Two Fluid Drop Snap-Off Problem: Experiments and Theory

Itai Cohen,[1] Michael P. Brenner,[2] Jens Eggers,[3] and Sidney R. Nagel[1]
[1]*James Franck Institute, University of Chicago, Chicago, Illinois 60637*
[2]*Department of Mathematics, MIT, Cambridge, Massachusetts 02139*
[3]*Universität Gesamthochschule Essen, Fachbereich Physik, 45117 Essen, Germany*
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We address the dynamics of a drop with viscosity $\lambda\eta$ breaking up inside another fluid of viscosity η. We track the time evolution of the drop near snap-off in the experiments and then compare with a scaling theory developed for $\lambda = 1$. The theory is in excellent agreement with both the experiments and the previous simulations of Lister and Stone. Finally, we also investigate the λ dependence of the drop shape and breaking rate, and develop a simple theory for the latter.

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When a fluid droplet breaks, as shown in Fig. 1, a singularity develops due to the infinite curvature at the point of snap-off [1]. Near such a singularity, the axial and radial length scales become vanishingly small. Such a separation of scales suggests that near snap-off the profiles may be self-similar: On rescaling by the axial and radial scales the profiles near the singularity should collapse onto a universal curve [2].

The character of the singularity depends on which terms in the Navier-Stokes equations are dominant at the point of breakup. If the drop breaks up in vacuum, surface tension, viscous stresses, and inertia are balanced asymptotically, although the motion may pass through other transient regimes, depending on viscosity [2–8]. In this paper, we investigate the situation where the viscous effects of the inner and outer fluid are included as are the pressure gradients produced by the curvature in the surface separating them; the inertial terms are taken to be insignificant so that we are in the Stokes regime [9–12]. Assuming that molecular scales are not reached first, this is the final asymptotic regime describing flows near snap-off for any pair of fluids even in the case of arbitrarily low viscosity. An understanding of this asymptotic regime is of crucial importance for the physics of mixing, since the fine scale structure of a dispersion is determined by the surface tension driven breakup of fluid filaments [13]. To that end, this paper uses experiments, simulations and theory to characterize the self-similar approach to snap-off in this regime.

We consider the rupture of a fluid of viscosity $\lambda\eta$ surrounded by another fluid of viscosity η. The interface between the two fluids has surface tension γ. At a time t^* shortly before rupture, dimensional analysis suggests that all length scales have the form $H(\lambda)v_\eta t^*$, where $v_\eta \equiv \gamma/\eta$ and $H(\lambda)$ is the universal function yet to be determined. Hence, if drop profiles near rupture are rescaled by $v_\eta t^*$, they should collapse onto a universal curve, independent of the initial conditions. However, Lister and Stone [11] noticed that the long-ranged character of the Stokes interaction leads to logarithmic corrections in the velocity field. They simulated Eqs. (1)–(3) below for drops having various unstable initial conditions, and demonstrated collapse if the logarithmic term was subtracted. (See also Loewenberg *et al.* [12].) In this present paper, we demonstrate this collapse experimentally. We then construct a scaling theory to explain the profile shapes, by incorporating the nonlocal contributions into a local similarity description.

The experiment used 9.5 St glycerin dripping through 10 St PDMS. The viscosities are large enough that the experiment is in the Stokes-flow regime even at macroscopic scales. The surface tension γ was measured using the pendant drop method [14], and the viscosity was measured using calibrated Cannon-Ubbelohde viscometers. We used a Kodak Motion Corder Analyzer to capture ten thousand frames per second. These images were then analyzed using an edge-tracing program, and smoothed [15]. Figure 2 shows the collapse of rescaled profiles at $\lambda = 1$ for both experiments (solid circles) and numerical simulations

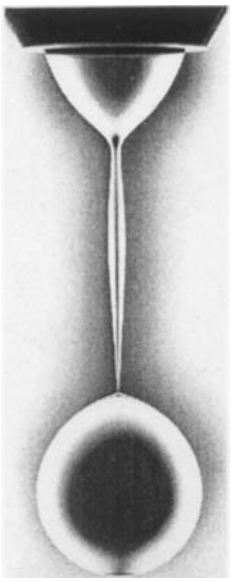

FIG. 1. A drop of 9.5 St (1 St $\equiv$ 1 cm^2/s) glycerin dripping through 10 St polydimethylsiloxane (PDMS) near snap-off. The nozzle diameter is 0.48 cm.

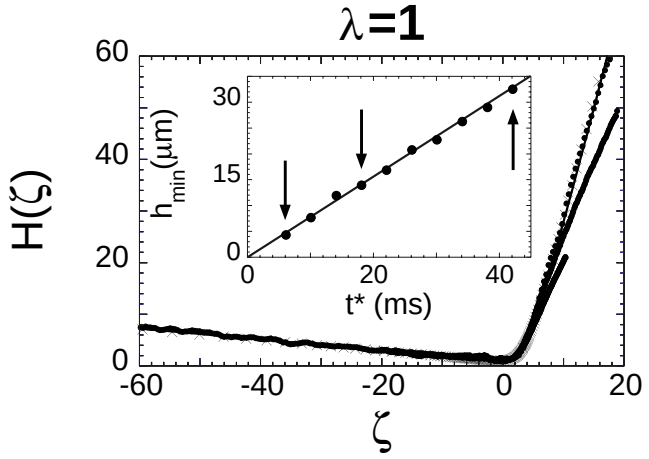

FIG. 2. The inset shows the minimum radius, $h_{\min}(t)$, as a function of time for the drop shown in Fig. 1. The solid line is the theoretical prediction. The main figure shows the similarity function $H(\xi)$ [as defined by (4)], where ξ is the axial coordinate. The dots are rescaled experimental profiles corresponding to the times indicated by the arrows in the inset. The solid line is the theory, and the $\times$'s mark the final simulation profile. The two points of deviation indicate the transition from the self-similar regime to the spherical regime of the drop.

($\times$'s) using a numerical technique similar to that of Lister and Stone. They are superimposed on the scaling theory developed below (solid line). In rescaling the experimental profiles, we shifted each rescaled profile in the axial, ξ, direction to minimize the cumulative deviation in $H(\xi)$. This lined up the minima locations.

The inset of Fig. 2 shows that near snap-off $h_{\min}(t)$ is a linear function of t^*. By fitting the prefactor of this linear dependence, we obtain $h_{\min} = (0.031 \pm 0.008)\nu_\eta t^*$, in excellent agreement with the result $h_{\min} = 0.0335\nu_\eta t^*$ from numerical simulations [11] and the scaling theory constructed below.

Scaling Theory.—Near the singularity, the axial and radial length scales become vanishingly small. Simulating the final stages of breakup should therefore become prohibitively expensive to compute numerically. Fortunately, the separation of scales between the local and the large scale motion leads to universal behavior independent of initial conditions. This self-similarity leaves two options available for the theoretical analysis. First, one can simulate the full equations to the point where the drop starts displaying the self-similar behavior. Alternatively, one can plug into the full equation a similarity ansatz for the drop profile which incorporates the time dependences. This latter procedure reduces the governing equations into ones that can be solved through numerical integration. The first method is discussed in [11]. Below, we explain the details of the latter method.

Since the Stokes equation is linear, the fluid surface velocity can be expressed as an integral over the surface of the fluid-fluid interface. At $\lambda = 1$, the equation is [16]

$$\mathbf{v}^{(S)}(z,t) = -\gamma \int \kappa(z')\mathbf{J}(z,z')\mathbf{n}(z')\,dz', \qquad (1)$$

where $\mathbf{n}$ is the outward normal, κ is the curvature, z is the axial coordinate, and the tensor $\mathbf{J}$ is

$$\mathbf{J}(z,z') = \frac{1}{8\pi}\int_0^{2\pi}\left[\frac{\mathbf{I}}{r} + \frac{\mathbf{rr}}{r^3}\right]d\theta\,, \qquad (2)$$

where $\mathbf{r}$ is the vector between the two points on the surface, $\mathbf{I}$ is the identity matrix, and the integration is over the azimuthal angle θ. Physically, Eq. (2) represents the response of the surface tension forcing the interface. For unequal viscosities $\lambda \neq 1$, Eq. (2) must be amended by an additional term, which accounts for the jump in viscosity. Given the radial v_r and axial v_z components of the surface velocity, the interface advances according to

$$\partial_t h(z,t) + v_z\partial_z h = v_r\,, \qquad (3)$$

which states that the surface at a given axial position can deform by radial motion and axial advection.

Motivated by the simulations of Lister and Stone [11], we try the similarity ansatz,

$$h(z,t) = \nu_\eta t^* H(\xi)\,,$$
$$\xi = \nu_\eta^{-1}(z^*/t^*) + b\ln t^* + \xi_0\,, \qquad (4)$$

where z^* is the axial distance from the singularity, b is a constant, and the factors of ν_η have been inserted to make H and ξ dimensionless. The shift $b\ln t^*$ in the similarity variable ξ results from the logarithmic divergence of the axial velocity field [11], and ξ_0 will be shown to be an arbitrary constant which depends on the boundary conditions. Since the solution near snap-off must match onto the outer profile, which varies slowly on the time scale t^*, $H(\xi) \sim s_\pm\xi$, as $\xi \to \pm\infty$. Here we define s_- as the slope (relative to the azimuthal axis) of the shallow side of the pinch region, which by convention we place to the left of the minimum, and s_+ as the steep slope.

The subtle feature of this problem is the interplay of the local singularity with the nonlocal fluid response from the Stokes flow. The principal nonlocal effect is that the surface tension force from the cones produces a logarithmically diverging axial velocity field at the pinch point, $\xi = 0$ [11]. For a local scaling theory, this singularity must be absorbed. We fix two points ξ'_- and ξ'_+ within the linear part of the solution to the left and right of $\xi = 0$. Splitting the contributions to the velocity on the surface into a contribution from $\xi_- < \xi < \xi_+$ and from the rest of the drop, and converting to similarity variables, we find

$$\mathbf{V}^{(S)}(\xi,t^*) = \left[-\int_{\xi'_-/t^*}^{\xi'_+/t^*}\kappa(\xi')\mathbf{J}(\xi,\xi')\mathbf{n}(\xi')\,d\xi' - b\ln t^*\mathbf{e}_z\right], \qquad (5)$$

where $\mathbf{V}^{(S)} = (\mathbf{v}^{(S)} - b \ln t^* \mathbf{e}_z)/v_\eta$, and $\mathbf{e}_z$ is the unit vector along the axial direction. Because of the cones, the axial component of the $\mathbf{J}$ integral in angular brackets diverges logarithmically as $t^* \to 0$. For the special choice $b = -[s_+(1 + s_+^2)^{-1} + s_-(1 + s_-^2)^{-1}]/4$, the singularity cancels and the term in angular brackets remains finite for $t^* \to 0$. It is straightforward to extend this scaling theory to arbitrary λ; in this case the amplitude of b depends only on λ through s_+, s_-. The remaining constant in (5) depends on the detailed shape of the drop as well as on the choice of ξ'_-, ξ'_+.

Inserting the similarity form (4) into the equation of motion for the interface (3) gives

$$-H + \frac{dH}{d\xi}(V_z + \xi - \xi_0 - b) = V_r, \qquad (6)$$

where we have absorbed the constant advection velocity $b \ln t^*$ into V_z.

The system [(5) and (6)] now has to be solved in the limit $\xi'_+/t^* \to \infty$, $\xi'_-/t^* \to -\infty$, for the interval $-\infty < \xi < \infty$ and with boundary conditions $H(\xi) \sim s_\pm \xi$, as $\xi \to \pm\infty$. Changing the constant $\xi_0 + b$ results only in a constant shift of the similarity function $H(\xi)$. The computation involves solving an integrodifferential equation with a nonlocal constraint: The parameter b in (5) must be determined self-consistently with the solution $H(\xi)$ according to relation (5). The difference between this scaling theory and others developed for fluid rupture is that here the *parameters* in the similarity equation must be determined self-consistently with the solution to the similarity equation.

We solved this system by discretizing $H(\xi)$ in an interval $\xi \in [-\xi_{\text{in}}, \xi_{\text{in}}]$ and approximated all derivatives and the integral by second-order formulas. At $\xi = -\xi_{\text{in}}, \xi_{\text{in}}$ we demand $H'' = 0$. Using a linear approximation for H outside the interval $[-\xi_{\text{in}}, \xi_{\text{in}}]$, the logarithm is subtracted explicitly. A simulation of the full equations provided an initial condition for Newton's iteration, which converged in a few steps. The iteration always converges to the same solution for any given ξ_0. The calculation gives

$$H_0(\lambda = 1) = 0.0335, \qquad s_- = -0.105,$$
$$s_+ = 4.81,$$

where $s_{+,-}$ are the asymptotic slopes at $\pm\infty$. These results are in good agreement with both the experiments (Fig. 2) and the simulations of [11]. Although the theory has been solved only for $\lambda = 1$, by continuity we expect that solutions exist for a range of λ and that h_{min} obeys the law $h_{\text{min}} = H_0(\lambda) v_\eta t^*$.

Arbitrary λ.—We begin with the experimental results for arbitrary λ. Using glycerin/water mixtures (1 St $< \eta <$ 9.5 St) and silicone oils (1 St $< \eta <$ 600 St), we were able to cover a range of λ between 0.002 and 30 [9]. The same procedure as used above verified self-similar data collapse in experiments with $0.02 < \lambda < 30$ [15]. The rescaling analysis for $\lambda = 1$ can be seen in Fig. 2. The point at which the experimental profiles deviate from one another in the conical profile associated with s_+ marks the transition from the self-similar regime to the spherical regime of the drop. As the drop breaks, the profiles are rescaled by a shrinking radius. Therefore the point of deviation moves away and the region of self-similarity grows with time.

Figure 3 shows the cone slopes s_+ and s_-, and the dimensionless breaking rate H_0 as a function of λ. As shown by Fig. 3a, the cone slopes for s_+ appear to obey a power law over an extended range of λ: $s_+ \sim \lambda^{0.22 \pm 0.07}$. Figure 3b shows that the cone slopes for s_- peak near $\lambda = 0.5$ and approach 0 as $\lambda \to \pm\infty$. The s_- slopes for $\lambda = 30$, $\lambda = 0.002$, and $\lambda = 0.004$ were too small to analyze [15]. Within error (and with the exception of the $\lambda = 0.002$ experiment [15]), the analyses performed on both the snap-off event near the nozzle and the snap-off event near the bulb lead to the same results. This agreement implies that the results are robust and independent of small variations in the surrounding flows. Note that our findings are in qualitative disagreement with lubrication-type scaling arguments [10,11], which predict s_+ and $s_- \sim \lambda^{-1/2}$ for the slope on either side of the minimum. On the other hand, the trends in our data are consistent with recent numerical simulations of the full Stokes equations by Zhang and Lister [17]. This is yet another indication of the breakdown of one-dimensional models which use a lubrication approximation in describing the dynamics of two fluid rupture.

A Simple Theory for $H_0(\lambda)$.—This follows by comparing the breaking rate $\partial_t h_{\text{min}}/h_{\text{min}}$ of the singular solution with the linear growth rate of disturbances on the interface. For the two fluid case the maximum linear growth rate $\Omega(r_0, \lambda)$ of a small disturbance on a fluid cylinder of radius r_0 was calculated by Tomotika [18] using the assumption of slow flow. Since s_- is always quite small, the linear growth rate $\Omega(h_{\text{min}}, \lambda)$ is well approximated by Tomotika's formula. It is clear that h_{min} cannot shrink faster than the fastest growing *linear* disturbance, and thus we have the upper bound

$$\Omega(h_{\text{min}}, \lambda) > \partial_t h_{\text{min}}/h_{\text{min}} = 1/t^*. \qquad (7)$$

Using Tomotika's explicit form for Ω this equation turns into an upper bound for $H_0(\lambda)$, which is compared with experimental data in Fig. 3c. All of the data obeys the bound; moreover, in the range $0.1 < \lambda < 10$ the agreement is nearly exact. Note that while most of the experimental data [and the upper bound Eq. (7)] can be fit with a power law of exponent -0.53 ± 0.05, a significant trend with an overall negative curvature is observed in the experimental deviations.

The agreement of the experiments with the upper bound in the range $0.1 < \lambda < 10$ is reminiscent of the marginal stability hypothesis, as formulated for the selection of traveling waves propagating from a stable to an unstable state [19]. Both experiments and numerical simulations

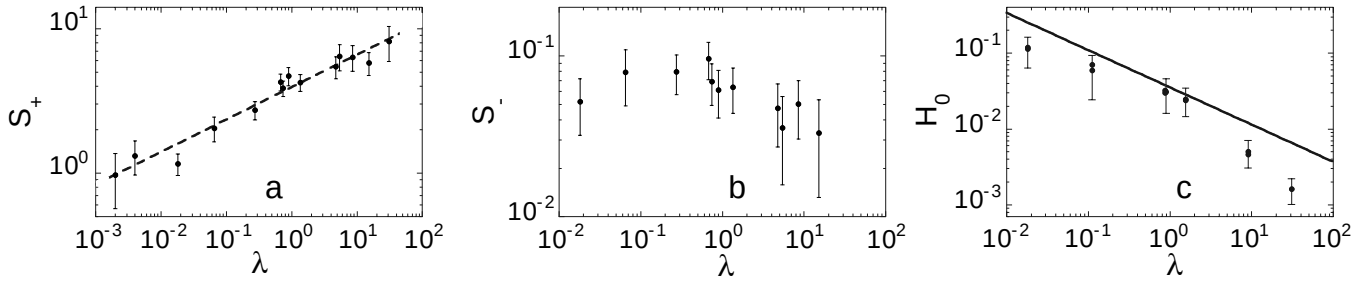

FIG. 3. The asymptotic slopes s_+, s_-, and the rescaled minimum radius H_0 as a function of viscosity contrast λ. The dashed line is a fit to the experimental data; in the rightmost graph the solid line is the result of our stability argument.

show that the breaking rate is approximately equal to the growth rate of linear instabilities around a cylinder of radius $h_{\min}$. The upper bound in Eq. (7) should apply to all problems involving singularity formation in a system with a local instability. We have tested this upper bound on similarity solutions from several other examples including spherically symmetric gravitational collapse [20] and chemotactic collapse of bacteria [21]; the upper bound is obeyed in each case, giving a reasonable estimate for the prefactor. Hence, this principle appears to be of rather general applicability.

In conclusion, we have (i) shown through experiments that near the singularity the drop profiles are self-similar; (ii) constructed a similarity solution for rupture at $\lambda = 1$, which agrees quantitatively both in the form of the profile and its time dependence with the experiments and previous numerical simulations [11]; and (iii) presented a simple argument which quantitatively predicts the breaking rate as a function of λ. Experiments have also shown self-similar behavior for the range $0.02 < \lambda < 30$. There are many unresolved issues: Among them, there is no solid simple argument for the λ dependence of the slopes s_-, s_+. Finally, our results suggest that the scaling (4) holds even in the limit $\lambda \to \infty$, while a different set of scaling exponents is found for a Stokes fluid breaking up in vacuum ($\lambda = \infty$) [4]. In addition, the profiles are asymmetric for $\lambda \to \infty$, as also found in a recent numerical simulation [22], but are symmetric for breakup in vacuum [4], making this a singular limit. Preliminary experimental results of an inviscid fluid breaking up in a viscous fluid suggest that the snap-off shape is different from that in Stokes flow with $\lambda \to 0$ implying that this limit is singular as well.

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