

## From Bouncing to Floating: Noncoalescence of Drops on a Fluid Bath

Y. Couder,<sup>1,2,\*</sup> E. Fort,<sup>1,3</sup> C.-H. Gautier,<sup>1</sup> and A. Boudaoud<sup>4</sup>

<sup>1</sup>*Physique Expérimentale, UFR de Physique, Université Paris 7 Denis Diderot, 2 Place Jussieu, 75 252 Paris Cedex 05, France*

<sup>2</sup>*Laboratoire Matière et Systèmes Complexes, UMR 7057 CNRS - ENS - Université Paris 7, 24 rue Lhomond, 75 005 Paris, France*

<sup>3</sup>*Laboratoire Matériaux et Phénomènes Quantiques, UMR 7162, CNRS - Université Paris 7,  
2 Place Jussieu, 75 252 Paris Cedex 05, France*

<sup>4</sup>*Laboratoire de Physique Statistique, 24 rue Lhomond, 75231 Paris Cedex 05, France*

(Received 22 June 2004; published 5 May 2005)

When a drop of a viscous fluid is deposited on a bath of the same fluid, it is shown that its coalescence with this substrate is inhibited if the system oscillates vertically. Small drops lift off when the peak acceleration of the surface is larger than  $g$ . This leads to a steady regime where a drop can be kept bouncing for any length of time. It is possible to inject more fluid into the drop to increase its diameter up to several centimeters. Such a drop remains at the surface, forming a large sunk hemisphere. When the oscillation is stopped, the two fluids remain separated by a very thin air film, which drains very slowly ( $\sim 30$  min). An analysis using lubrication theory accounts for most of the observations.

DOI: 10.1103/PhysRevLett.94.177801

PACS numbers: 68.15.+e, 47.55.Dz, 68.03.Cd

Intuition expects the merging of two volumes of the same fluid when they come into contact with each other. This is also expected by physics. As the two free surfaces come close, a van der Waals attractive force tends to bring them into contact. As soon as this contact is effective, surface tension leads to a minimization of the interface area and to merging. This coalescence is usually slightly delayed by the need for air to flow out of the intermediate region. It was first observed by Lord Rayleigh [1] that the collision of two drops can lead to a rebound, a situation revisited since [2]. The stabilization of drops on the surface of a bulk fluid was first reported by Walker [3] in experiments using vertically vibrated soap solutions. Several more recent experiments have been concerned with other situations in which coalescence is inhibited [4–7]. Several effects have been shown to delay or inhibit merging. When the interstitial air is forced into flowing, a lubrication effect can generate a lift strong enough to keep the surfaces apart. This was observed in the vicinity of a hydraulic jump [4] or induced by either thermocapillary convection or isothermal shear [5,6]. Other experiments have shown a delayed coalescence in the presence of surfactants [7]. In all these situations the importance of the dynamics of the air layer has been invoked and studied. However, these situations are often complex, due to either the active internal motions of the two fluids or the surface elasticity induced by surfactants.

Here we investigate the situation of a drop placed on the surface of a vertically oscillating bath [8], a configuration similar to that of Ref. [3] but where the noncoalescence is obtained below the Faraday instability threshold without surfactant. This simplified situation lends itself to quantitative measurements and theoretical analysis of the basic role of the air flow.

A container ( $5 \times 5 \times 3$  cm<sup>3</sup>) is filled with silicon oil and placed on a vibration exciter driven by a low frequency

generator. Silicon oils are used to avoid surfactant effects. We explored the phenomena described below in a large range of viscosities:  $5 \times 10^{-3} < \mu < 1$  Pa s. The reported quantitative results were obtained with Rhodorsyl oil 47V 500, which has a viscosity  $\mu = 500 \times 10^{-3}$  Pa s, surface tension  $\sigma = 20.9$  mN/m, and density  $\rho = 965$  kg/m<sup>3</sup>. In our experiments, the amplitude of the imposed oscillation is below the Faraday instability threshold so that the surface of the bath is stable. Care is taken for the drop and the bath to be at the same temperature. The motion can be observed in stroboscopic light or, for transients, with a fast video camera (1000 images/s). We measure the forcing acceleration  $\gamma = \gamma_m \cos \omega t$ .

A drop deposited on a motionless bath vanishes in a few tenths of a second. If we oscillate the substrate vertically, several regimes are observed where coalescence is either completely inhibited or strongly delayed. With small drops and large enough forcing, a stationary regime is observed where the drop lifts up from the surface at each period (Fig. 1). In this bouncing regime, the air separating the drop from the film is constantly renewed: there is no aging of the system. As a result, the lifetime of these drops appears unlimited. For instance, using an oil ( $\mu = 20 \times 10^{-3}$  Pa s at  $\omega/2\pi = 80$  Hz and  $\gamma_m = 6g$ ) we kept a drop of diameter  $d \sim 1$  mm bouncing for three days. During this motion the smaller drops ( $d < 1$  mm) remain nearly spherical. As observed in Fig. 1, the larger drops become oblate when pressed onto the liquid surface and return towards sphericity when they lift up. These deformations do not generate oscillations [9–11] because the drops are smaller than the viscous length scale  $R^* = \mu^2/\sigma\rho$ . In our experiments since  $R^* \sim 1$  cm (for  $\mu = 500 \times 10^{-3}$  Pa s), even the first eigenmode is strongly damped.

Except for large drops on very viscous fluids, the bouncing is a necessary condition for the inhibition of coalescence. It occurs when the amplitude  $\gamma_m$  of the acceleration

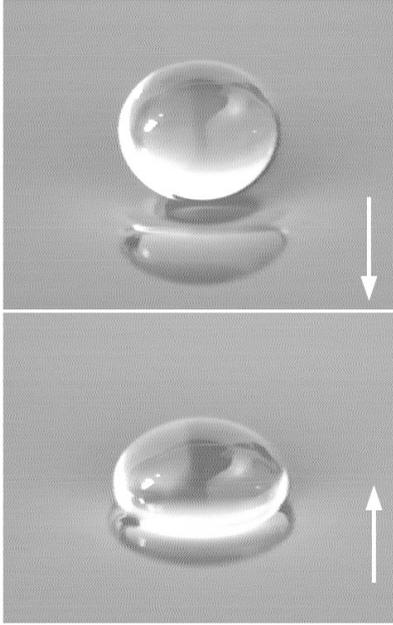


FIG. 1. Two photographs showing a drop of radius 2 mm as it bounces on the liquid surface. The arrows show the direction of the bath motion.

is larger than a threshold  $\gamma_m^C$ . Using oil with  $\mu = 500 \times 10^{-3}$  Pa s we measured  $\gamma_m^C$  as a function of the frequency, for drops of various sizes. The results [Fig. 2(a)] show that  $\gamma_m^C$  roughly grows as  $\omega^2$  for drops of a given size and that larger accelerations are needed for larger drops. A simple limit is observed for small drops at low frequencies where  $\gamma_m^C = g$ . At each period, the drop lifts when the substrate moves downward with an acceleration exceeding  $g$ . This limit is identical to that of lift-off for an inelastic solid sphere deposited on a vibrating solid plate [12–14].

Stability requires that the air film resists squeezing during the half period of upward motion and that, during the downward half period, air has time to penetrate the film to allow lift-off. We must thus consider the dynamics of the thin air film, and seek if it can sustain the drop bouncing and be renewed. At a given time the thickness of this film is  $h$  and its radius  $r_F$  [see Fig. 2(c)]. In the following, we assume that the viscosity of the liquid  $\mu$  is much larger than that of air  $\mu_a$ , so that the airflow does not entrain the liquid. At the drop landing, the film of air resists squeezing only when a viscous regime is reached, i.e., the Reynolds number becomes small enough:  $\text{Re} = \rho_a h_0^2 \omega / \mu_a < \text{Re}_c$ , with  $\rho_a$  and  $\mu_a$  being the density and viscosity of air. This condition sets a scale for a typical film thickness  $h_0$ . Reynolds lubrication theory shows that the film resists squeezing with a force of magnitude  $F \sim \mu_a r_F^4 \omega / h_0^2$  (using the vibration period as a time scale). Let  $m = 4/3\pi\rho R^3$  be the drop mass and  $\gamma$  the imposed acceleration. At landing the balance of forces gives  $-mg + F = m\gamma$ , and at lift-off  $mg + F = m\gamma$ . The lift-off condition being more restrictive determines the critical acceleration  $\gamma_m^C$  needed for

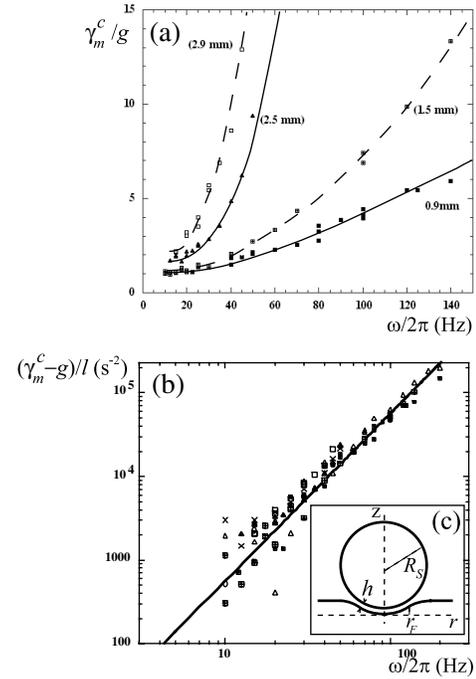


FIG. 2. (a) The observed acceleration thresholds for the bouncing of drops of various sizes as a function of the forcing frequency. (b) The rescaling of the data by Eq. (1): plot of  $(\gamma_m^C - g)/l$  as a function of the frequency. The symbols corresponding to drops of various sizes are the same as in (a). (c) Inset: Sketch of the geometry of the drop and of the separating film.

bouncing (using the scale for the film thickness found above):

$$\gamma_m^C - g = \frac{1}{\text{Re}_c} \frac{\rho_a}{\rho} l \omega^2 \quad \text{with } l = \frac{r_F^4}{R_S^3}. \quad (1)$$

Qualitatively, this means that a larger acceleration is needed to squeeze or fill the air film at a higher frequency or for a more extended film. To close the analysis, we need to estimate the film radius  $r_F$ . At the bouncing threshold, the drop lifts at a maximum of the bath oscillation, when  $\gamma = \gamma_m$ . Its free vertical motion has an acceleration  $g$ , while the bath acceleration is  $\gamma = \gamma_m \cos \omega t$ . The drop lands at a time  $t_L$ , at which the bath acceleration  $\gamma_L$  is determined only by the kinematics of the relative motion of the drop and the bath. For the maximum bath accelerations  $\gamma_m$  in the range  $2g$ – $8g$  used in the experiment, we find it in the range  $2g$ – $3g$ . The results being insensitive to its exact value, we use  $\gamma_L = -2.5g$  in the following. The shape of the drop is set by the balance between the capillary forces and the apparent inertia  $m(g - \gamma_L)$ . The relevant length scale is then the effective capillary length defined as  $\lambda = [\sigma/\rho(g - \gamma_L)]^{1/2}$ . Dimensional analysis requires  $r_F = R_S f(R_S/\lambda)$ . The function  $f$  was determined numerically by solving the capillary-hydrostatic ordinary differential equations for the equilibrium of a drop of given volume resting on a bath of the same liquid, under an effective

gravity of acceleration  $g - \gamma_L$  (a multidimensional shooting method was used). Between the drop and the bath, we used a surface tension equal to twice the liquid-air surface tension accounting for the thin film of air. This way we obtained the length scale  $l$  defined in Eq. (1). It allows the rescaling of the experimental data for the lift-off acceleration  $\gamma_m^C$ , as shown in Fig. 2(b), thus confirming that the viscous dynamics of the film prevents coalescence and sets the threshold for bouncing.

We now examine the conditions for which bouncing inhibits coalescence. The air film sustains the drop as long as it does not entrain the liquid; i.e., the shear stresses in the air  $\mu_a U/h$  are smaller than those in the drop  $\mu U/R_s$ . The drop radius is used in this last estimate as it is smaller than the penetration depth  $(\mu/\rho\omega)^{1/2}$ . The drop adapts its shape to the forcing if its relaxation time  $\mu R_s/\sigma$  is smaller than the period of vibration  $1/\omega$ . These two conditions result in a viscosity range  $\mu_a R_s/h \sim 10^{-2}$  Pa s  $< \mu < \sigma/(\omega R_s) \sim 1$  Pa s (for  $h \sim 1$   $\mu$ m,  $R_s \sim 1$  mm), which is almost the same as in the experiments. Indeed, bouncing was observed for all viscosities  $5 \times 10^{-3} < \mu < 500 \times 10^{-3}$  Pa s but in slightly different ranges of drop diameters and forcing frequencies. Typically for  $\mu = 500 \times 10^{-3}$  Pa s, drops of diameter  $d = 2R_s < 3$  mm bounce in a frequency range  $15 < \omega/2\pi < 150$  Hz. For a lower viscosity  $\mu = 20 \times 10^{-3}$  Pa s smaller drops are stable  $d = 2R_s < 1$  mm in a frequency range  $40 < \omega/2\pi < 300$  Hz.

When the amplitude of the vibration becomes smaller than what is needed for bouncing, the small drops coalesce almost instantaneously. However, for drops of intermediate sizes (e.g.,  $d \sim 3$  mm) a second regime is reached, for which the drop no longer lifts off but only oscillates with a weak amplitude. Here the main effect of the vibration is to delay the coalescence because it has the effect of “molding” the shape of the drop and the trough of the bath surface onto each other. It leads to a configuration in which an air film of relatively large area and approximately constant thickness separates the two fluids. In this regime the coalescence occurs after a few minutes delay. During this time, it is possible to insert the tip of a hypodermic needle into the drop and to inject more oil into it, a technique first used in Ref. [3]. The drop diameter can be enlarged considerably, up to, e.g., 3 cm (only limited by the depth of our cell).

A third regime is thus reached where the drop has a flat upper surface and is deeply sunk into the underlying fluid, separated from it by a thin air film (see Fig. 3). Once such a large drop has been formed, the oscillation can be switched off; the drop will survive for the same length of time with or without oscillation. This time  $\tau$  is of the order of half an hour for an oil with  $\mu = 0.5$  Pa s. With  $\mu = 0.1$  Pa s,  $\tau$  is reduced to a few minutes. For smaller viscosities, such drops become very difficult to form. This is in agreement with the lower bound for viscosity  $\mu_a R_s/h \sim 10^{-1}$  Pa s derived above. The shape of the drop is easy to understand as it depends on the Bond number  $Bo = \rho g R_s^2/\sigma$ , the ratio

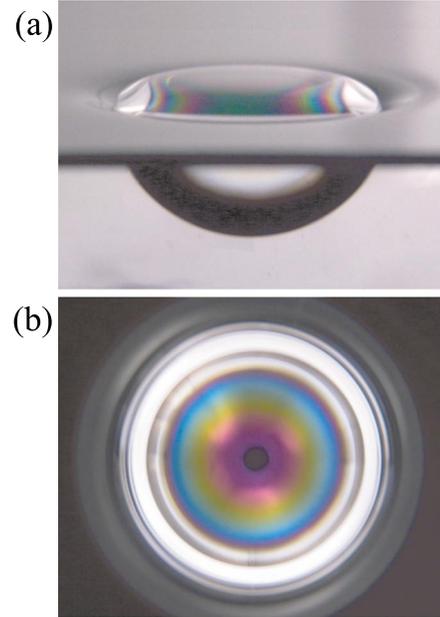


FIG. 3 (color). (a) A large floating drop, as seen from the side. (b) The interference fringes of the air film as observed when the drop is lit from the top with white light. The black dot at the center is the reflection of the camera’s lens.

of gravitational to surface tension forces. The role of gravity dominates for large drops (large  $Bo$ ), so that the upper surface of the drop becomes flat (as in the Leidenfrost experiments [11] where large drops become puddles). In the immersed part the gravity effects are exactly balanced by hydrostatic pressure so that the shape, determined by surface tension is hemispherical of radius  $R_H$  [Fig. 3(a)]. The drop and the bath having the same density, the flat part of the upper surface of the drop comes to coincide with the surface of the bath.

We thus obtain a “negative bubble” where a hemispherical air film separates two liquids. Observed with light the film exhibits colored interferences [Fig. 3(b)]. The film thickness can be measured using a monochromatic sodium lamp. As time elapses, the concentric fringes move towards the center, showing the thinning of the film. The fringe of zero order is finally reached. The order of all the observed fringes can then be deduced. Taking into account the variation of the incidence angle due to the sphericity, we can deduce the local thickness  $h$  of the film as a function of the distance to the drop axis  $r/R_H$  [Fig. 4(c)]. In the region where it can be measured,  $h$  is found to be spatially uniform [Fig. 4(a)] and to thin down with time [Fig. 4(b)].

The following interpretation can be given. Each point of the film is submitted to the hydrostatic pressure of the liquid above it generating a pressure gradient which drives the air flow out of the gap. In the limit of large viscosities the oil is not entrained by the air flow [15]. We thus have an air film flowing in a Hele-Shaw cell of variable thickness  $h(\theta, t)$ , depending on the angle  $\theta$  to the axis of symmetry

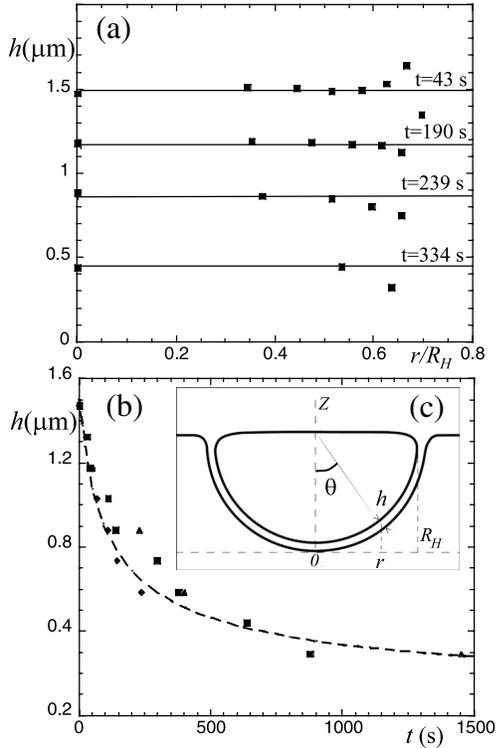


FIG. 4. (a) The local thickness of an air film (as measured from the interference fringes) as a function of the nondimensional distance  $r/R_H$  to the drop axis. The evolution at four different times shows the early stages of the film thinning. (b) The thinning air films as a function of time for three different drops with  $R_H = 1.2$  cm and for which the initial film thickness was  $1.5 \mu\text{m}$ . The dashed line is the thinning estimated by Eq. (3). (c) Inset: Sketch of the geometry of the floating drop.

[Fig. 4(c)]. The velocity averaged over the film thickness is given by  $\langle \mathbf{V} \rangle = \frac{h^2}{12\mu_a} \nabla \mathbf{P}$ , and mass conservation leads to the following lubrication equation:

$$\frac{\partial h}{\partial t} = -\frac{1}{4\tau R^2 \sin\theta} \frac{\partial}{\partial \theta} (h^3 \sin^2\theta) \quad \text{with } \tau = \frac{3\mu_a R}{\rho g}, \quad (2)$$

which has similarity solutions of the form

$$h(\theta, t) = \left( \frac{\tau}{t + t_0} \right)^{1/2} c(\theta) \quad (3)$$

$$\text{with } c(\theta) = \left( \frac{4}{3\sin^{4/3}\theta} \int_0^\theta \sin^{1/3}\varphi d\varphi \right)^{1/2}.$$

The resulting thickness is almost constant as observed experimentally [ $c(\theta)$  varies only slightly:  $c(0) = 1$ ,  $c(\pi/4) = 1.06$ ]. The time dependence predicted by Eq. (3) provides also a good fit to the measurements [Fig. 4(b)]. The drainage time does not depend on whether or not the vibration has been stopped. Drops of the same diameter have practically the same lifetime, even with

different initial thicknesses of the film. The flow in the initial thick film is fast so that, during most of the lifetime of the drop, the film is thin and the flow slow. The lifetime is limited by the bursting of the film. It was not possible to measure directly the film thickness just before its bursting. However, the extrapolation of the experimental data for the thinning suggests that the bursting occurs systematically when the film is approximately  $0.2 \mu\text{m}$  thick. This is the typical range of the attractive van der Waals forces.

In summary, whenever coalescence is inhibited, the role of the air cushion is crucial. We obtained two results: (i) by renewing the air film there can be unlimited levitation of drops; (ii) large floating drops can be formed, due to the slowness of the drainage of large air films. In both these simple situations an understanding of the dynamics of the air films is reached so that our quantitative measurements are well fitted by their theoretical analysis.

We thank A. Roger, J. Jovet, L. Quartier, and S. Protière for their help in setting up this experiment and O. Ronsin, K. Moffat, and D. Quéré for useful discussions.

\*Corresponding author.

Electronic address: Yves.Couder@lps.ens.fr

- [1] Lord Rayleigh, *Philos. Mag.* **48**, 321 (1899).
- [2] J. Quian and C. K. Law, *J. Fluid Mech.* **331**, 59 (1997).
- [3] J. Walker, *Sci. Am.* **238**, No. 6, 123 (1978).
- [4] K. R. Sreenivas, P. K. De, and J. H. Arakeri, *J. Fluid Mech.* **380**, 297 (1999).
- [5] P. Dell'Aversana, V. Tontodonato, and L. Carotenuto, *Phys. Fluids* **9**, 2475 (1997).
- [6] G. P. Neitzel and P. Dell'Aversana, *Annu. Rev. Fluid Mech.* **34**, 267 (2002).
- [7] Y. Amarouchene, G. Cristobal, and H. Kellay, *Phys. Rev. Lett.* **87**, 206104 (2001).
- [8] Alternatively, it is also possible to obtain the inhibition of coalescence with a drop hanging from a vertically vibrating pipette, set into contact with a motionless bath.
- [9] M. Papoular and C. Parayre, *Phys. Rev. Lett.* **78**, 2120 (1997).
- [10] M. Pérez, Ph.D. thesis, Institut National des Sciences Appliquées de Lyon, 2000.
- [11] A. L. Bianco, C. Clanet, and D. Quéré, *Phys. Fluids* **15**, 1632 (2003).
- [12] P. Pieranski, *J. Phys. (Paris)* **44**, 573 (1983).
- [13] N. B. Tuffillaro and A. M. Albano, *Am. J. Phys.* **54**, 939 (1986).
- [14] It can be noted that the same period doubling cascade, observed in Refs. [12,13], is obtained in our experiment with small drops, when the forcing amplitude is increased.
- [15] For weaker viscosities, the interference fringes are observed to move more rapidly. Correlatively the drop has a reduced lifetime. This faster thinning originates in the entrainment of the oil by the air motion. The Hele-Shaw approximation is no longer valid.