



Bifurcation from Bubble to Droplet Behavior in Inviscid Pinch-off

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High-speed video is used to analyze the pinch-off of xenon in water. By varying the pressure in the xenon from 0.05 to 68 atm, the mass density ratio $D = \rho_{\text{int}}/\rho_{\text{ext}}$ of the interior gaseous xenon ρ_{int} to the exterior liquid water ρ_{ext} can be varied by over 3 orders of magnitude. Both the shape of the pinch region and the power law that governs the collapse of the minimum neck radius are distinctly different for the low D (bubblelike) and high D (dropletlike) cases. We show that there is a rather abrupt transition between these types of behavior near $D \approx 0.25$.

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The separation of one fluid mass into two disjointed pieces involves the formation of a finite-time singularity in the flow where quantities such as pressure, velocity, and curvature diverge. A recent review of fluid break-up phenomena can be found in Ref. [1]. This pinch-off process generally involves both an interior and an exterior fluid characterized by different mass densities ρ_{int} and ρ_{ext} , and perhaps other physical properties such as surface tension σ and viscosity η . Viscosity is important at length scales smaller than $\eta^2/(\sigma\rho)$, but for many low viscosity fluids such as air and water, this viscous length is a few nanometers, so most of the pinch-off process can be described as an inviscid flow. Inviscid pinch-off can be characterized by the density ratio $D = \rho_{\text{int}}/\rho_{\text{ext}}$ where $D \rightarrow 0$ corresponds to a low density bubble surrounded by a dense fluid, while the limit $D \rightarrow \infty$ corresponds to a dense liquid drop pinching off in a low density vapor. Each of these limiting cases has recently received considerable theoretical and experimental attention. The singularity structure in the droplet case is an example of self-similarity of the first kind, in which singular quantities diverge with power laws with rational exponents; for example, the minimum neck radius of an inviscid droplet scales as $R_{\text{min}} \sim (t_0 - t)^{\alpha_{\text{drop}}}$, where $\alpha_{\text{drop}} = 2/3$ and t_0 is the time at which the singularity occurs [2–9]. In inviscid drop pinch-off, an additional constraint requires the radial and axial lengths to scale in the same way, so the shape of the singular region is preserved, and is universal in the sense that it is independent of initial conditions such as the size of the droplet.

The singularities in bubble pinch-off ($D \rightarrow 0$) are qualitatively different. The neck radius has a power-law form $R_{\text{min}} \sim (t_0 - t)^{\alpha_{\text{bubble}}}$, but $\alpha_{\text{bubble}} \approx 1/2$ [10–12]. The shape of the singular region is not constant, but rather evolves from a conical shape to a slender cylinder as the pinch-off proceeds. This evolution takes place over many decades in time, which is related to the fact that α_{bubble} is not strictly constant, but rather has logarithmic corrections [13–16]. Detailed experiments yield an approximate effective exponent with $\alpha_{\text{bubble}} \approx 0.53\text{--}0.57$ [15,17,18]. Furthermore, bubble pinch-off is extremely sensitive to nonaxisymmet-

ric perturbations [12,18], and retains a memory of the initial conditions.

Pinch-off in both the droplet limit ($D \rightarrow \infty$) and bubble limit ($D \rightarrow 0$) represents singular asymptotic solutions to the partial differential equations of evolution of the fluid. For dynamical systems described by ordinary differential equations, asymptotic solutions are associated with geometrical objects in the phase space such as fixed points, limit cycles, and strange attractors. For partial differential equations, the phase space is infinite dimensional, but the phase space picture is nevertheless useful for classifying types of singular behavior [19,20]. From the dynamical systems point of view, for each value of the density ratio parameter D there is a fixed point with a basin of attraction and local properties which determine the singular exponents. As D is varied, the properties of the fixed point can vary either smoothly, or abruptly via a bifurcation. The region near both the bubblelike fixed point [16,21] and the dropletlike fixed point [4,7] have been investigated using various numerical techniques, but the nature of the transition from one fixed point to the other has not been explored. In this Letter, we use high-speed video to analyze the pinch-off of xenon from a nozzle submerged in water to experimentally investigate the parameter range $6.5 \times 10^{-4} < D < 0.7$. Xenon is particularly suitable for this purpose because the critical point is slightly below room temperature, so the density can be continuously varied over a wide range with only modest increases in the pressure. At low values of D , pinch-off resembles that of a collapsing vacuum cavity, and at high densities the pinch-off resembles that of a droplet. We observe a sharp increase in the scaling exponent from $\alpha \approx 0.57$ to $\alpha \approx 2/3$ at a density ratio $D \approx 0.25$.

The apparatus consisted of a stainless steel cell filled with water with optical access through two parallel sapphire windows of diameter 3.81 cm and thickness 0.635 cm. The windows were press sealed to the cell with viton o-rings. The xenon emerged from a Teflon nozzle with an inner diameter of 0.47 cm. The nozzle was connected to a high-pressure (~ 170 atm) xenon

source. The flow rate of the gas was controlled by a fine metering valve, so that the bubbles were produced at approximately 1 bubble per 20 sec. A mechanical vacuum pump was attached to the cell through a valve so that subatmospheric pressures could be achieved. The pressure in the cell could be adjusted from the vapor pressure of water (~ 0.03 atm at 25°C), to a maximum of 100 atm, although in practice, we were limited to lower pressures because xenon forms a solid clathrate with water at room temperature and tens of atmospheres of pressure [22]. In order to avoid this problem, all experiments were performed at slightly elevated temperatures ($\sim 37^\circ\text{C}$), which increased the clathrate formation pressure to $P \sim 68$ atm and allowed us to reach values of $D \approx 0.7$. The pressure and temperature of the xenon were measured, and the density was computed from data in Ref. [23]. To obtain a large dynamic range of measurements of R_{\min} , videos were taken with two levels of optical magnification. For the images in Fig. 1, the high-speed camera (Phantom v7.2, Vision Research) was attached to a macro lens, allowing a full view of the bubble evolution. For the high-magnification videos in Fig. 2, the camera was attached to a photographic bellows with a $5\times$, long working distance microscope objective. The optical resolution for this setup was fixed at $2.4\ \mu\text{m}/\text{pixel}$. Typical frame rates for the low (high) magnification videos were 40 000 (100 000) fps. All bubble images were captured using illumination from behind with a $2\ \mu\text{s}$ exposure time. Back illumination produced bright, white optical artifacts in the images due to focusing of the light as it passes through the bubble (Figs. 1 and 2). The images in the videos were processed so that the minimum neck radius R_{\min} could be measured for each frame. In order to estimate the effects of viscosity during pinch-off, the Reynolds number Re was calculated using R_{\min} and the axial velocity near R_{\min} ; the value obtained was $\text{Re} \geq 100$, indicating an inertia-dominated flow. Images of quiescent bubbles were used to measure the xenon-water surface tension as a function of D using axisymmetric drop-shape analysis; the surface tension varied smoothly from 73 ± 1 dyne/cm at $D = 6.5 \times 10^{-4}$ to 34 ± 1 dyne/cm at $D = 0.695$.

Figure 1 shows low-magnification images of the bubble evolution at four different density ratios. For very low densities ($D \leq 0.001$), such as in Fig. 1(a), the pinch-off is nearly perfectly symmetric in the vertical direction. As the density increases, there is a visible asymmetry in the vertical direction [Fig. 1(b), 3rd frame]. This is presumably due to an axial jet of gas from the nozzle into the connecting bubble. As the density is raised even more [Figs. 1(c) and 1(d)], this asymmetry intensifies until the pinch-off looks like that of an inverted droplet at $D = 0.7$. After the initial pinch-off event, the connecting filament begins to retract towards the nozzle. At higher densities, this filament can be sufficiently elongated to result in another pinch-off event prior to retraction, which creates a satellite bubble. At $D = 0.217$ in Fig. 1(c), the satellite bubble is approxi-

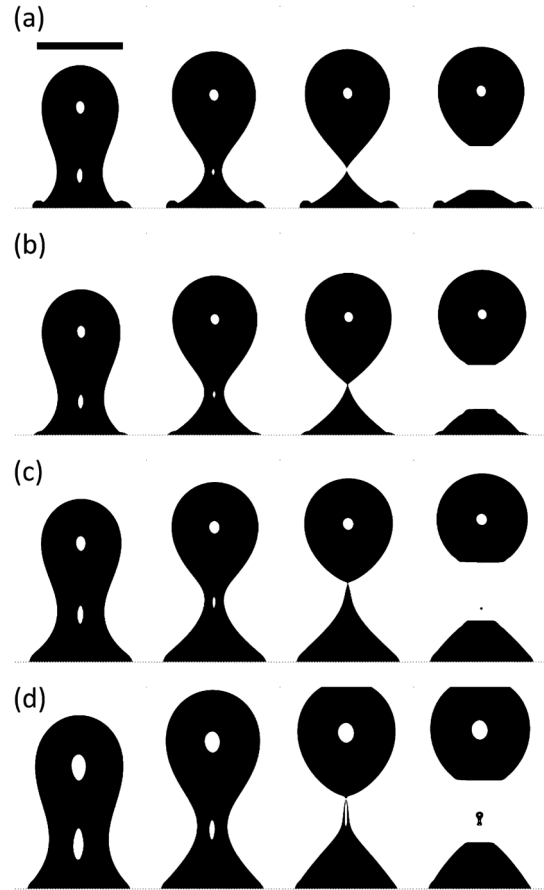


FIG. 1. Images extracted from the high-speed video showing the pinch-off of xenon rising in water at four different mass density ratios (specific gravity): (a) 6.5×10^{-4} , (b) 0.047, (c) 0.217, (d) 0.695. The scale bar in the first frame of (a) represents 5 mm, and applies for all images in (a)–(d). The time stamp on each frame relative to the moment of pinch-off is (from left to right) (a): -8.95 ms, -1.025 ms, 0 ms, 1.275 ms; (b): -9.50 ms, -1.35 ms, 0 ms, 1.525 ms; (c): -13.15 ms, -2.1 ms, 0 ms, 2.6 ms; (d): -58.625 ms, -11.55 ms, 0 ms, 3.225 ms. The bright, white spots in the images are optical artifacts due to illuminating the bubbles from behind. The bubbles are larger at higher densities because of decreased buoyancy as $D \rightarrow 1$.

mately $120\ \mu\text{m}$ in diameter and at $D = 0.695$ in Fig. 1(d), the satellite bubble diameter is $500\ \mu\text{m}$.

High-magnification images of the upper pinch-off from the main bubble are shown in Fig. 2 for identical values of D . At low densities [$D = 6.5 \times 10^{-4}$, 2(a)], the pinch-off is highly symmetric and collapses very quickly to zero diameter. The slight rotational asymmetry in the last frame of Fig. 2(a) is due to the extreme sensitivity to tilts and perturbations in the nozzle [18]. At slightly higher densities [$D = 0.047$, 2(b)], the pinch-off is less sensitive to axisymmetric perturbations; however, the vertical symmetry is lost due to the axial jet of gas flowing into the main bubble. The increase in vertical asymmetry continues until the pinch-off geometry finally becomes reentrant [2(d)], where the pinch-off occurs below the optical horizon. This

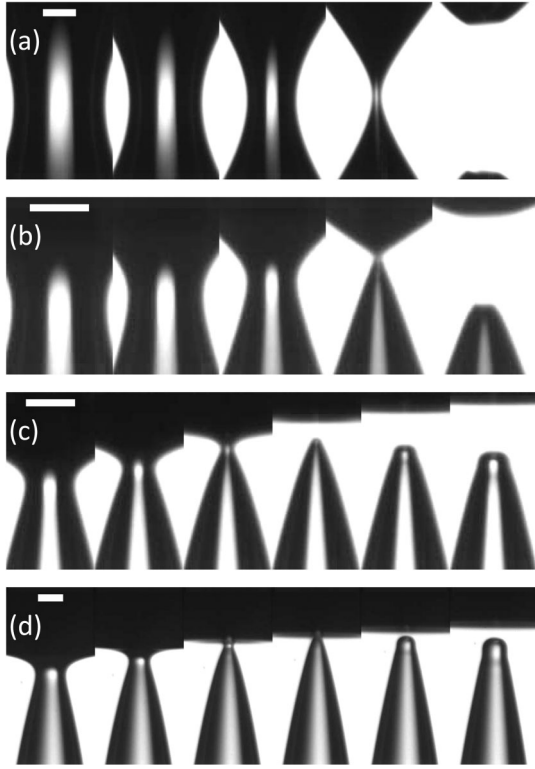


FIG. 2. Images extracted from the high-speed video showing the pinch-off of xenon in water at four different mass density ratios (specific gravity): (a) 6.5×10^{-4} , (b) 0.047, (c) 0.217, (d) 0.695. The white bar at the top of the first image in each sequence represents $100 \mu\text{m}$. The time between each successive image is $30.25 \mu\text{s}$, $13 \mu\text{s}$, $15 \mu\text{s}$, and $20 \mu\text{s}$ for sequences (a)–(d), respectively.

is the expected behavior for purely inviscid droplet pinch-off [3,6,8]. In contrast to previous experimental studies of bubble pinch-off [12,17,18], we do not observe the creation of satellite bubbles for $D \leq 0.2$. We note that this difference may be due to the fact that water used in our experiment was degassed prior to the addition of the xenon. Since negative pressures develop near the singularity due to high fluid velocities, dissolved air in water used in [12,17,18] may have contributed to the creation of satellite bubbles through nucleation.

To quantify the transition from bubble to dropletlike pinch-off, we have measured the minimum neck radius R_{\min} of the collapsing bubble as a function of time using a combination of the low and high-magnification video images. For each value of D , measurements of R_{\min} vs t for both sets of images were simultaneously fit to the form $R_{\min} = A(t_0 - t)^\alpha$ using a χ^2 method, assuming the same values of A and α , but a different t_0 for each video. The data used in the fitting procedure were restricted to values of R_{\min} for which $(t_0 - t) \leq 200 \mu\text{s}$, since the functional form is strictly valid only near the singularity [24]. Once the values of t_0 were determined, the full range of data could be plotted, as shown in Fig. 3. The minimum and maximum values of the density ratio $D = 6.5 \times 10^{-4}$ and $D = 0.695$

are unambiguously in the bubblelike and dropletlike regime, respectively, and the power law fits yield values of the exponents $\alpha_{\text{bubble}} = 0.57$ and $\alpha_{\text{drop}} = 0.66$ in agreement with previous measurements, as shown in Fig. 3(b).

Figure 4 shows values of the power-law scaling exponent α as a function of the density ratio for values of D that span the bubblelike and dropletlike regimes. The value and error estimates for the exponent α were taken from the fitting procedure, assuming a normally distributed pixel error with a standard deviation of 0.5 pixels. The error bars on the values of α represent the 95% confidence interval from our fitting procedure. α is essentially constant with the value expected for a vacuum bubble $\alpha \approx 0.57$ over a wide range of density ratios until $D \sim 0.1$, at which point a relatively sharp transition is made to $\alpha \approx 0.66$. The $\alpha = 2/3$ regime is expected to extend indefinitely beyond our maximum value of $D = 0.695$. This has been experimentally verified for $D \approx 14000$ [9].

For density ratios $D \geq 0.16$ in the dropletlike regime, boundary-integral numerical simulations of inviscid fluids

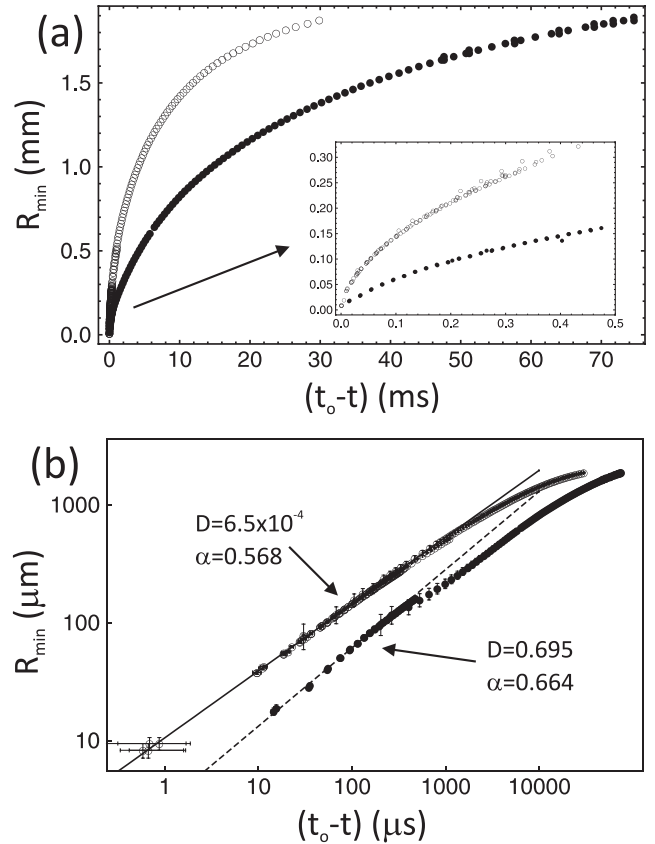


FIG. 3. (a) Linear and (b) $\text{Log}_{10} - \text{Log}_{10}$ plots of R_{\min} vs $(t_0 - t)$ for a bubblelike density $D = 6.5 \times 10^{-4}$ (open circles) and a dropletlike density $D = 0.695$ (closed circles). Both sets of data consist of measurements from one high-magnification video and multiple low-magnification videos. The inset in (a) shows data very close to the singular region. Error bars in (b) represent 0.5 pixels in the measurements of R_{\min} and $1 \mu\text{s}$ in the measurements of $(t_0 - t)$. The solid line in the logarithmic plot represents $(t_0 - t)^{0.568}$ and the dashed line represents $(t_0 - t)^{0.664}$.

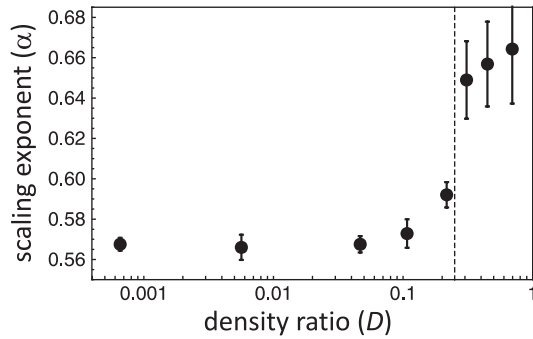


FIG. 4. Plot of the power-law scaling exponent α of the minimum neck radius R_{\min} vs the density ratio D . Error bars represent a 95% confidence interval. There is a sharp transition from $\alpha \approx 0.57$ to $\alpha \approx 2/3$ when the density ratio is near 0.25 (dotted line). This transition represents a bifurcation of the solutions of the equations of motion from bubblelike to dropletlike pinch-off.

[4,7] and the corresponding similarity solutions [4] show that the power law $R_{\min} \sim (t_0 - t)^{2/3}$ is valid for a range of values of D . However, for $D \leq 0.16$, the numerical solutions become linearly unstable to oscillatory modes, producing striking fractal-like shapes near pinch-off. These simulations do not address density ratios below 0.16 due to the high computational costs of resolving the fine structure in shapes with high curvature. Experimentally, at these intermediate density ratios [Fig. 2(c)], we see no evidence of this oscillatory instability; the fluid profile evolves smoothly from the sharply pointed filament characteristic of drops to the hyperboloids characteristic of bubbles. Although it is possible that this instability may occur under the optical horizon of the drop at small length scales, we believe that the viscous boundary-layer of gas at the free surface (which is not accounted for in the inviscid simulations) damps out any oscillatory behavior. Numerical techniques have also been used to study the bubblelike regime for $D < 0.01$ [13,14,21]. The results show that the exponent α is independent of D at sufficiently long times, but at extremely short times and high Reynolds numbers a crossover to $\alpha = 1/3$ is expected; apparently this regime is outside the dynamical range of our experiment.

In conclusion, we have observed the transition from bubble to droplet pinch-off using xenon rising in water at pressures up to 68 atm. The behavior of the minimum neck radius R_{\min} is determined by the density ratio $D = \rho_{\text{int}}/\rho_{\text{ext}}$. Assuming a power law of the form $R_{\min} \sim (t_0 - t)^\alpha$, the scaling exponent α is approximately constant $\alpha \approx 0.57$ for $D < 0.1$, while $\alpha \approx 2/3$ for all $D > 0.25$. The bubble and dropletlike regimes represent stable fixed points of the dynamical system. The transition between the two fixed points is quite abrupt, and is probably related to the inviscid instabilities discussed in Ref. [4], broadened by weak viscous and boundary-layer effects.

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