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[The Physics of Pattern Formation at Liquid Interfaces]

A SURVEY OF RECENT AND CURRENT WORK ON THIS GRANT

In this report we describe the recent research done under this grant and, for each topic, we prepare the way for the discussion of proposed future work which will be presented in a later report.

A. Pattern Formation at Liquid Interfaces

Much has been learned in recent years about pattern formation in Hele-Shaw cells. In particular, the onset of the Saffman-Taylor instability has been carefully studied as has the shape and stability of the steady state in Saffman-Taylor flow[1,2,3]. However, little is known about how the initial pattern, formed on the initially flat interface, develops into the steady state. This intermediate region of pattern formation is quite important to understand, not just to complete our knowledge of this simplest of pattern-forming problems but also because many of the interesting pattern formation problems have no well defined steady state and are effectively in the intermediate regime forever (with interesting questions of whether there are universal aspects (intermediate asymptotics) to these transients[4]). In addition, it is important to make connection between simple pattern formation problems and the more commonly occurring cases of interest for materials technology. Most pattern formation problems of technological interest have something in common with Saffman-Taylor flow, at least in the linear regime[5]. The differences between problems normally involve extra terms added to the basic nonlinear differential equation which governs the growth of an interface. Thus our recent research along with the work to be proposed in Chapter III is aimed at the two goals of understanding the nonlinear growth regime and discovering the effect on the

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pattern forming process of adding well-controlled terms to either the growth equation or to the boundary conditions.

During the past year we have reported the results of four projects[3,6,7,8] aimed at different aspects of the development of nonlinear patterns. (Reprints of these papers are attached as appendices 1,2,3,and 4.) We give only brief summaries of these results here (referring the interested reader to the appended reprints), along with somewhat more detailed summaries of two very recent projects which have not as yet led to publications.

1. A racetrack for Competing Viscous Fingers.

We summarize the results[6], since they are available in Appendix 1: we formed an array of approximately equal-sized viscous fingers in narrow channels and brought these fingers simultaneously into a wider Hele-Shaw cell. This allowed us to study the dynamics of finger competition. We observed several regularities: 1) there was a strong tendency, predicted by Kessler and Levine[9], to kill off every other finger very early in the pattern development. While the problem is noisy, and killing off only a third of the fingers in the first stage is a real possibility, the most probable pattern for a spatial correlation function is that of Figure 2b of appendix 1. 2) There was a second stage of competition wherein every other one of the fingers which had survived the first stage were typically killed off. This suggests a process which proceeds in stages of approximately locally-dominated interactions, killing off roughly every other finger until only one finger survives. 3) We could construct trajectories (position vs. time, in dimensionless units for which we at present have no theoretical justification) for the fingers which finish in each place in the race. These trajectories gave a good account of the finger patterns for all observed flow conditions, independent of which of the initial channels contributed the finger which finished the race in the given position. This suggests that there are universal features to intermediate stages of the pattern evolution and that we can give an interesting phenomenological account of the pattern left behind as the winning finger moves on to form the steady state.

2. An Experimental Realization of Periodic Boundary Conditions.

Side-wall interactions are known to be quite important in Saffman-Taylor flow. The walls of the cell not only set the length scale of the eventual steady state finger[1,2] but they also provide a source of friction which causes the initial fingers near the wall to grow earliest[1,3]. Since the latter of these effects would not be included in normal theoretical formulations with periodic boundary conditions, it is of interest to try to eliminate side-wall-friction effects in at least one experiment to learn how profoundly the flows are being affected by wall friction. We have constructed a cell which, to a reasonable approximation, realizes periodic boundary conditions. This cell is an annulus formed by joining two coaxial right circular cylinders of glass. The cell is 26 cm long and 3.6 cm in diameter, with a gap of 1 mm. We filled the cell with a critical mixture of isobytyric acid + water and initiated the flows at temperatures where the mixture was in the two phase regime by abruptly rotating the (vertical) axis of the cell through 180 degrees. We placed four mirrors to allow observation of the parts of the circumference of the cell which were not visible on a direct line of sight and obtained photographic images of the cell and the four mirrors with a tv camera. After digitizing these images and mathematically projecting them back onto the surface of the original cylinder (thus forming patterns of interface position as a funtion of azimuthal angle), we could make direct comparison of these flows with our earlier work on flows in a plane rectangular cell under otherwise identical conditions. Figure 1 shows a typical pattern. The results of the experiment are as follows: 1) most features of the flow are unaffected by the change to periodic boundary conditions. 2) Since our annular cell was much longer than our original rectangular cell, we were able to observe the pattern development to much later stages than was possible earlier. As the earlier work[3,8] hinted, here we observe no tendency for the length scale of the pattern to grow with time toward the expected steady state. Instead the number of fingers stays constant while the longest fingers pinch off behind their finger-tips, forming a rich variety of bubbles only after their widths have been reduced to less than the gap size (i.e., after the flow can no longer be regarded as two dimensional). This tendency of shorter fingers, to fatten and pinch off the tips of the leaders suggests a different dynamic from the one observed in high contrast flows where the shorter fingers stop growing altogether. Even though the same steady state should be available and stable for all viscosity contrasts, it could be that the mathematical zone of attraction of this steady state is much smaller for low contrast flows. None of our present results can distinguish between this possibility and the less interesting one that some as yet unforeseen three dimensional effect drives the weakly-driven low-contrast case away from the steady state. In the course of studying this problem, we have constructed a narrow cell whose width matches the wavelength of the initial pattern; this cell essentially forces the initial pattern to look like the steady state. Under these conditions we can form the steady state and see that is is indeed stable.

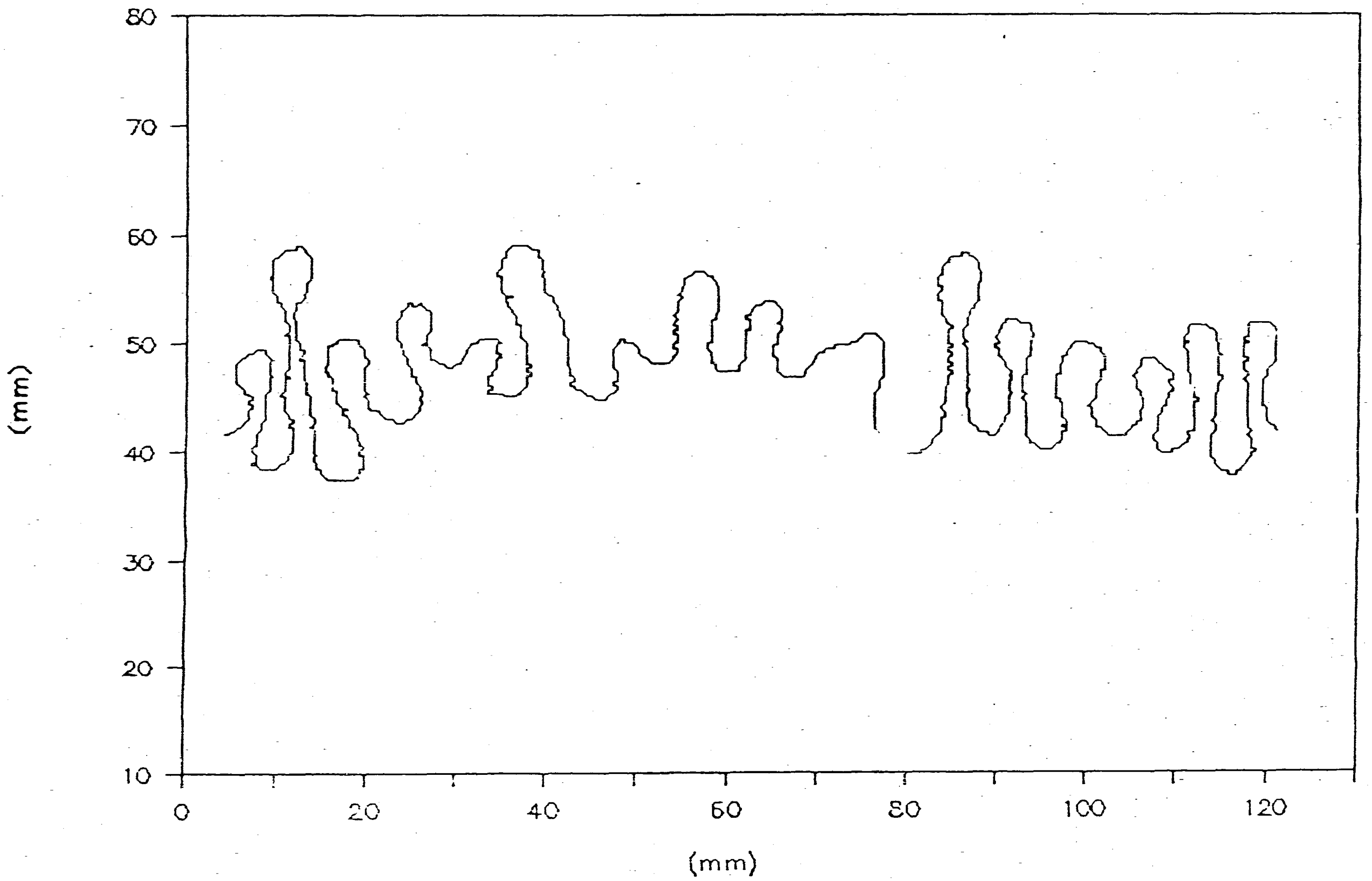

Figure 1: Pattern formed in an annular Hele-Shaw cell between the two phases of a critical mixture of isobutyric acid and water. The ordinate represents the vertical position of the interface while the abscissa is the position on the circumference of the annulus. This is a computer reconstruction of the pattern by mathematically projecting a tv view of the cell and 4 mirror images back onto the surface of the annulus.

3. What Sets the Length Scale for Patterns between Miscible Liquids?

The initial length scales for patterns between immiscible liquids are reasonably well understood in terms of a capillary length set by a combination of dynamics and the equilibrium interfacial tension[1,2,3]. However, important questions remain about the origin of length scales for patterns between miscible liquids, even though some of the most interesting patterns have been observed in such cases[10]. Using a maximum-entropy- production argument along with an assumption of zero interfacial energy for miscible Hele-Shaw flows, Paterson[11] has suggested that the length scale for the miscible case should be set by the gap in the Hele-Shaw cell. DeGennes[12] has considered the effect of polymer solution visco elasticity on length scales for the case of patterns between water and an aqueous polymer solution, a case which has been shown to show striking fractal patterns[10]. In DeGennes' formulation[12], the interfacial energy is also assumed to be zero or sufficiently low to be unimportant in the problem. An experiment to test DeGennes' formulation will be presented in Section III. A. 4 below. The experiment reported in the present section is relevant only to Paterson's formulation since it involves simple Newtonian liquids.

We have produced interfaces between miscible liquids by forming an interface between the two phases of a critical mixture of isobutyric acid+water at a temperature in the two-phase region and then raising the temperature into the one phase region. While the system comes to temperature equilibrium within a few minutes, the interface will persist and remain shiny for many hours because particle interdiffusion is so slow a process. We then initiate flow and measure the resulting fingering pattern as the interfacial advance proceeds too rapidly for diffusion to mix the liquids. (The rapid evolution of the pattern adds interface area while the interface continues to appear sharp, thus raising the possibility that the interfacial composition-gradient energy involved may lead to dynamical effects which act like an interfacial tension, even though the equilibrium state would have neither an interface nor a surface tension.) We find variations in the typical finger width, with a dependence on temperature and suggesting that the gap in the cell is not the controlling length scale. If this is indeed the case, a plausible explanation would be that the gradient energy which provides the most important contribution to the interfacial energy at two-phase equilibrium is still present until diffusion visibly blurs the interface and that the presence of this energy has an important effect on the pattern formation. Unfortunately, we were not able to vary this length scale enough to make a very convincing case that the cell gap is not the important length, and so rather than publish the experiment in its present form, we propose in the next chapter (Section III.A.3) an auxiliary experiment to measure the effective interfacial energy directly with surface light scattering.

4. The Fractal Dimension of Radial Hele-Shaw Patterns.

Several pattern-forming systems (e.g., electrodeposition and amorphous annealing) which have a mathematical kinship to radial Hele-Shaw flow have been shown to go over at late stages of their development into a space filling dense-branching morphology[13,14]. It would be of interest to understand the origin of this dense-branching transition and in particular to know which feature(s) of the differential equation governing the pattern growth give rise to the phenomenon. One group formulates the problem to say that dense-branching is already present in the underlying Hele-Shaw flow[15]. We have measured[7] the fractal dimension of high-contrast radial Hele-Shaw flows at constant flow rate over a reasonably large range of driving forces (roughly equivalent to looking at successively later stages of development) and have not seen dense branching. There are many details of importance to this problem, and our published paper is appended (appendix 2) for the interested reader. Our conclusion is that, for constant flow rate, and within the range of driving forces we can attain without beginning to distort the plates of our cell, we cannot see evidence of dense-branching. Yeung and Jasnow[16] have subsequently suggested that dense branching should not occur in Hele-Shaw flow at constant flow rate. Grier et al.[17] have suggested an explanation for the dense-branching seen in electrodeposition in terms of the finite conductivity of the solid phase. An analogy may exist in radial Hele-Shaw flow in which weakening the viscosity contrast between the fluids might take the Hele-Shaw problem over closer to the electrodeposition case, yielding a dense branching. In section III.A.6 below we propose an experiment to test this.

5. Detailed Analyses of Low-Contrast Saffman-Taylor Flows.

We have recently finished and published our extensive analysis of low contrast Saffman-Taylor flow[3,8]. The two resulting and interdependent papers are attached as appendices 3 and 4. The main results of these papers are as follows: 1) most features of the flows can be reproduced to within 20% even though the instability is broad-band. (Fluctuations in the Fourier transform of the developing pattern are, however, much larger if one looks at a single mode). 2) Several quantitative measures of the developing patterns collapse to characteristic curves when the results from flows at very different control conditions are put in the dimensionless form suggested by the Hele-Shaw equations. Wetting effects have kept this from happening in all observed high-contrast studies[1,2].

The results of these papers form a background of information for some of the more complicated pattern formation problems we wish to study in the future, but we do not plan to do any more work on this problem in its original, simple form.

B. Interfaces within Disordered Materials

A few years ago we found that random field theories gave a rather good account of several of the properties exhibited by swollen polymeric gels whose solvent was a binary liquid mixture with a nearby consolute point[18,19]. In a subsequent experiment[20] we attempted to make the random field behavior go away by making gels whose characteristic length for the random field (polymer spacing) would be larger than the solvent correlation length at all temperatures except those exceptionally close to the renormalized critical temperature. Interestingly, most of the light scattering behavior expected of the random field system persisted in these larger-pore-size gels. While it was hard to interpret this result because of the possibility that the gels did in fact have a very short characteristic length even though they had a greater incidence of large pores, the experiment also raised interesting questions about the behavior of demixing liquids inside a disordered medium. Recently we have been using dynamic light scattering to study how domains of phase separating liquid grow inside such a medium[21] and also to learn how wetting and adsorption layers are affected by the radius of curvature of the wall to which they are attracted (i.e., will the phase of the demixing liquid which has greater affinity for the polymer form a layer on the polymer strand much as it would if the polymer formed a flat wall?) A reprint of our work[21] on the first issue is attached as appendix 5, and this will not be discussed further here because we do not propose any follow-up experiments at the present time. The second issue has led us to study the behavior of polystyrene spheres at very low concentration in binary liquid mixtures.

1. The Wetting/Adsorption Properties of Polystyrene Spheres in Binary Liquid Mixtures

We have used static and dynamic light scattering to study the behavior of charge-stabilized polystyrene spheres in critical and off-critical mixtures of 2,6lutidine+water(LW). This binary liquid system has an inverted coexistence curve with a lower critical point near 34C. By tuning the system temperature we can make the liquid correlation length as large as 500nm (our temperature stability is 0.2mK over periods of a few hours and 2 mK over a few days). We have used polystyrene spheres of radii 30nm, 50nm, 250nm, 500nm and 1500nm, all at a very low volume fraction of 6×10^{-5}. This range of particles gives us great flexibility in varying the ratio of particle size to solvent correlation length. We know from the work of Pohl and Goldburg[22] that pure LW has a wetting transition for the walls of the glass sample tube near the demixion critical point, and we might also expect effects analogous to those seen in LW + glass beads by Esteve and Beyssens[23]. We can hope to avoid the flocculation effects seen by Esteve and Beyssens, and thus study wetting/adsorption layers on isolated spheres both because their glass particles are known to flocculate easily and because our charge-stabilized spheres should be rather unwilling to aggregate.

In our first attack on this problem (which we reported recently at the APS March Meeting in Anaheim) we have seen the following effects: 1) there is no tendency to aggregate in the one phase region at any LW concentration (on or off-critical). 2) As soon as the solvent enters the two phase region (the critical temperature is shifted by at most a few mK by the presence of the spheres), the upper phase (lutidine rich) expels most of its polyballs. The lower phase (water rich) takes on a slightly higher density of polyballs than in the one phase region, but most of the polyballs expelled from the upper phase go to the liquid-liquid interface and to a water-rich layer which coats the side wall of the sample cell around the edges of the upper phase. Near the critical point this water rich surface layer is only about 1 cm high (the upper phase is typically about 3 cm high) but the height of the coating layer grows as the system temperature is placed deeper in the two- phase region and eventually completely wets the glass. The existence of the wetting layer is no surprise in light of the work of Pohl and Goldburg[22]; however, this layer not only holds polyballs, but the polyballs pack in tightly and form a crystalline structure along the wall. The geometry of our samples has thus far prevented us from measuring the number of polyball layers packed against the glass wall, but we have measured the radius and thickness of the first ring of the characteristic transmission diffraction pattern and learned that the polyball microcrystals are ordered along the glass wall over at least 5 polyball diameters with a mean interparticle spacing of approximately one diameter. All of the above effects are equilibrium effects in the sense that they can be observed for many days after the sample has been brought to apparent equilibrium and that they will return reliably after the sample is stirred. There is a rich literature about molecular and particle ordering at interfaces and walls and between walls[24], but we are not aware of any other case where a layer has spontaneously formed in an ordered state between a wall and a free interface. 3) We have measured the effective sizes of polyballs in the water-rich phase by combining dynamic light scattering (autocorrelation function measurements) with drawing off and measuring the viscosity of the lower phase from a pure liquid sample. This has shown no change in apparant polyball size from that seen when the same size polyballs are measured as they move diffusively in pure water (both measurements agree very well with the manufacturer's stated size which was determined with electron microscopy). It is in the lutidine-rich phase that we hope to see evidence of an adsorption layer since this is the phase which clearly rejects the polyballs; we have not yet managed to extract reliable sizes for the effective scatterers in the lutidine-rich phase because their number density is so low. We hope soon to have results on this. 4) We observe peculiar effects in the apparant correlation length of the critical LW fluctuations near the critical point in the one-phase region. This is a difficult determination to make because the autocorrelation functions involve two exponential terms which have similar time constants for at least part of the temperature range for each size of polyball (i.e., there is one characteristic time from the diffusive decay of LW concentration fluctuations and another time from the Brownian motion of the polyballs). However, the data are good enough to give a rather good separation of the two signals over a significant temperature range, particularly for the largest polyballs. The resulting length scale for the composition fluctuations shows a power law in the reduced temperature with an exponent near 0.5 instead of the expected value of 0.62 (using a pure liquid critical mixture, we have no trouble reproducing the published value[46] of 0.58 for this system in our temperature range, whereas we measure 0.48 with 1500nm polyballs present). It is extremely unlikely that the critical exponent for the correlation length has really been changed by the presence of such a small concentration of polyballs (Fisher renormalization[25] would drive the exponent in the other direction and would not be expected to be observable until the polyball density was raised by several orders of magnitude[26]). Rather we suspect that a dynamical effect is involved, changing the effective viscosity for the fluctuating volumes which are trapped between polyballs whose separation is, at the smallest reduced temperatures, only about ten correlation lengths.

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