Oscillation of liquid drops under gravity: Influence of shape on the resonance frequency

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Abstract. – A recently demonstrated technique of aerodynamic levitation has been used to study the resonance frequency of liquid droplets. A thin pressurized gas layer supports the droplet avoiding any contact with the environment. Due to the external gravitational field, the measured frequency is different from the one predicted by the classical models for a spherical drop. An ellipsoidal shape has been introduced to take into account the competition between surface and gravitational energies. Results of this model are in good agreement with experiments in terms of volume, as well as density and surface energy, dependence of the oscillation frequency.

Introduction. – In order to measure physical properties of liquids (such as surface energy and viscosity) one can study droplet oscillations. Indeed, when excited at various frequencies, the resonance of the oscillating droplet can be characterized by the frequency at which the peak occurs, and by the width of the resonance peak. In the Earth's gravitational field, the position of the resonance is governed by surface energy (restoring force) and density (inertial effect), whereas the width of this peak is related to the dissipative terms, namely the viscosity of the liquid. In this paper, we focus our attention on the determination of the resonance frequency of an oscillating droplet in an external gravitational field.

Lamb [1] (see also ref. [2]) gave the resonance mode frequencies for a drop of volume V, surface energy σ, density ρ in the absence of gravity. For the mode ℓ, one gets

$$f_{\rm r} = \sqrt{\ell(\ell-1)(\ell+2)\frac{\sigma}{3\rho\pi V}} \, . \tag{1}$$

Chandrasekhar [3] solved the full linear problem of the self-gravitational oscillations of a liquid globe, having in mind astrophysical problems. Later, Wang, Anilkumar and Lee [4] emphasized the nonlinear character of damped oscillations in microgravity environment as a cause of frequency shift. The nonlinearity of large-amplitude free oscillations has been numerically [5], analytically [6,7] and experimentally (for the case of a free falling drop) [8] studied. Bisch, Lasek and Rodot [9] showed experimentally that the sessile drop boundary

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conditions had a large influence on the resonance frequency in terms of contact surface between the rod and the sessile drop. They cancelled the gravity effects by embedding the drop within a fluid with the same density. However, most of the experimental situations on Earth are performed under terrestrial gravity, and therefore one needs a theoretical approach including this aspect.

Some authors tackled the problem numerically. Wikes and Bassaran [10] with a finite-element method describe the dynamics of oscillations of pendant or sessile drops. Boundary integral methods have been used to study the static and dynamics of drops within an acoustic standing wave field (Shi and Apfel [11]). Boichon [12] developed this boundary integral method to predict the static shape and the eigenmodes of an aerodynamically levitated drop.

Very few systematic experiments on the dependence of the oscillating modes under gravity with volume and nature of the liquid droplet have been performed [13]. Moreover, finite element methods are numerically very heavy and hardly adaptable.

In the present paper, systematic experiments with droplets of varying volumes are performed and the results are compared with a simple analytical model. To allow this simple description, a variational approach is introduced to calculate the equilibrium shape and the natural frequency of the droplet.

Sessile drop geometry has been extensively studied [10]. The drawback of this configuration is the necessity to take into account a contact angle between the rod and the sessile drop. The complexity of wetting hysteresis [14] added to the singular boundary conditions make the interpretation difficult. In order to bypass the difficulty, we have used a new configuration recently demonstrated by Parayre *et al.* [13]: the droplet is in levitation on a gas layer coming from a porous diffuser. In order to avoid translational modes of the droplet, the diffuser is slightly curved (its radius of curvature is much larger than the droplet radius R).

Compared with standard electromagnetic levitation methods, the present one is well adapted to insulating liquid materials. Moreover, dissipation measurements, even in a liquid metal drop, will hit directly the viscosity parameter instead of a combination of viscosity and Joule dissipation due to eddy currents.

Experimental device and choice of material. – The schematic of the apparatus is shown in fig. 1.

The drop is excited through an electromagnetic vibrator providing vertical oscillations to the system (diffuser/drop). The frequency and amplitude of the excitation are adjustable in a wide range (from 1 to 100 Hz for the frequency and 0 to 100 μm for the amplitude). A video system allows drop profile measurements with an accuracy of 10 μm (around 0.25% of the levitated droplet height). The amplitude of excitation is chosen high enough for the motion to be detectable and low enough to stay in the linear approximation and avoid anharmonicity (Papoular and Parayre [15]). The droplet's north-pole vibration amplitude at resonance is usually around 300 μm (around 7.5% of the levitated droplet height). A scanning in frequency allows to measure the resonance frequency: the one at which the oscillation amplitude at the north pole of the droplet is maximum. Resonance frequency measurement is reasonably accurate (around 0.1 Hz) and very reproducible. Only the first oscillating mode has been studied, the following ones being much smaller and occurring at much higher frequencies.

Because of the gas flux, the drop undergoes a Kelvin-Helmholtz instability [3]. To avoid this problem, the viscosity of the liquid must be higher than 1 mPa · s. In this case, the dynamical drag does not influence neither the static (the equilibrium shape of the drop can be appropriately described by solving just the Laplace equation) nor the dynamics (the gas film thickness has been proved to remain constant by levitating a solid sphere and the droplet resonance frequency is independent of the gas flow). In this work, experimental measurements have been performed on drops of various liquids, whose physical parameters (viscosity μ,
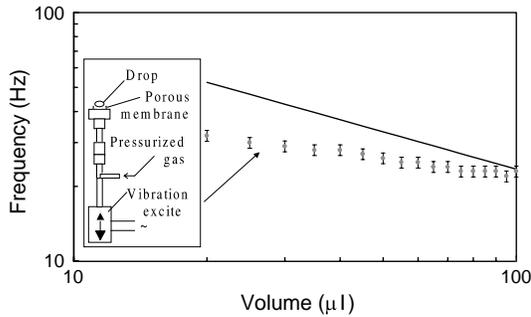
Fig. 1

Fig. 2

Fig. 1. – Experimental (dots) and theoretical (line, from eq. (1)) resonance frequency as a function of the volume of the drop.
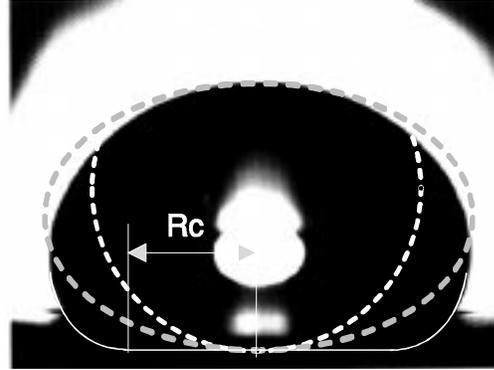
Fig. 2. – Real shape, ellipsoidal approximation (dark gray) and spherical shape (light gray) of equivalent volume for a 50 μl levitated water/glycerol (30%) drop.

volume V, density ρ, surface energy σ, and capillary length $l_c = \sqrt{(\sigma/(\rho \cdot g))}$ are listed in table I.

Concerning the water/glycerol mixture, the surface tension of glycerol is almost the same as water ($\sigma_{\text{water}} = 0.073$ J/m^2 and $\sigma_{\text{glycerol}} = 0.063$ J/m^2) so that Marangoni effects are negligible. A possible drawback could be the evaporation of water but the time needed for a resonance frequency measurement is short enough (30 s) to neglect evaporation consequences. This latter assumption is checked by the stability of the resonance peak position.

Experimental results and empirical fit. – The first vibration mode experimentally observed has been compared with the first vibration mode of eq. (1), *i.e.* the $\ell = 2$ mode, because of their geometrical similarities. The resonance frequency as a function of the water/glycerol droplet volume is shown in fig. 1. Equation (1) with no gravity effects is unable to describe the results. Both the power law exponent (-0.22 instead of -0.5) and the absolute value of $f_r(V)$ are different from the predicted result. It must be noted that the radius of the drop is close to the capillary length for water/glycerol ($l_c = 2.5$ mm). Therefore, gravity effects have to be taken into account. Lambs formula neglects gravity and it is not surprising to observe a discrepancy with experimental results. Indeed, the shape of the droplet is far from spherical. As shown in fig. 2, the aspect ratio is around 1.2 for a 50 μl water/glycerol (30%) droplet. Moreover, boundary conditions due to the presence of the diffuser below the drop also affect the dynamics and shift the resonance frequency because as the south pole is forced, the center of gravity oscillates and the experimental resulting oscillation mode is a mixture of a pure $\ell = 2$ oscillating mode and a $\ell = 1$ translatory mode. The apparent agreement of the

TABLE I. –

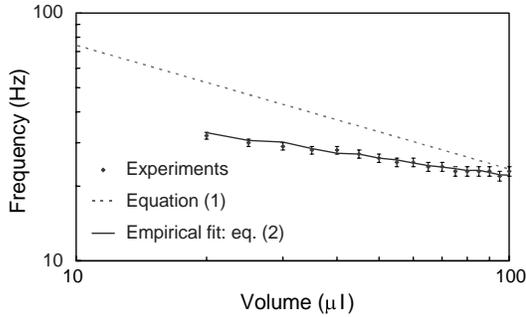| Liquid	Viscosity (mPa · s)	Volume (μl)	Density (kg/m^3)	Surface Energy (J/m^2)	Capillary length (mm)
Water/Glycerol (30%)	5	10 to 100	1078	0.066	2.5
Water/Glycerol (30%) + Surfactant	5 to 20	50	1078	0.66 to 0.032	1.8 to 2.5
Silicon Oil	10	20 to 100	900	0.017	1.4

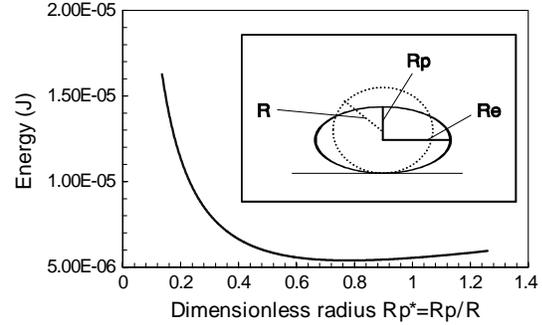Fig. 3   Fig. 4

Fig. 3. – The empirical fit (eq. (2)), experiments and eq. (1) *vs.* the volume of the droplet.

Fig. 4. – Variation of the drop energy as a function of the dimensionless parameter $R_{\rm p}^*$ for a 50 μl droplet.

experimental result with formula (1) for a volume of 100 μl is coincidental since experimental and theoretical curves cross at that point.

In simulated microgravity, C. Bisch *et al.* [9] have proposed an empirical formula which allows to bring all the results on a single master curve. On the same empirical basis, another empirical formula, taking into account the nonspherical shape has been derived:

$$f_{\rm r} = \frac{\sqrt{3}}{2} \cdot \sqrt{\frac{8}{3} \cdot \frac{\sigma}{\rho \pi V}} \cdot \sqrt{\frac{R_{\rm c}}{R} \cdot \frac{R_{\rm e}}{R_{\rm p}}} \,. \tag{2}$$

For the $\ell = 2$ mode, $R_{\rm c}$ is the radius of the (essentially horizontal) disc at the base of the droplet (see fig. 2); R the radius of the drop if it were spherical; $R_{\rm e}$ the equatorial radius; and $R_{\rm p}$ the polar radius. The first factor is eq. (1) like, the second is due to boundary conditions, and the third takes into account the nonspherical shape. The numerical coefficient $\sqrt{3/2}$ optimizes the agreement with experiments. The resulting master curve is shown in fig. 3. In spite of the good agreement between experiments and this formula, the approach is not satisfactory. One would like to have a fully consistent derivation of both vibration frequency and droplet ellipticity.

Vibrating droplet under gravity and capillary force: a variational approach. – The drop shape at equilibrium is complex (see fig. 2). Moreover, the vibrating shape can have different modes. However, keeping the amplitude small enough allows to observe a pure $\ell = 2$ mode. In what follows, only this mode is studied in the framework of a variational approach for the shape of the droplet.

Variational approach: an ellipsoid is chosen as an approximation of the drop shape. Its parameters are the polar radius $R_{\rm p}$, and the equatorial radius $R_{\rm e}$. The volume is given by

$$V = \frac{4}{3}\pi \cdot R_{\rm p} \cdot R_{\rm e}^2 = \frac{4}{3}\pi \cdot R^3 \,. \tag{3}$$

Here, R is the radius of the drop if it were spherical. V is kept constant so that the droplet shape depends only on one parameter, chosen here to be $R_{\rm p}$ (see fig. 4).

The shape of the droplet is driven by two counteracting forces: the surface tension tends to give a spherical profile to the drop ($R_{\rm p} \to R$); the gravitational force tends to flatten the drop ($R_{\rm p} \to 0$). The competition between those two forces gives the equilibrium shape.

The total energy of the system at rest can be calculated as the sum of gravitational potential and surface energy (S is the ellipsoid surface):

$$E = \sigma \cdot S + \rho \cdot V \cdot g \cdot R_{\rm p} \,. \tag{4}$$

The energy of the drop *vs.* the dimensionless parameter $R_p^* = R_p/R$ is plotted in fig. 4. The minimum of energy E for $R_p = R_{p0}$ corresponds to a stable equilibrium position. R_{p0} is a function of $t^* = \sqrt{(\rho V/\sigma)}$. It can be observed that the energy curve is strongly asymmetric around its minimum. Such a fact was qualitatively explained by Wang, *et al.* [4] noticing that an oblate ellipsoid has its maximum total curvature along the equator whereas a prolate ellipsoid concentrates the maximum curvature at the two poles.

Considering the drop as an oscillator with non-harmonic potential, its dynamics is simplified as follows: 1) the vibration amplitude is imposed small enough to stay in the linear domain where the potential is harmonic ($0.7 < R_p^* < 0.9$ in fig. 4), 2) the stiffness of the energy curve, is taken as the second derivative of E with respect to the center of gravity position, and 3) the mass M of the liquid considered in the oscillations is taken into account to evaluate the frequency: the square of the resonance frequency is given by the ratio of the stiffness over the mass M. In the case of free oscillations, only a fraction of the total mass of the droplet is concerned ($M = \phi \cdot \rho \cdot V$), where ϕ depends on the vibration mode. But if the oscillations are forced, *i.e.* if the south pole is essentially fixed and the center of gravity oscillates, all the mass has to be taken into account ($M = \rho \cdot V$): the resulting oscillation mode is a mixture of a pure $\ell = 2$ oscillating mode and a $\ell = 1$ translatory mode.

The vibration frequency of the droplet in the harmonic forced ellipsoidal oscillation regime is then

$$f_r = \frac{1}{2\pi}\sqrt{\frac{\partial^2 E/\partial R_p^2|_{R_p=R_{p0}}}{\rho \cdot V}} = \frac{1}{2\pi} \cdot \sqrt{\partial^2 S/\partial R_p^2} \cdot \frac{1}{t^*} \,. \tag{5}$$

We then can get f_r as a function of $t^* = \sqrt{(\rho V/\sigma)}$.

Limiting case:

If the volume is small enough to allow a power expansion in terms of

$$\left(\frac{R_p}{R_e} - 1\right) \approx \varepsilon \ll 1 \,, \tag{6}$$

gravity cancels out and the drop oscillates with elliptical modes around its spherical shape. The frequency will now be

$$f_{r\,\lim} = \frac{1}{2\pi}\sqrt{\frac{\partial^2 E/\partial R_p^2|_{R_p=R}}{\rho \cdot V}} = \sqrt{\frac{4}{5}\frac{\sigma}{\rho \cdot \pi \cdot V}} \,. \tag{7}$$

As a matter of fact, comparing with eq. (1), the exponents of σ, ρ and V are unchanged. This equation differs from eq. (1) only by a coefficient $\sqrt{(3/10)} \approx 0.5$. As we have just explained, this stems from the difference in effective mass between free and forced oscillations. We are in the latter case and, clearly, the experimental results are more accurately described by our model than by eq. (1). In the case of free oscillations, eq. (1) is obtained with $M = \phi \cdot \rho \cdot V$ and $\phi \approx 1/4$. In the limiting case of small volume and small amplitude of oscillations (compared to the gas layer thickness ($100 \, \mu$m)), both gravity and the diffuser boundary condition would have no effect, thus justifying spherical harmonic analysis, leading back to formula (1) for the resonance frequency.

Real shape of the droplet

As can be seen in fig. 2, the ellipsoidal shape is not the exact representation of the droplet profile. However, as the simplest way to flatten a sphere is to turn it into an ellipsoid, the ellipsoid has been chosen, as a first approximation. The same variational approach taking into account a more realistic shape (described by an Archimedian spiral arc as we shall explain elsewhere [16]) leads to similar results.
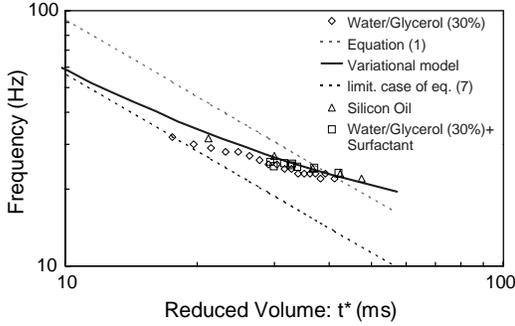
Fig. 5

Fig. 6

Fig. 5. – Comparison between variational model (continuous line), experiments ($\diamond$, $\triangle$ and $\square$), eq. (1) (gray dashed line), and limiting case of eq. (7) (black dashed line).
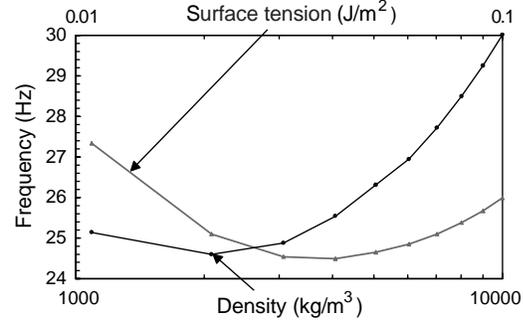
Fig. 6. – The σ- and ρ-dependence of the resonance frequency from eq. (5). The volume is fixed at $50\,\mu$l.

Discussion. – An interpretation of both experimental and theoretical results can be given as follows.

Figure 5 shows resonance frequency *vs.* the t^* parameter ($t^* = \sqrt{(\rho V/\sigma)}$). The calculated frequency is in good agreement with experiments.

Since the typical size of our systems is of the order of the capillary length, the system lies in an intermediate region for which no effect is totally dominating the behavior. For small enough t^* ($t^* < 20$ s) surface forces dominate and the frequency tends to the $f_{\mathrm{r\,lim}}$ calculated above (eq. (7)). For larger volumes, gravity becomes significant. The equilibrium shape is an ellipsoid even more oblate since t^* is larger. Compared to the t^* dependence of the limiting case of eq. (7), (dashed curve in fig. 5), the frequency is higher. This could be understood as follows: in an oblate position, the equatorial curvature is higher than in a spherical position. Oscillating around a more curved shape will involve more energy and then shift the frequency toward higher values. Those results are consistent with the numerical ones of Wilkes and Bassaran [10] concerning the effect of the drop size on its resonance frequency: the frequency shift increases as the flattening increases. The limiting case of eq. (7) (small droplets) is consistent with Wilkes and Bassaran's results for $\alpha = 0.5$ (α is their dimensionless drop size parameter). For this value of α, the oscillation mode can be considered as elliptic.

Whereas for the oscillating sphere in the absence of gravity (eq. (1)) the dependence of the frequency on ρ and σ is monotonous, the interplay between gravity and surface tension leads to a more complex dependence.

The surface energy plays two different roles. On the one hand, it weighs the importance of surface forces relative to gravity forces. On the other hand, it also makes the system stiffer. For small values of σ, when the drop shape is very oblate, an increase in σ has two conflicting effects : it will bring the drop back to a spherical shape, which tends to decrease the frequency as explained above. But it will also increase the stiffness of the system, which tends to increase the frequency. As can be seen in fig. 6, in the transition zone, the frequency is not very much σ-dependent. But once σ is high enough in order for the drop to be spherical, only the second effect dominates: the frequency becomes strongly σ-dependent.

As the surface energy, the density plays also two conflicting roles: the first is gravitational: it tends to flatten the drop and thus to increase the frequency, the second one is inertial: it tends to decrease the resonance frequency. Figure 6 shows that for low-density drops, the second effect is dominant and for high-density drop, the first one is dominant.

A comparison is made between the real and ellipsoidal shape in fig. 2. It is obvious that the model over-estimates the oblate shape of the drop. For gravitational reasons, the matter at the equatorial extremities tends to accumulate at the bottom of the drop, which does not change much the surface energy. That could explain the slight difference between the variational model and the experiments (see fig. 5).

Conclusion. – For a vibrating droplet under gravity, the variational approach presented here gives a good agreement, without any adjustable parameters, with the present experiments performed in gas-levitation geometry. This formulation, in contrast with empirical formulae previously proposed [9], allows a description of the full range of sizes, from small sizes where capillarity is dominant ($R \ll l_c$) to large ones where gravity governs the system ($R \gg l_c$).

The experimental set-up described in this paper provides us with a tool to measure surface tension accurately. For instance, segregation effects at liquid surfaces and the associated decrease in the surface tension could be studied by this method as it has been done with magnetic levitation technique [17].

Dissipation too can be introduced in this simple analytical framework. A suitable estimation of the velocity field leads to an assessment of the viscosity within the same, containerless, method [16]. The asymmetric profile of the energy curves points to the nonlinear character of the oscillations due to anharmonicity [15], and could lead to a detailed description of the parametric resonance that may occur in those oscillations [18, 16].

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