# Surface-Tension-Driven Nonlinear Instability in Viscous Fingers

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We show analytically and numerically that the linearly stable flat interface in the Saffman-Taylor problem becomes unstable against finite amplitude perturbations when the surface tension is modified in proportion to the local curvature. This nonlinear instability is identified with unstable saddle points in a flow diagram in a weakly nonlinear approximation. Numerical simulations confirm this prediction and reveal that this instability leads to spiky cellular patterns.

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In studies of the formation of patterns in nonequilibrium dissipative systems [1], it is essential to examine the stability of the advancing front against perturbations. Theoretically, the instability is usually detected by a linear analysis, and, to the best of our knowledge, most instabilities reported thus far, excluding perhaps turbulence, have been found essentially by this method. Examples along this line include the most widely studied instabilities in the pattern formation field, including the Mullins-Sekerka instability in solidification [2], the Saffman-Taylor instability in the viscous fingering problem [3], and the convective instability in the Bénard problem [4]. Even though these instabilities all give rise to very complex patterns, we recognize that they are all triggered by infinitesimal perturbations. We refer to these as *linear* instabilities, since one can essentially determine their existence by a simple linear analysis.

Often there is a control parameter, say $\beta$, with a critical value, $\beta_c$, beyond which the instability sets in. In some cases, nonlinear analysis reveals a finite amplitude instability when $\beta$ is below $\beta_c$, with a threshold amplitude which vanishes as $\beta$ reaches critical. Such an instability is still linear, at least in the sense that its existence can be detected by linear analysis, even though nonlinear analysis is necessary to show that it persists as a finite amplitude instability even when the interface is stable against infinitesimal perturbations.

In this Letter, we report an intrinsically nonlinear instability in the viscous fingering problem with a polymeric surfactant at the interface. Unlike the linear instabilities mentioned above, this instability always has a nonzero threshold amplitude, say $A_c$. Thus, a simple linear analysis will miss it completely. As a control parameter $\beta$ is increased, $A_c$ becomes smaller going to zero as $\beta$ approaches infinity, but the critical value is infinity—the instability is never a linear one. For large parameter values, the threshold amplitude is quite small, and so it is likely that the instability will in fact occur in the real system. Linear stability analysis, however, will predict that the interface should be stable, and will give no clue as to the nature of the instability that would be observed.

Considering the fact that all the known instabilities are linear ones [2-4], we find it unusual to discover such a finite amplitude instability in the context of the Saffman-Taylor (ST) problem, which continues to play an important role in pattern formation [1,5,6]. Numerical simulations confirm that the flat interface indeed becomes unstable and breaks into a cellular pattern. These cells, however, are *not* similar to those seen in directional solidification (DS); instead they are arrays of spikes, with the sharp points extending forward [Fig. 4(b)]. Thus the instability we have found is not an instance of the known analogy [7-9] between the ST problem and DS. Moreover, these patterns arise only as a result of a finite amplitude instability, whose onset can only be detected by a nonlinear analysis.

Our starting point is a set of equations of motion for the fingering instability in a linear Hele-Shaw cell with impermeable side walls. The dynamic field is the pressure $P$ that satisfies the Laplace equation everywhere in the cell $\nabla^2 P=0$, and the velocity is given by $v_n=-(\nabla\cdot \mathbf{P})_n$. The pressure at the interface is $P=-\gamma\kappa$, with $\gamma$ the surface tension and $\kappa$ the curvature. Suppose now that surface tension itself is locally proportional to the curvature $\kappa$. This might be realized by introducing diblock copolymers or surfactants, consisting of hydrophobic and hydrophilic groups, into the system. The effect of this new species at the interface on the dynamic instability might be complex. But in the limit where the segment length of one species is much larger than the other, it is entropically unfavorable to pack polymers in a region with negative curvature (Fig. 1) and so the polymer concentration might be locally perturbed in proportion to the curvature. Further, if polymer distribution relaxes on a time scale which is short compared to that of the interface motion, then the polymers will always be in

 

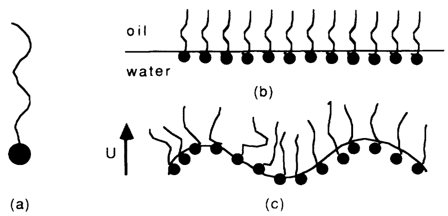


FIG. 1. (a) Polymers with hydrophobic tail and hydrophilic head group. (b) They arrange themselves at the oil-water interface. (c) Because of entropic reasons, the region of positive curvature (bump) has slightly more polymers where the surface tension is locally reduced, enhancing the instability. In this case, narrow fingers are observed. $U$ is the pushing velocity in the direction of the arrow. The nonlinear instability discussed in the paper occurs when oil pushes water.

local equilibrium and the surface tension, which depends on the concentration of polymer, will in turn depend on the local curvature. Ignoring higher-order terms in $\kappa$, the effective surface tension is then $\gamma' = \gamma + \beta\kappa$, so we have at the interface

$$P = -\gamma\kappa - \beta\kappa^2 . \tag{1}$$

A negative value of $\beta$ means that the surface tension is reduced in regions with positive curvature, which tends to enhance the fingering instability. If the system is reversed, so that the more viscous fluid pushes the less viscous fluid, the $\beta$ term helps suppress fingering. On the other hand, a positive $\beta$ tends to suppress the instability when the less viscous fluid pushes the more viscous one, but tends to produce fingering in the opposite situation.

With $\beta < 0$, instability is enhanced and we expect narrow fingers to emerge if the water pushes oil. We think this is the reason why the narrow finger was observed only for the *first* time when the cell was washed by soap containing surfactant molecules [10]. To be more specific, we briefly present the solvability analysis [1,5]. The function $\Psi(\eta)$ in the solvability function $\Lambda$ in Hong and Langer [5] will contain an additional term in the denominator, $[1+\alpha(1+\beta^2\eta^2)/(1+\eta^2)^{3/2}]^{1/2}$ with $\alpha = \beta\pi(1-\lambda)/\lambda^2\gamma = \epsilon(1-\lambda)^2 < 0$, because the surface tension $\gamma$ has been changed into $\gamma + \beta\kappa$. There is now a new branch point in the imaginary axis located at $z = \imath - \imath\delta/2$ with $\delta = [|\epsilon|(1-\beta^2)]^{2/3}$ and thus a narrow finger solution of $\lambda < \frac{1}{2}$ exists. The steepest contour that runs from $-\infty$ to $\imath$ must bend to include $z$, coming back to $\imath$, and then runs away to $-\infty$. Applying the resolution criterion described in Hong and Langer [5], we obtain the following scaling relation between $\alpha$, $\beta$, and $v = \pi b^2\gamma/12UW^2(1-\lambda)^2$:

$$v/\lambda^4 \approx |\epsilon|^{7/3}(1-2\lambda)^{1/3} . \tag{2}$$

Since $\beta$ enters through the $\kappa^2$ term, it will have no effect on the linear stability analysis and thus we must perform a nonlinear analysis. This can be done in the weakly nonlinear regime, in which the linear growth of the most unstable (or least stable) mode is small. The weakly nonlinear regime can be realized by making $k_c = 1/\sqrt{\gamma}$ only slightly larger than $\pi/2$, the smallest perturbation wave number allowed by the boundary conditions, where $k_c$ is the most dangerous wave number obtained by the linear analysis. By adjusting the surface tension $\gamma$ via polymer concentration at the planar interface, the weakly nonlinear regime can be realized. In this limit, all modes except the lowest one will be linearly stable, and since the linear growth rate of the lowest mode is small we expect that it will saturate at a small amplitude. We may then expand the interface position $\xi(y,t)$ in Fourier modes, $\xi(y,t) = t + \sum_n \xi_{\text{in}}(t)\cos[nq(y+1)]$, with $q = \pi/2$. The pressure field can be expanded similarly,

$$P(x,y,t) = -(x-t) + \sum_n P_q(t)e^{-nq(x-t)}\cos[nq(y+1)] .$$

The next step is to impose the two boundary conditions at the interface and expand in the amplitudes to obtain the evolution equation for the $\xi_{\text{in}}(t)$.

Our most interesting results are obtained in the case when the *more* viscous fluid pushes the *less* viscous one, for which the amplitude equations can be obtained from those usual cases of the less viscous fluid pushing the more viscous one by simply replacing $t$, $\gamma$, and $\beta$ by $-t$, $-\gamma$, and $-\beta$. After expanding to third order and truncating to the lowest two modes, we obtain

$$\dot{x} = -(1+\Gamma)x - (1+\Gamma+3B)xy + (1+5\Gamma/2)x^3 + (1/2+13/2-3B)xy^2 , \tag{3a}$$

$$\dot{y} = -2(1+4\Gamma)y - 3Bx^2 + 4(1+4\Gamma)x^2y + 2(1+7\Gamma)y^3 , \tag{3b}$$

where $x = q\xi_1/2$, $y = q\xi_2/2$, $\Gamma = \gamma q^2$, and $B = 4\beta q^3/3$ and the overdot represents a rescaled time derivative, $q^{-1} \times d/dt$. We now seek for the fixed points of this pair of equations. There is always one trivial fixed point $(x,y) = (0,0)$, corresponding to the linearly stable flat interface solution. There are, however, additional unstable fixed points. Two are located on the $y$ axis, at $(x,y) = (0, \pm a)$ with $a^2 = (1+4\Gamma)/(1+7\Gamma)$, and the other fixed points are found by solving

$$x^2 = [(1+\Gamma) + (1+\Gamma+3B)y - (1/2+13\Gamma/2-3B)y^2]/[1+5\Gamma/2] = 2y[(1+4\Gamma) - (1+7\Gamma)y^2]/[4(1+4\Gamma)y - 3B] . \tag{4}$$

Cross multiplying yields a cubic in $y$, so there exist at most up to three additional fixed points. However, $x$ must be real and positive, and so not all three solutions of the cubic will necessarily be solutions of (4).

We first consider the limit $B \ll 1$, flow diagrams of which are shown in Fig. 2. The unstable fixed points alternate between saddles and repellers, and so the insets of

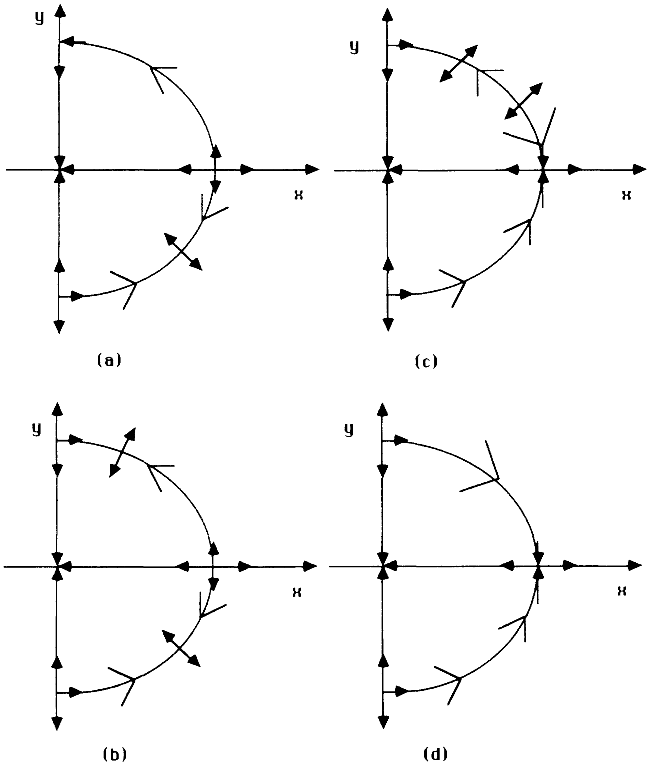


FIG. 2. Flow diagrams for the two-mode approximation (3) with boundary condition given by (1). $\Gamma$ is a control parameter that contains surface tension, which increases from (a) to (d): (a) $0 \le \Gamma < 0.608$, (b) $0.608 \le \Gamma < 2$, (c) $2 \le \Gamma < 3.314$, (d) $3.314 \le \Gamma$.

the saddles mark the limit of a finite amplitude instability of the flat interface fixed point. Experimental results have always indicated that a flat interface is completely stable when the less viscous fluid is displaced by the more viscous one, so it is clear that the threshold instability is quite large in practice. It might be interesting to devise external perturbations that could trigger this instability.

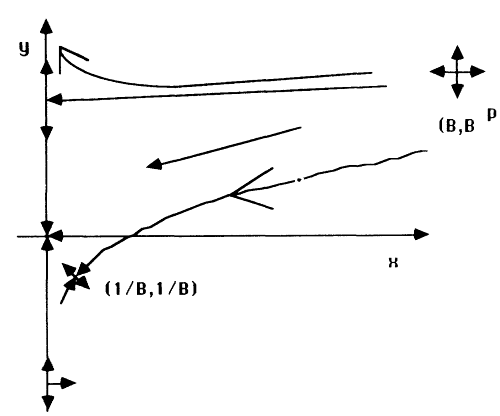


FIG. 3. Flow diagram with $B \to \infty$. Note the appearance of two unstable fixed points: one at infinity $(B,B^p)$ with $p=\frac{3}{2}$ and the other one near the origin $(1/B,1/B)$. In the limit $B \to \infty$, the flat interface is thus unstable to finite amplitude perturbations.

As $B$ increases, the fixed points which delimit the basin of attraction of the flat interface fixed point will move. Their behavior for large $B$ is quite interesting. In the asymptotic limit of $B \to \infty$, (4) has two solutions. One has $x \approx B$ and $y \approx B^{3/2}$; the other one is near the origin, with both $x$ and $y$ of order $1/B$. The former is a repeller while the latter is a saddle. The flow diagram is shown in Fig. 3. Note that the distance between the flat interface and the new nearby fixed point is finite for any finite $B$, even though it approaches zero as $B$ increases. Therefore, the flat interface is only unstable to a finite amplitude perturbation, although the threshold amplitude goes to zero for large $B$. For a sufficiently large $B$, it is then pos-

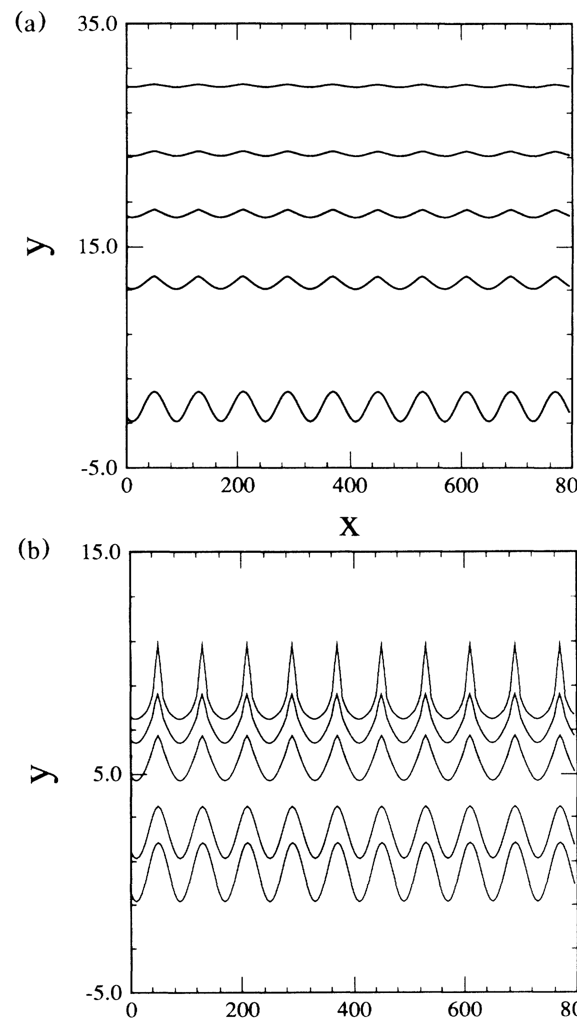


FIG. 4. Simulations with $\beta \neq 0$. (a) For small $B=-200$, the interface is stable. (b) For large $B \approx -400$, cellular patterns with sharp tips show up. The parameters used in this run are $q=10$, $U=-0.025$, $\gamma=1.0$, $B=-400$, and amplitude $A=0.8$.

sible that fluctuations may be large enough to trigger the instability. Note that this instability can *only* be found theoretically through nonlinear analysis; linear stability analysis can never even hint at its existence. The criterion for the occurrence of this nonlinear instability is $B \gg \Gamma$ or equivalently $\beta q \gg \gamma$. It is not difficult to satisfy this criterion experimentally by controlling, say, surface tension.

In order to test the predictions of the above analysis, we have carried out careful numerical simulations, integrating the governing equations forward in time. The numerical code used for this simulation is a modification of that reported in Ref. [11]. As initial conditions, we perturb the flat interfaces by a sinusoidal function of a given wave vector $q$. The time evolution of this perturbation is then followed. For fixed external parameters such as the pushing velocity and the surface tension, the interface is stable for small values of $B$ or small values of the amplitude of perturbation [Fig. 4(a)]. It becomes unstable, however, for large $B$ ($\approx -400$) and breaks into cellular patterns [Fig. 4(b)]. Note that the patterns are quite different from those seen in directional solidification in that the shape around the tip of each unit is quite spiky. Fixing the value of $B$, we observed similar behavior by increasing the amplitude of the initial perturbation. Finally, fixing both $B$ and the initial amplitude, we observed this instability by increasing the initial wave vector $q$. These observations are in qualitative agreement with the results of the weakly nonlinear analysis. Note that these cellular patterns arise because of changes in the surface tension, and thus we expect that any perturbation that changes the pressure boundary condition, like (1), could produce the same effect [12].

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