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A note on the damping and oscillations of a fluid drop moving in another fluid

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The oscillations of a drop moving in another fluid medium have been studied at low values of Reynolds number and Weber number by taking into consideration the shape of the drop and the viscosities of the two phases in addition to the interfacial tension. The deformation of the drop modifies the Lamb's expression for frequency by including a correction term while the viscous effects split the frequency into a pair of frequencies—one lower and the other higher than Lamb's. The lower frequency mode has ample experimental support while the higher frequency mode has also been observed. The two modes almost merge with Lamb's frequency for the asymptotic cases of a drop in free space or a bubble in a dense viscous fluid but the splitting becomes large when the two fluids have similar properties. Instead of oscillations, aperiodic damping modes are found to occur in drops with sizes smaller than a critical size ($\sim \hat{\rho}\hat{\nu}^2/T$). With the help of these calculations, many of the available experimental results are analyzed and discussed.

1. Introduction

A study of the oscillations of drops and bubbles moving in background media finds important applications in many physico-chemical and technological problems. The oscillations are affected by the shape of the drop, the inertial effects caused by the drop motion, the interfacial tension and the two phase parameters like the viscosities. The complexity of the interactions among these factors has often restricted the analysis into certain limiting cases like a spherical drop at rest in an inviscid fluid (Lamb 1932). Such calculations are of limited applicability and have failed to explain many of the observational details (Kintner 1963).

In order to attempt a full discussion of the large amount of experimental data, we have calculated the oscillational modes of drops taking into account all the four factors, namely shape of the drop, interfacial tension, terminal velocity and viscosities. The analysis is confined to the familiar Lamb's oscillation modes where the surface distortion is expressed in terms of Legendre polynomials. The damping of the oscillations is also considered. The present analysis brings out a number of new features and gives a much better account of the experimental observations than the existing limiting cases treated by many workers. However, it may be added that the calculations are limited to low values of Reynolds number and Weber number, such that the deformation is small and the oscillations considered here are the normal symmetric modes. We outline here the main features of the calculations, whose full details will be reported in due course (Subramanyam 1968). A recent paper by Miller & Scriven (1968) deals with the oscillations of stationary viscous drops in background fluid media. They include the contributions from interfacial viscosity and elasticity and study both the frequency and damping of the oscillations in various limiting cases. Here we are mainly concerned with the oscillations of drops moving with a uniform velocity in a fluid medium. However, if we consider the limiting case of the terminal velocity approaching a very small value, then the present results agree with those of Miller & Scriven as discussed in detail elsewhere (Subramanyam & Gopal 1969).

2. Formulation of the problem and analysis

The problem is basically one of solving the perturbed equations of motion for the drop and the continuous phase with matched boundary conditions. The equations of motion are written by choosing the origin of the spherical polar co-ordinate system at the centre of the drop and the equations are made dimensionless by taking the radius of the undeformed spherical drop a as a characteristic length, the uniform stream velocity U as a characteristic velocity, a/U as a characteristic time, Ua^2 as a characteristic stream function and $\frac{1}{2}\rho U^2$ as a characteristic pressure. Then on account of the drop surface being perturbed, the stream function changes from a value ψ_0 to ψ where

$$\psi = \psi_0 + \Delta\psi_1 e^{i\sigma' t} + \Delta\psi_2 e^{2i\sigma' t} + \ldots, \tag{1}$$

where the non-dimensional frequency σ' is related to the oscillation frequency σ, as yet unknown, of the drop by the equation $\sigma' = \sigma a/U$; $\Delta\psi$ are the perturbations of the stream functions. A similar expansion can be written for the drop phase also. These expansions of the stream functions can be substituted in the equations of motion

$$\frac{\partial}{\partial t}(D^2\psi) + E(\psi, \psi) = \frac{1}{R} D^4\psi, \tag{2}$$

where

$$\left.\begin{aligned} R = \frac{Ua}{\nu}, \quad \xi = \cos\theta, \quad D^2 \equiv \frac{\partial^2}{\partial r^2} + \frac{1-\xi^2}{r^2}\frac{\partial^2}{\partial \xi^2}, \\ E(\psi, \hat{\psi}) = \frac{1}{r^2}\left[\frac{\partial(\psi, D^2\hat{\psi})}{\partial(r,\xi)} + \left(\frac{2\xi}{1-\xi^2}\frac{\partial\psi}{\partial r} + \frac{2}{r}\frac{\partial\psi}{\partial\xi}\right) D^2\hat{\psi}\right].\end{aligned}\right\} \tag{3}$$

Then the coefficients of $e^{i\sigma' t}$, $e^{2i\sigma' t}$, ... give the following: zeroth, first, ..., ordered perturbation equations

$$E(\psi_0, \psi_0) = \frac{1}{R} D^4\psi_0, \tag{4}$$

$$D^4\Delta\psi_1 - R[E(\psi_0, \Delta\psi_1) + E(\Delta\psi_1, \psi_0)] - i\sigma' R D^2\Delta\psi_1 = 0. \tag{5}$$

Similar equations can be written for the drop phase also by changing ψ to $\hat{\psi}$ and $R\,(= Ua/\nu)$ to $\hat{R}\,(= Ua/\hat{\nu})$. The former equations, that is (4), were studied by Taylor & Acrivos (1964) in connexion with the flow past a spherical drop.

Their calculations revealed that inertial effects of motion past the drop causes the drop shape to become oblate spheroidal

$$r = 1+\epsilon_1 P_2(\xi), \tag{6}$$

where ϵ_1, linear in W ($= \rho a U^2/T$) at low Weber numbers, determines the deviation of the drop from the spherical shape. The full value of ϵ_1 has been given by Taylor & Acrivos (1964). The above expression for the shape of the drop is found to agree with experimental observations for low Reynolds and Weber numbers (Wellek, Agrawal & Skelland 1966; Hayashi & Matunobu 1967).

The normal symmetric modes of oscillation of such a deformed drop due to external perturbations can be represented by the equation

$$r = 1+\epsilon_1 P_2(\xi)+\epsilon_0 e^{i\sigma' t} P_l(\xi). \tag{7}$$

ϵ_0 can finally be expressed in terms of other parameters (Subramanyam 1968) but our interest here is confined to a discussion of σ'. The equations (5) can be discussed in the following three limiting cases.

Case (i): $\sigma' = \sigma a/U = (a^2\sigma/\nu)(1/R) \gg 1$

The highest derivative term (i.e. $D^4\Delta\psi_1$) and the term with σ' are to be retained and the inertial terms can be neglected. Then the non-singular solutions of the resulting equations can be written in a suitable form as

$$\left.\begin{aligned} \Delta\psi_1 &= [Br^{-l}+Dr^{\frac{1}{2}}J_{-(l+\frac{1}{2})}(hr)]F_l(\xi),\\ \Delta\hat{\psi}_1 &= [Ar^{l+1}+Cr^{\frac{1}{2}}J_{l+\frac{1}{2}}(\hat{h}r)]\,F_l(\xi), \end{aligned}\right\} \tag{8}$$

where $$F_l(\xi) = \int_{-1}^{\xi} P_l(\xi)\,d\xi, \quad h^2 = -i\sigma' R = -i\sigma a^2/\nu,$$
$$\hat{h}^2 = -i\sigma'\hat{R} = -i\sigma a^2/\hat{\nu}.$$

Actually, in the expression for $\Delta\psi_1$, the Hankel function of first kind $H'_{+\frac{1}{2}}(hr)$ will have to be used instead of $J_{-(l+\frac{1}{2})}(hr)$. But the latter has been chosen for purposes of numerical computations as tables of $J_{\pm(l+\frac{1}{2})}(x)$ are more readily available. As a consequence, the present results deviate from those using Hankel functions by a magnitude $O(1/h^2)$, which turns out to be less than 1% in practice. Otherwise the characteristic equation (11), for example, agrees in the limiting case of a stationary drop with that obtained by Miller & Scriven (1968). This is being discussed elsewhere (Subramanyam & Gopal 1969) and will not be analyzed any further here. The four constants will have to be eliminated by the use of the boundary conditions

$$u_r = \hat{u}_r, \quad u_\theta = \hat{u}_\theta, \tag{9}$$

$$T_{r\theta} = \hat{T}_{r\theta}, \quad T_{rr} = \hat{T}_{rr} - \frac{2}{W}\left(\frac{1}{x_1}+\frac{1}{x_2}\right), \tag{10}$$

at $r = 1+\epsilon_1 P_2$, x_1 and x_2 being the principal radii of curvature and $W = \rho a U^2/T$ is the Weber number of flow (Scriven 1960). A calculation of the velocity components from (8) and substitution in the condition (10) give four homogeneous equations in A, B, C and D. In order that they be satisfied simultaneously, the characteristic determinant should vanish.

For the sake of simplicity, let us first consider the oscillations of a spherical drop ($\epsilon_1 = 0$), because many of the special features are brought out in a convenient form. For a spherical drop, $\epsilon_1 = 0$, and the characteristic equation is simplified to

$$\begin{vmatrix} -\hat{h}Q_{l+\frac{1}{2}}(\hat{h}) & 2l+1 & hQ_{-(l+\frac{1}{2})}(h) \\ k\left[-\frac{\hat{h}^2}{2}+\hat{h}Q_{l+\frac{1}{2}}(\hat{h})\right] & k(l^2-1)-l(l+2) & \frac{h^2}{2}-hQ_{-(l+\frac{1}{2})}(h) \\ -\frac{1}{l}-\frac{2\hat{h}}{i\hat{R}}Q_{l+\frac{1}{2}}(\hat{h}) & \begin{array}{l}\left[\frac{1}{l}+\frac{1}{\gamma(l+1)}+\frac{2(l-1)}{i\hat{R}}\right. \\ \left.+\frac{2(l+2)}{ik\hat{R}}-\frac{(l-1)(l+2)}{\hat{W}}\right]\end{array} & \frac{2hQ_{-(l+\frac{1}{2})}}{ik\hat{R}}-\frac{1}{\gamma(l+1)} \end{vmatrix} = 0, \quad (11)$$

where $\gamma = \hat{\rho}/\rho, \quad k = \hat{\mu}/\mu = \gamma\hat{\nu}/\nu, \quad Q_{l+\frac{1}{2}} = \frac{J_{l+\frac{3}{2}}}{J_{l+\frac{1}{2}}}, \quad Q_{-(l+\frac{1}{2})} = \frac{J_{-(l-\frac{1}{2})}}{J_{-(l+\frac{1}{2})}}.$

With the help of this equation, the frequencies of oscillation can be computed for any fluid–fluid system provided the dimensionless parameter

$$\sigma' = \left(\frac{\sigma a^2}{\nu}\right)\left(\frac{1}{R}\right) \gg 1.$$

This holds good for drops in very slow motion. Postponing the discussion of a typical system like an *o*-nitrotoluene drop oscillating in water, we shall instead simplify the equation in several limiting cases like (*a*) a drop in free space, (*b*) a bubble in a dense viscous liquid, and (*c*) a drop in a similar fluid.

(*a*) For a spherical drop in free space ($\gamma \to \infty$, $k \to \infty$), the above equation reduces to the form

$$\sigma_{01}^2 = \frac{l(l-1)(l+2)}{\hat{W}\sigma'^2} = 1+\frac{2l(l-1)}{i\hat{R}}-\frac{2(l^2-1)}{\hat{h}^2-2\hat{h}Q_{l+\frac{1}{2}}}-\frac{4l(l^2-1)Q_{l+\frac{1}{2}}}{i\hat{R}(\hat{h}-2Q_{l+\frac{1}{2}})}. \quad (12)$$

If we want to study the oscillations of a drop which is relatively at rest in the background fluid medium, we will have to make an important change. In this discussion, we have chosen a/U as a characteristic time to non-dimensionalize the basic equations. For the study of a stationary drop, this unit should be replaced by $1/\sigma_L$ where σ_L is Lamb's frequency $(\rho a^3/T)^{\frac{1}{2}}$, so that in the equations we have to replace a/U by $1/\sigma_L$. The resulting equation agrees with the results of Reid & Chandrasekhar (Chandrasekhar 1961) and Miller & Scriven (1968).

(*b*) Similarly, if we consider the oscillations of a bubble in a dense viscous fluid ($\gamma \to 0$, $k \to 0$),

$$\sigma_{02}^2 = \frac{(l-1)(l+1)(l+2)}{W\sigma'^2} = 1+\frac{2(l+1)(l+2)}{iR} -\frac{2l(l+2)}{h^2-2hQ_{-(l+\frac{1}{2})}}-\frac{4l(l+1)(l+2)hQ_{-(l+\frac{1}{2})}}{iR[h^2-2hQ_{-(l+\frac{1}{2})}]}. \quad (13)$$

(*c*) For the oscillations of drop in a similar fluid, ($\gamma = 1$, $k = 1$).

$$\sigma_{03}^2 = \frac{l(l-1)(l+1)(l+2)}{(2l+1)W\sigma'^2} = 1-\frac{2l+1}{h[Q_{l+\frac{1}{2}}-Q_{-(l+\frac{1}{2})}]}. \quad (14)$$

From equations (12)–(14) it is evident that the frequencies are altered by the viscous terms in the right-hand side of the equations. The deviation from Lamb's result due to viscous effects is clearly seen here. The left-hand side is merely the square of the ratio of Lamb's to the modified frequency. The presence of the terms containing h on the right-hand side of (12)–(14) makes σ an implicit parameter in these equations as $h^2 = -i\sigma' R$. Hence a direct evaluation of σ is not possible. The relative order of magnitudes of various terms may be seen for example in (12), right-hand side, as $O(1)$, $O(1/R)$, $O(1/h^2)$ and $O(1/h^2R)$ (since $Q_{l+\frac{1}{2}}(h) \rightarrow O(1/h)$ as $h \rightarrow \infty$). So the second and third terms need be retained only as correction terms for $R \ll 1$. Since we are considering the shape of the drop to be either spherical or slightly deformed, it requires $W \ll 1$ (Taylor & Acrivos 1964). Hence the results of case (i) are valid as long as $W \ll 1$, $R \ll 1$.

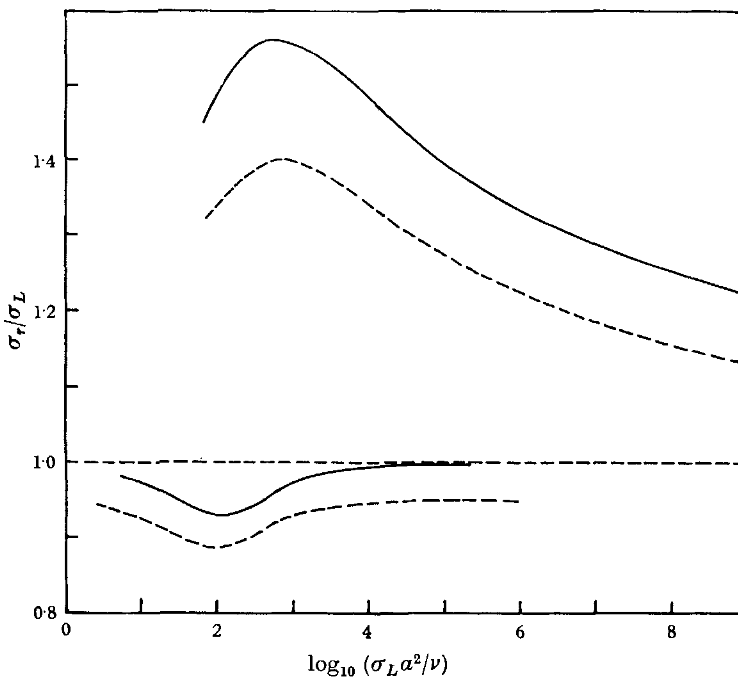

FIGURE 1. Oscillation modes of a drop in a similar fluid ($\gamma = 1$, $k = 1$). (Full curve corresponds to a spherical drop, dashed curve refers to a deformed drop, $\epsilon_1 = 0{\cdot}2$.)

In the first two limiting cases of a drop in free space or a bubble in a dense viscous liquid, the deviation from Lamb's result is very small. But deviation of the order of 10–15 % from Lamb's is found to occur in the practically important case of a drop oscillating in a similar fluid.

Since the characteristic equation (11) is an implicit equation in σ, σ can be written as

$$\sigma = \sigma_r + i\sigma_i \quad \text{and} \quad \hat{h}^2 = -i\sigma a^2/\hat{\nu} = \alpha^2(\beta - i),$$

where $\alpha^2 = \sigma_r a^2/\hat{\nu}$, $\beta = \sigma_i/\sigma_r$, α determines purely the oscillatory part, whereas β gives the damping of the oscillations. In order to evaluate σ, we first separate the real and imaginary parts of (11). For a given α, the value of β is determined by solving the imaginary part and using these α, β in the real part, the value of σ

is computed. In the absence of high-speed computational facilities, the calculations have proved to be very difficult. As illustrated in the table, the imaginary part of σ_L^2/σ_r^2 is still about 10 % of the real part and has not yet vanished. We have estimated approximately the error caused in α, β, σ by this deficiency to be about 2–5 %. The alternate signs of $Q_{l+\frac{1}{2}}$ generate a pair of frequencies, one higher and the other lower than Lamb's for each drop size. For the case $\gamma = 1$, $k = 1$, these values are plotted in figure 1, against the drop size. The table gives the calculated values of the frequencies of oscillations as a function of drop size for an *o*-nitrotoluene drop in water using the full equation (11).

α	β	σ_L^2/σ_r^2	σ_r/σ_L	a (cm)
15	0·0070	0·7508 + 0·0641i	1·154	0·1464
25	0·0068	0·8054 + 0·0774i	1·114	1·461
50	0·0024	0·9035 + 0·076i	1·052	21·74
100	0·0006	0·9546 + 0·0414i	1·024	367·4
α	β'	$\sigma_L^2/\sigma_r'^2$	σ_r'/σ_L	a (cm)
15	0·0062	1·2576 + 0·109i	0·8917	0·2452
25	0·0084	1·1790 − 0·0869i	0·9206	1·797
50	0·0035	1·0952 − 0·064i	0·9554	26·3
100	0·0010	1·0482 − 0·0394i	0·9768	403·5

TABLE 1. Damped oscillations of *o*-nitrotoluene drop in water for $R \ll \sigma a^2/\nu$ ($\gamma = 1{\cdot}160$, $k = 2{\cdot}7526$, $l = 2$, $T = 26{\cdot}6$ dyne cm^{-1})

Reid & Chandrasekhar (Chandrasekhar 1961) first pointed out that the perturbations on the drop surface can be damped aperiodically. For this to occur σ will have to be purely imaginary. For a typical case of a drop in free space, these aperiodic damping modes are calculated and plotted in figure 2. It shows that there are two possible damping modes for a given drop size (the lower one being favoured because of energy considerations), and that above a certain critical drop size, defined by the quantity $\sim \sigma_L a^2/\hat{\nu}$ or $\hat{\rho}\hat{\nu}^2/T$, aperiodic damping cannot occur. This means that for oscillations to start, the drop size must be larger than this critical size (a_c). Though this critical size is very small ($\sim 10^{-6}$ cm) for a water drop in air, it assumes values as large as a few mm for a drop or a bubble in a dense viscous liquid. Such a critical size for oscillations to start has not yet been explicitly observed to our knowledge.

We can now discuss the oscillations of a deformed drop (for the case (i), $\sigma' \gg 1$). The general equation is quite complicated (Subramanyam 1968). As a special case, we can study how deformation modifies the Lamb's expression for frequency of oscillation. As in Lamb's treatment, if the viscous terms are neglected from the expressions (8), then a very simple expression such as

$$\sigma^2 = \sigma_L^2(1 - \tfrac{1}{4}\epsilon_1), \tag{15}$$

correct to the first order in ϵ_1 is obtained. This can profitably be used in the analysis of experimental results for highly deformed drops. Another simple case is $\gamma = 1$, $k = 1$, $l = 2$, the prolate–oblate oscillations of a drop in a similar fluid. The contribution from the ϵ_1 term turns out to be

$$\sigma_{03}^2 = 1 - \frac{5}{h(Q_{\frac{5}{2}} - Q_{-\frac{5}{2}})} + \frac{\epsilon_1}{4} + O\left(\frac{1}{h^2}\right), \tag{14a}$$

which corrects (14) for the deformation effects. The contributions from the viscous and deformation factors are coupled in the general case but in the limiting cases they turn out to be additive correction factors.

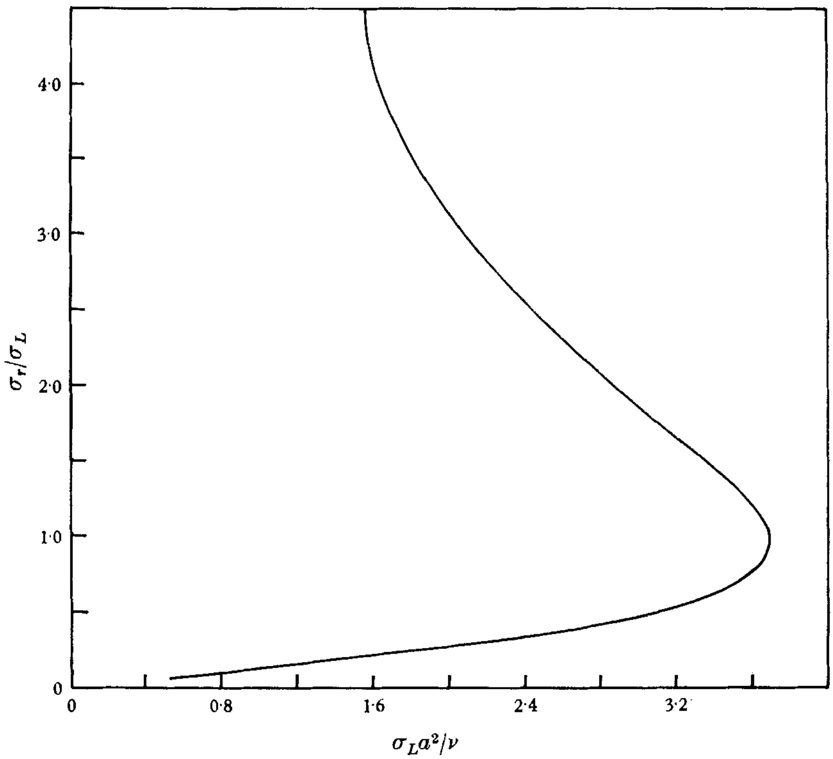

FIGURE 2. Aperiodic damping modes of a drop in free space ($\gamma \to \infty$, $k \to \infty$).

Case (ii): $\sigma' \ll 1$

Equation (5) becomes

$$D^4\Delta\psi_1 - R[E(\psi_0, \Delta\psi_1) + E(\Delta\psi_1, \psi_0)] = 0.$$

A similar equation for the drop phase can also be written. In this case, the time-dependent terms are relatively small and the resulting equations form the basis of the study of deformation, rather than oscillations, of the drop (Taylor & Acrivos 1964).

Case (iii): $\sigma' \sim 1$ or $\sigma a^2/\nu \sim R$

In this case the calculations can be carried out by taking R as an expansion parameter.

$$\left.\begin{aligned} \Delta\psi_1 &= \Delta\psi_{10} + R\Delta\psi_{11} + \ldots, \\ \Delta\hat{\psi}_1 &= \Delta\hat{\psi}_{10} + R\Delta\hat{\psi}_{11} + \ldots. \end{aligned}\right\} \quad (16)$$

Zeroth ordered equations give

$$D^4\Delta\psi_{10} = 0, \quad D^4\Delta\hat{\psi}_{10} = 0, \quad (17)$$

with non-singular solutions

$$\left.\begin{aligned}\Delta\psi_{10} &= (Br^{-l}+Dr^{-l+2})\,F_l(\xi),\\ \Delta\hat{\psi}_{10} &= (Ar^{l+1}+Cr^{l+3})\,F_l(\xi).\end{aligned}\right\} \tag{18}$$

The boundary conditions (10) can be used at $r = 1+\epsilon_1 P_2$, the surface of the deformed drop, to eliminate the constants A, B, C and D. The final characteristic equation, for the $l = 2$ mode, turns out to be, after some lengthy simplifications (Subramanyam 1968).

$$\frac{-40(k+1)}{\sigma'^2\hat{W}}+\frac{38k^2+89k+48}{i\sigma' k\hat{R}}\left(1-\frac{\epsilon_1}{4}\right)+\left(1+\frac{\epsilon_1}{4}\right)\left[\frac{26k+29}{4}+\frac{16k+14}{\gamma}\right] = 0. \tag{19}$$

This characteristic equation can be used to discuss the damping and oscillations of a drop moving in a background fluid medium. The relative order of magnitudes of the various terms is $O(1/\sigma'^2\hat{W})$, $O(1/\sigma'\hat{R})$ and $O(1)$. Now $\sigma' \sim 1$ and $\hat{R} \ll 1$, so the second term is large and hence the first term must also be large to balance it, i.e. $\sigma' = O(\hat{R}/\hat{W})$ and $\hat{W} = O(\hat{R}) \ll 1$. This is consistent with the assumption $\epsilon_1 \ll 1$. Hence in this case also, the results are valid for $\hat{R} \ll 1$ and $\hat{W} \ll 1$.

Equation (19) exhibits all the qualitative features described earlier in connexion with (11). There is a splitting of the frequencies which is pronounced when the two liquids are similar. This equation again reveals the presence of a critical size of the order of $(\hat{\rho}\hat{\nu}^2/T)(1-3\epsilon_1/4)$, above which oscillations can occur. Perturbations on drop surfaces with sizes smaller than this critical size are damped aperiodically. This critical size is of the same order as that given by Reid & Chandrasekhar, and that obtained in the previous case. Damped oscillations for drops with larger size are well explained by equation (19).

In addition, this equation is useful for purposes of calculation and comparison with experimental results. It is applicable to the flows with a definite Reynolds number. With the help of this equation, the oscillation frequencies are computed for the cases of a water drop in air, *m*-cresol drop in water, *o*-nitrotoluene drop in water and the results are in qualitative agreement with the available experimental results.

3. Brief discussion of experimental results

The general features of the calculated results can be discussed as follows. The various factors like the deformation of the drop, the viscosities of the two phases, the inertial effects caused by the drop motion give nearly comparable effects to the oscillations and it is not justified to ignore any one of them.

There is experimental evidence that, for instance, the deformation of the drop affects the oscillation frequency even in the stationary case. Schoessaw & Boumeister (1966) studied the oscillation frequencies of a stationary drop for the modes $l = 2$ to $l = 8$. The experiment was conducted for drops supported by their own superheated vapour over a hot plate. The presence of large temperature gradients can cause oscillations, but it is supposed that the frequency of oscillation is independent of temperature. Observed results were lower than Lamb's by about 10–15 % and this can easily be explained with the help of equation (15). If the drop shape in their experiment is assumed to be oblate spheroidal of the form $r = 1+\epsilon_1 P_2$, then $\epsilon_1 \sim 0{\cdot}8$ to $1{\cdot}3$. Then $\sigma/\sigma_L \sim 0{\cdot}84$ to $0{\cdot}9$ explaining the observational results, although strictly one cannot apply the present calculations for such large ϵ_1.

There are a number of experimental observations on moving drops. However the lack of information on the several associated physical parameters makes any attempt at a detailed comparison futile. The viscosities of the two phases tend to damp the perturbation on the surface of the drop in two different ways depending on the size of the drop. If the drop size is smaller than a critical value, approximately determined by the factor $\sigma_L a^2/\hat{\nu}$ or $\hat{\rho}\hat{\nu}^2/T$, then aperiodic damping occurs. Though this critical size is as small as 10^{-6} cm for a system such as water drops in air, it assumes values as large as a few mm for a drop in a dense viscous liquid. This type of aperiodic damping mode has not been clearly observed though Winnikow & Chao (1966) have mentioned the absence of oscillations in such systems.

If the drop size is larger than this critical size, then the viscous effects split the frequency mode into a pair of permissible frequencies, one lower and the other higher than Lamb's. The deviation of these from the Lamb's result is small for the asymptotic cases of a drop oscillating in free space or a bubble in dense viscous fluid. But it acquires practically important magnitudes for systems of similar fluids.

The lower mode (lower than Lamb's by about 10–15 %) has ample experimental support. If the Reynolds number of the flow is very small, the results of case (i) are to be applied, but if $R \sim \sigma a^2/\nu$, results of (iii) are applicable. The former case gives almost Lamb's values for a system such as water drops in air and slightly reduced values for a liquid–liquid system, while the latter case gives very much lower values. Experiments on water drops in air (Lane 1957), *o*-nitrotoluene drop in water, *m*-cresol drop in water (Kaparthi & Licht 1962) and several other systems (Constan & Calvert 1963) all give frequencies lower than Lamb's by about 10–20 % in general agreement with our present calculations. Schroeder & Kintner (1965) also find the frequencies to be about 16 % lower than Lamb's from a study of nineteen liquid–liquid systems. Another observation of Constan & Calvert on the oscillations of propylene glycol and ethylene glycol drops in gaseous media is that to a first approximation the frequencies are independent of R, again in qualitative agreement with our calculations. All these experimental results are for drops in terminal motion in the background media. In most of the cases, the drop shape will deviate from spherical to an oblate spheroidal form. In the experimental observations on systems the shape of the drop has not been clearly defined. In addition to this the experimental results are widely scattered because of the varying amplitudes of oscillation of different drops, wall effects and, above all, impurities. It is for this reason that a detailed comparison cannot be made between these observed values and calculated results. However the general trend of the experiments seems to be in better agreement with our calculations than with Lamb's.

The higher frequency mode (larger than Lamb's by about 20%) is not easily observed because it is energetically less favourable. However it has an indirect evidence in the experimental results of Valentine, Sather & Heideger (1965). They observed oscillation frequencies larger than Lamb's result by about 30–40% in rather special circumstances. Small drops of cyclohexanol were made to coalesce with large drops of a solution of benzene and carbon tetrachloride. Frequencies of the large benzene-carbon tetrachloride drops before and after coalescence were obtained. Normally one expects, as mentioned earlier, only the lower frequency modes. A simple calculation using the drop sizes given by Valentine *et al.* shows that the low frequency of the small drop is approximately equal to the high frequency of the large drop. Perhaps this has set up a resonant excitation of the high-frequency mode of the large drop.

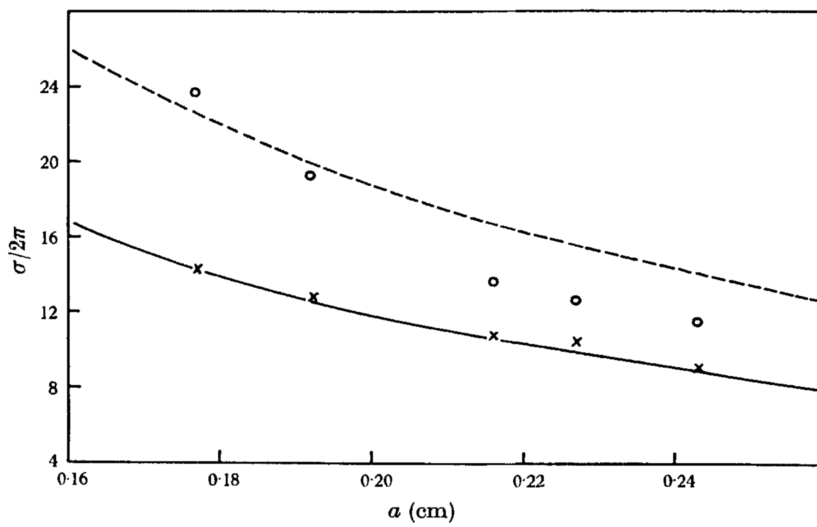

FIGURE 3. Comparison of the calculated results with the measured frequencies of Winnikow & Chao (1966) for a pure liquid–liquid system, nitrobenzene drop in water. – – –, Lamb's theory; ——, present calculations; × × ×, measured oscillation frequency; ○ ○ ○, measured eddy discharge frequency.

There is one refreshing exception to all these experiments which, although in qualitative agreement, do not permit a quantitative comparison. Winnikow & Chao (1966) have measured the frequencies of nitrobenzene drops moving in water taking great care to eliminate all impurities and to keep the physical parameters clearly defined. This has enabled us to perform the calculations in detail. In figure 3 the experimentally observed frequencies are compared with our calculations and with Lamb's expression. There is gratifying agreement with our calculations. The magnitude of the deviations from Lamb's frequencies is also clearly seen. Accurate measurements on pure liquid–liquid systems, such as the above one are badly needed for a definitive comparison between theory and experiment.

Lastly the effect of eddy discharge frequencies should also be noted. Winnikow & Chao have also measured these eddy frequencies which are shown in figure 3.

The intersection of the eddy discharge curve and the oscillation line will result in a resonance, which has been noticed for instance by Gunn (1949) in the case of water drops in air. A striking maximum in the frequency-drop size curve observed by Kaparthi & Licht (1962) is also most probably due to such a process.

In conclusion, it appears that experiments reveal distinct deviations from the classic analysis of Lamb. These discrepancies undoubtedly arise from the neglect of viscous and inertial effects in Lamb's calculation. Experimental studies on clearly defined systems would help to check the validity of the present calculations.

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