Oscillations of liquid drops: results from USML-1 experiments in Space

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Oscillations of low-viscosity drops were studied in the microgravity environment of a Space shuttle flight. From the damped oscillation data, the inviscid frequency shift, due to nonlinearity, has been extracted using a central-averaging scheme. For the classical case of the oscillations of a free low-viscosity drop, it has been found that the frequency shift agrees well with the predictions of the inviscid nonlinear theory of Tsamopoulos & Brown (1983) for $\epsilon < 0.3$. But for the oscillations of a rotating low-viscosity drop, under acoustic levitation, the frequency shift is smaller, and the percentage of time spent in prolate displacement is significantly less than that for the classical case.

1. Background

Consider a free liquid drop of spherical radius R, density ρ, and surface tension σ. In the inviscid linear limit (Rayleigh 1879; see also Lamb 1945), for an axisymmetric normal mode of oscillation, its surface is given by

$$F(\theta, t) = R[1 + \epsilon \cos \omega t\, P_n(\theta)], \tag{1.1}$$

where θ is the polar angle from 0 to π, t is time, ϵ is much less than 1 and represents the wave amplitude; $n = 2, 3, 4, \ldots, P_n$ is a Legendre polynomial, and the angular resonant frequency ω is given by

$$\omega^2 = \frac{\sigma}{\rho R^3} n(n-1)(n+2). \tag{1.2}$$

More generally, each mode n can have $2n+1$ non-axisymmetric modes m, where $m = 0, \pm 1, +2, \ldots, \pm n$, with $m = 0$ corresponding to the axisymmetric mode in (1.1). Here, the non-axisymmetric modes are degenerate in the sense that they all have the same frequency, given in (1.2).

If the kinematic viscosity ν of the liquid is small, then the oscillation amplitude is damped in time like $\exp(-\lambda t)$, where λ is given by (Lamb 1945)

$$\lambda = \frac{\nu}{R^2}(n-1)(2n+1). \tag{1.3}$$

Chandrasekhar (1961) showed that the normal mode of oscillation of a viscous free drop is a damped oscillation if $\omega R^2/\nu$ is larger than a constant M, and is an aperiodic decay if it is smaller than M. For the fundamental mode of oscillation ($n = 2$), the value of M is 3.6902. In our work, since we are primarily interested in seeing nonlinearity in drop oscillations, we have made viscous damping small by choosing ν to be low enough such that $\omega R^2/\nu$ is much larger than M.

Prosperetti (1974, 1980) considered the initial-value problem for the small-amplitude oscillation of a viscous drop. For a drop with a zero vorticity distribution, and with a static deformation along one of the axes at $t = 0$, the ensuing motion has three phases. In the initial phase, before vorticity becomes important, the oscillation frequency is given by

$$\omega' = (\omega^2 - \lambda^2)^{1/2}, \tag{1.4}$$

where ω and λ are given by (1.2) and (1.3), and the decay rate is λ. If $\lambda/\omega < 1$, the motion is oscillatory, but if $\lambda/\omega > 1$, ω' is imaginary and the oscillation is aperiodic, at least initially. After the initial phase, vorticity is generated by viscosity at the drop surface, where the tangential stress is required to be zero, and diffuses viscously into the drop. This intermediate non-equilibrium phase is associated with heightened viscous dissipation because of the generation and spreading of vorticity. When the vorticity distribution reaches equilibrium, the drop motion is in its final phase. The oscillation can then be described in general by the least-damped normal mode (Prosperetti 1977) given by Chandrasekhar (1961), which may or may not be periodic. In the low-viscosity limit, it is given approximately by (1.2) and (1.3), which is periodic.

Since after an initial adjustment, which covers the initial and intermediate phases, the drop oscillations can be described by least-damped normal modes, it is useful to estimate the time taken for the adjustment. The vorticity can penetrate to a depth of $\delta \sim (\nu/\omega)^{1/2}$ from the drop surface if $\delta < R$, and the whole drop if $\delta \gtrsim R$. The two cases are related to Chandrasekhar's criterion for $\omega R^2/\nu$: in the first case $\omega R^2/\nu > 1$ and the motion is damped oscillatory, and in the second case $\omega R^2/\nu \lesssim 1$ and it is aperiodic. In the first case, the adjustment time is of the order of δ^2/ν, i.e. an oscillation period. In the second case, it is of the order of R^2/ν. In our experiment with low-viscosity drops, we are only concerned with the first case. The initial adjustment is over after about one oscillation such that it is appropriate to consider least-damped normal modes in our data analysis.

Tsamopoulos & Brown (1983) studied the nonlinear but inviscid oscillations of drops using a perturbation technique. For $n = 2$, the shift δf due to nonlinearity in the oscillation frequency f ($f = \omega/2\pi$) is given by

$$\delta f/f = -\tfrac{1}{2} C \epsilon^2, \tag{1.5}$$

where $C = 1.17037$ (*corrected* value from Tsamopoulos 1989), such that the frequency decreases quadratically with wave amplitude.

The frequency shift can be understood as follows. At large amplitudes, the motion of the drop from its extreme prolate shape to spherical, or vice versa, is governed by the high pressure at the curved poles. Similarly, the motion during the oblate part of the cycle is governed by the high pressure at the curved equator. However, as the poles cover a smaller neighbourhood than the equator, the restoring force for the prolate part of the cycle is weaker. This brings about an asymmetry in oscillation, with relatively more time spent in the prolate form than in the oblate. Moreover, around the prolate extremum, the drop is almost cigar-shaped and the rebound of the drop to a spherical shape is opposed and slowed by the strong action of surface tension at the equator, which takes on an almost cylindrical shape at large amplitude. The result of this is an increase in the period of oscillation, or a decrease in the frequency, due to nonlinearity.

Since the drop oscillation has a linear resonance, the frequency shift should not be dependent on ϵ near $\epsilon = 0$, which rules out a linear dependence of the frequency shift on ϵ. Furthermore, assuming that the dependence on ϵ is analytical, the decrease in

frequency should be proportional to an even power of ϵ near $\epsilon = 0$, because it should be independent of the sign of ϵ, which reflects the choice of the origin of time. Hence to the lowest order, for small ϵ, the frequency shift is proportional to ϵ^2.

Trinh & Wang (1982) experimentally studied the large-amplitude oscillations of quasi-neutrally buoyant drops of silicone oil and carbon tetrachloride levitated in distilled water. They reported that, with increasing oscillation amplitude, the frequency decreases and the prolate phase lasts longer than the oblate phase in an oscillation period. This latter aspect was confirmed in the calculations of Tsamopoulos & Brown (1983).

Becker, Hiller & Kowalewski (1991) studied the nonlinear oscillations of freely falling liquid drops, after pinching off from a nozzle in air, both experimentally and theoretically. Their numerical prediction of the inviscid frequency shift, due to nonlinearity, is close to the analytical result (1.5) of Tsamopoulos & Brown, and the numerical result of Dürr & Siekmann (1987), for ϵ up to 0.3. They found experimentally that the frequency shift decreases quadratically with the oscillation amplitude, as predicted theoretically, but at a rate somewhat larger than the theoretical value, and also with a variation for different drops. Because of the generation process of the drops, the initial drop shapes are necessarily complex, requiring Fourier analysis. The complications arising from having to handle many modes on the experimental side may have contributed to the difference between theory and experiment. In our experiments, the situation is simpler because a drop could oscillate in a single mode. More recently, Becker, Hiller & Kowalewski (1994) have included the effect of viscosity in their theory, and have found that viscosity has a large effect on mode coupling, which is the nonlinear interaction between two modes.

Lundgren & Mansour (1988) included small viscosity in their calculation on drop oscillations, using the boundary-integral method, considering mostly higher modes and their interactions, which are beyond the scope of our work.

Patzek *et al.* (1991) theoretically studied the nonlinear oscillations of inviscid drops for ϵ up to about 0.8. The frequency shift shows reasonable agreement with that of Tsamopoulos & Brown (1983) for ϵ up to about 0.3, but they found significant deviation from the parabolic dependence on ϵ for larger ϵ.

Basaran (1992) also calculated drop oscillations including viscosity as an initial-value problem. A Reynolds number $Re = (1/\nu)(\sigma R/\rho)^{1/2}$ is defined, which is proportional to Chandrasekhar's $\omega R^2/\nu$. The results show that for $Re \gg 1$, the drop undergoes most rapid changes during the first cycle, signifying the initial adjustment of the drop, in agreement with our earlier comments on Prosperetti's work. After the first cycle, change continues but much more slowly, as the drop relaxes further into equilibrium. Hence the first cycle is not a typical cycle, but is transient, sensitive to the initial conditions, and more dissipative than other cycles. However, in Basaran's work, emphasis has been placed on the first cycle. For example, the important comparison with the result (1.5) of Tsamopoulos & Brown, has been made using the first cycle.

In our experiments, we have also studied the oscillations of a rotating drop. In the linear inviscid limit, for an axisymmetric oscillation about the rotation axis, the frequency shift from Rayleigh frequency (1.2) due to a rotation rate Ω is given by (Busse 1984)

$$\frac{\delta f_r}{f} = B_r \left[\frac{n^4 + 4n^3 + n^2 + 26n + 48}{64n(n+2)(2n-1)(2n+3)} \right], \tag{1.6}$$

where δf_r is the frequency shift due to rotation, and $B_r = \rho \Omega^2 D^3/\sigma$ is the rotational Bond number in which $D = 2R$ is the spherical diameter of the drop. The full theory

of Busse considers the inviscid oscillation of a liquid drop in an immiscible fluid, with the whole system rotating at a constant rate. It has been verified experimentally using a drop in an immiscible liquid (Annamalai, Trinh & Wang 1985). More recently, Patzek *et al.* (1995) calculated the nonlinear oscillations of two-dimensional rotating drops, finding some interesting patterns of non-axisymmetric running capillary waves.

The damping coefficient of the axisymmetric oscillation of a rotating drop is given by (Lee, Lyell & Wang 1985)

$$\lambda_r = \lambda\left[1 + \frac{\beta^2 n(4n^5 + 16n^4 + 15n^3 - 106n^2 - 211n - 78)}{48(2n+1)(2n-1)(2n+3)}\right], \qquad (1.7)$$

where $\beta = 2\Omega/\omega$; ω and λ are given by (1.2) and (1.3), respectively. The damping coefficient decreases with rotation rate for $n = 2$, but increases with rotation rate for higher n. Rotation deflects motion through the Coriolis force, thereby producing vorticity which enhances viscous dissipation. But near the drop surface, as the normal oscillatory motion is deflected by the Coriolis force, the fluid changes direction and moves along the surface. Since the surface is free from tangential stress for a liquid drop in air, the deflection directs the liquid into a relatively stress-free environment, and hence helps to reduce viscous dissipation. What rotation does to dissipation is the sum of these two opposite effects. Equation (1.7) suggests that the second effect is important only for $n = 2$, but is overpowered by the first effect for higher n.

Since our experiments involved acoustic levitation with imposed flattening, it is also necessary to know the frequency shift due to acoustic flattening (Marston 1980). In the linear inviscid limit, for an axisymmetric $n = 2$ oscillation, the frequency shift from Rayleigh's value (1.2) is (Suryanarayana & Bayazitoglu 1991)

$$\delta f_a/f = \tfrac{1}{2}c_0 B_a, \qquad (1.8)$$

where δf_a is the frequency shift due to acoustic flattening, $c_0 = 0.08036$, and B_a is the acoustic Bond number given by $B_a = (DA^2/\sigma\rho_0 c_0^2)[1 + 7(kD)^2/20]$, in which ρ_0 and c_0 are respectively the density of and sound speed in air, A is the acoustic pressure amplitude, and k is the wavenumber of the sound wave. There has been no experimental verification of this theory. However, the validity of the model is suspect, because in the formulation the deformation of the drop is treated as a consequence of the balance between surface tension and acoustic flattening, whereas in the calculation of drop oscillations the restoring force comes only from the perturbation of the deformed surface, while the part due to the perturbation in the radiation pressure is missing.

In the experiments to be described below, we are interested in observing nonlinearity in oscillations, therefore we want to work with low-viscosity drops. 'Low viscosity' means that the drop oscillation suffers from small decay in one period, i.e. $(\omega/2\pi)(1/\lambda) \gg 1$. This is equivalent to $Re \gg 1$, using Basaran's (1992) definition.

2. Experimental procedure

The experiments were performed in the Drop Physics Module (DPM) of the United States Microgravity Laboratory-1 (USML-1) onboard Space shuttle Columbia (STS-50) during its flight between June 25 and July 9, 1992, by Astronauts Eugene Trinh and Bonnie Dunbar. Briefly, the DPM is a triaxial acoustic chamber in which standing sound fields are established in the orthogonal directions (x, y, z fields) for positioning and manipulating the liquid drops. The x- and y-fields are operated at the same

frequency. For a complete description of the experimental apparatus, the readers should refer to our earlier two papers (Wang *et al.* 1994 *a, b*).

To achieve high temporal resolution, the drop oscillations are recorded with high-speed film running at 200 or 400 frames per second. We have chosen for presentation those drops which satisfy the following criteria: (i) clean deployments, with no evidence of contamination, (ii) accurate estimation of drop volume, (iii) axisymmetric drop shape, both during levitation and during oscillations, through balanced x and y sound fields, (iv) sufficiently large oscillation amplitude ($\epsilon \gtrsim 0.3$), and (v) stable levitation prior to and during oscillations. 'Stable levitation' here means that the drop is positioned at the centre of the chamber with minimal translational oscillations in the acoustic potential well, and more importantly, with no uncontrolled rotation, which happened to be predominantly along the y-axis.

We are interested in the $n = 2$ free-decay oscillation of a drop after its stable levitation. This was achieved using two different approaches.

In the first approach, the drop was flattened along the z-axis by raising the z-field relative to the other two fields (Marston 1980). Stable levitation was attained when the aspect ratio, i.e. the ratio of the equatorial diameter to polar diameter, was greater than about 1.5. On the other hand, given the drop size relative to the wavelength of the z-wave, the aspect ratio should not be larger than about 3, or the drop will disintegrate (Anilkumar, Lee & Wang 1993; Lee, Anilkumar & Wang 1994). The drop was then set into a free oscillation by suddenly turning off all acoustic fields, thereby freeing the drop of all forces and allowing it to regain its spherical shape. It is important that the recovery through viscous relaxation could occur on a time scale shorter than that for the drop to drift out of the window view, as levitation was no longer in effect. This required that the viscosity be relatively high (~ 10 cS).

In the second approach, the drop was gyrostabilized through an imposed rotation along the z-axis. It is noted that, in practice, achieving this rotation always requires a finite flattening of the drop along the z-axis. The rotation rate Ω scaled by the $n = 2$ Rayleigh frequency ω in (1.2), denoted by Ω^*, turned out to have to be greater than 0.3 for this purpose. But Ω^* cannot be as large 0.56 for that would lead to the $n = 2$ rotational bifurcation of the drop (Brown & Scriven 1980). The working value of Ω^* was about 0.4. Oscillations of the drop were started by using an imposed z-field, which was the third harmonic of the original z-wave, modulated in amplitude at the $n = 2$ oscillation frequency of the drop (Marston 1980). A free-decay oscillation of a rotating drop ensured after the modulated field was turned off, while the original levitation fields were still on, keeping the drop in position. Unlike the first approach, this experiment is not constrained by viscosity, and can be used for low-viscosity drops (1–2 cS). The maximum oscillation amplitude was dictated by the limits of the modulated z-drive.

Using the selection criteria listed at the beginning of this Section, we ended up with two drops, one from each approach, oscillating in the free-decay modes. The one from the first approach was truly free, but that from the second approach was a rotating drop, and was subjected to a little additional acoustic flattening at the poles. For the second drop, there was also the small acoustic levitation force, but such a force plays a negligible role in affecting the drop shape or oscillations. A free-decay oscillation lasted for only a short time, such that any temperature variation occurring in the chamber during the process was negligible.

3. Data analysis

The data analysis was conducted with a high-resolution video converted from high-speed film.

In observing a drop shape oscillation, it is noted that the drop does not stay still but gently translates in the acoustic potential well, or in the case in which all acoustics is turned off, slowly drifts toward the walls. Hence instead of measuring the absolute dimensions of the drop, it is more convenient to characterize the shape of a drop at any time by the ratio of the height to the width or vice versa. Thus the oscillation process is described by a dimensionless ratio versus time.

For the initially flattened drop 1, the ratio chosen is width to height W/L, where W is the equatorial diameter and L is the polar diameter, because it reflects the drop initial conditions better. The oblate extrema, especially at higher amplitudes, are much sharper than the prolate ones, thereby providing better frequency resolution. Since the drop is not concave at the poles to begin with, it could never be concave in an oscillation. The wave amplitude ϵ has been evaluated from the W/L data not by (1.1), but by the more accurate drop shape with a nonlinear correction of $O(\epsilon^2)$ from Tsamopoulos & Brown (1983).

For the rotating drop 2 the ratio is chosen to be height to width (L/W), because the prolate extremum shape is well defined, but the oblate extremum shape can be actually concave at the poles. Approximating the drop as spheroidal and basing on (1.1), the wave amplitude is modelled as $\epsilon = 2\{[L/W-1)/(L/W+2)]_{max}-[L/W-1)/(L/W+2)]_r\}$, where the subscript '$max$' means evaluated when the drop is in its extremum prolate shape, and 'r' means evaluated when the drop is in static rotation.

While the oscillation frequency depends on the wave amplitude due to nonlinearity, the wave amplitude decays in time. If the decay of the amplitude is noticeable within one cycle, then the value of the amplitude to be used for evaluating the frequency needs to be clarified. In this context, before proceeding with the frequency shift analysis, one needs to explain how to define the frequency. We have used the following approach.

First, the curve (W/L or L/W versus t) is converted into an array of ϵ versus t, with ϵ defined at the peaks of the original curve in terms of the ratio. Consider any three consecutive peaks A, B, and C of the curve. At the central peak B in particular, using the defined ϵ, we can apply (1.5) to evaluate the theoretical nonlinear frequency shift. The actual frequency can be defined in a central scheme as $2/T_{AC}$, where T_{AC} is the time interval between peak A and peak C. The frequency shift from the data is this frequency minus f, which is also experimentally determined as the asymptotic limit of this frequency when amplitude becomes very small, and the peaks are uniformly spaced. The shift, scaled with f, is compared with the prediction of (1.5).

Alternatively, the frequency at peak B can be defined 'forwardly' as $1/T_{BC}$, where T_{BC} is the time interval between peak B and peak C, or 'backwardly' as $1/T_{AB}$, where T_{AB} is the time interval between peak A and peak B. But these inevitably will be biased by viscosity, because the amplitude decays in the forward direction and grows in the backward direction. However, within one cycle, at large Re, the change of amplitude with time is approximately linear and is inversely proportional to Re. Therefore the central averaging we have described above cancels the time, and thus viscosity, dependence. In other words, what comes out of our analysis will be an inviscid result, although drop we are working with is in fact a low-viscosity drop.

Basaran (1992) found that the nonlinear frequency shift has a dependence on Re even at relatively high Re. In view of our argument above, and his emphasis on the first cycle, the claim probably arose from his choice of defining the frequency forwardly. We

believe that he could have recovered the inviscid result of Tsamopoulos & Brown if he had used central averaging and avoided the first cycle. The question is: given a damped oscillation curve of experimental or numerical origin what is the right way to extract the frequency? Of course, one can always consider this as a matter of definitions. But at large Re, at least for the purpose of comparing with an inviscid theory, central averaging seems to be more appropriate. However, when Re is low, the frequency shift should exhibit a real dependence on Re no matter how we define it.

For drop 1, we also evaluate the percentage of time that the drop spends in oblate form. The percentage assigned to a peak of the W/L versus time curve is determined as the percentage of time the curve is above 1 between the adjacent minima on both sides of the peak.

For analysing the oscillation of the rotating second drop we have a complication in that the drop was also inevitably flattened by the sound wave. We first examine the data at large time, which means the linear limit. In this limit, the frequency f' does not depend on the wave amplitude. The frequency shift $\delta f'$ due to rotation and flattening is the difference between f' and f (1.2). Then $\delta f'/f$ is compared with the sum of the contributions from the corresponding theories (1.6) and (1.8).

Next we use this as an opportunity for testing the effect of nonlinearity on the frequency shift, regardless of the outcome of the preceding test. To do this, for any three consecutive peaks A, B and C, the actual frequency f_1 at B is 2 divided by the time interval between A and C. The actual nonlinear frequency shift at B is given by $\delta f = f_1 - f'$. Then $\delta f/f$ is compared with the prediction of (1.5). Of course we do not expect an agreement, but we shall see whether we can make some observations about it.

For the second drop which is flattened by rotation and acoustic stress, we define a 'prolate displacement' as that part of the oscillation cycle where L/W is greater than the equilibrium value, and evaluate the percentage of time that the drop spends in prolate displacement versus ϵ. The percentage assigned to a peak of the L/W versus time curve is determined as the percentage of time the curve is above the equilibrium value between the adjacent minima on both sides of the peak.

4. Results and discussion

The oscillations are followed until ϵ decreases to about 0.1, below which the resolutions for ϵ ad δf are poor. Owing to the high temporal and spatial resolution of the drop imaging, the measurement errors are less than 0.5% for frequency and about 0.5% for linear dimensions.

The first drop, which is initially flattened, is a nominal 65/35 glycerin/water drop of volume of 4.3 ± 0.1 cm^3 with a viscosity of about 12.2 cS at 25 °C. The other measured properties of the drop liquid are listed in table 1. The initial flattening aspect ratio (W/L) is about 1.72. The free-decay oscillation takes place at 31.7 °C. The value of Re during free decay is about 80. The free decay of the oscillations was studied with high-speed film recorded at 400 frames per second.

The measured frequency after the amplitude has become small is 3.26 Hz, comparing well with Rayleigh's value from (1.2), evaluated at the free-decay temperature by applying a correction to the surface tension measured at 25 °C at -0.16 dyn cm^{-1} °C^{-1}.

The results for the first drop are presented in figures 1–4 (the error bars, whenever bigger than the plot symbols, have been typically indicated in the figures). In figure 1, the ratio W/L (equatorial to polar diameter) is plotted versus time. In figure 2, the amplitude ϵ evaluated at successive peaks is plotted versus time, showing an exponential decay. The decay time τ (the inverse of the decay rate λ) is about 2.05 s,
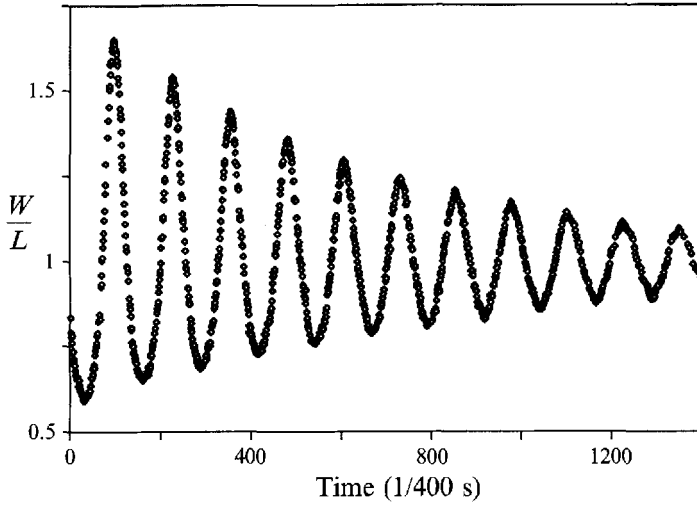
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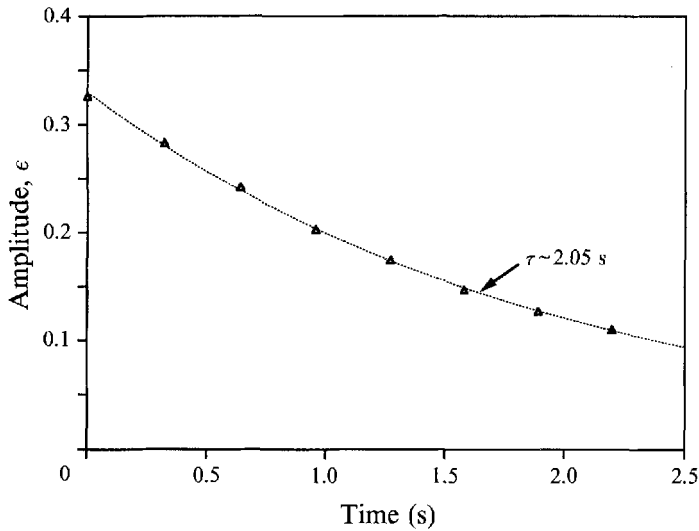FIGURE 1. Free-decay oscillation of the initially flattened drop (drop 1): W/L versus time.

FIGURE 2. Decay of wave amplitude ϵ in time (drop 1). The dotted curve is a fitted exponential curve, with a decay time of 2.05 s. The amplitude ϵ has been evaluated from W/L, using the drop shape with a nonlinear correction of $O(\epsilon^2)$ from Tsamopoulos & Brown (1983). The time origin corresponds to the first oblate extremum following the release of drop flattening.

Drop	ν (cSt)	ρ (gm cm^{-3}) @ 25 °C	σ (dyn cm^{-1}) @ 25 °C
(1) Glycerine/water (65/35)	12.2 ± 0.1 @ 25 °C	1.168	64 ± 1
	9.2 ± 0.1 @ 31.7 °C	—	—
(2) Silicone oil (DC 200 series)	2.10 ± 0.05 @ 25 °C	0.872	18.8 ± 0.2

TABLE 1. Properties of the drop liquids used in the experiments. σ was measured from the leftover flight fluids and ν and ρ from retained samples of the flight fluids.
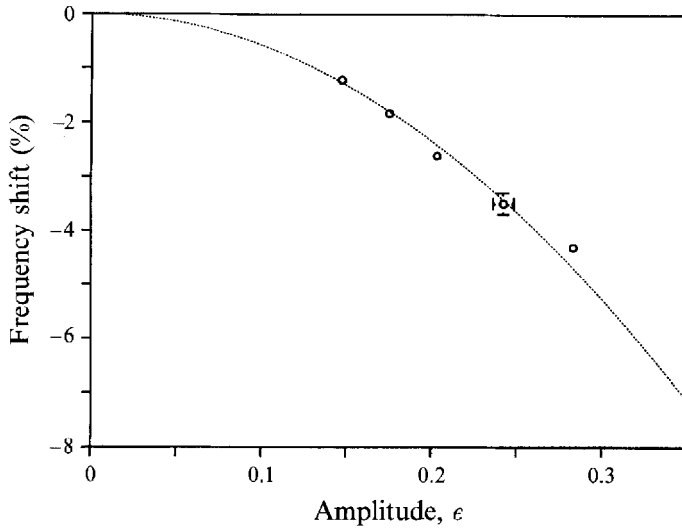
FIGURE 3. Frequency shift versus amplitude ϵ (drop 1). The dotted curve is from (1.5) of Tsamopoulos & Brown (1983).
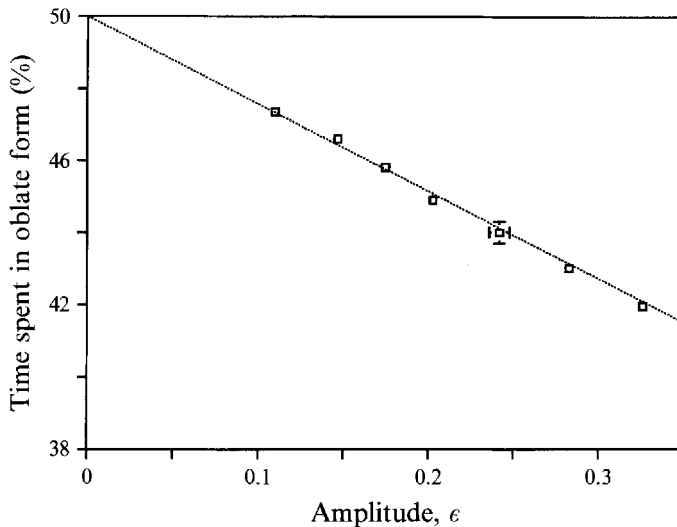

FIGURE 4. The percentage of time spent in oblate form versus amplitude ϵ (drop 1). The dotted line is from the theory of Tsamopoulos & Brown (1983).

agreeing well with Lamb's value from (1.3) of about 2.2 s. In figure 3, the nonlinear frequency shift is plotted versus ϵ, in good comparison with the prediction of Tsamopoulos & Brown (1983). In figure 4, the percentage of time spent in oblate form is plotted versus oscillation amplitude ϵ, also in good agreement with their prediction.

The second drop, which is oscillating while rotating and is a little flattened by the levitation field, is a silicone oil drop (DC 200 series) of volume 3.35 ± 0.06 cm^3 and viscosity of 2.10 ± 0.05 cS at 25 °C. The other measured properties of the drop are listed in table 1. The value of Re is about 210. The equilibrium aspect ratio L/W (polar to equatorial diameter), measured when the oscillations have completely stopped, is 0.77. The drop rotates at 0.91 r.p.s. and this results in an aspect ratio of about 0.85 (Brown & Scriven 1980). The acoustic flattening, which is the difference between the
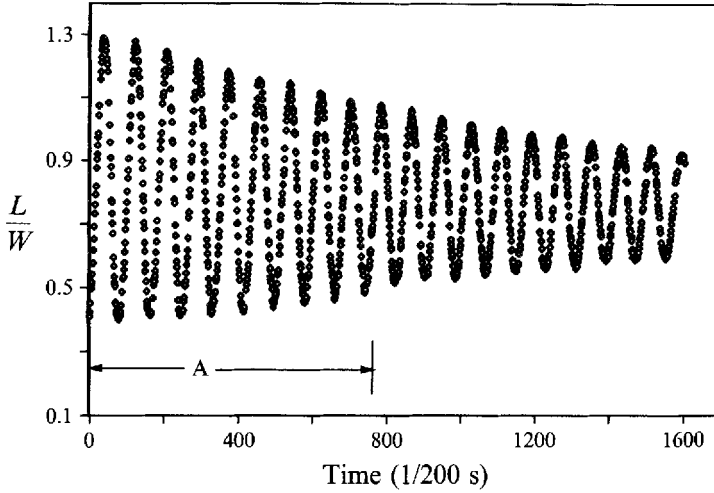
FIGURE 5. Free-decay oscillation of a rotating drop with some acoustic flattening (drop 2): L/W versus time.
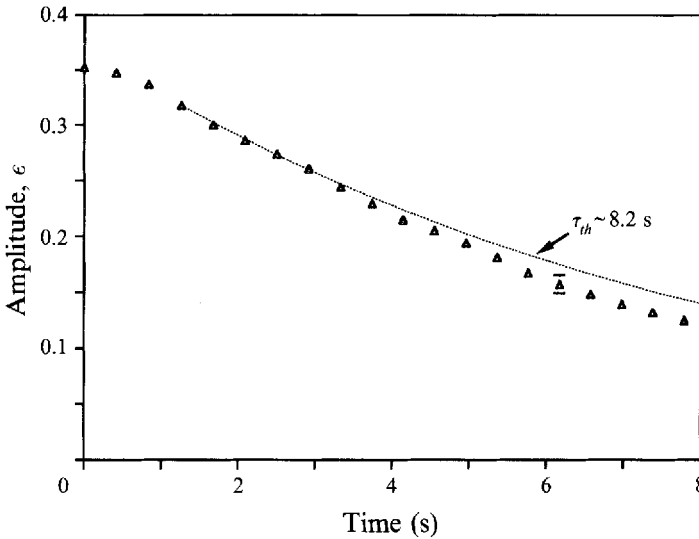

FIGURE 6. Decay of wave amplitude ϵ in time (drop 2). The dotted curve is an exponential curve, with a decay time of 8.2 s, from Lamb (1945). The amplitude ϵ is evaluated from L/W using (1.1), after subtracting the equilibrium shape.

total aspect ratio while the drop is not oscillating and the aspect ratio due to rotation alone, is thereby about 8%. The free-decay oscillation occurred at 25.6 °C and was studied with high-speed film recorded at 200 frames per second.

Figure 5 is a plot of the ratio L/W versus time for the second drop ('A' in the figure denotes the region where the oblate extremum shape is concave at the poles). The measured frequency shift $\delta f'$ from the Rayleigh frequency at large time, when oscillation is linear, is about 8%. The predicted frequency shift due to rotation alone (equation (1.6)) is about 14%. If it is assumed that the frequency shifts due to rotation and acoustic flattening are simply additive, then it appears that the frequency shift due to additional acoustic flattening is negative, contradicting the trend predicted by Suryanarayana & Bayazitoglu (1991). Figure 6 shows the exponential decay of
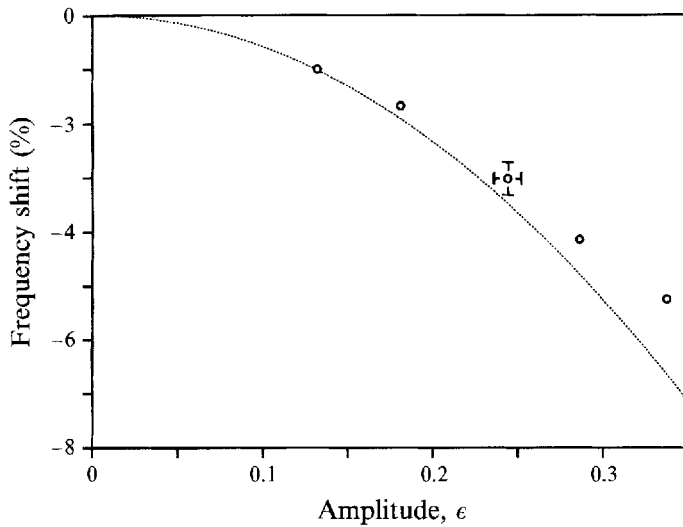
FIGURE 7. Frequency shift versus amplitude ϵ (drop 2). The dotted curve is from (1.5) of Tsamopoulos & Brown (1983) for a spherical drop.

amplitude ϵ in time. The decay time is shorter than the 8.2 s predicted by Lamb from (1.3) for a spherical non-rotating drop. In contrast, according to (1.7), the $n = 2$ decay time for a rotating drop should be longer than that from (1.3). It is not clear what role acoustic flattening may play in changing the dissipation rate.

In figure 7, the frequency shift due to nonlinearity is plotted versus amplitude ϵ for the second drop. It is seen that the dependence is weaker than that predicted by Tsamopoulos & Brown (1983) for spherical drops. As discussed in §1, for a spherical drop, the nonlinear frequency shift is mainly a consequence of the relatively slower motion of the drop, when it is in the prolate phase of an oscillation. For the second drop under consideration, both rotation and acoustic flattening suppress the prolate part of its oscillation, thereby reducing the main cause of the nonlinear frequency shift.

According to figure 5, the equilibrium value for L/W is 0.77. The explanation for the weaker nonlinearity for the second drop is corroborated by figure 8, where the percentage of time spent in prolate displacement is plotted. First it is noted that the equilibrium shape of the drop corresponds to an ϵ of about 0.17. This means that the drop can attain a true prolate shape, during an oscillation only when ϵ is greater than 0.17. Accordingly, the data in figure 8 have been demarcated into two regions: region I corresponding to larger amplitudes ($\epsilon \geqslant 0.17$) where the oscillations are truly oblate–prolate, and region II corresponding to smaller amplitudes ($0 < \epsilon < 0.17$), where the oscillations are oblate–oblate. At the demarcation point $\epsilon = 0.17$ between regions I and II, the drop spends the same length of time in prolate and oblate shapes within a cycle. On comparing the data in region I with theory it is seen that the oscillations of this drop, which is flattened through rotation and acoustic stress, show less bias toward the prolate shape. The data at smaller amplitudes (region II) show that the drop spends less than 50 % of the time in the prolate half of a cycle when ϵ is below 0.17. It is a little surprising that the percentage of time decreases with decreasing ϵ more rapidly than that in region I, because we expect the curve to turn up back to 50 % as ϵ approaches zero. This has also been previously observed by Trinh & Wang (1982), in their experiments on drop oscillations in an immiscible levitator.

For both drops, the oscillation curves look very much like damped sinusoidal
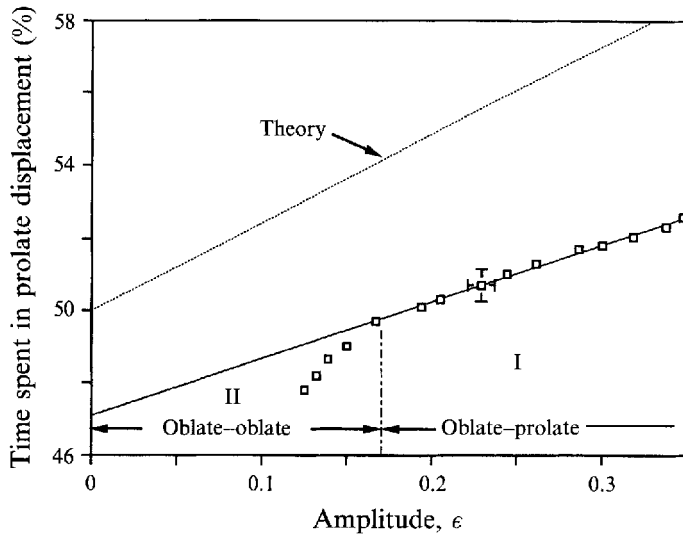
FIGURE 8. The percentage of time spent in prolate displacement versus the amplitude ϵ (drop 2). The dotted line is from Tsamopoulos & Brown (1983) for a spherical drop.

functions. It is unlikely that a detailed fast Fourier transform would reveal any additional information like the presence of non-axisymmetric modes and the occurrence of mode coupling. Since the oscillations of the free drops decayed to low amplitudes quickly, mode coupling, if there was any, did not have a chance to appear.

5. Conclusions

Oscillations of low-viscosity drops were studied in the microgravity environment of a Space shuttle flight. From the damped oscillation data, the inviscid frequency shift, due to nonlinearity, was extracted using a central-averaging scheme. In the context of a free low-viscosity drop, it has been found that the frequency shift agrees well with the predictions of the classical inviscid nonlinear theory of Tsamopoulos & Brown (1983), in the range of experimental amplitudes ($\epsilon < 0.3$). However, in the context of a rotating low-viscosity drop, under acoustic levitation, it has been found that the frequency shift is smaller, and the percentage of time spent in prolate displacement is significantly less than that for the classical case.

The central-averaging scheme employed in our data analysis has effectively eliminated the role of viscosity in nonlinear frequency shift. According to Basaran (1992), viscosity influences the resonance frequency in two opposite ways. It reduces the frequency by slowing down the fluid motion, which is a linear effect. At large Re, from (1.4), the effect is inversely proportional to Re^2 and is negligible. Viscosity also reduces the amplitude and hence increases the frequency by reducing the negative nonlinear shift. But this effect of viscosity is implicit, because the frequency shift is still given by (1.5) in terms of the amplitude. This implicit effect, given by the decay of the amplitude with time, is inversely proportional to Re at large Re, and is eliminated during the evaluation of the frequency shift by central averaging.

The question remains as to what large Re means in the central-averaging scheme. The oscillation amplitude decays by a factor of $\exp(-2\pi\lambda/\omega)$ in one cycle. In order that the frequency shift can be considered as inviscid, the decay has to be approximately

linear within one cycle, such that central averaging is applicable. Therefore we need $2\pi\lambda/\omega \ll 1$. From (1.2) and (1.3), it means

$$Re \gg 2\pi(2n+1)\left(\frac{n-1}{n(n+2)}\right)^{1/2} \approx 10 \quad \text{for} \quad n = 2. \tag{5.1}$$

Re for the two drops we worked with is 80 and 210, respectively.

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