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DYNAMICS AND PATTERN SELECTION AT THE CRYSTAL-MELT INTERFACE

Progress Report #5

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PI: Herman Z. Cummins

Research Foundation of CUNY
City College of the City University of New York

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DYNAMICS AND PATTERN SELECTION AT THE CRYSTAL-MELT INTERFACE

DOE Grant #DE-FG02-84ER45132

A. Progress Report for the Current Grant Period (Beginning March 1, 1990)

1. Dendritic Sidebranching

We completed a study of the sidebranching dynamics of dendrites in a succinonitrile-acetone mixture following the application of a single brief laser pulse close to the tip. Analysis of the interface profile by digital video image analysis showed that the perturbation grows with the $1/4$ power of distance along the dendrite, in agreement with the theoretical predictions of Pieters and Langer.[Ref.3]

Because the image analysis was complicated by the weak optical contrast between solid and liquid, we have explored an alternative binary system, rhodamine 6G dye in succinonitrile, which yields very high contrast images. Characterization of this system is in progress, and preliminary runs of periodically perturbed dendritic growth are just beginning.

2. Anisotropic Surface Tension

We are able to trap isolated microcrystals of succinonitrile inside our cylindrical high precision thermostat, capture video images, and extract the interface coordinates digitally. We have also developed computer programs which compute theoretical profiles and fit to the experimental data. This procedure yields preliminary numerical results for the surface tension anisotropy of about 0.7%, in the same range as the only published value.

3. Light Scattering at the Crystal-Melt Interface

We completed our study of the growth of bubbles during prolonged solidification runs in salol [Ref. 1]. Subsequently, we extended our studies to a new material, succinonitrile, which also exhibits intense dynamic interfacial scattering. We have explored the effects of purification by distillation and zone refining, of vacuum pumping and the intentional injection of gases. Currently, we are studying the angular dependence of the scattered intensity.

4. Succinonitrile-Dye Mixtures

We began a study of two new two-component systems recently: succinonitrile-rhodamine 6G, and succinonitrile-coumarin 152. These mixtures produce solid-liquid interfaces with excellent optical contrast which is ideal for our digital image analysis procedures. In order to characterize these binary systems, we have carried out measurements of their phase diagrams, diffusion constants, and partition coefficients.

B. Research Plan for the Next Grant Period

1. Dendritic Sidebranching

Theoretical analysis of the sidebranching instability has shown that sidebranches result from selective amplification of noise present at the tip, within a relatively narrow range of frequencies. We will carry out a systematic study of this effect by utilizing a periodically pulsed laser perturbation, and studying the response as the pulse frequency, pulse amplitude, and growth speed are varied. Preliminary measurements have been performed which indicate that rhodamine-succinonitrile is well suited for this study. We are currently developing computer programs needed for extracting appropriate interface correlation parameters from the digital images.

2. Anisotropic Surface Tension

Because anisotropy plays a central role in current theories of pattern formation, we plan to refine our current succinonitrile experiments to produce a more precise anisotropy value, and to then carry out numerical studies of the microscopic solvability theory to test the consistency of its predictions, and to compare these with experimental values for dendritic tip radius and growth velocity. Subsequently, we plan to extend these experiments to two other materials: pivalic acid and camphene.

3. Light Scattering at the Crystal-Melt Interface

First, we will complete our study of the angular intensity of scattering in order to resolve a controversy of whether $I(\theta)$ is best described by the Mie form or by the Ornstein-Zernike form as reported by Bilgram. We will explore the effect on $I(\theta)$ of changing the bubble size by pumping out residual gas.

Subsequently we will study the systematics of onset radius and delay time as functions of growth velocity. Our ultimate goal is to pin down the primary determinant of the bubble radius at onset. This part of the program should be completed during the next grant period.

4. The Planar to Cellular Transition

Directional solidification of a binary alloy leads, with increasing growth velocity V , to a morphological instability of the planar interface. Theoretically, the transition from planar to cellular morphology can be either supercritical or subcritical, depending on the relevant material parameters. Two previous experimental studies have shown subcritical bifurcations, based on hysteresis in response to changes in V .

We believe that such experiments are inconclusive, however, because steady state is not easily achieved. We have designed an alternative experiment in which growth will be stabilized near the threshold of linear stability, and a localized laser pulse will be applied to perturb the interface. The growth or decay of the perturbation will then directly test the sub- or supercritical nature of the instability.

In this experiment, we plan to use one of the dye-succinonitrile systems currently under investigation in our laboratory.

D. Publications and Presentations

1. **Direct Observation of Microbubbles in Directional Solidification of Salol**, L.M. Williams, M.R. Srinivasan and H.Z. Cummins, *Phys. Rev. Lett.* 64, 1526 (1990).
2. **Pattern Forming Instabilities at the Crystal-Melt Interface**, H.Z. Cummins in: Fundamental Problems in Statistical Mechanics VII (Altenberg, F.R.G. 18-30 June 1989), edited by H. van Beijeren (Elsevier Science Publishers, 1990) p. 11.
3. **Dendritic Sidebranching Initiation by a Localized Heat Pulse**, X.W. Qian and H.Z. Cummins, *Phys. Rev. Lett.* 64, 3038 (1990).
4. **Dynamic Light Scattering at the Nonequilibrium Crystal-Melt Interface**, H.Z. Cummins and L.M. Williams, in: Light Scattering by Liquid Surfaces edited by D. Langevin (Marcel Dekker - in press).
5. Papers presented at the March 1990 meeting of the American Physical Society
 - a) Investigation of the Origin of Dynamic Light Scattering at the Crystal-Melt Interface of Salol. L.M. Williams and H.Z. Cummins
 - b) Experimental Observation of Dendritic Sidebranch Generation in Response to a Pulsed Perturbation. X.W. Qian, M. Muschol and H.Z. Cummins
6. Abstracts submitted for presentation at the March 1991 meeting of the American Physical Society
 - a) Experimental Studies of Dendritic Sidebranches Generated by Periodic Perturbations in Succinonitrile-dye Systems. X.W. Qian, M. Muschol and H.Z. Cummins
 - b) Measurements of the Angular Intensity Dependence of Dynamic Light Scattering at the Crystal-Melt Interface in Succinonitrile. L.M. Williams and H.Z. Cummins
 - c) Surface Tension Anisotropy of Dendritic Growth Materials. M. Muschol, X.W. Qian and H.Z. Cummins

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