extent on the analysis of Brattkus & Davis[12], but to a much greater extent build on the foundation of the WKBJ studies of Tanveer[7,8], in which the essential scale analysis has been given. The two approaches have proved to be complementary, each reinforcing the results of the other.

Numerical approaches outlined above have also yielded important insights into the nonlinear steady-state solutions[10], but also some preliminary information with regard to what circumstances must exist at the ampoule ends in order that anything like a 'steady state' can be attained. Below is an itemized summary or our conclusions to date, in both analytical and computational arenas.

- Imposition of a no-stress rather than no-slip on the side walls of the Bridgman ampoule makes no difference to the leading-order flow/segregation at large Rayleigh numbers and small Biot numbers.[9,10].

- A minimization of material segregation can be achieved at small Biot number, if either the thermal Rayleigh number, or the solutal Rayleigh number are large, or both are large and of comparable order [7,8,9]. Simple optimization formulae can be found in both axisymmetric [7,8] and two-dimensional cases[9] in the two limit cases, but not when both Rayleigh numbers are of comparable size, though optimization has been demonstrated in this case as well[9]. Small interfacial surface tension has no effect on the optimization[9].

- For Biot numbers that are comparable to the largest Rayleigh number to the '-1/6' power, the flow becomes nonlinear in the melt boundary layer that is adjacent to the solid interface–remaining linear elsewhere. Such nonlinearity can also arise even at small Biot numbers if the Prandtl number in the melt is small–which is often the case for materials of interest.

- The usual quasi-steady hypothesis employed by countless investigators in analyzing the flow in the melt is problematic. The thermal boundary conditions at the top and bottom of the moving ampoule must be arranged in a particular way in order to establish such a quasi-steady state. Numerical and analytical results indicate that such a steady state may be difficult to achieve in practice. We have found that the Biot numbers play a crucial role in determining when an ampoule is long enough so that the interface will grow steadily without feeling the influence of the conditions at the top and bottom of the ampoule[13].

4.2 Dendrite problem

Results for the temporal evolution of a dendrite in a weakly undercooled melt, in the limit as the surface energy goes to zero involve three papers, which incorporate material from the PhD Dissertation of Kunka[14], but also include a great deal of additional material. We summarize below the principal points of those three papers, all of which we expect to submit for publication by the end of the summer of 1996.

- In Part I we first carry out a formal asymptotic expansion for small Peclet number, P, and determine where such an assumed expansion ceases to be consistent. For a dendrite that is initially Ivantsov-like in the far-field, we show that there are three asymptotic regions at small P, with different governing equations and scales to the leading order. For an $O(1)$ region around the tip, the temperature is harmonic to the leading order in Peclet number, with boundary and far-field conditions are very similar to those for the problem of unstable viscous fingering in a Hele-Shaw cell. When initial shape deviation (not necessarily small or localized) from an Ivantsov parabola is confined to the tip region, the temperature and interfacial shape in other regions, to the leading order, remains that given by the Ivantsov solution-at least so long as the time, $t \ll P^{-1}$. For t satisfying this constraint, the growth and advection of initially localized disturbances superposed on an arbitrary time evolving state is investigated, in both linear and nonlinear regimes. The linear results essentially agree with previous well known results of Barber, Barbeiri & Langer[15], though careful examination of the fully nonlinear problem indicates that the results of the linearized analysis give a false impression of the dynamics of dendrite evolution. That is because the dynamical equations for the interface evolution without surface energy are mathematically ill-posed, so it cannot be deduced that the solutions for surface energy tending to zero are necessarily solutions of the zero-surface-energy equations[16,17].

To understand and predict dendritic evolution properly, one is therefore, as in the Hele-Shaw case, to investigating the dynamics in an extended complex plane, where the zero-surface-energy equations are well-posed[18]. The complex plane specification of initial conditions, while apparently artificial from the viewpoint of an experimentalist who is only in a position to determine initial interface shape to a finite precision, has the theoretical advantage of removing all sensitivity of the dynamics to initial condition. In this formulation, the actual results of an experiment are to be understood by studying a random ensemble of initial conditions in the complex plane subject to the constraint that the corresponding initial interface shapes for each of these initial conditions are the same interface shape, to within errors of measurement.

- In Part I, we restrict discussion to the various features of the zero-surface-energy dynamics in the complex plane, and the corresponding features observed at the interface. In terms of complex-plane zero-surface-energy dynamics involving particular classes of singularities, we also present a possible mechanism for nonlinear coarsening. However, in Part II, surface-energy effects are considered in the extended complex plane, examining in particular the nature of the singularity structure at finite, but small, surface energy, and the response of the dendritic interface as singularities approach the real domain.

- In Part III, we identify and amplify a specific mechanism for how dynamics in the limit of small surface energy differ from the zero-surface-tension dynamics. We also consider the effect of anisotropy on the impact of 'daughter singularities'. We determine that, unlike the case for isotropic surface energy, where no locally-steady tip region forms, the impact time of a daughter singularity can theoretically predict the formation of a locally-steady tip region, at the same time that the rest of the dendrite continues to be unsteady. A scaling analysis complements a highly accurate computation that supports the scalings, in the context of both the dendrite and the mathematically analogous Hele-Shaw finger, with anisotropic surface energy.

References

1. R. A. Brown (1988) *AICHE J.* **34**, 881.
2. D. Kessler, J. Koplik & H. Levine (1988) *Adv. Phys.*, **37** 255.
3. P. Pelce (1988) *Dynamics of Curved Fronts*, Academic Press, NY. 4. W. Kurz & R. Trivedi (1990) *Acta Metall. Mater.* **38**, 1.
5. E. Coutsias & H. Segur (1991) In *Asymptotics Beyond All orders*, NATO ARW Proceedings (Ed. Segur et al), Plenum.
6. J.J. Xu, (1991) *Phys. Rev. A* **43**, 930.
7. S. Tanveer (1994) *Phys. Fl* **6**, 2270..
8. S. Tanveer (1995) *Phys. Fl* , under review.
9. M.R. Foster (1996), submitted to *J. Crystal Growth.*
10. G. Koester (1996) PhD Dissertation, The Ohio State University.
11. S. Tanveer (1993) *Phil. Trans. R. Soc. London A* **343**, 155.
12. K. Brattkus & S.H. Davis (1988) *J. Crystal Growth* **91**, 538-556.
13. G. R. Baker & D. Vompe (1996), in preparation.
14. M. Kunka (1995) PhD Dissertation, The Ohio State University.
15. Barber, Barbieri & Langer (1987) *Phys Rev A* **36** 3340.
16. M. Siegel & S. Tanveer (1996) *Phys Rev Lett*, to appear.
17. M. Siegel, S. Tanveer & W.S. Dai (1996) *J. Fluid Mech*, to appear.
18. G.R. Baker, Michael Siegel & S. Tanveer (1995) *J. Comp. Phys.*, **120**, 348.

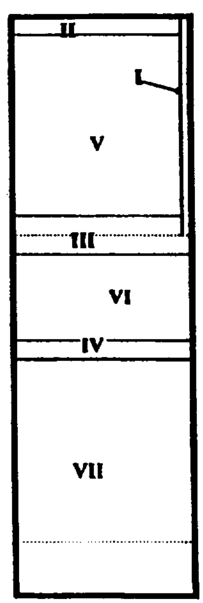

Figure 1 Flow structure in the Bridgman ampoule at large Rayleigh number and small Biot number, B. If Ra represents the larger of the thermal and solutal Rayleigh numbers, then the region I boundary layer scales with $Ra^{-1/4}$, and regions II, III and IV scale with $Ra^{-1/6}$. The solid occupies region VII. The flow in V and VI is inviscid but diffusive. The deflection at the IV-VII interface scales with either $Ra^{-1/6}$ (Solutal dominance[8,9]) or with $BRa^{-1/6}$ (Thermal dominance[7,9]).