A STUDY OF THE OSCILLATIONS OF A VISCOUS FLUID DROP MOVING IN A BACKGROUND FLUID MEDIUM

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ABSTRACT

The oscillations of a drop moving in a background fluid medium have been studied by including the contributions from the viscosities of the two phases, the inertial effects caused by the drop motion and the deformation of the drop. The nonspherical shape of the drop alters the expression for frequency of a spherical drop by including correction terms explaining many of the experimental results on highly deformed drops. The viscous terms split the mode of frequency into two-one lower and the other higher than Lamb's. Experimental evidence exists for both the lower and the higher frequency modes. If the size of the drop is smaller than a certain critical value, ($\sim \hat{\rho}\, \hat{\nu}^2/T$, $\hat{\rho}$ *the density,* $\hat{\nu}$ *the kinematic viscosity and* T *interfacial tension between the two phases) the perturbations on the drop surface will be damped aperiodically. This critical size which is very small* ($\sim 10^{-6}$ cm.) *for a system like a liquid drop in free space, becomes prominent* (*~ a few mm*) *for a fluid drop in a dense viscous liquid medium This can be a possible explanation for the nonobservance of oscillations in highly dense viscous systems. Detailed calculations are given in the limiting cases* ($\sigma' >> 1$, $\sigma' \sim 1$, $\sigma' << 1$) *of an oscillation parameter* σ' ($= \sigma a/U$, σ *being the oscillation frequency,* a *the radius of the drop and* U *the terminal velocity of the drop in the fluid medium*).

1. INTRODUCTION

An analysis of the motion of drops and bubbles is vital for a better understanding of certain physico-chemical processes. For example, the oscillations of the drop contribute to such interfacial phenomena like heat and mass transfer. But a complete theoretical study of the oscillations is hampered by inadequate knowledge about the shape of the drop and the contributions from the inertial effects caused by the drop motion, the interfacial tension and the two phase parameters like the viscosities.

Lamb[1] studied the limiting case of the oscillations of a spherical liquid drop at rest in an inviscid fluid. When the surface disturbance can be expressed in terms of Legendre polynomials, he found that the frequency of oscillation is given by the expression

$$\sigma_L^2 = \frac{l(l-1)(l+1)(l+2)\,T}{[(l+1)\hat{\rho} + l\,\rho]\,a^3} \qquad [1]$$

where $\hat{\rho}$ is the density of the drop phase, ρ the density of the external phase, a is the drop radius and the mode of frequency l is related to the perturbed drop surface

$$r = a\,[1 + \epsilon_0\, P_l(\cos\theta)\, \exp(i\,\sigma'\,t)]$$

and ϵ_0 is the amplitude of oscillation. $l=2$ corresponds to the familiar prolate-oblate oscillations. Such calculations are of limited applicability and have failed to explain many of the observational details[2].

In an earlier paper[3] the oscillations of fluid drops relatively at rest in background media were discussed by including the viscous effects and the deformation of the drop. Independent of our work Miller and Scriven[4] have also discussed the oscillations of stationary viscous drops in background fluid media. They include the terms from interfacial viscosity and elasticity and study both the frequency and damping of the oscillations in various limiting cases. But in many of the practical situations, the drop will be in a steady terminal motion in a background medium. Consequently the above calculations are not strictly comparable with the experiments.

In order to remove this lacuna, we have undertaken a study of the oscillations of a drop moving with a steady terminal velocity in another fluid medium. The contributions from the deformation of the drop and the viscous terms have been taken into account. The analysis is restricted to the Lamb's oscillation modes where the disturbance is expressed in terms of Legendre polynomials. The calculations are valid for low *Re* and *We*.

Oscillations of a spherical drop in terminal motion in another fluid medium are discussed in the next section whereas section 3 deals with a study of the oscillations of a deformed drop in motion. Finally the theoretical results are compared with the available experimental results in section 4.

2. Oscillations of a Spherical Liquid Drop moving with a Steady Terminal Velocity in Another Fluid Medium

2.1 *Formulation of the Perturbation Equations*:

The equations of motion will here correspond either to a drop moving with a uniform velocity U in a fluid medium (the coordinate system being fixed in the continuous phase) or to the fluid having a uniform velocity U at infinity flowing past a drop (the coordinate system being fixed in the drop and conveniently the origin at the centre of the drop). The latter coordinate system can be chosen as it simplifies the calculations. The equations can be made dimensionless by choosing a the radius of the spherical drop as a characteristic length, U the uniform stream velocity as a characteristic velocity, Ua^2 as a characteristic stream function, $\frac{1}{2}\rho U^2$ as a characteristic pressure and a/U as a characteristic time. The dimensionless equations can be written in the form

$$\partial (D^2 \psi)/\partial t + E(\psi, \psi) = (1/Re)\, D^4 \psi \quad \text{for} \quad a \leq r < \infty$$

$$\partial (D^2 \hat{\psi})/\partial t + E(\hat{\psi}, \hat{\psi}) = (1/\hat{Re})\, D^4 \hat{\psi} \quad \text{for} \quad 0 \leq r \leq a \qquad [2]$$

where

$$Re = Ua/\nu, \quad \hat{Re} = Ua/\hat{\nu}, \quad \nu = \mu/\rho, \quad \xi = \cos\theta,$$

$$E(\psi, \hat{\psi}) = \frac{1}{r^2}\left[\frac{\partial(\psi, D^2\hat{\psi})}{\partial(r, \xi)} + \left(\frac{2\xi}{1-\xi^2}\frac{\partial\psi}{\partial r} + \frac{2}{r}\frac{\partial\psi}{\partial\xi}\right) D^2\hat{\psi}\right]$$

$$D^2 = \frac{\partial^2}{\partial r^2} + \frac{1-\xi^2}{r^2}\frac{\partial^2}{\partial\xi^2}$$

If the time dependent part $[\partial(D^2\psi)/\partial t]$ of the above equations is left out (corresponding to the condition $\sigma a^2/\nu << 1$) then the equations describe the steady flow of a fluid past a drop. This case has been fairly well analysed at low Reynolds numbers by Proudman and Pearson[6] for a rigid sphere and Taylor and Acrivos[5] for a fluid sphere.

On such a steady flow of a fluid past a drop, a perturbation causing the drop to oscillate can be superimposed. Then the resulting drop surface can be written as

$$r = 1 + \epsilon P_l(\xi) \qquad [3]$$

where ϵ is a function of time [such as $\epsilon_0 \exp(i\sigma' t)$], ϵ_0 being the amplitude of oscillation, $P_l(\xi)$ is a spherical harmonic of order l, and $\sigma' = \sigma a/U$ is a dimensionless frequency.

The perturbation is assumed to be quite small compared to the size $(=1)$ of the drop so that $\epsilon_0^2, \epsilon_0^3, \cdots$ can be neglected. Then the stream function undergoes a change from $\psi_0, \hat{\psi}_0$ to $\psi_0 + \Delta\psi(t)$ and $\hat{\psi}_0 + \Delta\hat{\psi}(t)$ respectively. The stream function can be expanded in the form

$$\psi = \psi_0 + \Delta\psi_1 \exp(i\sigma' t) + \Delta\psi_2 \exp(2i\sigma' t) + \cdots \qquad [4]$$

Substituting these expansions in the equations of motion (2) and collecting coefficients of exp $(i\sigma' t)$, exp $(2i\sigma' t)$, $\cdots$ and time independent terms, the equations for $\Delta\psi_1$, $\Delta\psi_2$, $\cdots$ and ψ_0 are obtained. The zeroth (steady) and I order perturbation equations can be written as

$$E(\psi_0, \psi_0) = (1/Re)\, D^4 \psi_0$$

$$E(\hat{\psi}_0, \hat{\psi}_0) = (1/\hat{Re})\, D^4 \hat{\psi}_0 \qquad [5]$$

$$i\sigma' D^2 \Delta\psi_1 + E(\psi_0, \Delta\psi_1) + E(\Delta\psi_1, \psi_0) = (1/Re)\, D^4 \Delta\psi_1$$

$$i\sigma' D^2 \Delta\hat{\psi}_1 + E(\hat{\psi}_0, \Delta\hat{\psi}_1) + E(\Delta\hat{\psi}_1, \hat{\psi}_0) = (1/\hat{Re})\, D^4 \Delta\hat{\psi}_1 \qquad [6]$$

2.2 *Solutions of the perturbation equations* :

The solutions of the above equations will have to satisfy the following boundary conditions [5,7]. As $r \to \infty$, $u_r = -\xi$, $u_\theta = \sin\theta = (1-\xi^2)^{1/2}$ and hence $\psi \to \frac{1}{2}(1-\xi^2)\, r^2$. At the interface of the drop and the continuous phase, u_r, u_θ, tangential stress $(T_{r\theta})$ and the radial stress (T_{rr}) should be continuous taking into account the surface stresses. Physically meaningful solutions only are to be considered at $r = 0$ and at $r = \infty$.

The steady flow equations [5] have been discussed by Taylor and Acrivos[5] and Matunobu[8] for flow past a liquid drop at low *Re*. In the creeping flow region (inertial contribution neglected), the solution shows that the drop should remain spherical for all Weber numbers. Inertial effects can then be studied by expanding ψ in terms of *Re*. Then the shape of the drop departs from spherical into a spheroidal shape.

$$r = 1 - \epsilon_1\, We\, P_2(\xi), \quad \hat{We} = \hat{\rho}\, a\, U^2/T$$

at low Weber numbers, and into a spherical cup shape

$$r = 1 - \epsilon_1\, We\, P_2(\xi) - \epsilon_2\, (We^2/Re)\, P_3(\xi)$$

at higher Weber numbers.

Now let us study the I order perturbation equations [6]. The dimensionless oscillation parameter $\sigma' = \sigma\, a/U$, σ being the complex oscillation frequency can have any value in a wide range. Let us consider three limiting cases.

Case (i): $\sigma' >> 1$. This can happen at very low *We*. The inertial contribution in eq. [6] can be neglected as it is $O(Re)$. The time dependent terms are predominating and the terms in $D^4\Delta\psi$'s are taken since they are the highest derivative terms. Hence the equations become

$$D^2(D^2 + h^2)\,\Delta\psi_1 = 0; \quad h^2 = -i\sigma' Re = -i\sigma\, a^2/\nu$$

$$D^2(D^2 + \hat{h}^2)\,\Delta\hat{\psi}_1 = 0; \quad \hat{h}^2 = -i\sigma' \hat{Re} = -i\sigma\, a^2/\hat{\nu}$$

It is obvious that these are just the basic equations of I order perturbation for an oscillating liquid drop, relatively at rest in a background fluid medium[3]. The solutions can be discussed as before with a difference in the boundary conditions for the stress components.

$$T_{r\theta}=\frac{2}{Re}\left[\frac{1}{r}\ \frac{\partial u_r}{\partial\theta}-\frac{u_\theta}{r}+\frac{\partial u_\theta}{\partial r}\right]$$

$$\hat{T}_{r\theta}=\frac{2\gamma}{\hat{Re}}\left[\frac{1}{r}\ \frac{\partial\hat{u}_r}{\partial\theta}-\frac{\hat{u}_\theta}{r}+\frac{\partial\hat{u}_\theta}{\partial r}\right] \qquad [8]$$

$$T_{rr}=p_0+\delta p-\frac{4}{Re}\ \frac{\partial u_r}{\partial r}$$

$$\hat{T}_{rr}=\hat{p}_0+\delta\hat{p}-\frac{4\gamma}{\hat{Re}}\ \frac{\partial\hat{u}_r}{\partial r} \qquad [9]$$

where $\gamma=\hat{\rho}/\rho,\ k=\hat{\mu}/\mu,\ \nu=\mu/\rho.$

Physically meaningful solutions of [7] can be written.

$$\Delta\ \psi_1=[Br^{-l}+Dr^{\frac{1}{2}}\ J_{-(l+\frac{1}{2})}(hr)\ ]\ F_l(\xi)$$

$$\Delta\ \hat{\psi}_1=[Ar^{1+l}+Cr^{\frac{1}{2}}\ J_{(l+\frac{1}{2})}(\hat{h}r)\ ]\ F_l(\xi) \qquad [10]$$

where $F_1(\xi)=\int_{-1}^{\xi}P_l(\xi)\,d\xi$ are the Gegenbauer functions.

A more general solution satisfying the boundary conditions would involve Hankel functions H′ instead of Bessel functions. But the latter have been chosen for purpose of easy computation as tables of $J_n(x)$ are readily available.

The constants A, B, C, and D can be eliminated by the interfacial conditions ; namely

$$u_r=\hat{u}_r,\ \ u_\theta=\hat{u}_\theta.$$

$$T_{r\theta}=\hat{T}_{r\theta},\ \text{and}\ T_{rr}=\hat{T}_{rr}+(2/We)\,[1/x_1+1/x_2] \qquad [11]$$

at the interface $r=1$; where $u_r=-\dfrac{1}{r^2}\ \dfrac{\partial\psi}{\partial\xi}$, $u_\theta=-\dfrac{1}{r\,(1-\xi^2)^{1/2}}\ \dfrac{\partial\psi}{\partial r}$

x_1, x_2 are the principal radii of curvature and $We = \rho\, aU^2/T$. The resulting four equations for A, B, C and D can hold good only if their secular determinant vanishes. This can be further simplified[11].

$$\begin{vmatrix} -\hat{h}\, Q_{l+\frac{1}{2}}(\hat{h}) & (2l+1) & h\, Q_{-(l-\frac{1}{2})}(h) \\ k\left(-\dfrac{\hat{h}^2}{2} + \hat{h}\, Q_{l+\frac{1}{2}}\right) & k(l^2-1) - l(l+2) & \dfrac{h^2}{2} - h\, Q_{-(l-\frac{1}{2})} \\ -\dfrac{1}{l} - \dfrac{2\hat{h}\, Q_{l+\frac{1}{2}}}{i\hat{Re}} & \left(\dfrac{1}{l} + \dfrac{1}{\gamma(l+1)} + \dfrac{2(l-1)}{i\hat{Re}} + \dfrac{2(l+2)}{ik\hat{Re}} - \dfrac{(l-1)(l+2)}{\hat{We}}\right) & \left(\dfrac{2h\, Q_{-(l+\frac{1}{2})}}{ik\, Re} - \dfrac{1}{\gamma(l+1)}\right) \end{vmatrix} = 0 \qquad [12]$$

where $\qquad Q_{l+1/2} = J_{l+3/2}/J_{l+1/2}, \; Q_{-(l+1/2)} = J_{-(l-1/2)}/J_{-(l+1/2)}$

With the help of this equation, the frequencies of oscillation can be computed for any fluid-fluid system provided the dimensionless parameter $\sigma' = \sigma a/U >> 1$. This holds good for drops in very slow motion. For the general case like a nitrotoluene drop oscillating in water, the full characteristic equation [12] will have to be solved for the damping and oscillatory parts. The results of such a calculation[3] have been plotted in the Figure 4 against the observed frequencies.

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

$$\sigma_{01}^2 = \frac{l(l-1)(l+2)}{\sigma'^2\, \hat{We}} = 1 + \frac{2l(l-1)}{i\hat{Re}} - \frac{2(l^2-1)}{\hat{h}^2 - 2\hat{h}\, Q_{l+\frac{1}{2}}} - \frac{4l(l^2-1)\, Q_{l+\frac{1}{2}}}{i\hat{Re}(\hat{h} - 2Q_{l+\frac{1}{2}})} \qquad [13]$$

For the case $\hat{Re} \to a\sigma \cdot a/\hat{\nu}$ (*i.e.*, the drop relatively at rest, this agrees with the result of Reid[10] and Chandrasekhar[9]. From the equation it is evident that the frequencies are altered from the Lamb's by the introduction of viscous terms. L.H.S. is merely the ratio of Lamb's to the modified frequency.

If σ is taken to be purely imaginary, then the aperiodic damping modes discussed by Reid and Chandrasekhar result. These have been calculated using equation [13] and plotted in Fig. 1 for the case of a drop in free space. 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 $\hat{\rho}\,\hat{\nu}^2/T$

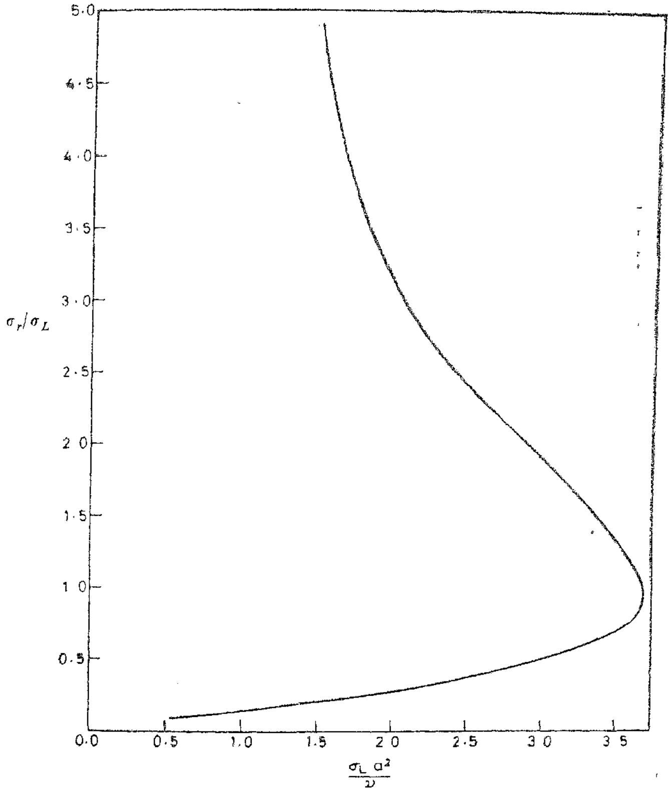

FIG. 1

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

aperiodic damping cannot occur. This means that for oscillations to start, the drop size must be larger than this critical size($a_C = C\hat{\rho}\,\hat{\nu}^2/T$, C a constant ~ 1). Though this critical size is very small ($\sim 10^{-6}$ cm.) for a system like water drop in air, it assumes as large values as a few *mm* for a drop or a bubble in a dense viscous liquid.

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

$$\sigma_{02}^2 = \frac{l\,(l-1)\,(l+1)\,(l+2)}{\sigma'^2\,(2\,l+1)\,We} = 1 - \frac{2\,l+1}{h\,[Q_{l+\frac{1}{2}} - Q_{-(l+\frac{1}{2})}]} \qquad [14]$$

The frequencies of oscillation compiled from this equation have been plotted as a function of drop size in Fig. 2. It shows that the contribution from viscous terms split the mode of frequency into two branches—one lower and the other higher than Lamb's. The deviation from Lamb's result is small when the two phases are dissimilar like a drop in free space or a bubble in a dense viscous liquid, but in this case of two similar phases the departure is significant. Even here the splitting becomes pronounced when the drop size is not too small or not too large.

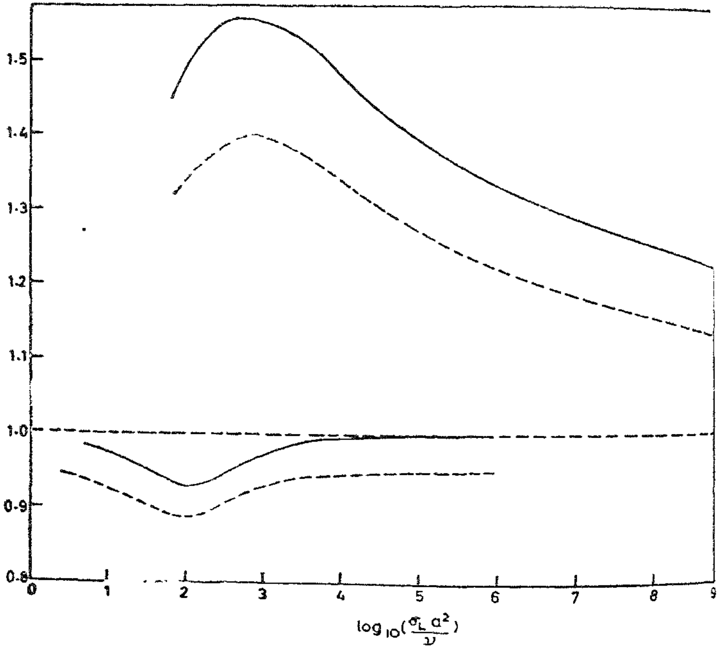

FIG. 2

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 ($\epsilon_1 = 0\ 2$).

Case (ii) : $\sigma' < < 1$. This case does not have many applications and hence will not be discussed here.

Case (iii) : $\sigma' \sim 1$

By far, the very interesting limiting case seems to be that for which $\sigma' = O(1)$. This can be studied at low *Re* by the well known expansion methods. Expanding $\Delta\psi_1$ and $\Delta\hat{\psi}_1$ in powers of *Re*,

$$\Delta\psi_1 = \Delta\psi_{10} + Re\ \Delta\psi_{11} + \cdots$$

$$\Delta\hat{\psi}_1 = \Delta\hat{\psi}_{10} + Re\ \Delta\hat{\psi}_{11} + \cdots \qquad [15]$$

and substituting these expansions in the equations [6] and collecting the coefficients of Re^0, Re^1, $\cdots$ we obtain

$$D^4\ \Delta\ \psi_{10} = 0\ ; \quad D^4\ \Delta\ \hat{\psi}_{10} = 0 \qquad [16]$$

$$D^4\ \Delta\ \psi_{11} - [\,E(\psi_{00},\ \Delta\ \psi_{10}) + E(\Delta\psi_{10},\ \psi_{00})\,] - i\ \sigma'\ D^2\ \Delta\ \psi_{10} = 0$$

$$D^4\ \Delta\ \hat{\psi}_{11} - [\,E(\hat{\psi}_{00},\ \Delta\ \hat{\psi}_{10}) + E(\Delta\hat{\psi}_{10},\ \hat{\psi}_{00})\,] - i\ \sigma'\ D^2\ \Delta\ \hat{\psi}_{10} = 0 \qquad [17]$$

etc.

These pairs of equations are then to be solved satisfying the boundary conditions.

Equations [16] have the physically meaningful solutions

$$\Delta\psi_{10} = (Br^{-l} + Dr^{-l+2})\ F_l(\xi)$$

$$\Delta\hat{\psi}_{10} = (Ar^{l+1} + Cr^{l+3})\ F_l(\xi) \qquad [18]$$

The constants, A, B, C, and D can be eliminated by the use of the four interfacial conditions at $r = 1$, namely the continuity of radial and tangential velocity components, continuity of tangential stress and the equality of excess radial stress with that due to the interfacial tension. The resulting equations for A, B, C, and D at $r = 1$ are[11]

$$A + C - B - D = 0$$

$$A\,(l+1) + C\,(l+1) + B\,l + D\,(l-2) = 0$$

$$k\,[A\,(l+1)\,(l-1) + C\,l\,(l+2)] - B\,l\,(l+2) - D\,(l^2-1) = 0$$

$$A\left(\frac{1}{l}+\frac{2(l-1)}{i\,\sigma'\,\hat{Re}}-\frac{(l-1)(l+2)}{\sigma'^2\,\hat{We}}\right)+C\left(\frac{1}{l+2}-\frac{2(2l+3)}{i\,\sigma'\,l\,\hat{Re}}+\frac{2(l+1)}{i\,\sigma'\,\hat{Re}}\right.$$

$$\left.-\frac{(l-1)(l+2)}{\sigma'^2\,\hat{We}}\right)+B\left(\frac{1}{\gamma\,(l+1)}+\frac{2(l+2)}{i\,\sigma'\,k\,\hat{Re}}\right)+D\left(\frac{1}{\gamma\,(l+1)}\right.$$

$$\left.+\frac{2(2l-1)}{(l+1)\,i\,\sigma'\,k\,\hat{Re}}+\frac{2l}{i\,\sigma'\,k\,\hat{Re}}\right)=0 \qquad [19]$$

In writing the last equation of [19], use has been made of the expressions

$$(1/x_1)+(1/x_2)\approx 2+(l-1)(l+2)\,\epsilon\,P_l$$

$$(\delta p)_{r=1}=2\,P_l\,i\,\sigma'\left(-\frac{B}{l+1}-\frac{D}{l-1}\right)-\frac{2}{Re}\,DP_l\frac{2(2l-1)}{l+1}$$

$$(\delta\hat{p})_{r=1}=2\,P_l\,\gamma\,i\,\sigma'\left(\frac{A}{l}+\frac{C}{l+2}\right)-\frac{2\gamma}{\hat{Re}}\,\frac{2\,C\,P_l\,(2l+3)}{l}$$

Since the four interfacial conditions [19] should hold good simultaneously and since they are homogeneous in A, B, C, and D, their secular determinant should vanish. This can be simplified to

$$\begin{vmatrix} 2 & 2l+1 & 0 \\ k(2l+1) & k(l^2-1)-l(l+2) & (k+1)(2l+1) \\ \left(-\frac{2}{l(l+2)}-\frac{6}{i\,\sigma'\,l\,\hat{Re}}\right) & \left(\frac{1}{l}+\frac{1}{\gamma\,(l+1)}+\frac{2(l-1)}{i\,\sigma'\,\hat{Re}}+\frac{2(l+2)}{ik\,\sigma'\,\hat{Re}}-\frac{(l-1)(l+2)}{\sigma'^2\,\hat{We}}\right) & \left(-\frac{2}{l(l+2)}+\frac{2}{\gamma(l^2-1)}-\frac{6}{i\,\sigma'\,\hat{Re}\,k\,(l+1)}\right) \end{vmatrix}=0 \qquad [20]$$

In practice, since the prolate-oblate oscillations predominate over others, only $l=2$ mode need be considered. In such a case the determinental equation gets reduced to

$$\frac{-40\,(k+1)}{\sigma'^2\,\hat{We}}+\frac{38k^2+89k+48}{i\,\sigma'\,k\,\hat{Re}}+\frac{26k+29}{4}+\frac{16k+14}{\gamma}=0 \qquad [21]$$

This equation exhibits all the qualitative features described by the characteristic equation in the case of oscillations of a stationary spherical drop. The difference in the form arises because of the range of σ' considered. However both the results merge with each other when we consider, say, for example the aperiodic damping modes for $k \to \infty$.

As the characteristic equation [21] describes oscillations for drops in terminal motion in other fluid media, it is more useful for purposes of comparison with experimental results. But it cannot give an accurate description of the oscillation frequency for all Re as it is derived for the case $\sigma' \sim 1$.

(*a*) For the oscillations of a drop in free space, $\gamma \to \infty$, $k \to \infty$ Then

$$\sigma' = \frac{38}{13\,\hat{Re}}\left[1 \pm \left(\frac{13 \times 80\,\hat{Re}^2}{38^2\,\hat{We}} - 1\right)^{1/2}\right]$$

For
$$\frac{13 \times 80\,\hat{Re}^2}{38^2\,\hat{We}} > 1, \text{ ie., } a > a_c = \frac{18\,\hat{\rho}\,\hat{\nu}^2}{13\,T} \qquad [22]$$

σ' consists of a damping and an oscillatory term. This represents the usual damped oscillations of a liquid drop. For a typical case of water drop oscillating in air, the present results are compared with those given by Lamb's equation ; in Fig. 3. A detailed discussion of these will be taken up in section 4.

When the radius of the drop is almost the critical size, a_c, the oscillations commence. This can be compared with the results obtained by Reid[10],

$$a_c = \frac{3.69^2}{8}\,\frac{\hat{\rho}\,\hat{\nu}^2}{T}$$

The two results agree fairly well.

For drops with sizes smaller than this critical size, σ' has two imaginary solutions and this results in two different modes of aperiodic damping.

(*b*) The oscillations of a bubble in a viscous liquid is given by the limiting values of γ and k tending to zero. Then

$$\sigma' = \frac{48}{28\,Re}\left\{1 \pm \left(\frac{14 \times 160\,Re^2}{48^2\,We} - 1\right)^{1/2}\right\}$$

As in the previous case, oscillations set in at a critical drop radius $a_c \approx \rho\,\nu^2/T$. For drops larger than a_c, damped oscillations take place whereas for smaller drop sizes, two modes of aperiodic damping are present.

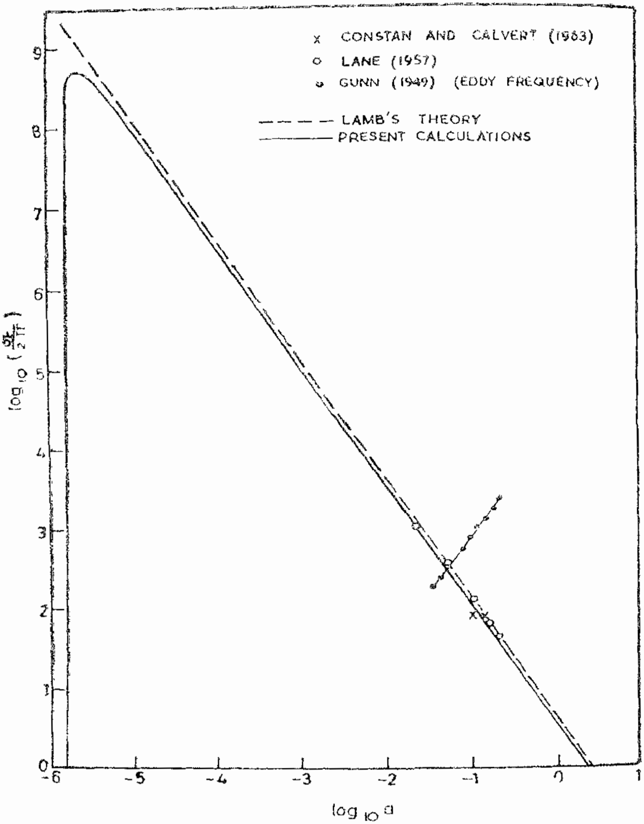

FIG. 3

Oscillations of a water drop in air with a uniform relative velocity. Also are plotted the eddy discharge frequencies.

(*c*) The third limiting case of a fluid sphere oscillating in a similar fluid is very important from practical considerations. In this case $\Upsilon = 1$, $k = 1$. Then

$$\sigma' = \frac{2}{Re}\left[\, i \pm \left(\frac{16\, Re^2}{35\, We} - 1\right)^{1/2}\right]$$

Again, as before, oscillations set in at a critical drop size $a_c \approx 35\,\rho\,\nu^2/16\,T$. This is about twice the critical drop size for oscillation in free space. Oscillations will be aperiodically damped for sizes less than the critical size (a_c), For drop sizes larger than a_c, damped oscillations take place. Using the full equation [21] the oscillation frequency of *o*-nitrotoluene drop in water has been studied as a function of drop size. Fig, 4 shows the present calculated values with those of Lamb. The results are consistently lower than Lamb's values by more than 30%, clearly indicating the inadequacy of the limited calculations of Lamb. Part of the abnormally large deviation from the experimetal results is due to the nonspherical shape of the drop.

The critical size at which a drop can execute oscillations is approximately $C\rho\,\nu^2/T$ for all fluid-fluid systems. (The numerical constant C is of the order of 1). Hence Winnikow's reasoning that this critical size may be increased by a few orders for liquid-liquid systems proves to be not correct. However for very viscous liquids like *m*-cresol, cyclohexanol, Glycerine, castor oil etc., this critical size assumes practical magnitudes. Some of the calculated critical sizes for such viscous liquids are shown in Table 1. Absence of oscillations noticed in several viscous liquid systems is most probably because of this reason.

TABLE 1

Critical Size (a_c) for Oscillations to start in some viscous Liquids

$a_c \approx C\,(\hat{\rho}\,\hat{\nu}^2/T)$ which C is a constant factor ~ 1

(All values approximate)

Liquid drop in water	$\hat{\mu}$ Poise	$\hat{\rho}$ gm/cc	T with air dyne/cm.	T with water dyne/cm.	$(\hat{\rho}\,\hat{\nu}^2/T)$ cm.
m-cresol	0.21	1.034		4.0	0.01
Cyclohexanol	0.68	0 94	25.3	3 92	0.12
Glycerine (in oil)	14.9	1.26	60	28.6	6.2
Castor oil	9.86	0.96	39	20.0	5.0
Machine oil (light)	1.14	1.0	35	15.0	0.08
Olive oil	0 84	0.92	35 8	22.9	0.03
Oleic acid	0.26	0.895	32 5	15.59	0.004
Phenol	0.127	1.072	40.9	0.34	0.05

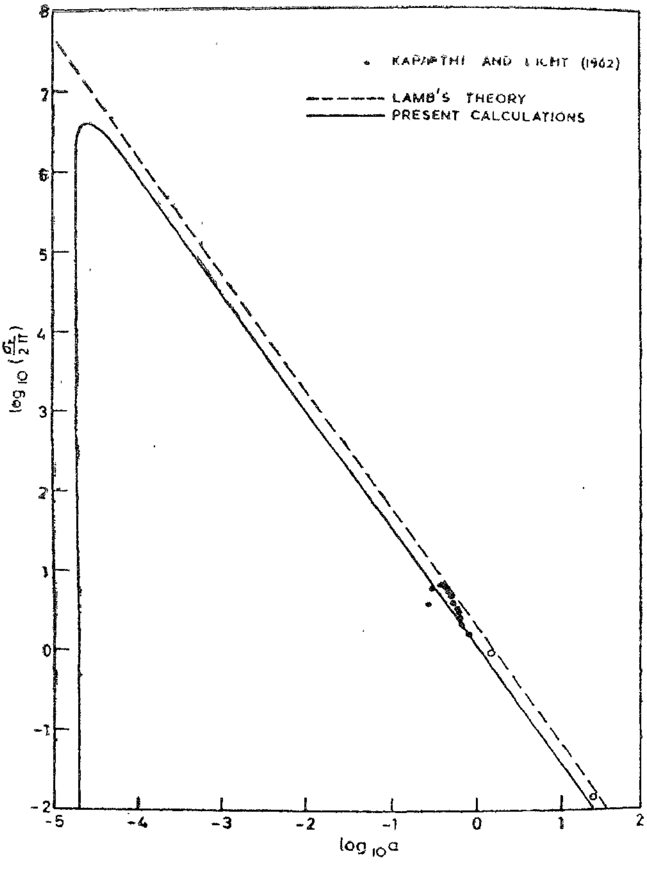

FIG. 4

Frequencies of oscillation of an 0-nitrotoluene drop falling with its terminal velocity in water. (00 . . . calculated frequencies if the drop motion is very slow. Full line ——— calculated frequencies if $\sigma a/U \sim 1$).

3. Oscillations of a Deformed Drop in Motion

The deformation of a drop moving in a liquid medium has been attributed mainly to the inertial effects[5]. Taylor and Acrivos showed that to a first approximation the shape of the drop can be taken as that of an oblate spheroid.

$$r = 1 + \epsilon_1 P_2(\xi) \qquad [23]$$

where $\epsilon_1 = -\dfrac{We}{4(k+1)^3}\left[\left(\dfrac{81}{80}k^3 + \dfrac{57}{20}k^2 + \dfrac{103}{40}k + \dfrac{3}{4}\right) - \dfrac{\gamma - 1}{12}(k+1)\right]$

ϵ_1 which is linear in We at low Weber numbers determines the deviation of the drop from the spherical shape. For higher We, the drop shape changes into that of a spherical cup, given by

$$r = 1 + \epsilon_1 P_2(\xi) + \epsilon_2 (We/Re) P_3(\xi) \qquad [24]$$

where $$\epsilon_2 = \frac{3(11k + 10)We}{70(k+1)}\epsilon_1.$$

These equations do not fully explain many of the experimental results at high We and Re[13,14]. But at small values of We and Re, they satisfactorily explain the observed shapes of drops. Further they are the best calculations available so far and therefore form the basis for our study of the effect of deformation on oscillations of a drop. We consider the drop shape to be an oblate spheroid given by [23] for the present calculations.

A perturbation on the surface of the drop which causes it to oscillate can be considered as usual to consist of spherically symmetric Legendre displacements. Hence

$$r = 1 + \epsilon_1 P_2(\xi) + \epsilon P_l(\xi) \exp i\sigma' t \qquad [25]$$

The equations of motion can be written and split into various time ordered terms. The first of these correspond to time independent steady flow and the latter to various ordered perturbations. The perturbation equations of *I* order are those given by [6]. Various ranges of σ' can be considered as before.

Case (i) : $\sigma' >> 1$.

In this case, the inertial contribution can be neglected as the time dependent terms predominate and together with terms in $D^4 \Delta\psi_1$, the equations [7] are obtained. These equations which are similar to those obtained in the study of oscillations of a stationary liquid drop[3] are to be solved subject to the boundary conditions using equations [8] and [9] at the deformed surface $r = 1 + \epsilon_1 P_2(\xi)$. Results very similar to those for a stationary drop are obtained. These have been briefly summarised in case (*i*) of section (2). Especially if one neglects the viscous effects, then the resulting equations

$$D^2 \Delta \psi_1 = 0, \; D^2 \Delta \hat{\psi}_1 = 0$$

satisfying the boundary conditions give

$$\sigma^2 = \frac{l(l-1)(l+1)(l+2)\,T}{\hat{\rho}\, a^3 [l+1+l/\gamma]} \left(1 - \frac{\epsilon_1}{4}\right)$$

$$= \sigma_L^2 \left(1 - \frac{\epsilon_1}{4}\right)$$

With the introduction of viscous terms, the characteric equation [12] is altered by the contribution from the deformation of the drop. For example, eq. [14] becomes

$$\sigma_{02}^2 = 1 - \frac{5}{h\,(Q_{5/2} - Q_{-5/2})} + \frac{\epsilon_1}{4} + O\left(\frac{1}{h^2}\right)$$

Case (ii) : $\sigma' << 1$.

In this case, terms containing σ' can be neglected from the basic equations [6]. The resulting equations are devoid of these time dependent terms. Hence they are not useful in discussing the oscillations of drops in media.

Case (iii) : $\sigma' \sim 1$.

$\Delta \psi_1$ and $\Delta \hat{\psi}_1$ can be expanded in powers of Re and the solutions obtained for small Re. Equations [16], which will be obtained in this case also can be solved.

$$\Delta \psi_{10} = (Br^{-l} + Dr^{-l+2})\, F_l(\xi)$$

$$\Delta \hat{\psi}_{10} = (Ar^{l+1} + Cr^{l+3})\, F_l(\xi)$$

The constants A, B, C and D can be eliminated by using the interfacial conditions [11] for the velocity and the stress components at the deformed surface. Then as in the previous case we get four homogeneous equations in A, B, C and D. For a simultaneous fulfillment of these conditions, the secular determinant should vanish. For $l = 2$ mode, the following characteristic equation can then be obtained.

$$-\frac{40\,(k+1)}{\sigma'^2\,\hat{We}} + \frac{38\,k^2 + 89\,k + 48}{ik\,\sigma'\,\hat{Re}} \left(1 - \frac{\epsilon_1}{4}\right) + \left(1 + \frac{\epsilon_1}{4}\right)\left[\frac{26\,k + 29}{4} + \frac{16k + 14}{\gamma}\right] = 0 \qquad [26]$$

As before the characteristic equation for oscillations of a deformed drop in motion can be studied for various limiting cases of γ and k.

(*a*) $\gamma \to \infty$, $k \to \infty$. This corresponds to oscillations of a drop in free space. Then

$$\sigma' = \frac{38\,(1-\epsilon_1/2)}{13\,\hat{Re}}\left[\, i \pm \left\{\frac{260\ \hat{Re}^2}{361\ \hat{We}}\left(1+\frac{3\epsilon_1}{4}\right)-1\right\}^{1/2}\right] \qquad [27]$$

$\sigma' = \sigma\, a/U$ consists of a damping and an oscillatory term. There exists a critical size a_c

$$a_c \approx \frac{361}{260}\ \frac{\hat{\rho}\,\hat{\nu}^2}{T}\left(1-\frac{3\,\epsilon_1}{4}\right) \qquad [28]$$

at which oscillations start. For drop sizes smaller than a_c, two modes of aperiodic damping occur. The critical size below which the drop cannot execute oscillations at all is smaller than that for a spherical drop by a factor of $(1-3\,\epsilon_1/4)$.

(*b*) $\gamma \to 0$, $k \to 0$. This can be used for the study of oscillations of a bubble in a dense viscous liquid medium. Drops with sizes larger than a_c

$$a_c \approx \frac{36\,\rho\nu^2}{35\,T}\left(1-\frac{3\,\epsilon_1}{4}\right)$$

execute damped oscillations, but disturbances on smaller drops will be aperiodically damped.

(*c*) $\gamma \sim 1$, $k \sim 1$. In this case, similar behaviour results except that the critical size is

$$a_c \approx \frac{35\rho\nu^2}{16\,T}\left(1-\frac{3\,\epsilon_1}{4}\right)$$

Numerical result for the frequencies of oscillation of a deformed drop moving in a background medium are given in Table 2, for a system of water drop oscillating in air. It appears that the deformation does not affect the frequency in a major way, but alters it by a correction factor which is however appreciable.

4 Results and Discussion

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 etc., give nearly comparable effects to the oscillations and it is not justified to ignore any one

TABLE 2

Damped Oscillations of a Deformed Water Drop in Air

$\gamma = 841.74$, $k = 48.31$, $T = 71.97$ dyne cm^{-1}

Shape of the drop is assumed as

$$r = 1 + \epsilon_1 P_2 = 1 - (k'/4)\ We\ P_2 \approx 1 - 0.0155\ We$$

a cm.	σ_r for Weber number				
	0	0.4	0.8	1.2	1.4
10^{-4}	21×10^6	21.26×10^6	21.5×10^6	21.78×10^6	21.9×10^6
10^{-3}	6.64×10^5	6.72×10^5	6.8×10^5	6.89×10^5	6.93×10^5
10^{-2}	21×10^3	21.26×10^3	21.5×10^3	21.78×10^3	21.9×10^3
10^{-1}	6.64×10^2	6.72×10^2	6.8×10^2	6.89×10^2	6.93×10^2
2×10^{-1}	234.7	237.6	240.5	243.4	244.9
4×10^{-1}	83.0	84 0	85 1	86.1	86.6
6×10^{-1}	45.2	45.8	46.3	46.9	47.2
8×10^{-1}	29.35	29.71	30.1	30.4	30 6
1	21.0	21.26	21.5	21.78	21.9
10	0.664	0.672	0.68	0.689	0.693

of them. It is also not possible to simplify the calculations by sepearately studying the individual effects from each of these factors, as in a practical situation all the different factors will give a net resultant effect.

(i) The deformation of the drop modifies the Lamb's expression for frequency and the resulting expression can be used to explain the experimental results of highly deformed drops. Sch saw and Boumeister[15] 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 independeut of temperature. Observed results were lower than Lamb's by about 10 to 15% and this can easily be explained with the help of the equation

$$\sigma^2 = \frac{l\,(l-1)\,(l+1)\,(l+2)\,T}{\hat{\rho}\,a^3\,[l+1+(l/\gamma)]}\left(1 - \frac{\epsilon_1}{4}\right) = \sigma_L^2\left(1 - \frac{\epsilon_1}{4}\right)$$

If the drop shape in their experiments is assumed to be cblate spheroidal of the form $r = 1 + \epsilon_1 P_2$, then $\epsilon_1 = 0.8$ to 1.3. Then

$$\sigma/\sigma_L = 0.837 \text{ for } \epsilon_1 = 1.3$$

$$= 0.9 \text{ if } \epsilon_1 = 0.8$$

Hence for $\epsilon_1 = 0.8$ to 1.3, $\sigma/\sigma_L \approx 0.9$ to 0.84. To a fair degree, this agrees well with the observational frequencies, although strictly one cannot apply the present calculations for such large ϵ_1.

(ii) 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, then aperiodic damping occurs. In order to study these damping modes, the perturbation was expressed in the form

$$r = 1 + \epsilon\, P_l(\xi) \exp(-\sigma t)$$

Then the characteristic equation obtained can be studied in the three limiting cases—drop in free space, bubble in a dense viscous liquid and a fluid drop in a similar medium. First let us consider the case $\sigma' >> 1$. The case of the aperiodic damping modes of a drop in free space has been studied by Chandrasekhar[9] and Reid[10]. The principal conclusion that can be drawn from Figure 1 is that there are two modes of aperiodic decay. That with the smaller decay constant usually predominates, because it requires smaller energy. In addition to the lowest modes of aperiodic decay, which has been described above, there are an infinity of other higher order modes. These have larger damping constants and can be derived from the characteristic equation by considering later intervals of $\hat{h}$. Considering the first interval only, it is evident from the figure 1 that the characteristic equation allows aperiodic modes so long as $\sigma_L a^2/\hat{\nu}$ is less than a certain maximum value. And for larger values, a different type of damping namely oscillatory damping results. For the principal mode $l = 2$, the critical point is $a_c \approx 2.34 \times 10^{-6}$ cm for a system of water drop in air. Similar aperiodic damping modes have been studied for the cases of a bubble in a viscous liquid and a liquid drop in a liquid medium. For drops with sizes smaller than this critical size, oscillations cannot occur. This may very well account for the nonobservance of oscillations in some viscous systems[12, 16], although such an aperiodic damping mode has not been explicitly observed. Very similar conclusions about the critical size can easily be drawn for the case $\sigma' \sim 1$. The magnitude of critical size does not differ much from that of the previous case. If the drop is deformed into an oblate spheroid of the form $r = 1 + \epsilon_1 P_2(\xi)$, then the critical size for the two modes will be approximately $(C \hat{\rho}\, \hat{\nu}^2/T) \times (1 - 3\epsilon_1/4)$.

(iii) 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 Lamb's result is small for the asymptotic cases of a drop oscillating in free space and a bubble in a dense viscous liquid. But it acquires large values for systems of similar fluids. Even in this case, as shown in Fig. 2, the splitting is small for drops with extreme sizes like very small and very large drops. But the splitting is very pronounced for drops of intermediate sizes in similar fluid media.

The higher frequency mode larger than Lamb's by about 20 to 30% has an indirect evidence in the experimental results of Valentine *et al.*[17]. They observed oscillation frequencies larger than Lamb's result by abount 30-40% in rather special circumstances. Small drops of cyclohexanol were made to coalesce with a solution of benzene and carbon tetra chloride reducing the interfacial tension by about 10-20 dyne cm^{-1}. Frequencies of benzene-carbon tetra chloride drop before and after coalescene with cyclohexanol were obtained. The frequeneies after coalescence were about 30% larger than the values calculated from Lamb's equation. This would happen if the higher frequency mode of these drops were resonantly excited by the natural frequency of the smaller drops. An approximate analysis, using the parameters of these experiments, shows that such a type of excitation of the higher mode is quite possible. In these experiments of Valentine *et al.*[4] the 73% benzene and 27% carbon tetrachloride drop (A) had a density of 1 $gm\ cm^{-3}$ and an interfacial tension of 35 dynes cm^{-1} with respect to water. Using these values we can compute the Lamb's frequeney of oscillation for the prolate-oblate mode ($l=2$) by using

$$\sigma_{AL}^2 = (8\, T_A/\rho_A\, a_A^3)$$

The higher mode has a frequency larger than Lamb's and can approximately be written as $\sigma_{AH} \approx \sigma_{AL} \cdot K$ where $K \approx 5/4$. The 80% cyclohexanol drop (cyclohexanol : density = 0.945 gm/cc, interfacial tension with respect to water = 3.92 $dyne/cm$) possesses a natural oscillation frequency σ_{BL} given by

$$\sigma_{BL}^2 = k'^2\ (8\, T_B/(\rho_B\, a_B^3)$$

where $k' \approx 9/10$ corrects the Lamb's frequency for viscous effects. For the volumes of the drops used (A – 0.25 to 0.75 cc, B – 0.005 to 0.013 cc) we can estimate $\sigma_{AH} / \sigma_{BL}$.

$$\frac{\sigma_{AH}^2}{\sigma_{BL}^2} = \left(\frac{K^2}{k'^2}\right)\left(\frac{T_A}{T_B}\right)\left(\frac{\rho_B}{\rho_A}\right)\left(\frac{V_B}{V_A}\right) \approx 0{\cdot}4 \text{ to } 1{\cdot}3.$$

Hence the higher mode frequency of the A drop matches with the natural frequency of the B drop for a number of drops used in the experiments. This plausible explanation for the higher frequencies observed by Valentine *et al.* provides an indirect justification for the predicted higher mode of oscillation. However it is highly desirable to test this prediction explicitly.

The lower frequency mode (lower than Lamb's by about 10-15%) requires comparatively smaller energy to be excited and hence is usually favoured in practical systems. This mode can be studied either in a general fluid-fluid system or in limiting cases like a drop oscillating in free space, a bubble in a dense viscous liquid and a fluid drop in a similar fluid. The introduction of the shape parameter into these expressions does not significantly alter the results, but the form of the expressions is slightly changed as though a correction term has been introduced. The results were obtained in several limiting cases of an oscillation parameter $\sigma' = \sigma\, a/u$.

If the drop is moving at low Weber number, then $\sigma' >> 1$. In such a case, the characteristic equation [12] will have to be split into real and imaginary parts to find the oscillatory and damping part of the oscillations. The results are very similar to those obtained in the case of a stationary drop[3].

If the drop is moving with a Reynolds number of the order of $\sigma a^2/\nu$ i.e., $\sigma' = 0\,(1)$, then the final characteristic equations determining the frequency are eq. [21] and eq. [26]. The numerical results in this case depart much more from Lamb's values than in the last case. The oscillation frequencies of (i) water drop in air, (ii) o-nitrotoluene drop in water, (iii) m-cresol drop in water and (iv) o-nitrobenzene drop in water have been computed and compared with the experimental results[12, 18, 19, 20] in the Figs. 3, 4, 5 and 6. The absence of a uniform sphericity of the drop and a considerable amplitude of oscillation caused a large scatter of the experimental data. The general trend of the results seem to be in somewhat better agreement with the present calculated values than with Lamb's. A word should be said about this comparison of the theoretical results with the available experimental data. Much of the earlier experimental investigations on the oscillations of the drop lack such details as the shape of the drop as a function of drop size and and also the purity of the systems used. The absence of these details make a real comparison futile. Impurities largely affect the drop oscillations[12]. Recently there has been some reliable experimental data[12] on pure liquid-liquid systems, which can be used as a real check of the analysis. However we first use the earlier results for a qualitative comparison and afterwards make a real quantitative check with the results on pure liquid-liquid systems.

The oscillations of water drops in air have been experimentally studied by both Constan and Calvert[19] and by Lane[18]. The Fig. 3 shows a comparison of the observed results with the present calculated and Lamb's values. The other result of Constan and Calvert on the oscillations of propylene glycol and ethylene glycol drops in gaseous media show that to a first approximation. the frequencies are independent of *Re* and this is well borne out by the present calculations also.

The results of Schroeder and Kintner[16] could not be fully utilised since the results are for nineteen liquid-liquid systems put together. However their conclusion that the frequencies of oscillation are lower than Lamb's by about 16% is well supported from these results. The dependence of the frequency on the amplitude of oscillation is also studied by Schroeder and Kintner and this may also be taken into account.

In the Figs. 4 and 5, the experimental findings of Kaparthi and Licht[20] are plotted against the present calculated values and Lamb's results. For the system of nitrotoluene drop oscillating in water, the results for the case $\sigma' >> 1$, i.e., very small *We* are also plotted. These values agree very well with the experimental findings. Even in the other case of the values calculated for large *Re*, the continuous variation of the shape of the drop and the amplitude of oscillation make the comparison of the calculated and observed results very difficult.

The absence of impurities from the system is very important for a better comparison and in this connection the results of Winnikow and Chao[18] are of special interest, since a critical check can be made on the observed and

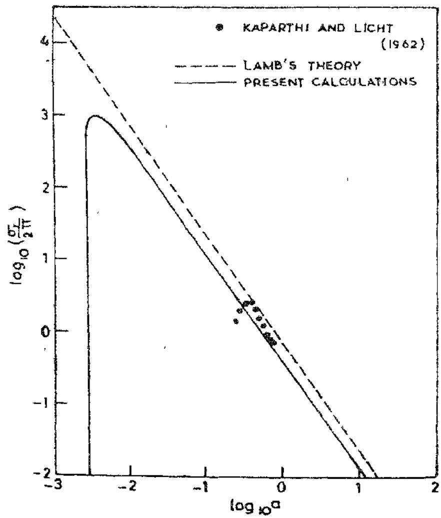

FIG. 5

Frequencies of oscillation of an m-cresol drop moving with its terminal velocity in water.

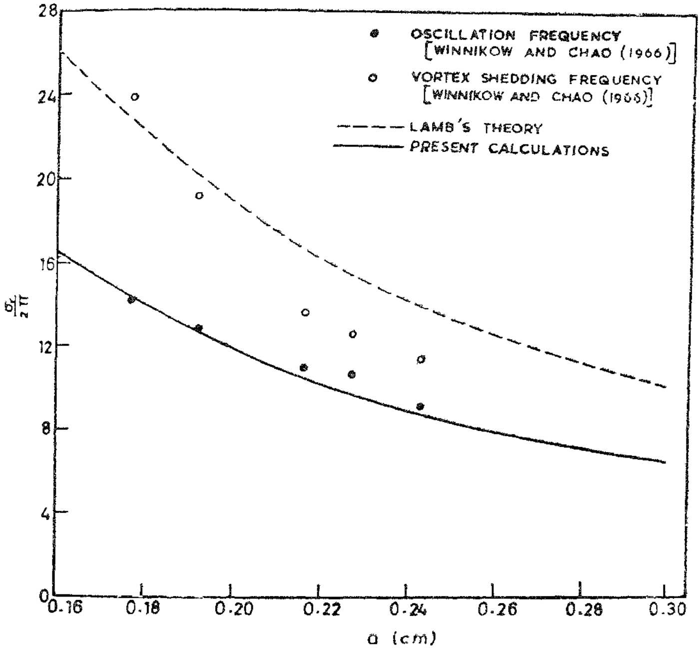

FIG 6

Oscillation frequencies of a pure liquid-liquid system (0-nitrobenzene drop oscillating in water)

calculated results. The observed frequencies and those obtained by the present calculations are compared with the Lamb's result in Fig. 6. It clearly establishes the limited applicability of Lamb's results. Accurate measurements on pure liquid-liquid systems, such as the above one are badly needed for a definite comparison between theory and experiment.

The eddy discharge frequency as measured by Gunn[21] for water drop in air and Winnikow and Chao[12] for a nitrobenzene drop in water is also shown in the Figs. 3 and 6. The intersection of the eddy discharge curve and the oscillation line will result in a resonance which have been noticed by several workers. A striking maximum in the frequency-drop size curve observed by

Kaparthi and Licht[20] is also most probably due to such a process. But if the oscillation of the drop is caused by Vortex discharge, then the problem will have to be treated as a forced oscillation problem and this has not been done so far.

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 caused by the drop motion. We have extended Lamb's analysis by including these effects. As a result most of the discrepancies are qualitatively accounted for and indeed in the one case where very careful measurements on pure systems have qeen performed, there is even a quantitative agreement. Further experimental studies on clearly defined systems would help to elucidate the validity of the present calculations.

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