Break-up dynamics of fluctuating liquid threads

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The thinning dynamics of a liquid neck before break-up, as may happen when a drop detaches from a faucet or a capillary, follows different rules and dynamic scaling laws depending on the importance of inertia, viscous stresses, or capillary forces. If now the thinning neck reaches dimensions comparable to the thermally excited interfacial fluctuations, as for nanojet break-up or the fragmentation of thermally annealed nanowires, these fluctuations should play a dominant role according to recent theory and observations. Using near-critical interfaces, we here fully characterize the universal dynamics of this thermal fluctuation-dominated regime and demonstrate that the cross-over from the classical two-fluid pinch-off scenario of a liquid thread to the fluctuation-dominated regime occurs at a well-defined neck radius proportional to the thermal length scale. Investigating satellite drop formation, we also show that at the level of the cross-over between these two regimes it is more probable to produce monodisperse droplets because fluctuation-dominated pinch-off may allow the unique situation where satellite drop formation can be inhibited. Nonetheless, the interplay between the evolution of the neck profiles from the classical to the fluctuation-dominated regime and the satellites’ production remains to be clarified.

critical fluids | singularity formation

For a drop to detach from a capillary or a faucet, the liquid thread connecting them must thin and break. This break-up, or pinch-off, is an example of a singularity with well-established scaling laws and similarity solutions (1–5). Different regimes and scaling laws have been predicted and observed. For small liquid viscosities, the balance between inertia and capillarity leads to the so-called inertial thinning regime, with the thread radius vanishing as time to pinch-off to the power 2/3. When the radius of the thinning thread becomes smaller than the so-called viscous length scale \( R_\eta \sim \eta^3 / \eta_0 \) (where \( \eta, \eta_0, \text{ and } \rho_0 \) are respectively the surface tension, the shear viscosity, and the density of the fluid), viscous forces become important and the neck radius decreases linearly vs. time (1) as \( R(t) = CV(t^\ast - t) \), where \( V_\ast = \gamma / \eta_0 \) is a capillary velocity, \( C \) is a constant, and \( t^\ast \) is the break-up time at neck pinch-off; a viscous time scale can be defined as \( \tau_\eta = R_\eta / V_\eta \). Two thinning regimes have been predicted and observed in this case: the so-called visco-capillary regime at low Reynolds numbers exhibiting symmetric necks with \( C = 0.071 \) (6) and the visco-capillary-inertial regime emerging when further thinning significantly increases the inner fluid velocity and thus inertia (7). In this latter case, the constant is \( C = 0.030 \) and the neck profiles are asymmetric. Note that more recently, other symmetric break-up dynamics have been found for a class of non-Newtonian fluids for which thinning is dictated by the rheological properties of the fluids (8, 9).

When the viscosity of the outer fluid is no more negligible, as in the present investigation, the thinning dynamics is dominated by visco-capillary stresses when the radius of the rupturing neck \( R(t) \leq \eta_\text{out} \) (1), where \( \eta_\text{out} \) is the shear viscosity of the fluid outside the thread. The variation of the radius again obeys a linear scaling law \( R(t) = H V(t^\ast - t) \), where \( H(\eta_\text{out} / \eta_\text{in}) \) is a function that was experimentally (2) and theoretically evaluated (10, 11). In this two-fluid visco-capillary regime, the thinning neck is asymmetrical, eventually leading to the formation of satellite droplets.

Though these different regimes consider the interface as smooth even at the smallest scales examined, recent simulations of nanojet break-up (12, 13), as well as theoretical (14) and experimental work (15), revealed that the interface roughness due to the interfacial thermal fluctuations may play a dominant role in liquid column break-up when \( R(t) < L_T \), where \( L_T = \sqrt{\kappa T / \gamma} \) is the so-called thermal length scale estimated by comparing the thermal energy \( \kappa T \) to the surface tension \( \gamma \). In this case, the thinning neck is predicted to be symmetrical with respect to the break-up location, thus minimizing the formation of satellite drops at pinch-off (13, 14). Moreover, the thinning of the neck is predicted to follow the scaling law \( R(t) \propto (t^\ast - t)^{0.42} \) (14), where the proportionality factor remains to be determined theoretically and experimentally (14, 15). Because the thermal length is classically in the range of a few molecules, observation of this thinning regime in a laboratory-scale experiment requires a significant increase of \( L_T \). A route for fulfilling this condition is the use of near-critical binary fluids with strongly fluctuating interfaces (16), which offer the unique opportunity of reaching strongly fluctuating hydrodynamic regimes.

Results

The experiment is performed in a near-critical phase-separated water-in-oil micellar phase of a microemulsion whose mass composition is adjusted to be critical at a temperature \( T_C = 308 \text{ K} \). The fluid preparation and properties are detailed in SI Text. For a temperature \( T > T_C \), the mixture separates in two coexisting phases of different micellar concentrations separated by an interface that has large thermally induced interfacial fluctuations near \( T_C \). Two main reasons motivated this choice of system. (i) Due to the supramolecular nature of the micelles, the bulk correlation length of density fluctuations \( \xi = v_*^\circ \eta_0 \left( T - T_C \right)^{-0.63} \) is intrinsically large, allowing interfacial fluctuations to be observable optically. (ii) It follows from the universal ratio \( \eta / \kappa T = v_*^\circ \eta_0 \left( T - T_C \right)^{1.26} \) (17) that the interfacial tension \( \gamma = \eta_0 \left( T - T_C \right)^{2.08} / \kappa T \) is extremely weak compared with that of usual liquid mixtures. For \( T - T_C = 0.1 \text{ K} \), one finds \( L_T = 46 \text{ cm} \) and \( \tau_\eta = 1.5 \times 10^5 \text{ s} \) on the one hand, and \( L_T = 1 \mu\text{m} \) as \( L_T = \xi / \sqrt{\kappa T} = 3 \xi \) on the other. Moreover, considering \( \xi \) as the relevant length scale, the corresponding relaxation time scale \( \tau_\xi = \eta_\xi / (3 \xi) \) is of the order of magnitude \( \sim 0.2 \text{ s} \) is orders of magnitude larger than in usual molecular liquids. Neck thinning driven by thermal fluctuations thus becomes experimentally accessible.

The second key point of the experiment requires starting with an initially stable and well-controlled liquid column to properly fix initial conditions and boundary effects before further destabilization. Though large-aspect-ratio liquid columns are

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known to be unstable due to the Rayleigh–Plateau instability (3), it is briefly shown in SI Text that this fundamental limitation can be circumvented using the radiation pressure of a continuous laser beam to deform the meniscus separating the coexisting phases (18), which is located in the middle of the sample due to near criticality. For a sufficient beam power, the surface deformation ends up connected to the bottom glass face of the cell, thus forming a large-aspect-ratio liquid column whose diameter is controlled by the incident beam power. Hydrodynamic stabilization is provided through the radiation pressure exerted by the beam propagation inside the column (19). Fig. 1A illustrates the regularity of such a laser-sustained liquid column. Once formed, the column is left to relax after turning off the laser beam, as illustrated in Fig. 1B and C. To avoid boundary effects, we focus on a midcolumn rupture event, as that indicated by the horizontal arrow in Fig. 1C. We record the neck-thinning dynamics with a 50 × Olympus microscope objective (N.A. = 0.45) coupled to a video camera (2.560 × 1.600 pixels) operating at variable frame rates. Fig. 1D–F illustrates a typical thinning morphology with particular emphasis on the observed neck symmetry and the inhibition of satellite drop formation in this case. Note, however, the occurrence of a more-elongated neck below the one selected for a close-up; such a neck may lead to satellite drop formation because it may break at more than one location. Fig. 2 presents the thinning dynamics of a column for \( (T - T_C) = 0.23 \) K. The determination of the neck radius is detailed in Materials and Methods. After the first stages of the Rayleigh–Plateau instability, the neck radius \( R(t^* - t) \) starts to decrease linearly in time, as expected for two-fluid visco-capillary thinning (Fig. 2, Inset). Beyond a cross-over at \( t = 0.34 \) s and \( R(t^* - t) \approx 0.8 \) μm, i.e., \( (t^* - t)/\tau_C \approx 8 \) and \( R(t^* - t)/\xi_C \approx 4 \), the thinning dynamics switches to a power law behavior, well approximated by \( R(t) \sim (t^* - t)^{0.42} \), up to break-up; a power law fit gives 0.43.

To confirm the robustness of the observed thinning dynamics, we first investigate the two-fluid visco-capillary behavior. The variation of \( R(t^* - t) \) measured for each \( (T - T_C) \) is fit linearly to extract the break-up time \( t^* \) expected for pure two-fluid visco-capillary thinning. Data are then reanalyzed in terms of reduced length and time scales, respectively \( R/L_0 \) and \( (t^* - t)/\tau_C \), to focus on this regime. Note that we should have used the length scale \( \eta \sim \xi_C \), but close to the critical point \( \eta \sim \eta_{cr} \sim \xi_C \), the viscosity is critical. As shown in Fig. 3, the measurements all fall onto a single master straight line over more than one order of magnitude in rescaled length and time scales. We extract \( H(\eta_{cr}/\eta_{out} = 1) \approx (2.9 \pm 1) \times 10^{-2} \) from the fit of the whole data set exhibiting a linear regime, illustrated in Fig. 3 Inset, which is in agreement with previous measurements \( H(\eta_{cr}/\eta_{out} = 1) \approx (3.3 \pm 1) \times 10^{-2} \) (2). Note that this rescaling requires confidence in the value of the interfacial tension \( \gamma(T - T_C) \), which is here deduced from \( R^* \) and the set value of \( (T - T_C) \). However, weak temperature variations around the set point as well as minor deviations from the set composition of the sample can produce large relative variations of \( \gamma \) for temperatures close to \( T_C \). Consequently, some experiments were preceded by in situ contactless measurements of the interfacial tension from the meniscus deformation by the optical radiation pressure at a very low beam power. As briefly discussed in SI Text, this method leads to a relative uncertainty \( \gamma H(1)/\eta_{cr} \) for the viscous regime.
Eventually, the neck thinning deviates from the linear viscocapillary behavior and accelerates during the last instants, indicating the presence of an additional more-efficient mechanism operating at small length scales and expected when interfacial fluctuations play a role. Although small scales may be difficult to determine (Materials and Methods), the fact that this deviation appears systematically for a wide range of conditions signals the onset of a different rupture regime. To further investigate this regime, the data set for all \((T-T_C)\) examined is now rescaled with the correlation length and time scales \(R_{\xi}^t\) and \((t^* - t)/t_{\xi}^t\). Fig. 4 shows that data rescaling leads to a single behavior up to a systematic variation in \(t^* - t\). (Inset) Fit of the whole set of data belonging to the viscocapillary regime.

Upper Inset and gives \(R = A_{\xi} \xi^g \left(t^* - t_{\xi}^t\right)^{0.42}\), with \(A_{\xi} = 1.61\), when forcing the exponent to its predicted value 0.42. A free-parameter nonlinear fit leads to \(A_{\xi} = 1.71\) and an exponent 0.37. The existence of a universal thermal fluctuation-dominated regime is thus firmly demonstrated by finding, over two orders of magnitude in rescaled time, a robust exponent close to the numerically calculated one, 0.42. Our data also allow a measurement of the amplitude \(A_{\xi}\) over a wide range of conditions, and show that the relevant length and time scales for this pinching regime are indeed the correlation length and its relaxation time.

Fig. 4 also shows that the cross-over from the viscocapillary to the thermal fluctuation-dominated regime appears as an inflection point at a well-defined range in rescaled time and radius, centered around \((t^* - t)_{\text{cross-over}}/t_{\xi}^t \approx 6\) and \(R_{\text{cross-over}}/\xi^t \approx 3\), i.e., \(R_{\text{cross-over}} \approx L_{\xi}\). The cross-over to the fluctuation-dominated regime therefore occurs at scales comparable to the height fluctuations of the interface, which in this case are simply proportional to the bulk correlation length \(\xi^t\). Again and despite the difficulty of extracting such small values of the neck radii, a systematic variation in \(\xi^t\) of the cross-over radius is observed. One may argue that this cross-over originates from a balance between the driving capillary pressure and the additional pressure in the neck due to the fluctuating interface. Neglecting the axial curvature, the Laplace pressure inside the neck is \(\gamma/R\).

Besides, the thermal energy density in an elementary cylinder of radius \(R\) and length \(L\) is given by \(\frac{\kappa \rho T}{2 R^2}\), where \(\xi^t\) represents the correlation length of the interface fluctuations, which is proportional to \(\xi^t\): (20). Because \(k_B T_C/\gamma = (\xi^t)^2/2 R^2\), this balance occurs for \(R\) proportional to \(\xi^t\). We identify this radius as the cross-over between the two regimes. Fig. 4 Lower Inset shows that \(R_{\text{cross-over}}(\xi^t)\) is indeed consistent with a linear variation \(R_{\text{cross-over}} = 3.5\xi^t\).

**Discussion**

Besides being of importance as it tackles the difficult problem of hydrodynamics of strongly fluctuating media (21), the thermal fluctuation-dominated pinch-off regime is supposed to have clear repercussions on the formation of satellite drops. To shed light on this issue, we considered different \((T-T_C)\), to tune the amplitude of fluctuations, and different beam powers and waists to modify the mean radius \(R_0\) of the initial light-sustained liquid column. This column ends up breaking into a number of main droplets due to the Rayleigh–Plateau instability (4) when light is turned off. Besides these main drops, smaller droplets may appear in between, which are referred to as satellite drops. Fig. 5 A–C shows that the number of satellites depends on the ratio between the initial radius \(R_0\) and the correlation length \(\xi^t\): the smaller this ratio the smaller the number of satellites. Satellite droplets are basically absent in Fig. 5A, where only the main drops are present, whereas farther away in temperature from the critical point, they are systematically present. To quantify this...
fl may coexist with symmetric ones in the formation. Nonetheless, the exact details of the evolution of the major role of thermal fluctuations in preventing satellite drop formation, the subtle interplay between the temporal evolution of the neck shape and the production of these satellites in the presence of fluctuations calls for additional theoretical and experimental work.

In conclusion, we have demonstrated the robustness of the signature of thermal fluctuations on the thinning dynamics of liquid necks, as well as the existence of a well-defined cross-over to this regime. Our measurements bring a quantitative description of this regime in a near-critical fluid. Because the existence of this regime requires self-similar solutions, our results bring support to their relevance even in strongly fluctuating systems. We have also shown that the consequences of such fluctuation-dominated thinning can be quite important for the production of satellite observations, we present in Fig. 5D the fraction of satellite drops, defined as the ratio of the number of satellite drops to the number of necks between main drops, vs. \( R_0 \). This figure shows that the mean satellite fraction is roughly unity for \( R_0/\xi^\ast \sim 50 \) (an example of an almost bidisperse situation can be seen in Fig. 5B), and strongly fluctuates in the range \( R_0/\xi^\ast \sim 60–80 \) from one experiment to another, and along the same liquid thread as illustrated in Fig. 5C, where zero, one-, and two-satellite events are present in the same snapshot.

Most important is that this satellite fraction is a decreasing function of \( R_0 \) and goes to zero when \( R_0 \) becomes smaller than a cutoff value \( R_c \approx 10^{-2} \) (a linear fit leads to \( R_c/\xi^\ast \approx 13 \)). This decrease and the fact that \( R_c \) is close to \( R_{cross} \) both point to the major role of thermal fluctuations in preventing satellite drop formation. Nonetheless, the exact details of the evolution of the neck profile from asymmetric in the two-fluid viscoplastic regime to symmetric in the fluctuation-dominated regime and its link to the decrease of the satellite fraction in Fig. 5D remains to be elucidated. In addition, as noted in Fig. 1C, elongated necks may coexist with symmetric ones in the fluctuation-dominated regime, leading to the presence of a small number of satellite drops [as for \( (T-T_c) = 0.1 \, \text{K} \) in Fig. 5D]. Though the fluctuation-dominated regime does inhibit satellite drop formation, the substantial decrease and the fact that \( \mu_c/\xi^\ast = 200 \, \text{mW} \), leading to \( R_0/\xi^\ast = 55 \); note the production of an almost bidisperse drop distribution. (C) \( (T-T_c) = 1 \, \text{K} \), \( \omega_0 = 3.0 \, \mu\text{m} \), and \( P = 200 \, \text{mW} \), leading to \( R_0/\xi^\ast = 1134 \, \text{mW} \); note the simultaneous presence of zero, one-, and two-satellite events keeping a satellite fraction close to 1. The interface roughness increase due to the interfacial thermal fluctuations can also be noticed from C to A when the critical point is neared. (Scale bars: A–C, 20 \, \mu\text{m}).

Fig. 5. (A–C) Typical liquid thread before and after destabilization for different \((T-T_c)\). (A) \((T-T_c) = 0.1 \, \text{K}\), and a laser beam of waist \( \omega_0 = 1.4 \, \mu\text{m} \) and power \( P = 130 \, \text{mW} \) to produce a liquid column of rescaled radius \( R_0/\xi^\ast = 15.8 \); note the absence of satellite drops. (B) \((T-T_c) = 0.5 \, \text{K}\), \( \omega_0 = 3.0 \, \mu\text{m} \), and \( P = 200 \, \text{mW} \), leading to \( R_0/\xi^\ast = 55 \); note the production of an almost bidisperse drop distribution. (C) \((T-T_c) = 1 \, \text{K}\), \( \omega_0 = 3.0 \, \mu\text{m} \), and \( P = 200 \, \text{mW} \), leading to \( P = 1134 \, \text{mW} \); note the simultaneous presence of zero, one-, and two-satellite events keeping a satellite fraction close to 1. The interface roughness increase due to the interfacial thermal fluctuations can also be noticed from C to A when the critical point is neared. (Scale bars: A–C, 20 \, \mu\text{m}).

Fig. 6. (A–E) Photographs extracted from an experiment performed at \((T-T_c) = 0.3 \, \text{K}\) show a close-up of the neck region to be analyzed to measure the minimum neck diameter and therefore the minimum neck radius used in this study. (Scale bar: A, 5 \, \mu\text{m}). The images correspond to (A) \( 0.86 \, \text{s}, \) (B) \( 0.56 \, \text{s}, \) (C) \( 0.17 \, \text{s}, \) (D) \( 0.08 \, \text{s}, \) and (E) \( 0.02 \, \text{s} \) before estimated rupture time. The associated intensity profiles are obtained by averaging along the direction perpendicular to the symmetry axis of the neck over the few pixels of the window indicated in A. These profiles are fit to a Gaussian whose minimum width is obtained by moving the window up and down. (F) Illustration of fits, respectively, denoted a–e in correspondence to A–E and arbitrarily shifted in intensity; the corresponding widths are (a) 19 pixels, (b) 17 pixels, (c) 11 pixels, (d) 7.5 pixels, and (e) 6 pixels. The minimum diameter, from which the minimum neck radius is obtained to plot the thinning dynamics of the neck vs. time, is taken to be the width at half-maximum of the Gaussian fit, i.e., the minimum width of the Gaussian is multiplied by \( \sqrt{n/4} \).
drops, as predicted (12, 14), even though how this works precisely remains to be addressed. This property may be useful to produce monodisperse drops at very small scales, with examples ranging from nanojet devices such as carbon nanotube channels (22) to the fragmentation of nanowires by thermal annealing (23) for creating chains (24) or patterns (25) of monodisperse nanoparticles.

Materials and Methods

The minimum neck radius measurement is carried out using movies of the break-up of the chosen liquid neck taken at frame rates between 100 and 500 frames per second. An example is given in Fig. 6 for $T = T_0 + 0.3$. First, and as depicted by the rectangular window in Fig. 6A, the images are inspected to visually delimit the region of minimal neck diameter. The intensity profile in the direction perpendicular to the neck is then measured and averaged over the few pixels of the depicted window along the direction of the neck.

This intensity profile is fit to a Gaussian up to the black stripes associated with the interface. By translating the window along the neck over which the intensity profile is measured and averaged, different Gaussian widths can be measured, and we select the minimum width at half-maximum as the minimum neck diameter. The resulting intensity profile corresponding to Fig. 6A is reported vs. distance in pixels in Fig. 6F, row a. At the used magnification, each pixel corresponds to 0.1 μm. Intensity profiles are then measured at successive times. Several images as well as their associated profiles are shown in Fig. 6 at different times before rupture; rows a–e in Fig. 6F correspond to snapshots A–E. As the diameter becomes smaller and smaller, this determination becomes more and more difficult because the profiles become less well-defined. Still, such a procedure remains quite reasonable down to at least 0.6 μm in diameter, as shown in Fig. 6E. Diameters below 0.4 μm become very difficult to measure, because the contrast between the neck and the outer medium becomes very small. Further inspection of the rupture region, nevertheless, allows estimating the final rupture time.

Supporting Information

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SI Text

Near-Critical Micellar Phases of Microemulsion. We used a near-critical phase-separated micellar solution of a microemulsion because, as illustrated below, supramolecular liquids have many advantages when investigating hydrodynamics near a critical point. The chosen microemulsion is a four-component liquid mixture composed of water, oil (toluene), surfactant (SDS), and cosurfactant (l-butanol-1). At low concentrations in water and surfactant, the quaternary mixture can in fact be considered as binary because it organizes at thermodynamic equilibrium as a suspension of surfactant-coated water nanodroplets, the micelles, dispersed in a continuum mainly composed of toluene. In the considered part of the phase diagram, a line of low critical points exists, the coexistence curves being inverted compared with the usual case (1). For the chosen composition [toluene: 70% (wt), water: 9% (wt), SDS: 4% (wt), and butanol: 17% (wt)], the micelle size, given by the amplitude factor of the correlation length of density fluctuations, is \( \xi_0 = 40 \) Å (2). This value is sufficiently small for the mixture to be transparent in the visible range and large enough, typically 10x that of classical fluids, to facilitate observation of critical opalescence. This micellar phase of microemulsion is isotropic and belongs to the universality class \((d = 3, n = 1)\) of the Ising model (3), as most of classical liquid mixtures. For the chosen composition, the critical temperature is \( T_C \approx 35^\circ C \). Above \( T_C \), the mixture separates in two micellar phases of different micelle concentrations \( \Phi_{i=1,2} \), as indicated in the schematic phase diagram shown in Fig. S1A. Many fluid properties present scaling-law behavior in \((T - T_C)\) near the critical point. Of interest for the present investigation are the following:

1. \( i \) The bulk correlation length of density fluctuations in the two-phase region: \( \xi = \frac{\xi_0}{(T - T_C)^{\nu}} \) with \( \nu = 0.63 \) and \( \xi_0 = 40 \) Å/1.9 = (21 ± 1) Å.
2. \( ii \) The coexistence curve, assumed to be symmetric close to the critical point: \( \Phi_{i=1,2} = \Phi_C + (-1)^i \frac{\Delta \Phi}{2} \left( \frac{T - T_C}{T_C} \right)^{\beta} \) with \( \beta = 0.325 \), \( \Phi_C = 0.11 \), and \( \Delta \Phi = 0.42 \).
3. \( iii \) The density of the coexisting phases: \( \rho_{1,2} = \rho_{mic} + \rho_{cont}(1 - \Phi_i) \), where \( \rho_{mic} = 1.045 \) kg/m\(^3\) and \( \rho_{cont} = 850 \) kg/m\(^3\) are the densities of the micelles and the surrounding oil continuum, respectively.
4. \( iv \) The interfacial tension between the coexisting phases: \( \gamma = \gamma_0 \left( \frac{T - T_C}{T_C} \right)^{\delta} \) with \( \gamma_0 = 0.108 \) x \( k_B T_C / (\xi_0^{\gamma})^2 = 10^{-4} \) N.m\(^{-1}\) (ref. 4).
5. \( v \) The optical absorption at the used wavelength: \( \alpha_0(k_A = 532 \text{ nm}) \approx 0.03 \text{ m}^{-1} \), which prevents the mixture from laser heating at the used beam powers.

Generating Liquid Columns. The experimental procedure to produce stable liquid columns is presented in Fig. S1B. A laser beam is focused on the meniscus of the phase-separated mixture (contained in a 1- or 2-mm-thick sealed Hellma cell) using a 10× Olympus microscope objective (N.A. = 0.25). The beam power and the beam waist \( \omega_0 \) can be changed using various optical components. Because the interfacial tension of near-critical interfaces is extremely weak, meniscus deformations are induced by the radiation pressure of the continuous laser beam, here a frequency-doubled Nd\(^3+\) : YAG (wavelength in vacuum \( \lambda_0 = 532 \) nm) in the TEM\(_{00}\) mode. The interface bending direction does not depend on beam propagation because photons gain momentum when crossing the interface from a low to a large refractive index medium. Consequently, momentum conservation always leads to a meniscus bending toward the fluid of smallest index of refraction, i.e., here from \( \Phi_2 \) to \( \Phi_1 \), because \( i \) \( \Phi_2 \) is the liquid phase of lowest micellar concentration and \( ii \) the index of refraction of toluene is larger than that of water. Nonetheless, the generation of stable liquid columns (Fig. S1 C and D) requires a beam incidence from the fluid of largest index of refraction (6), i.e., the liquid phase \( \Phi_2 \). The beam should then propagate downward as indicated by the arrow in Fig. S1C. Above a beam power threshold, the bended meniscus becomes unstable, forms a jet, and produces a stable liquid column when the jet tip reaches the bottom face of the cell containing the sample. Comparison between Fig. S1 C and D shows that the column radius can be tuned by varying the beam power. Observations are performed from the side using a focused white-light source for illuminating the sample and a 50× Olympus microscope objective (N.A. = 0.45) for imaging using a video camera and recording the destabilization of the liquid columns when the laser is turned off. A spectral filter is also placed between the microscope and the camera to eliminate the laser light scattered by the micellar phases near the critical point.

Contactless Measurement of the Interfacial Tension. The measurement of ultralow interfacial tensions is very difficult by usual contact techniques. We thus used the radiation pressure of the laser beam at low beam power to deform the meniscus separating the two coexisting phases and deduce \( \gamma \) at a given \((T - T_C)\) from the stationary deformation height \( h(r = 0) \) on beam axis (7). In the weak deformation regime \((\partial h / \partial r < 1)\), this height is given by \( h(r = 0) = \left( \frac{\pi \rho_0}{\rho_{mic}} \right) \left( \frac{\mu}{\rho_{mic} \gamma} \right)^{1/2} \exp \left( \frac{-4}{\pi \rho_{mic} \gamma} \right) \), where \( \left( \partial h / \partial r \right)_T \) is the refractive index variation with density, \( l_c = \sqrt{\frac{6}{\pi \rho_{mic} \gamma}} \) is the capillary length, and \( E_1(x) \) is the exponential integral function (6). For example, we find \( h(r = 0) \approx -5.5 \pm 0.3 \) µm for an experiment performed at \((T - T_C) = 0.3 \) K, \( \omega_0 = 7.48 \) µm, and \( P = 33 \) mW; and \( h(r = 0) = -8.0 \pm 0.3 \) µm at \((T - T_C) = 0.4 \) K, \( \omega_0 = 7.48 \) µm, and \( P = 66 \) mW; the uncertainties on \( h(r = 0) \) results from the use of a 20× objective, instead of 50×, for imaging the entire interface deformation. We respectively deduce \( \gamma \approx 1.8 \times 10^{-8} \) N/m and \( \gamma \approx 2.6 \times 10^{-8} \) N/m. Taking into account the uncertainty on \( h(r = 0) \), the estimation of \( \left( \partial h / \partial r \right)_T \) from the Clausius–Mosotti relation, and the relative error \( \leq 10\% \) on \((T - T_C) \) at \((T - T_C) = 0.3 - 0.4 \) K, we find a deviation \( \leq 20\% \) on \( \gamma \) compared with the value calculated from the universal ratio \( \mathbb{R} \).


Fig. S1. (A) Schematic phase diagram of the micellar phase of microemulsion used. $T$ is temperature and $\Phi$ is the volume fraction of micelles; $T_C$ is the critical temperature and $\Phi_1$ and $\Phi_2$ are, respectively, the volume fractions of the micelle-rich and -poor phases in coexistence. (B) Experimental configuration for a temperature $T > T_C$. The optical bending of the meniscus of the phase-separated liquid mixture is driven by the optical radiation pressure of the laser beam, represented by the arrows. (C) Thinnest stable large-aspect-ratio liquid column obtained at $(T - T_C) = 4$ K for a beam power $P = 410 \text{ mW}$ and a waist $\omega_0 = 3.5 \mu\text{m}$. (D) Tuning of the liquid column diameter with further increase of the beam power to $P = 1.134 \text{ mW}$. The liquid column length is 334 $\mu\text{m}$.