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Controlling the partial coalescence of a droplet on a vertically vibrated bath

T. Gilet, N. Vandewalle, and S. Dorbolo
GRASP, Physics Department, University of Liège, B-4000 Liège, Belgium

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A method is proposed to stop the cascade of partial coalescences of a droplet laid on a liquid bath. The strategy consists of vibrating the bath in the vertical direction in order to keep small droplets bouncing. Since large droplets are not able to bounce, they partially coalesce until they reach a critical size. The system behaves as a low pass filter: droplets smaller than the critical size are selected. This size has been investigated as a function of the acceleration and the frequency of the bath vibration. Results suggest that the limit size for bouncing is related to the first mode of the droplet deformation.

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I. INTRODUCTION

Liquid droplets are studied more and more intensively in the framework of microfluidic applications [1]. Among possible applications, droplets can be used in chemical engineering in order to mix tiny amounts of reactive substances. To achieve such a goal, one has to invent some processes to manipulate a droplet: motion, binary collision, fragmentation,…. To prevent droplets from contamination, it is also important to avoid any contact with a solid element. This Rapid Communication introduces a method that could eventually realize all those operations. The basic idea is to combine two physical phenomena: the delayed coalescence on a vibrated interface and the partial coalescence. First, we describe both phenomena separately and identify the physical conditions required to achieve these effects.

A. Partial coalescence

When a droplet is gently laid on a liquid bath at rest, it very quickly coalesces with the bath. Because of the low air viscosity, the drainage of the air film between the droplet and the bath lasts less than a second (a similar drainage process can be found in antibubbles [2]). Suddenly, the film becomes thin enough to break and there is contact between the droplet and the bath. A droplet made of a low-viscosity liquid can experience a partial coalescence [3]: it does not fully empty, and a new smaller droplet is formed above the bath interface (as illustrated in the second image line of Fig. 1). This daughter droplet can also coalesce partially. The process is repeated until the droplet becomes small enough to coalesce totally. When the droplet is made of a nonviscous liquid surrounded by air, the daughter droplet has a radius R_{i+1} that is about half [4] of the mother one (R_i):

$$\frac{R_{i+1}}{R_i} \simeq 0.5. \tag{1}$$

This phenomenon was first reported in 1930 by Mahajan [5] and investigated by Charles and Mason in 1960 [6]. In their experiments, another immiscible liquid was surrounding the droplet (instead of air). Recently, many studies have focused on understanding this phenomenon [4,7–11]. In general, gravity and viscosity forces in both fluids tend to favor total coalescence [4]. Partial coalescence is only possible when surface tension is the dominant force. A measure of the relative influence of the droplet viscosity compared to surface tension is given by the Ohnesorge number

$$\mathrm{Oh} = \nu\sqrt{\frac{\rho}{\sigma R_i}}, \tag{2}$$

where ν is the viscosity of the liquid, ρ its density, and σ the surface tension between the droplet and the air. As shown in [4,7,9], the critical Ohnesorge corresponding to the transition from partial to total coalescence is approximately $\mathrm{Oh}_c \simeq 0.026$. Small droplets ($\mathrm{Oh} > \mathrm{Oh}_c$) undergo a total coalescence while larger ones ($\mathrm{Oh} < \mathrm{Oh}_c$) coalesce only partially.

B. Delayed coalescence

Typically, droplets cannot stay more than a second on the liquid-air interface due to gravity. In 2005, Couder *et al.* [12,13] found an original way to avoid coalescence. In their experiment, the liquid bath is vertically vibrated by an electromagnetic shaker. The sinusoidal vibration has a pulsation ω and an amplitude A. When the reduced acceleration $\Gamma = A\omega^2/g$ is sufficiently large, the droplet bounces periodi-

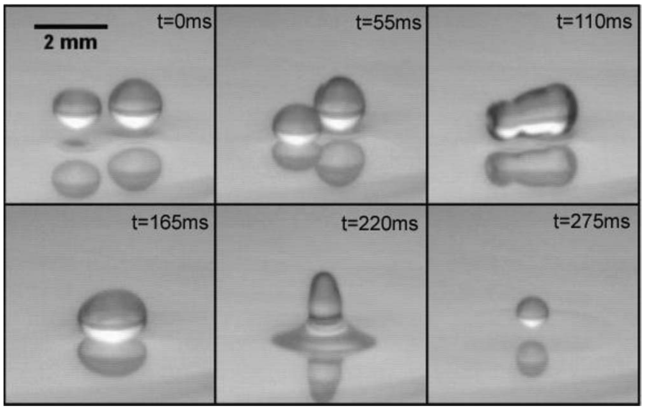

FIG. 1. Two droplets are bouncing on a vibrated liquid-air interface. They meet and coalesce. The new droplet is too large to take off from the interface and to bounce, so it eventually coalesces partially after 110 ms. The daughter droplet is small enough to bounce again for several tens of seconds. The six pictures in this sequence are taken every 55 ms.

cally on the vibrated interface. The minimum acceleration Γ_m for bouncing to occur has been estimated by Couder *et al.* [12] in the case of viscous droplets [23] as

$$\Gamma_m = 1 + \frac{1}{\mathrm{Re}_c}\frac{\rho_a}{\rho_l}\frac{\omega^2}{g}\frac{r_R^4}{R^3}, \tag{3}$$

where Re_c is a critical Reynolds number (constant), ρ_a the air density, ρ_l the liquid density, g the gravity acceleration, R the droplet radius (when spherical), and r_R the horizontal extension of the film between the droplet and the bath. Although r_R has no simple analytical form, it has been shown [12] that Γ_m is monotonically increasing with R. This means that for a given reduced acceleration Γ, there is a critical size $R_M(\Gamma)$ such as

$$R < R_M \rightarrow \text{bouncing} \rightarrow \text{stabilization},$$

$$R > R_M \rightarrow \text{no bouncing} \rightarrow \text{coalescence}. \tag{4}$$

One can reasonably think that this conclusion is still valid when droplets are not viscous. This point will be shown below. In reality, droplets experience a finite lifetime before coalescence, even when they bounce: the film between the droplet and the bath breaks and the droplet coalesces [14], sometimes after 1000 bounces. Moreover, when the vibration amplitude is too high, steady waves appear on the bath interface, due to the Faraday instability [15]. Under these conditions, the trajectory of the droplet generally becomes chaotic and the lifetime is considerably reduced. The more viscous the bath is, the higher is the Faraday acceleration threshold Γ_F. Couder *et al.* have noted that when the bath is vibrated just below Γ_F, the droplets are sometimes able to move horizontally on the interface [13,16]. In this regime, droplets can strongly interact among themselves or with obstacles [17].

In the present study, the key idea is to combine both effects: (i) the partial coalescence, that allows one to change the radius of the droplet according to Eq. (1) and (ii) the delayed coalescence that becomes efficient when $R_{i+1} < R_M$. Conditions on surface tension and viscosity have to be satisfied in order to observe both phenomena. According to the Ohnesorge criterion [24], the kinematic viscosity of the droplet liquid has to be lower than 3.7 centistoke (cSt) in order to ensure the partial coalescence of a millimetric droplet (with $\rho_l \simeq 1000\ \mathrm{kg/m^3}$ and $\sigma \simeq 20 \times 10^{-3}\ \mathrm{N/m}$). For such a low viscosity, $\Gamma_F \ll 1$ while $\Gamma_m > 1$ according to Eq. (3). Therefore it does not seem possible to stop a cascade of partial coalescences when using the same viscosity for the droplet and for the bath. However, one can satisfy Eqs. (2) and (4) by considering a droplet made of silicon oil with a low viscosity (0.65 cSt) bouncing on a bath with a large viscosity (1000 cSt).

II. EXPERIMENTAL SETUP

We apply vertical vibrations to a 7-mm-thick bath of 1000 cSt silicon oil in order to recover the daughter droplets of the partial coalescence. The frequency of the bath vibration is varied from 40 to 100 Hz. The acceleration is tuned from 0 to $2.5g$ and measured with an accelerometer; the precision is estimated below $0.4\ \mathrm{m/s^2}$, g being the gravity.

During a partial coalescence, the coalescing part of the droplet forms a pool at the bath surface. The liquid of this pool cannot immediately mix with the surrounding highly viscous oil. Consequently, the daughter droplet of the partial coalescence has unavoidably made its first bounces on the pool formed by its own mother droplet. This does not perturbe the bouncing too much. Sometimes, due to inertial effects, the daughter droplet does not succeed in quickly stabilizing its bouncing on the vibrating interface (it was observed even when $R < R_M$). In this case, another partial coalescence is necessary.

A fast video recorder (Redlake Motion Pro) is placed near the surface. Movies of coalescences have been recorded at a rate of up to 1000 frames per second. The droplet diameter is measured from the images with an absolute error due to the finite pixel size (about 30 μm). Since the droplet is constantly deforming, this measurement is made on the few images where the droplet appears the most spherical.

III. RESULTS

Figure 1 illustrates a typical scenario observed with droplets on a vibrated interface. Two small droplets are laid next to each other on the bath. They are small enough to take off and bounce. They approach each other and coalesce into a large droplet. This resulting droplet is now too large to take off, as the thin air film that separates it from the bath cannot be regenerated during the vibration. The droplet experiences a few oscillations before coalescing: it is unstable. The coalescence is partial and the daughter droplet is small enough to bounce again. The system behaves as a low pass filter: partial coalescences occur, decreasing the radius of the droplet, until the radius of the droplet respects the condition expressed in Eq. (4). This experiment is repeated several times, with various droplet sizes.

The radii R of the smallest unstable (open symbols) and the largest stable (closed symbols) observed droplets are plotted as a function of the bath acceleration Γ in Fig. 2, for various frequencies 50, 60, 75, and 100 Hz (see legend). Between both limit values of R, a mean critical value $R_M(\Gamma,f)$ may be defined as the maximum value of the radius for a stable bouncing droplet.

The minimum acceleration $\Gamma_{min}=0.35$ needed to stabilize droplets is lower than 1. It seems impossible to find a bouncing droplet on the left of the vertical dashed line. The on-off behavior of the stabilization process around Γ_{min} has to be nuanced. Indeed, the lifetime of a bouncing droplet should be considered close to the boundaries. Some combinations of the parameters probably stabilize droplets for a longer time than others, or allow one to stop the cascade of partial coalescences at an intermediate value between $R_M=0$ and 0.4 mm.

IV. DISCUSSION

In our experiments, low viscous droplets are bouncing on a high viscous bath. It means that the deformation of the droplet is larger than the deformation of the bath: the droplet

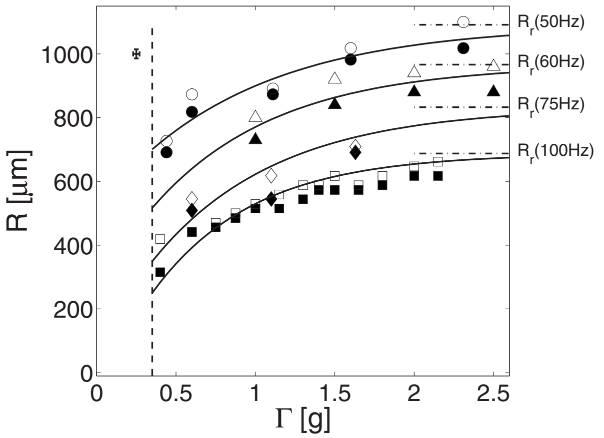

FIG. 2. Phase diagram (Γ-R). Critical radius R_M as a function of the bath reduced acceleration $\Gamma = A\omega^2/g$ for various frequencies: 50 Hz (●), 60 Hz (▲), 75 Hz (♦), and 100 Hz (■). Closed symbols correspond to the largest observed stable droplets while open symbols correspond to the smallest observed unstable droplets. The critical radius $R_M(\Gamma, f)$ is located between these two bounds. The dashed vertical line is the minimum acceleration for bouncing. The dashdot horizontal line corresponds to the asymptotic size $R_a(f)$, the maximum size of stable droplets, whatever the acceleration. This size approximately corresponds to R_r, given by Eq. (6). The typical error bar is illustrated in the upper left corner of the figure.

is more able to store and restore interfacial energy in order to ensure the bouncing. Conditions are comparable to an elastic ball bouncing on a rigid plate.

First, according to Eq. (3), $\Gamma_{min} = 1$ for viscous droplets. However, since nonviscous droplets can restore surface energy (the dissipation being limited), the bouncing is possible for reduced accelerations lower than 1: A restitution coefficient has to be taken into account. The analogy between the nonviscous droplet and the elastic ball (defined and analyzed in [19]) allows us to define this restitution coefficient α of the droplet, such as

$$\Gamma_{min} = \frac{1-\alpha}{1+\alpha} \simeq 0.35 \rightarrow \alpha \simeq 0.48. \tag{5}$$

This value is higher than the restitution coefficient of 0.22 found in previous works [20,21]. However, some aspects have to be taken into account: (1) In those experiments, small (less deformable) droplets bounce on a low-viscous (deformable) bath. The restitution mechanism is consequently not the same as in our case. (2) Moreover, for elastic beads, it is known that the restitution coefficient tends to 1 when the impact velocity tends to 0 [22]. In our case, impact velocities near the critical acceleration are about $v = \Gamma g/\omega \leqslant 2$ cm/s while in [21], impact velocities are at least ten times higher. This could also explain the difference between restitution coefficients in both configurations.

Second, experimental points of Fig. 2 define two regions in the Γ-R diagram. The boundaries that depend on the frequency are shown as continuous curves based on a qualitative fit. For a given frequency, droplets located above the curve are unstable while droplets located below are stable.

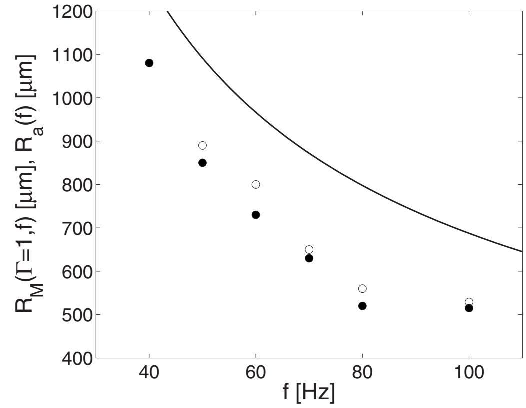

FIG. 3. Critical radius R_M as a function of the frequency when $\Gamma = 1$. Closed circles correspond to the largest observed stable droplets while open circles correspond to the smallest observed unstable droplets. The critical radius R_M is located between these two bounds. The continuous line corresponds to Eq. (6).

More precisely, when a droplet is created with a radius larger than the critical droplet radius $R_M(\Gamma, f)$, the droplet partially coalesces until it reaches the stable region (below the curves). The stability threshold $R_M(\Gamma, f)$ slightly increases with the forcing acceleration, as in Eq. (3). The curves seem to saturate at high accelerations towards an asymptotical value $R_a(f)$ that decreases with an increasing frequency.

Since the bath is much more viscous than the droplet, the droplet deformation is the main elastic mechanism able to ensure the bouncing. A characteristic size related to the deformation is given by the wavelength of the first normal mode. According to [18], for a sitting droplet, this wavelength λ satisfies to

$$\lambda^3 = (\pi R_r)^3 = \frac{2\pi\sigma}{\rho f^2\left(1+\sqrt{\frac{5}{4\pi}}\right)}. \tag{6}$$

The radius R_r corresponding to λ is represented in Fig. 2. It fits well to the asymptotical value R_a for high accelerations. This fact suggests that the first mode of droplets deformation is related to the maximum bouncing droplet size for a given frequency.

Finally, the critical radius R_M has been measured as a function of the frequency, the acceleration being fixed, $\Gamma = 1$ (Fig. 3). Open circles correspond to the smallest unstable droplets, while closed circles are related to the largest stable ones. The continuous line represents Eq. (6). The curve does not fit the experimental results because the asymptotic regime is not reached when $\Gamma = 1$. Moreover, sitting and bouncing droplet geometries are not exactly equivalent. However, we found that the decreasing of R_M with increasing frequencies (at $\Gamma = 1$) is coherent with the decreasing of R_r.

V. CONCLUSIONS

In summary, we have developed a method to stop a cascade of partial coalescences and to maintain the residual droplet alive on the interface. Our approach is based on the experiment of Couder [12] that consists of the bouncing of a droplet on a bath subjected to a vertical vibration. We have shown that it is essential to use a viscous liquid for the bath and a low-viscous one for the droplet. The threshold radius below which the droplet can bounce has been found to depend on the acceleration and on the frequency of the bath. By tuning these forcing parameters, it is possible to choose the maximum allowed size for bouncing droplets in a broad range. A concordance is observed between the radius of the largest stable droplet (for a given frequency) and the radius related to the first normal mode of deformation.

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[23] To obtain Eq. (3), Couder *et al.* assume that the droplet is not able to store interfacial energy during the bouncing. The excess of kinetic energy at impact is then dissipated by viscous forces.
[24] The Ohnesorge criterion is designed for interfaces at rest. However, it seems to prevail for interfaces in motion: only the critical Ohnesorge Oh_c should be slightly different.