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Laser-sustained liquid bridges

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Abstract. – The stabilization of free-standing liquid bridges encounters a fundamental limitation associated to the Rayleigh-Plateau instability. Classically in weightless conditions, a liquid column breaks when its length exceeds its circumference. We overcome this fundamental limitation using a new technique, based on the optical-radiation pressure of a continuous laser wave, to form and stabilize pure dielectric bridges far beyond the instability onset. Since control over aspect ratio and orientation are simply realized by adjusting the waist and the direction of the exciting laser, these laser-sustained liquid columns also behave as reconfigurable optical waveguides and provide an appealing example of self-adapted optical microsystem based on microfluidics.

Introduction. – Liquid columns are simple free-standing structures that play a major role in many physical areas as different as the floating-zone configuration of crystal growth [1], micro-powder adhesion [2] and capillary condensation [3], or nanolithography [4]. However, these processes encounter a fundamental limitation associated with the Rayleigh-Plateau (R-P) instability [5]. In weightless conditions, a liquid column breaks when its length exceeds its circumference because capillary forces tend to minimize the interfacial energy. Rupture occurs for aspect ratio (length/diameter) even smaller in the presence of gravity [6]. A great deal of efforts has therefore been devoted to bypass this intrinsic difficulty and repel the instability onset: compensation of gravity by magnetic field [7], stabilization under electric fields [8–11] for dielectric bridges or use of acoustic radiation pressure [12]. Here we present a new technique, based on optical radiation pressure, that allows to form and stabilize pure dielectric bridges far beyond the R-P instability onset. The control over the aspect ratio of the bridge is simply realized by adjusting the waist of the exciting laser and the relevant parameters for stability are identified. These laser-induced liquid columns behave as reconfigurable self-written optical waveguides and provide an appealing example of microfluidic-based devices for applications in adaptative optics.

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Optical tweezers [13] are nowadays common manipulation and characterization tools of biological and colloidal systems and represent even a way to tailor materials in three dimensions [14]. Surprisingly, the mechanical effects of light on liquid surfaces have attracted attention much more recently [15, 16], despite the seminal experiment made by Ashkin and Dziedzic [17] in 1973. The principle is the following. As photons experience a net change of momentum at the interface between two dielectric fluids of different refractive indices, the meniscus is forced to bend to ensure momentum conservation. The resulting Laser-Induced Surface Deformation (LISD) is always directed towards the medium of smaller optical density regardless of the direction of propagation of the exciting laser beam [15,17]. As the height of the deformation strongly depends on the capillary length l_C of the interface [16] ($l_\mathrm{C} = \sqrt{\frac{\sigma}{\Delta\rho g}}$, where σ is the surface tension of the interface and $\Delta\rho$ the density contrast between the two fluids), LISD represents a powerful non-invasive technique for measuring surface tensions, which is particularly accurate in the case of very soft interfaces [15]. The same forces have also been used to study the viscoelastic properties of biological cells [18]. Whereas these examples focus on probing applications, our purpose here is to show how optical radiation pressure effects can be extended to the creation of artificial liquid structures such as liquid columns of high aspect ratio that would be unstable otherwise.

Experimental. – Experiments are performed in phase-separated near-critical binary liquid mixtures. More detailed descriptions of the composition and methods have been given elsewhere for related experiments [16]. For a temperature $T > T_\mathrm{C}$ (where $T_\mathrm{C} \simeq 35\,^\circ\mathrm{C}$ for our system), the mixture separates into two phases Φ_1 (density ρ_1, refractive index n_1) and Φ_2 (ρ_2, n_2) of different composition. The resulting low surface tension of the meniscus separating the two phases renders this system particularly convenient for LISD because moderate laser beam power P becomes sufficient to induce micrometric interface deformations [16]. Generality of the purpose is nevertheless ensured because our mixture belongs to the universality class ($d = 3$, $n = 1$) of the Ising model that characterizes most of the classical liquids. Finally, another major advantage of near-criticality is the ability to continuously tune the relevant parameters of the experiment (such as surface tension, and refractive index and density contrasts) by simply varying the temperature gap $T - T_\mathrm{C}$. A single medium can thus be used to explore a large part of the parameter space.

A sketch of the experimental set-up is shown in fig. 1. As the more refractive phase Φ_2 wets glass better than Φ_1, we use the wetting films of Φ_2 that coat a glass capillary immersed in the thermo-regulated cell as disconnected liquid reservoirs. Then, in order to optically induce liquid columns, we exploit the general properties of optical-radiation pressure effects on liquid interfaces. First, as already mentioned, LISD is always directed towards the less refractive medium, *i.e.* the phase Φ_1 in our case. Thus, as illustrated in fig. 2, focusing of the beam within the capillary induces deformations of both wetting films which stand face to face and are disjoined at low beam power. The formation of a column is then ensured by the fact that the lower deformation becomes unstable when the incoming beam experiences total internal reflection at the edge of the induced deformation [19]. This "opto-hydrodynamic" instability occurs above a well-defined onset in laser beam power P_S (see snapshots c) and i) in fig. 2). It plays a role similar to the electro-hydrodynamic ejection of liquids from Taylor cones used to form liquid bridges by remote control in micro-gravity [10]. Once the bridge is formed, achievement of an almost cylindrical liquid column generally requires further increase in P. We should mention here that interface excitation by a downward laser beam leads exactly to the same result, instability occurring in this case on the upper deformation. The formation of liquid columns in a capillary is thus totally independent of the direction of propagation of the laser.

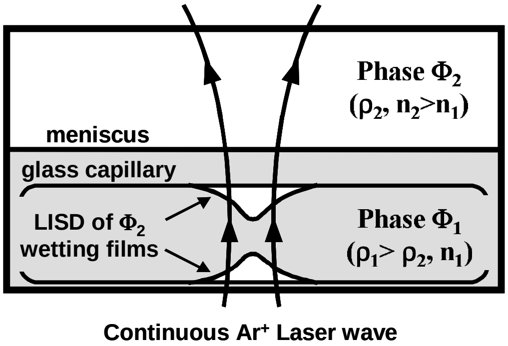

Fig. 1 – Schematics of the experiment. The phase-separated liquid mixture is regulated at a temperature T fixed above the critical temperature T_{C}. The less refractive phase Φ_1 (density ρ_1, refractive index n_1) is the denser one. A glass capillary of well-defined height (100 or 200 μm) stays at the bottom of the cell. Its surface is coated by a wetting film of the most refractive phase Φ_2 that is deformed by the radiation pressure of a focused linearly polarized TEM_{00} continuous Ar^+ laser (wavelength in vacuum $\lambda_0 = 514\,\mathrm{nm}$) propagating vertically from Φ_1 to Φ_2.

Results and discussion. – Liquid columns are classically characterized by defining the aspect ratio $\Lambda = \frac{L}{2R}$ as the ratio of their height L over their mean diameter $2R$. Another important quantity for liquid-bridges stability is the gravitational Bond number $B_{\mathrm{g}} = (\frac{R}{l_{\mathrm{C}}})^2$ which compares buoyancy to surface tension forces. It should be noticed that for both experiments shown in fig. 2, performed, respectively, at $T - T_{\mathrm{C}} = 6$ and $15\,\mathrm{K}$, the associated

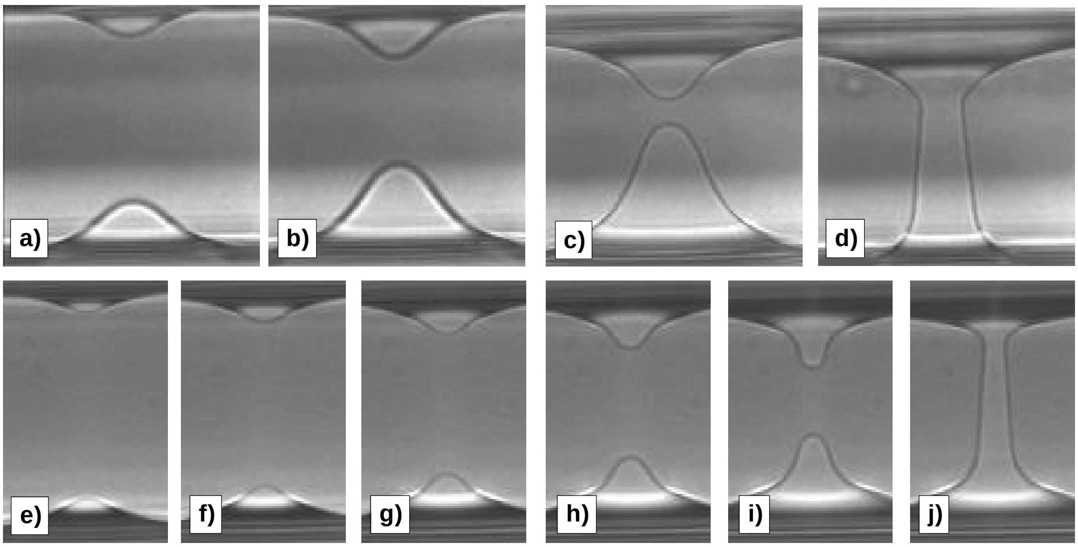

Fig. 2 – Liquid-bridge formation in a $100\,\mu$m height glass capillary. a) to d) $T - T_{\mathrm{C}} = 6\,\mathrm{K}$ and $\omega_0 = 15.3\,\mu$m. The increasing beam power values are, from left to right, $P = 1500$, 2000, 2150 ($= P_S$) and 2300 mW. The resulting liquid bridge has an aspect ratio $\Lambda = 3.6$ for a Bond number $B_{\mathrm{g}} = 0.21$. e) to j) $T - T_{\mathrm{C}} = 15\,\mathrm{K}$ with $\omega_0 = 5.3\,\mu$m. $P = 900$, 1100, 1400, 1600, 1750 ($= P_S$) and 1900 mW. The aspect ratio of the bridge is $\Lambda = 6$ and $B_{\mathrm{g}} = 3.2 \cdot 10^{-2}$.

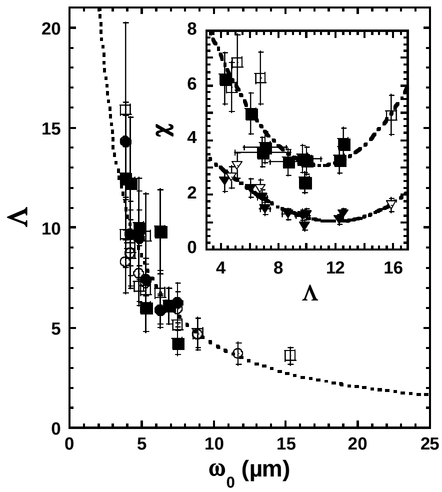

Fig. 3 – Liquid-bridge stability analysis. Aspect ratio Λ *vs.* beam waist ω_0 for liquid bridges stabilized in a $100\,\mu$m height capillary at four different temperature gaps: $T - T_{\rm C} = 6\,$K (□), $8\,$K (○), $10\,$K (●) and $15\,$K (■). The dotted line is a power law adjustment of the data leading to $\Lambda \propto \omega_0^{-1.02}$. Inset: $\chi_{\blacksquare}$ (squares) and $\chi_{\blacktriangledown}$ (triangles) corresponding to the extreme temperature gaps $T - T_{\rm C} = 6\,$K (empty symbols) and $15\,$K (filled symbols). Dash-dotted lines are guides to the eye.

numbers $B_{\rm g}$ are not negligibly small compared to one. Therefore, the density-matched condition is not fulfilled and the largest accessible aspect ratio Λ should be smaller than π in the absence of a stabilizing effect [6]. The liquid bridges presented in fig. 2 stand above this limit. Moreover, their aspect ratio seems to be controlled by the waist ω_0 of the exciting laser beam: the smaller the waist, the thinner the bridges.

To investigate this issue further, experiments were conducted in $100\,\mu$m height capillaries at four different temperature gaps ($T - T_{\rm C} = 6$, 8, 10 and 15 K) and beam waists ranging from $\omega_0 = 3.2$ to $15.3\,\mu$m. Results are summarized in fig. 3. All the data presented correspond to stable bridges greater than the limit predicted by the R-P instability in weightless conditions, even for the largest waist used. It can be observed that the bridge aspect ratio Λ does not depend on $T - T_{\rm C}$, while it is clearly a decreasing function of the beam waist. A power law fit of the full data set leads to $\Lambda = (45 \pm 3)\omega_0^{-1.02}$ when ω_0 is expressed in microns. Λ is therefore simply inversely proportional to ω_0. This is a field-dependent geometric factor that is not influenced by the thermodynamic properties of the fluids used. On the other hand, the proportionality constant measured is in very close agreement with the expected value $\frac{L}{2} = 50\,\mu$m, which means that the bridge radius is equal to the laser beam waist: $R \simeq \omega_0$. Consequently, to form a liquid bridge of given aspect ratio, one just has to adjust the waist to the suitable value and increase the beam power, provided that the onset P_S corresponding to the instability of the lower deformation can be exceeded.

We quantify this point by performing an experimental analysis of the bridges stability. To do so, we define another dimensionless number χ that measures the relative importance of the optical-radiation pressure on the interface $\Pi_{\rm rad}$ *vs.* the Laplace pressure $\Pi_{\rm Laplace}$. Optical-radiation pressure is a surface force and is defined at normal incidence as $\Pi_{\rm rad} = \frac{2n_1}{c}(\frac{n_2-n_1}{n_2+n_1})I_0$, where c is the light velocity in vacuum and $I_0 = \frac{2P}{\pi\omega_0^2}$ is the laser intensity on the beam axis [20].

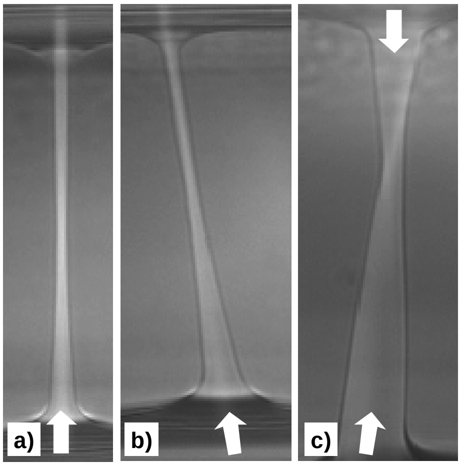

Fig. 4 – Artificial liquid structures created in a 200 μm height capillary. $T - T_C = 6$ K for the three pictures. a) Liquid bridge with an aspect ratio $\Lambda = 14$ stabilized by a laser beam of waist $\omega_0 = 3.2\,\mu$m and power $P = 1.4$ W. The corresponding Bond number is $B_g = 5.6 \cdot 10^{-2}$. The liquid column brightness evidences the self-trapping of light by the bridge. b) Same experimental conditions as in a), but the laser beam has been tilted as indicated by the arrow. c) Liquid elbow created in a two-laser beam configuration. The directions of propagation of each beam ($\omega_0 = 3.5\,\mu$m, $P = 800$ mW) are indicated by the arrows.

As $\Pi_{\text{Laplace}} = \frac{\sigma}{R}$, we get

$$\chi = \frac{\Pi_{\text{rad}}}{\Pi_{\text{Laplace}}} = \frac{2n_1}{c}\left(\frac{n_2 - n_1}{n_2 + n_1}\right)\frac{2P}{\pi\omega_0^2}\frac{R}{\sigma}\,. \tag{1}$$

By analogy with the experimental procedure used by Burcham and Saville [10] for the study of dielectric liquid bridges under electric field [21, 22], we first determine the minimum beam power $P_\blacksquare$, and the associated $\chi_\blacksquare$ value, required to sustain an almost cylindrical stable bridge after coalescence of the two deformations. Then, the power is lowered by small steps to ensure steady states and we locate the pinch-off transition. The corresponding values are noted $P_\blacktriangledown$ and $\chi_\blacktriangledown$. Results are presented in the inset of fig. 3 for the extreme temperature gaps $T - T_C = 6$ K and 15 K. The bridge stabilization and pinching occur for well-separated values of χ, which are moreover independent of $T - T_C$ for the range investigated. This point can be explained if we express χ as a function of P_S. It has been theoretically and experimentally demonstrated [19] that the interface instability onset P_S scales as

$$P_S \simeq \pi\frac{n_1}{n_2^2}\left(\frac{n_2 + n_1}{n_2 - n_1}\right)\sigma c\omega_0. \tag{2}$$

Since experiments are realized near the critical point with phases of close composition, we assume $n_1 \simeq n_2$ and χ reduces to

$$\chi \simeq 4\left(\frac{P}{P_S}\right)\left(\frac{R}{\omega_0}\right). \tag{3}$$

As $P_\blacksquare \geq P_S$ and $R \simeq \omega_0$, we find that $\chi_\blacksquare$ should take values close to 4, in good agreement with measurements. On the other hand, the formation of a liquid column strongly affects the propagation of the exciting laser. As illustrated thereafter, the bridge locally confines the beam and artificially increases its intensity. This feedback mechanism and the presence of wetting films support the hysteretic behaviour in bridge existence observed between $P_\blacktriangledown$ and $P_\blacksquare$, and thus the fact that $\chi_\blacktriangledown < \chi_\blacksquare$ for aspect ratio above R-P onset. The dimensionless number χ therefore abstracts the main features determining the bridge stability, although the question of the stabilization mechanism remains outstanding.

Indeed, it has been demonstrated that a strong axial electric field could theoretically yield to complete stabilization of pure dielectric liquid bridges with arbitrary aspect ratio [9], while the largest value experimentally achieved was $\Lambda \simeq 5$. Such an effect is nevertheless not relevant in our case because we used dielectric liquids under transverse electromagnetic field excitation. Moreover, the surprisingly high aspect ratios of the laser-sustained liquid bridges stand in contradiction with the limitations of stabilization via force fields recently underlined [23].

Concluding remarks. – The strong coupling between the induced bridge and its influence on the laser beam propagation is illustrated in fig. 4a), which depicts a liquid column of aspect ratio $\Lambda \simeq 14$ stabilized in a glass capillary of height $200\,\mu$m. The confinement of the beam inside the straight liquid column is clearly visible. Light is trapped by total reflection within the bridge because the refractive index of the liquid cladding (phase Φ_1) is smaller than that of the liquid core (phase Φ_2 with $n_2 > n_1$). One can therefore put forward the concept of liquid step-index optical fibre which provides a new approach towards self-induced wave-guiding. Indeed, contrary to optical waveguides written by photo-polymerization [24] or laser damage in glasses [25], those liquid fibres are non-permanent and thus totally reconfigurable. The laser creates its own channel that is automatically optimized to its waist. These tunable optical fibres can furthermore be oriented in any direction by on-demand tilting of the exciting beam. Figure 4b) gives an example of such an inclined liquid bridge stabilized by a laser beam propagating obliquely. Even more surprising are artificial structures like the stable liquid elbow created in a two-laser-beams configuration and presented in fig. 4c).

Optically induced liquid columns are thus particularly efficient to control beam propagation or to optimize light coupling devices because self-adaptation considerably reduces the sensitivity to precise mechanical alignments of the optical components used (see fig. 4c) for example). On the other hand, provided that radiation pressure effects drive microscopic flows in a contactless way [19], the optical analogue of electro-capillary pumping [26] of dielectric liquids can easily be accomplished and even extended to three dimensions since liquid confinement within micro-channels or by patterned surfaces [27, 28] is no longer required under optical excitation. Finally, while the results presented here were obtained with a single laser configuration, we are actually experimenting a parallel approach to form liquid-bridge patterns [29] and adaptative liquid gratings by tailoring the intensity distribution with interfering pump beams. Then, stabilization well above the R-P instability onset, smart tuning in diameter and versatility in orientation, make laser-sustained liquid bridges very promising to build microfluidic devices with optical forces [30] or, conversely, to anticipate new optical micro-systems based on microfluidics [31] for light coupling applications.

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