

## Pearling and Pinching: Propagation of Rayleigh Instabilities

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A new class of propagating fronts is proposed in which a spreading instability evolves through a singular configuration before saturating. As an example, we examine the viscous Rayleigh instability of a stationary fluid column, using the marginal stability criterion to estimate the front velocity, front width, and the selected wavelength in terms of the surface tension and viscosity contrast. Systems that may display this phenomenon include droplets elongated in extensional flows, capillary bridges, liquid crystal tethers, and viscoelastic jets. The related problem of propagation in Rayleigh-like systems that do not fission is also considered. [S0031-9007(97)02798-1]

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Front propagation problems are often divided into two broad classes: the invasion of one stable state by another, and the invasion of an unstable state by a stable one [1]. Here we point out the possibility of a third, qualitatively different type of front propagation: the invasion of an unstable state by one that develops discontinuously into its final configuration. Although we expect this class to be very broad, we focus on one particular example, the Rayleigh instability, wherein a stationary cylindrical body of fluid longer than its circumference is linearly unstable to breakup into droplets [2,3].

Recent work has shown Rayleigh instabilities in many experimental systems: laser-tweezed cylindrical lipid vesicles [4], the instability of liquid crystal tethers embedded in a polymer matrix [5], the peristaltic growth of shrunken polymer gels [6], and the dynamics of columns of viscoelastic fluid [7]. Traditional experiments on jets display an instability developing uniformly along the cylinder [8]. However, experiments on the “pearling instability” of laser-tweezed membranes reveal a different scenario—*front propagation*. The peristaltic shape deformation spreads out from the laser spot with a constant velocity [9]. The emerging explanation [10–12] for this instability is that it is driven by the tension induced in the membrane by the laser trap. Fissioning is ultimately prevented by the membrane elasticity.

Motivated by the phenomenology of the pearling instability, we ask, “Can the Rayleigh instability develop as a propagating front?” (as imagined in Fig. 1). Propagation in this context is problematic. As a droplet pinches off, the two tips of the broken neck recede from the pinching point; if the retracting neck overtakes the front, propagation will be spoiled. Classic experiments on the breakup and relaxation of elongated droplets [13] reveal that this competition depends on the viscosity contrast of the two fluids. This retraction and topological rearrangement of the interface is absent in more familiar systems exhibiting propagation, such as dendritic growth, viscous fingering, and reaction-diffusion systems. The mathematical structure of our problem differs from those just mentioned as

well, with a global conservation law from fluid incompressibility, nonlinearities from the two principal surface curvatures, and singular behavior near pinching. The former two features are also common to systems that, like membrane tethers, do not fission.

The novel aspects outlined above complicate analytical and numerical study of this problem. As a first step, we explore the hypothesis that the marginal stability criterion (MSC) [14] applies to the Rayleigh problem. The MSC states that the asymptotic front speed and shape are those of the marginally stable front; slower fronts are stable, faster fronts are unstable. Our results on front velocity, front sharpness, and selected wavelength should serve as a guide for further experimental and theoretical investigation. At issue are both the general question of propagation and the validity of the MSC, which in so many systems correctly predicts the front properties.

The three categories of front motion are illustrated by generalized Fisher-Kolmogorov (FK) equations [15],

$$u_t = -\frac{\delta \mathcal{E}}{\delta u}, \quad (1)$$

where  $\mathcal{E} = \int dx [u_x^2/2 - \mathcal{V}(u)]$ , with three types of potentials shown in Fig. 2. Recall the standard mechanical analogy for traveling-wave solutions: the ansatz

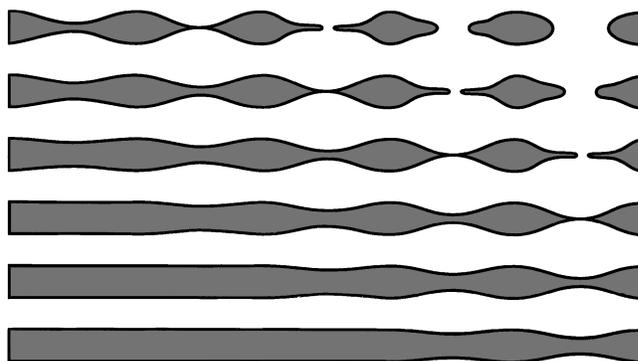


FIG. 1. Artist's conception of a propagating Rayleigh instability.

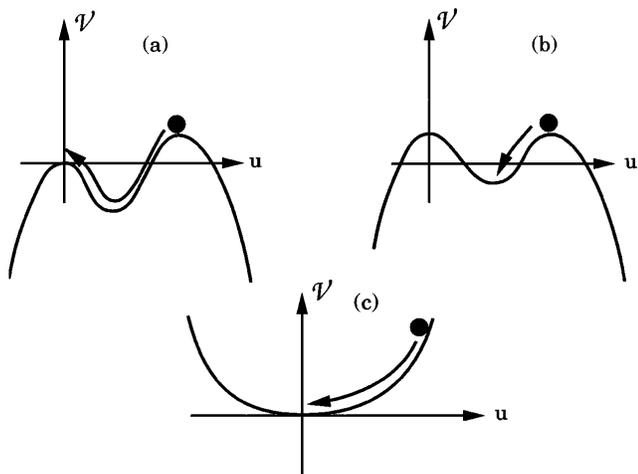


FIG. 2. Potentials illustrating different classes of front propagation in the mechanical analog for traveling-wave states.

$u(x, t) = f(x - vt)$  reduces (1) to Newton's equation for a particle of coordinate  $u$  at time  $z = x - vt$  in a potential  $\mathcal{V}(u)$  with friction coefficient  $v$ . In case (a) there is a unique  $v$  that allows the front to connect the two locally stable states. When a stable state invades an unstable one (b), the particle reaches  $u = 0$  at  $z = \infty$  for every friction coefficient  $v > 0$ . However, for sufficiently localized initial conditions, there is a unique front velocity that appears asymptotically, and is correctly predicted by the MSC. Finally, (c) displays a situation loosely analogous to the new category. A potential of this form, e.g.,  $\mathcal{V}(u) = u^2/2 + u^4/4$ , leads to a finite-time blowup of  $u$  [16]. Although this model does not describe a topological transition, it is like the Rayleigh problem in which the growing instability has a finite-time singularity.

This mechanical analogy suggests that case (c) would, like (b), possess a family of possible velocities. With this motivation, we examine the Rayleigh problem using the *linear* MSC, which yields predictions directly from the linear growth rate  $\omega(q)$  [17]. The front velocity,  $v^*$ , and the shape of the front's leading edge,  $u(x, t) \sim$

$\exp[\omega(q^*)t + iq^*x]$ , are given by the relations [14]

$$v^* = \frac{\text{Re } \omega^*}{\text{Im } q^*}, \quad \text{Re } \frac{\partial \omega^*}{\partial q} = 0, \quad -v^* = \text{Im } \frac{\partial \omega^*}{\partial q}, \quad (2)$$

where  $\omega^* = \omega(q^*)$  and  $q^* = q' + iq''$  is complex. In the standard MSC treatment, the selected wave number  $q_0$  of the saturated pattern is different from the selected wave number  $q'$  in the leading edge of the front;  $q_0$  is deduced by working in the rest frame of the front, and assuming that nodes are conserved as they pass through the front and into the saturated pattern. We do not expect  $q_0$  to be relevant to the Rayleigh problem since the evolution of the interface is not continuous.

The conservation of fluid in the Rayleigh problem is like that appearing in the invasion of the disordered phase of a block copolymer by the lamellar phase [18], with global conservation of the mean composition. In contrast to the FK equation, this leads to a dynamics of the form  $u_t + j_x = 0$ , with the flux  $j = -\partial_x(\delta \mathcal{E}/\delta u)$ , or

$$u_t = \frac{\partial^2}{\partial x^2} \frac{\delta \mathcal{E}}{\delta u}. \quad (3)$$

Here we focus on the *absolute* instability of a stationary fluid column; elsewhere we discuss the *convective* instability of a jet. Consider a thread of fluid of radius  $R$  and viscosity  $\eta^-$  in another fluid of viscosity  $\eta^+ = \eta^-/\alpha$ , with interfacial tension  $\Sigma$ . The growth rate of a volume-preserving axisymmetric sinusoidal perturbation with wave number  $q = k/R$  follows from the Stokes equations  $\eta^\pm \nabla^2 \mathbf{v}^\pm = \nabla p^\pm$ , subject to the boundary conditions of no-slip, no tangential stress, and the Laplace law for the jump in normal stress,  $(\sigma_{ij}^+ - \sigma_{ij}^-)n_j = -2\Sigma Hn_i$ . Here  $n_i$  is the outward surface normal,  $H$  is the mean curvature  $2H = r_{xx}(1 + r_x^2)^{-3/2} - r^{-1}(1 + r_x^2)^{-1/2}$ , and the stress tensors are  $\sigma_{ij}^\pm = \eta^\pm(\nabla_i v_j^\pm + \nabla_j v_i^\pm) - p^\pm \delta_{ij}$ . The growth rate is [19]  $\omega(k, \alpha) = (\Sigma/R\eta^+)\Lambda(k, \alpha)(1 - k^2)$ . The dynamical factor  $\Lambda(k, \alpha)$  (too lengthy to quote here) accounts for the dissipation, and the factor  $\Sigma(1 - k^2)$  is associated with the energy of a distortion.

We gain intuition about  $\Lambda(k, \alpha)$  from its small  $k$  form:

$$\Lambda \sim \frac{-k^2[-\frac{1}{8} + \alpha(\frac{1}{4} + \frac{1}{2}\gamma - \frac{1}{2}\ln 2) + \frac{1}{2}\alpha \ln k]}{\alpha - k^2(\alpha - 1)[\frac{1}{8} + \alpha(\frac{3}{4} + \frac{3}{2}\gamma - \frac{3}{2}\ln 2) + \frac{3}{2}\alpha \ln k]},$$

where  $\gamma$  is Euler's constant. For finite  $\alpha$  the asymptotic limit as  $k \rightarrow 0$  is  $\Lambda(k, \alpha) \sim -\frac{1}{2}k^2 \ln k$ . The logarithm is due to large-scale flows in the outer fluid, while the two powers of  $k$  are analogous to the two  $x$  derivatives in (3). They can be understood intuitively by noting that one power comes from the flux form of the dynamics,  $\pi \partial_t r^2 + \partial_x J = 0$  [12], with flux  $J \sim r^2 U$ , where  $U$  is the average axial velocity inside the tube. For small  $k$  the Stokes equations imply a Poiseuille flow of the inner fluid and thus the second power of  $k$  through Darcy's law for the average velocity,  $U \sim -r^2 p_x$ .

If we consider the case of  $\alpha = \infty$  (no outer fluid), we recover the small  $k$  limit of Rayleigh's stability analysis [3], which has a nonzero growth rate at  $k = 0$ . The inner fluid has plug flow since it does not have to entrain the outer fluid as the shape of the boundary changes. This, in turn, modifies the relationship between the average velocity  $U$  and the pressure so, for small  $k$ ,  $U_{xx} \sim p_x$  [20].

Figure 3 displays the results of the MSC applied to Tomotika's growth rate for a range of viscosity contrasts  $\alpha = \eta^-/\eta^+$  [21]. For example, the results for the castor-oil-eugenol mixture immersed in silicone fluid

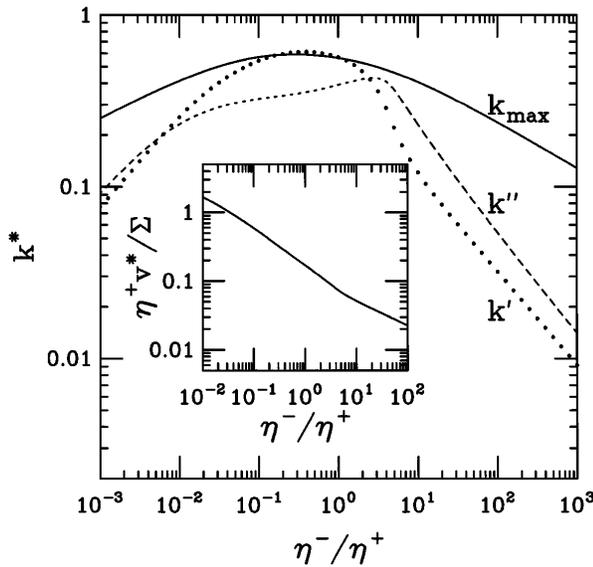


FIG. 3. Length scales and front velocity (inset) from the marginal stability criterion.

( $\alpha = 0.0315$ ) used in [22] are the following: the fastest-growing mode  $k_{\max} = 0.502$ ,  $k' = q'R = 0.404$ ,  $k'' = q''R = 0.291$ , and  $v^* = 1.04\Sigma/\eta^+$ . The MSC predicts a relatively sharp front; if we measure the front width in units of  $2\pi/k'$  we see that it is generically a fraction of a growing bulge, from about 0.25 at  $\alpha = 1$  to about 0.1 for large and small  $\alpha$ . We note also that  $v^*$  differs markedly from the naive estimate  $\omega(k_{\max}, \alpha)/k_{\max}$ . From these results we estimate the distance  $(k'/2\pi)[v^*/\omega(k')]$  the front moves in the time it takes a bulge to pinch off; it is about twice the front width for the range of  $\alpha$  shown, so the droplets pinch off right behind the front.

Now we crudely estimate the retraction velocity of the bulge at the end of a broken thread to see when we expect to find propagation. Assuming a spherical bulge, we balance the tension force with the drag of a liquid sphere of radius  $R$  [23] to deduce a velocity  $V = (\Sigma/\eta^+) \times (r/R)(\alpha + 1)/(2\alpha + 3)$ , where  $r < R$  is the radius of the neck attaching the bulge to the rest of the thread. Note that  $V$  does not depend sensitively on the inner viscosity. Comparing this expression with Fig. 3, we see that the front will outrun retraction at small  $\alpha$  but not for large  $\alpha$ . Therefore, we expect the MSC to describe front propagation at small  $\alpha$  if it applies at all [24].

These predictions are consistent with the experimental results of [13], which concern the relaxation and breakup of an initially elongated drop in an otherwise quiescent fluid. It is found that for large  $\alpha$  the bulbs on the ends of the dog-bone shaped droplet retract a substantial amount before they pinch off (if they pinch off at all); for lower values of  $\alpha$ , the retraction is relatively slower and the ends do not move as much before they pinch off. As alluded to above, this behavior occurs because the outer viscosity limits the retraction velocity, and the inner viscosity inhibits pinching [13]. The experimental results at low  $\alpha$  are tantalizingly close to our conception

of a propagating Rayleigh instability. We note that the mechanism of “end pinching,” in which the end of the extended drop bulges and pinches off while the rest of the drop is stationary [13], is consistent with the MSC prediction of a sharp front. It remains to be seen if these disturbances spread at a *constant* velocity [24].

Since the applicability of the MSC to our problem is speculative, it is important to check our predictions. A numerical computation of the interface evolution in the fissioning Rayleigh problem is a challenging task. As a first step to test the internal consistency of the MSC we studied a model partial differential equation for a Rayleigh-like instability that approaches pinching, with an energy functional with an elastic contribution such as that for membranes. Using a Poiseuille flow approximation to relate the fluid flux to the pressure gradient, we obtain

$$\partial_t r^2 = \frac{1}{4\eta} \partial_x \left( r^4 \partial_x \frac{1}{2\pi r} \frac{\delta \mathcal{E}}{\delta r} \right), \quad (4)$$

with  $\mathcal{E} = \int dS [\Sigma + (1/2)\kappa(2H)^2]$  and  $\kappa$  the bending modulus. Note the natural progression from (1) to (3) to (4). Although (4) has a nonsingular evolution, it has the conservation laws and nonlinear structure emphasized in the introduction. This structure is similar to thin-film dynamics [25] in which propagating Rayleigh-Taylor instabilities have been observed. In [12] we reported preliminary results that (4) supported front solutions. Now, as shown in Fig. 4, much more extensive simulations indicate close agreement between the numerical solution of (4) and the MSC. The discrepancy is likely due to the slow approach of the velocity to its asymptotic limit [14], coupled with computational limitations. Note in Fig. 4 how, as each bulge forms, it is pulled away from the thin tether, in accordance with our argument about tension and drag.

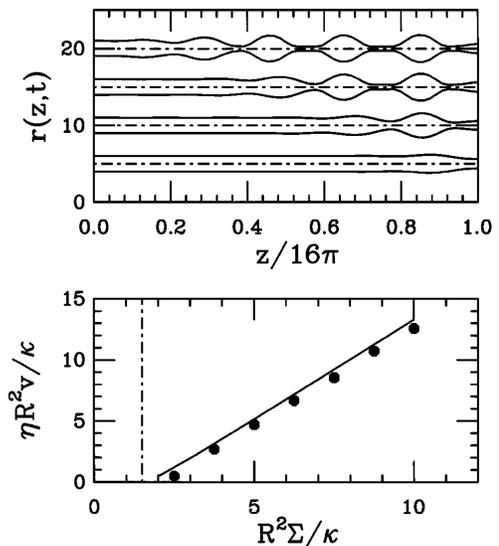


FIG. 4. Numerical results. Propagation of the Rayleigh instability with an elastic cutoff (top). Front velocity as a function of tension (bottom). The solid line is the MSC prediction. The dashed line indicates critical tension for the onset of instability.

We now turn to possible experiments. The examples we list here do not necessarily have a singular evolution; they illustrate the other novel features of propagation outlined in the introduction and raise, as well, the issue of the mechanism by which pinching is inhibited in each case. (i) In addition to droplets in extensional flow, another ideal candidate for propagation with pinching is the neutrally buoyant capillary bridge stabilized by an electric field [22]. (ii) Mather *et al.* [5] are studying shape instabilities of a thread of liquid crystal polymer embedded in a polymer matrix, with a competition between tension and nematic elasticity. (iii) Matsuo and Tanaka [6] have stretched a cylindrical gel between two glass plates and observed a peristaltic instability in the presence of an appropriate solvent. In addition to the long time scales, these systems have the advantage that the thermal fluctuations are small. These facts may conspire to allow one to see a propagating Rayleigh instability in the cases without a precisely controllable tension. (iv) Jets of viscoelastic fluid, as in the work of Renardy [7], may also display propagating Rayleigh instabilities. (v) Finally, we expect there are examples of front propagation with singular evolution that are not Rayleigh-like. For example, in surface-tension-driven Bénard convection [26], an initially flat layer of liquid distorts to expose dry spots when it is heated from below. We suggest that these dry spots can spread behind a front that moves at constant velocity, rather analogous to those seen in the Rayleigh-Taylor instability [25].

We have argued that front propagation in Rayleigh and Rayleigh-like systems is qualitatively different from more familiar examples of propagation. The differences are both mathematical and physical: the nonpinching Rayleigh-like systems have global conservation laws and a particular nonlinear structure due to the two-dimensional nature of the interface, whereas the Rayleigh problem, in addition, has singularities and the new physical ingredient of retraction. If these instabilities do indeed propagate, it will be important to determine whether the propagation can be described by the MSC calculations presented here or if some new picture is needed. There is a clear need for future experimental and theoretical investigations.

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