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The extended Rayleigh theory of the oscillation of liquid droplets

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The Rayleigh theory of oscillation of liquid drops is extended to include the effects of viscosity and a uniform external electric field. The resonant frequencies of the modes of the drop are shown to be shifted by the electric field. The magnitude and sign of the frequency shift depends on the dielectric constant of the drop. The condition for instability of drops in large electric fields is given and found to differ from that given by previous workers. This difference is attributed to the assumption by previous workers that the drops, under the influence of an electric field, distort into ellipsoids of revolution about the field direction. The dynamical equations are derived and the solution for small oscillations is given in an oscillating field and in an amplitude-modulated optical field.

1. Introduction

The first work on the dynamical theory of the oscillation of liquid drops appears to have been done by Lord Rayleigh (1879). In his investigation, Rayleigh considered small distortions of drops from equilibrium under the action of forces due to surface tension alone. Later, Lamb (1932) included the effects of damping of the small oscillations by internal viscous forces and showed that the rate of damping was dependent on the size of the drops, becoming extremely large for very small drops. The possibility of instability of the lower modes of oscillation was first investigated by Rayleigh (1882), who showed that the square of the resonant frequency of the nth mode of a charged drop is

$$\omega_n^2 = \frac{n(n-1)}{\rho a^3}\left[\gamma(n+2) - \frac{Q^2}{4\pi a^3}\right], \tag{1.1}$$

where ρ is the density of the liquid drop, a is the equilibrium radius, γ is the surface tension, and Q is the total charge on the drop. According to Rayleigh, instability occurs when $\omega_n^2 < 0$, or, when $|Q| > [4\pi a^3 \gamma(n+2)]^{\frac{1}{2}}$, the nth mode is unstable. The lower modes become unstable for the least amount of charge. The condition for instability is sometimes written in terms of the electric field at the surface of the drop, $E = Q/a^2$, so that the condition expressed above is frequently given as $|E| > (4\pi(n+2)\gamma/a)^{\frac{1}{2}}$ and has been used as a starting point by later investigators.

Considerably later than the above work, an investigation of the dynamics of small drops was begun by Thacher (1952) and O'Konski & Thacher (1953). They considered the distortion of drops by an electric field, assuming that the distorted drop shape was an ellipsoid of revolution about the direction of the electric field. They also discussed the possibility of enhancing the distortion of the drop by applying an alternating electric field but failed to observe that the frequency of the electric field should be half the natural resonant frequency of the drop. O'Konski & Harris (1957) extended this work to include in the analysis the effect of electrical conductivity of both the drop and the surrounding medium. No consideration was given to the possible effect of conductivity on the damping. In their analysis, they found the rather surprising result that, under certain appropriate choices of conductivities, the drop remained spherical in an applied field. As in their previous investigation, O'Konski & Harris assumed that the droplet, under the action of an electric field, becomes an ellipsoid of revolution about the field direction.

Later, Garton & Krasucki (1964) investigated the stability of bubbles in a static electric field and questioned the correctness of previous theoretical work on electrostriction. They showed actual photographs of bubbles in various stages of disintegration that vividly displayed the physical nature of bubbles breaking.

In a series of papers, Taylor (1964) discussed the stability of conducting drops in an electric field. He showed that it is necessary to introduce motion of the fluid inside a drop to attain the spherical (undistorted) solution of O'Konski & Harris (1957). Some experimental work is reported in Taylor's (1964, 1966) papers, and the results agree quite well with the theory. Sozou (1972) later extended Taylor's theory to include time-dependent electric fields. He gave a number of results in the form of equations with a few cases of numerical results, but further work seems necessary for comparison with experiment and with previous results.

Rosenkilde (1969) investigated the stability of drops in an electric field using methods of tensor calculus and Chandrasekhar's (1961) virial method. He showed that, under appropriate conditions, at least three different configurations can exist. He predicted that a drop could become unstable only if its dielectric constant was greater than 20·801.

Most of the above work is devoted to the development of the theory of equilibrium distortion of drops under the action of various forces. None of these workers attempted to develop their results using the original techniques introduced by Rayleigh (1879). In fact, the technique used by Rayleigh (1882) in his discussion of a charged drop was somewhat different from that used in his first paper. A detailed derivation of Rayleigh's result for a charged drop has been given by Hendricks & Schneider (1963) and later by Schneider (1964), along with many applications of the results. Dissipation by viscosity was not considered in either of these investigations.

In this work, we use the methods of Rayleigh's first paper. In §2, we find the energy due to surface tension, the kinetic energy of the fluid of the drop, and the Rayleigh dissipation function in terms of a set of generalized co-ordinates describing the distortion of the drop. From these quantities, the Lagrangian (as given by Rayleigh) is calculated and the generalized equations of motion are derived, including the dissipation terms. Lamb's result for the decay of oscillating drops is derived from these equations. In §3, we include in the Lagrangian the terms involving the energy due to an external uniform electric field. We discuss this aspect in considerable detail because we have not been able to find any reference to an extension of Rayleigh's result that includes an external field. Section 4 is devoted to a discussion of the results of the previous sections. The results are used to investigate the stability of drops under the action of a static external field as well as the dynamical behaviour of drops under the action of a time-varying field.

2. Equations of motion for free oscillating drops

In the dynamical theory of the oscillation of a liquid drop, we assume that the distance, r, from the centre of mass of the drop to a point on its surface can be expanded in a series of Legendre polynomials, $P_n(\cos\theta)$, as

$$r(\theta,t) = a_0(t) + \sum_n{}' a_n(t)\, P_n(\cos\theta). \tag{2.1}$$

The method of solving a given problem is to express the Lagrangian for that problem in terms of the variables $a_n(t)$ and to treat these $a_n(t)$ as generalized co-ordinates to obtain the equations of motion. The set of functions P_n is complete if we assume that the shape of the drop is symmetric about the z axis. At this point, the z axis can be chosen arbitrarily; later, when we include the electromagnetic energy, the electric field will be assumed to be along this axis. We shall assume symmetry about the z axis throughout the discussion. The prime on the sum in (2.1) indicates that the $n = 0$ term is omitted from the sum.

With the assumption of incompressibility, the constancy of the volume of the drop is a constraint on the $a_n(t)$ in (2.1). If a is the radius of the equilibrium sphere, this assumption leads to

$$a_0 \simeq a\left[1 - \frac{1}{a^2}\sum_n{}' \frac{a_n^2}{2n+1}\right], \tag{2.2}$$

which holds through order a_n^2 with $n \geqslant 1$. Since we are interested only in terms of second order in a_n, we treat (2.2) and subsequent results as equalities.

The potential energy, U_s, of the drop due to surface tension, γ, is the surface area of the drop multiplied by γ, or

$$U_s = 4\pi\gamma\left[a^2 + \sum_n{}' \frac{(n-1)(n+2)\,a_n^2}{2(2n+1)}\right], \tag{2.3}$$

where the constraint given in (2.2) has been used to obtain this result.

To calculate the kinetic energy, T, we need to evaluate the volume integral

$$T = \tfrac{1}{2}\int \rho \mathbf{v}^2\, d\tau, \tag{2.4}$$

where ρ is the density (uniform) and $\mathbf{v}$ is the velocity of the fluid. We assume that the fluid is incompressible and that there are no sources or sinks; under these assumptions, we obtain

$$T = 2\pi\rho a^3 \sum_n{}' \frac{\dot{a}_n^2}{n(2n+1)}. \tag{2.5}$$

This result is valid through terms of order a_n^2. If higher-order terms are retained in the derivation of (2.5), these terms give corrections of the form $\dot{a}_n^2 a_k$, thereby coupling the equations of motion of the a_n in a complicated manner, which we ignore in the present investigation. However, any serious investigation of dynamic instability should include these terms because they are of third order in the a_n and can be important in this and other terms.

To derive the effect of viscosity on the equations of motion, we use Rayleigh's

Radius (μm)	ν_2 (Hz)	τ_2 (s)	ν_4 (Hz)	τ_4 (s)	ν_6 (Hz)	τ_6 (s)
1	3·75(6)†	2·26(−7)	1·08(7)	4·19(−8)	1·88(7)	1·74(−8)
10	1·21(5)	2·26(−5)	3·60(5)	4·19(−6)	6·55(5)	1·74(−6)
100	3·82(3)	2·26(−3)	1·15(4)	4·19(−4)	2·09(4)	1·74(−4)
1000	1·21(2)	2·26(−1)	3·62(2)	4·19(−2)	6·61(2)	1·74(−2)

† The numbers in parentheses give the power of ten.

TABLE 1. Resonant frequencies, $\nu_n = \omega_n/2\pi$, and decay times, τ_n, for drops of various sizes oscillating on the first few modes.

dissipation function, R, as discussed by Goldstein (1950), instead of the procedure used by Lamb (1932). As will become apparent in the development, this method is much more compatible with the treatment presented here. A convenient form of the dissipation function given by Landau & Lifshitz (1959) is

$$R = \tfrac{1}{2}\eta \int (\text{grad}\,\mathbf{v}^2)\,.\,d\boldsymbol{\sigma}, \tag{2.6}$$

where η is the viscosity. As in calculating the kinetic energy, it is sufficient to assume that $d\boldsymbol{\sigma}$ is along $\mathbf{r}$. We obtain

$$R = 4\pi\eta a \sum_n{}' \frac{(n-1)\,a_n^2}{n}. \tag{2.7}$$

The equation of motion for $a_n(t)$ can now be readily found by forming the Lagrangian ($L_1 = T - U_s$) and using Lagrange's equation

$$\frac{d}{dt}\frac{\partial L_1}{\partial \dot{a}_n} - \frac{\partial L_1}{\partial a_n} = -\frac{\partial R}{\partial \dot{a}_n}, \tag{2.8}$$

together with (2.3), (2.5) and (2.7), to give

$$\frac{4\pi\rho a^3\,\ddot{a}_n}{n(2n+1)} + 8\pi\eta a\,\frac{(n-1)}{n}\,\dot{a}_n + \frac{4\pi\gamma(n-1)\,(n+2)}{(2n+1)}\,a_n = 0. \tag{2.9}$$

Thus, if the viscosity vanishes, we have Rayleigh's (1879) result

$$\omega_n^2(\eta = 0) = \frac{\gamma}{\rho a^3}\,n(n-1)\,(n+2). \tag{2.10}$$

If the viscosity is small enough, we obtain Lamb's (1932) result for the decay time of the nth mode

$$\tau_n = \frac{\rho a^2}{\eta(n-1)\,(2n+1)}. \tag{2.11}$$

The presence of viscosity shifts the resonant frequency so that

$$\omega_n^2 = \omega_n^2(\eta = 0) - (1/\tau_n)^2, \tag{2.12}$$

for $\omega_n^2 \geqslant 0$, otherwise, the drop does not oscillate. For the modes $n = 2$, 4, and 6, the frequency $\nu_n = \omega_n/2\pi$ and the decay time τ_n were calculated for drops of radius $a = 1$, 10, 100, and 1000 μm, where we have used $\gamma = 72{\cdot}0$ dynes cm^{-1} and $\eta = 0{\cdot}884 \times 10^{-2}$ P. The results of the calculation are given in table 1.

The above results have been previously derived by various techniques, and we have shown that they can be obtained by methods compatible with Rayleigh's original approach to the problem. Also, as we have shown, our method of obtaining the corrections to Rayleigh's results due to the viscosity are more transparent in this approach.

3. Effect of uniform external electric field

The inclusion of a uniform electric field into the analysis presents a problem of considerable complexity. Therefore, we present the approach and the solution to the problem in more detail than in our previous discussion. To be consistent with the preceding analysis, we need to obtain an expression for the electromagnetic energy of the drop, expressed in terms of the $a_n(t)$ as given in (2.1). This energy is then added to the existing Lagrangian, which can then be used to obtain the equations of motion for the problem.

A convenient form for the electromagnetic energy stored in the drop is given by Jackson (1975):†

$$U_E = -\frac{(\epsilon-1)}{8\pi}\int \mathbf{E}^{\mathrm{i}}.\mathbf{E}\,d\tau, \tag{3.1}$$

where $\mathbf{E}^{\mathrm{i}}$ is the electric field inside the drop, $\mathbf{E}$ is the field in the absence of the drop, and the integral is over the volume of the drop. In (3.1), the dielectric constant of the drop, ϵ, is assumed to be independent of electric field and pressure. Thus, our problem is to find a solution for $\mathbf{E}^{\mathrm{i}}$ before proceeding further.

For a dielectric body, we have from Maxwell's equations (assuming that the fields are slowly varying)

$$\operatorname{div}\operatorname{grad}\psi^{\mathrm{o}} = 0, \quad \operatorname{div}\operatorname{grad}\psi^{\mathrm{i}} = 0, \tag{3.2}$$

where ψ^{o} and ψ^{i} are the electric potentials outside and inside the drop, respectively. The electric fields are given by

$$\mathbf{E}^{\mathrm{o}} = -\operatorname{grad}\psi^{\mathrm{o}}, \quad \mathbf{E}^{\mathrm{i}} = -\operatorname{grad}\psi^{\mathrm{i}}, \tag{3.3}$$

where $\mathbf{E}^{\mathrm{o}}$ and $\mathbf{E}^{\mathrm{i}}$ are the electric fields outside and inside the drop. The appropriate solutions to (3.2) for our problem can be written

$$\psi^{\mathrm{o}} = \sum_{n=0}^{\infty}\frac{A_n}{r^{n+1}}P_n(\cos\theta) - Er\cos\theta, \tag{3.4}$$

for the exterior region and

$$\psi^{\mathrm{i}} = \sum_{n=0}^{\infty} r^n B_n P_n(\cos\theta), \tag{3.5}$$

for the interior region of the drop. The last term in (3.4) represents the potential of the uniform external field.

Before evaluating the coefficients A_n and B_n in the potential, we shall use (3.5) and (3.3) in (3.1) to obtain an expression for U_E in terms of the B_n. This procedure allows us to determine by inspection which terms must be retained in the solution of the boundary-value problem so that U_E is correct through order a_n^2.

The applied electric field can be written as

$$\mathbf{E} = E(\hat{r}\cos\theta - \hat{\theta}\sin\theta), \tag{3.6}$$

† The derivation of (3.1) is not trivial and Jackson gives considerable attention to it.

where $\hat{r}$ and $\hat{\theta}$ are unit vectors in their respective directions. From (3.3) and (3.5), we obtain

$$\mathbf{E}.\mathbf{E}^{\mathrm{i}} = -E \sum_{n=1}^{\infty} nB_n r^{n-1} P_{n-1}(\cos\theta), \tag{3.7}$$

where we have used a recursion formula for the P_n. Substituting (3.7) into (3.1), we have

$$U_E = \frac{(\epsilon-1)}{4} E\left[\frac{2B_1}{3}a^3 + \sum_{n=2}^{\infty} \frac{nB_n}{(n+2)} \int_0^{\pi} r^{n+2} P_{n-1}(\cos\theta)\sin\theta\, d\theta\right], \tag{3.8}$$

where we have separated the term $n = 1$ from the remainder since this term is the integral over θ of r^3, which is proportional to the volume and is contained in the constraining equation, (2.2).

To evaluate the terms in (3.8), we use (2.1) and expand the powers of a_0 through linear terms in a_n, or

$$\int_0^{\pi} r^{n+2} P_{n-1}(\cos\theta)\sin\theta\, d\theta = \frac{2(n+2)}{(2n-1)} a_0^{n+1} a_{n-1}, \tag{3.9}$$

for $n > 1$. The result (3.9) can be used in (3.8) to give

$$U_E = \frac{\epsilon-1}{2} E\left[\frac{B_1}{3}a^3 + \sum_{n=1}^{\infty} \frac{(n+1)}{(2n+1)} B_{n+1} a^{n+2} a_n\right]. \tag{3.10}$$

We have replaced the product $a_n a_0^{n+2}$ by $a_n a^{n+2}$, since the corrections to a_0 are of order a_n^2 and corrections to (3.10) would be of order a_n^3, which are neglected. Also, we have only expanded r through linear terms in a_n, because the B_n in (3.10) for $n > 1$ vanish for undistorted spherical drops. Therefore, to obtain an expression for the energy that is valid to order a_n^2, it is necessary to obtain B_1 to order a_n^2 and B_n for $n > 1$ to order a_n. To proceed, we have to return to the problem of evaluating the A_n and B_n of (3.4) and (3.5).

To evaluate A_n and B_n, we apply the boundary conditions on the fields at the surface of the drop. These are that the tangential components of the electric field are continuous, $E_{\parallel}^{\mathrm{i}} = E_{\parallel}^{\mathrm{o}}$, and that the normal components of the electric displacement are continuous, $D_{\perp}^{\mathrm{i}} = D_{\perp}^{\mathrm{o}}$, where the subscripts $\parallel$ and $\perp$ on the vectors denote tangential and normal components, respectively. The continuity of the tangential electric field can be easily shown to be equivalent to the continuity of the potential, so that $\psi^{\mathrm{i}} = \psi^{\mathrm{o}}$ at the surface. The boundary condition on the normal component of $\mathbf{D}$, however, requires some consideration. Since $\mathbf{D}^{\mathrm{i}} = \epsilon\mathbf{E}^{\mathrm{i}}$ and $\mathbf{D}^{\mathrm{o}} = \mathbf{E}^{\mathrm{o}}$, we have

$$\epsilon\hat{\mathbf{n}}.\mathrm{grad}\,\psi^{\mathrm{i}} = \hat{\mathbf{n}}.\mathrm{grad}\,\psi^{\mathrm{o}}, \tag{3.11}$$

at the surface of the drop, where $\hat{\mathbf{n}}$ is a unit vector normal to the surface and where we have used (3.3). To construct $\hat{\mathbf{n}}$, we note that the vector $d\mathbf{s} = \hat{\mathbf{r}}\,dr + \hat{\theta} r\,d\theta$ lies in the surface if r and θ are related by (2.1). The unit vector $\hat{\boldsymbol{\phi}}$ in the ϕ direction also lies in the surface, since the drop is symmetrical about the z axis. A vector normal to the surface can be formed by the cross product of these two vectors, or

$$\hat{\mathbf{n}} = \frac{d\mathbf{s}\times\hat{\boldsymbol{\phi}}}{|d\mathbf{s}\times\hat{\boldsymbol{\phi}}|} = \frac{\hat{\mathbf{r}} - \hat{\boldsymbol{\theta}}\, r^{-1}dr/d\theta}{[1+(r^{-1}dr/d\theta)^2]^{\frac{1}{2}}}, \tag{3.12}$$

which can be used in (3.11) to obtain the normal derivative. Thus, to reiterate, the boundary conditions are

$$\psi^{\mathrm{o}} = \psi^{\mathrm{i}}, \quad \epsilon\left[\frac{\partial\psi^{\mathrm{i}}}{\partial r} - \frac{1}{r^2}\frac{dr}{d\theta}\frac{\partial\psi^{\mathrm{i}}}{\partial\theta}\right] = \frac{\partial\psi^{\mathrm{o}}}{\partial r} - \frac{1}{r^2}\frac{dr}{d\theta}\frac{\partial\psi^{\mathrm{o}}}{\partial\theta}, \tag{3.13}$$

evaluated at the boundary with r given by (2.1).

To keep track of the order of the corrections, it is convenient to rewrite (2.1) as

$$r = a_0 + \delta \sum_n{}' a_n P_n, \tag{3.14}$$

and expand the various powers of r in powers of δ; we also expand

$$\begin{aligned} A_n &= A_n^{(0)} + \delta A_n^{(1)} + \delta^2 A_n^{(2)} + \ldots, \\ B_n &= B_n^{(0)} + \delta B_n^{(1)} + \delta^2 B_n^{(2)} + \ldots . \end{aligned} \tag{3.15}$$

Then, in the final result, we let $\delta = 1$, since it serves only as an artifice to keep the terms in order. We substitute (3.15) into (3.4) and (3.5) and insert these into (3.13) and equate the coefficients of δ^n on both sides to obtain

$$\left.\begin{aligned} A_n^{(0)} &= B_n^{(0)} = 0, \quad n \neq 1, \\ A_1^{(0)} &= \left(\frac{\epsilon-1}{\epsilon+2}\right) E a_0^3, \\ B_1^{(0)} &= -\frac{3E}{\epsilon+2}, \\ A_n^{(1)} &= \frac{3(\epsilon-1)E}{(\epsilon+2)}\left[\frac{n(n+1)(\epsilon-1)a_{n+1}}{(2n+3)[(\epsilon+1)n+1]} + \frac{n a_{n-1}}{(2n-1)}\right] a_0^{n+1}, \\ B_n^{(1)} &= -\frac{3(\epsilon-1)E}{(\epsilon+2)}\frac{(n+1)(2n+1)a_{n+1}}{(2n+3)[(\epsilon+1)n+1]a_0^n}, \\ B_1^{(2)} &= -\frac{9(\epsilon-1)E}{(\epsilon+2)^2 a_0^2}\sum_m{}' [G_m a_m a_{m-2} + H_m a_m^2], \end{aligned}\right\} \tag{3.16}$$

where

$$G_m = \frac{m(m-1)(2m-1)(\epsilon+2)}{(2m-3)(2m+1)[\epsilon(m-1)+m]}, \tag{3.17}$$

and

$$H_m = \frac{m\{\epsilon(m-1)[12m^3+10m^2-12m-1] - m[12m^3+2m^2-18m+10]\}}{(2m+1)^2(2m-1)(2m+3)[\epsilon(m-1)+m]}. \tag{3.18}$$

When the results given in (3.16) are substituted into (3.10), we get

$$U_E = -\frac{(\epsilon-1)E^2 a}{2(\epsilon+2)}\left[a^2 + \frac{6(\epsilon-1)a}{5(\epsilon+2)}a_2 + \frac{6(\epsilon-1)}{(\epsilon+2)}\sum_{m=1}^{\infty}\{G_{m+2}a_m a_{m+2} + H_m a_m^2\}\right]. \tag{3.19}$$

To form the full Lagrangian for the problem, we need only add U_E to U_s, so that $L = T - U_s - U_E$; the equation of motion for a_n is then given by

$$\begin{aligned} &\frac{4\pi\rho a^3}{n(2n+1)}\ddot{a}_n + 8\pi\eta a\frac{(n-1)}{n}\dot{a}_n + \frac{4\pi\gamma(n-1)(n+2)}{(2n+1)}a_n \\ &\quad = \frac{3(\epsilon-1)^2}{5(\epsilon+2)^2}E^2 a\delta_{n,2} + \frac{3(\epsilon-1)^2E^2 a}{(\epsilon+2)^2}[G_{n+2}a_{n+2} + G_n a_{n-2} + 2H_n a_n], \end{aligned} \tag{3.20}$$

where δ_{ij} is the Kronecker delta and G_n and H_n are defined by (3.17) and (3.18). The right-hand side of (3.20) is new.

Several aspects of (3.20) are as should be expected from the general symmetry of the problem. That is, the right hand side is dependent on the square of the electric field, so that a reversal of the field does not alter the results. Further, the $n = 2$ mode is the only mode driven directly by the electric field, and this mode couples to the $n = 4$ mode and subsequently to higher modes with n even. Therefore, if no further perturbations couple the odd and even modes, only those modes for n even need to be considered. Some of these results would have been immediately obvious if we had used Maxwell's stress tensor to evaluate the forces on the drop, but then the normal mode result of (3.20) would have been lost. If we ignore the coupling to the higher modes in (3.20), the resonant frequency of the nth mode is given by

$$\omega_n^2 = \frac{n(n-1)}{\rho a^3}\left[\gamma(n+2) - \frac{6(\epsilon-1)^2 E^2 a(2n+1)H_n}{4\pi(n-1)(\epsilon+2)^2}\right] - \left(\frac{1}{\tau_n}\right)^2, \tag{3.21}$$

where τ_n is defined by (2.11). This result is similar in appearance to that derived by Rayleigh (1882) for a charged drop (as stated in (1.1)), with $Q = Ea^2$. In Rayleigh's result, the frequency decreases as the field (charge) increases; in (3.21), this condition occurs only when $H_n > 0$. This is true if $\epsilon > 1{\cdot}405$ with $n = 2$, $\epsilon > 1{\cdot}119$ with $n = 4$, $\epsilon > 1{\cdot}070$ with $n = 6$, and $\epsilon > 1$ for $n = \infty$. Perhaps a fair comparison with Rayleigh's result is for a conducting drop, for which the resonant frequency can be obtained from (3.21) by letting the dielectric constant become infinite, or

$$\omega_n^2(\epsilon = \infty) = \frac{n(n-1)}{\rho a^3}\left[\gamma(n+2) - \frac{6E^2 a}{4\pi}\,\frac{n(12n^3+10n^2-12n-1)}{(n-1)(2n+1)(2n-1)(2n+3)}\right] - \left(\frac{1}{\tau_n}\right)^2. \tag{3.22}$$

This result shows the same behaviour as that of Rayleigh, but with a more complicated dependence on the mode number, n.

4. Applications of the Rayleigh formalism

The result given in (3.20) is at this point general in that no assumptions have been made concerning the nature of the electric field other than that the external field is uniform and that it can be derived from a scalar potential. In this section, we discuss the results obtained from (3.20) for three different types of fields; a static electric field, an alternating electric field, and an amplitude modulated high frequency field.

(a) *Static electric field*

In the case of a charged drop under the influence of its self-electric field (Rayleigh's 1882 result), the modes of oscillation are uncoupled, so that a simple criterion, $\omega_n = 0$, can be taken as the onset of instability. In the present case given in (3.20), we see that the modes are coupled, so that the condition for instability created by a large electric field becomes complicated. If we assume that $\ddot{a}_n$ and $\dot{a}_n$ in (3.20) vanish, then we can write

$$D_n x_n = \tfrac{3}{5}\lambda y \delta_{n,2} + 3\lambda y[G_{n+2}x_{n+2} + G_n x_{n-2}], \tag{4.1}$$

ϵ	y_c	y_4	y_6	y_8
1·1	6049	7009	6109	6054
1·3	720·0	817·6	728·2	720·5
1·5	278·1	310·9	280·4	278·2
2·0	82·78	90·16	83·71	82·80
5·0	12·39	12·87	12·42	12·40
10·0	6·362	6·511	6·367	6·362
50·0	3·555	3·603	3·556	3·555
78·2	3·363	3·406	3·364	3·363
100·0	3·291	3·332	3·302	3·291
∞	3·044	3·078	3·044	3·044

TABLE 2. Exact solutions, y_c, and approximate solutions, y_{2N}, of $\Delta(y) = 0$, $y = E^2a/\gamma$.

where $D_n = A_n - 6\lambda y H_n$, $x_n = a_n/a$, $y = E^2a/\gamma$, $\lambda = (\epsilon-1)^2/(\epsilon+2)^2$, and $A_n = 4\pi(n-1)(n+2)/(2n+1)$, where H_n and G_n are given in (3.17) and (3.18). For low fields ($y \sim 0$), the coupling between modes can generally be ignored; for larger fields, on the other hand (which would be required for the drop to become unstable), the coupling cannot be ignored. Thus, to investigate the instability of the system of equations in (4.1), it is necessary to consider the coupled equations in detail.

If we write (4.1) in matrix notation, then

$$\mathbf{T}\mathbf{x} = \mathbf{F}, \tag{4.2}$$

where $T_{n'n} = D_n\delta_{n'n} - 3\lambda G_{n'}\delta_{n',n+2} - 3\lambda G_n\delta_{n',n-2}$, and $\mathbf{x}$ and $\mathbf{F}$ are column matrices (vectors). The vector $\mathbf{F}$ has only one non-zero element, $\frac{4}{5}\lambda y$, for $n = 2$. The solution to (4.2) is given by

$$\mathbf{x} = \mathbf{T}^{-1}\mathbf{F}, \tag{4.3}$$

where $\mathbf{T}^{-1}$ is the inverse of $\mathbf{T}$. The inverse of $\mathbf{T}$ contains the determinant, Δ, of $\mathbf{T}$ in the denominator; when the determinant vanishes, the solution (4.3) becomes unstable. Thus, the lowest value of electric field (smallest positive y) for which the determinant vanishes gives the onset of instability of the drop. Since the $\mathbf{T}$ matrix is tridiagonal, it is a simple matter to obtain a recursion relation for Δ. If the matrix is truncated to contain N terms ($T_{nn'} = 0$, n or $n' > 2N$), then its corresponding determinant, Δ_{2N}, satisfies the recursion formula

$$\Delta_{2N} = D_{2N}\Delta_{2N-2} - (3\lambda y G_{2N})^2\Delta_{2N-4}, \tag{4.4}$$

with $\Delta_0 = 1$ and $\Delta_{2N} = 0$ for negative N. The determinant, Δ, of the original infinite matrix is obtained as the limit as $N \to \infty$ of Δ_{2N}. The result in (4.4) can be used to obtain the smallest positive value of y, y_{2N}, such that $\Delta_{2N}(y_{2N}) = 0$. The asymptotic value of y_{2N} as $N \to \infty$, which we call y_c, is the correct solution to the original problem of finding the lowest positive root of $\Delta = 0$. These roots have been found for a wide range of dielectric constants; the results are given in table 2.

The first two Δ_{2N} and y_{2N} are, from (4.4),

$$\Delta_2(y) = A_2 - 6\lambda y H_2, \quad y_2 = \frac{A_2}{6\lambda H_2}, \tag{4.5}$$

and

$$\left.\begin{aligned} \Delta_4(y) &= (A_4 - 6\lambda y H_4)(A_2 - 6\lambda y H_2) - (3\lambda y G_4)^2, \\ y_4 &= \frac{H_4 A_2 + H_2 A_4 - [(H_4 A_2 - H_2 A_4)^2 + A_2 A_4 G_4^2]^{\frac{1}{2}}}{3\lambda(4H_4 H_2 - G_4^2)}, \end{aligned}\right\} \tag{4.6}$$

which are the simplest roots for which algebraic expressions can be obtained. The sign of the square root in (4.6) is chosen so that y_4 is the smallest positive root in the range of dielectric constants $1 \leqslant \epsilon \leqslant \infty$. The result for y_2 in (4.5) is not physical ($y_2 < 0$) for $\epsilon < 52/37$ and becomes infinite for $\epsilon = 52/37$ (H_2 vanishes) and is, in general, not even a good estimate. This result is not surprising, since y_2 totally ignores coupling between modes. The result for y_4 given in (4.6) is, on the other hand, a good estimate even for small values of ϵ and can be used as an approximate analytic expression for y_c. The values of y_{2N} have been calculated for a few values of N and an extended range of ϵ and are given in table 2. As can be seen, the value of y_4 differs from y_c only by 16 % for $\epsilon = 1{\cdot}1$; for water ($\epsilon = 78{\cdot}2$), y_4 differs from y_c by 1·3 %; for larger ϵ the difference is ~ 1 %.

Upon comparing the results of table 2 with the results obtained by Brazier-Smith *et al.* (1971), we find a discrepancy. The value of $y_c = 2{\cdot}641$ ($E_c(a/\gamma)^{\frac{1}{2}} = 1{\cdot}625$) given by them (along with references to previous work) corresponds to our value of $y_c = 3{\cdot}044$ for $\epsilon = \infty$. This discrepancy is caused possibly by the fact that they assumed that the drop is constrained to be an ellipsoid of revolution. For water, we obtain $y_c = 3{\cdot}363$.

The amplitude of the x_2 mode is easily obtained by using (4.1) along with the determinant in (4.4) to give for the $2N$th approximate solution

$$x_2(2N) = \frac{Q_{2N}}{\Delta_{2N}}, \tag{4.7}$$

where the Q_{2N} obey the same recursion relation as Δ_{2N} given in (4.4), with the initial values

$$Q_2 = 3\lambda y/5, \tag{4.8}$$

and

$$Q_4 = 3\lambda y D_4/5, \tag{4.9}$$

which are sufficient to generate all the Q_{2N} from (4.4). The correct x_2 that satisfies (4.1) is obtained as the limit as $N \to \infty$ of $x_2(2N)$. Having determined the value of x_2, we can generate all the x_n using (4.1) and the condition $x_0 = 0$. The variations of x_2 and x_4 as functions of y are shown in figures 1 and 2 for two values of the dielectric constant, $\epsilon = 78{\cdot}2$ and $\epsilon = \infty$. The amplitudes of the x_n modes are approximately an order of magnitude larger than the x_{n+2} modes, for $y \lesssim 1$. The amplitudes for all the modes diverge at y_c.

When the x_n become large, the above results are invalid. A measure of the validity of the expansion procedure used in this work is the quantity

$$\frac{a_0}{a} = 1 - \sum_{n=1}^{\infty} \frac{x_n^2}{2n+1}. \tag{4.10}$$

When a_0/a becomes small, the entire procedure used here becomes questionable, because all the expansions used in the derivation of (3.20) assume that $x_n \ll 1$.

Many workers (see § 1) have calculated the drop distortion based on the assumption

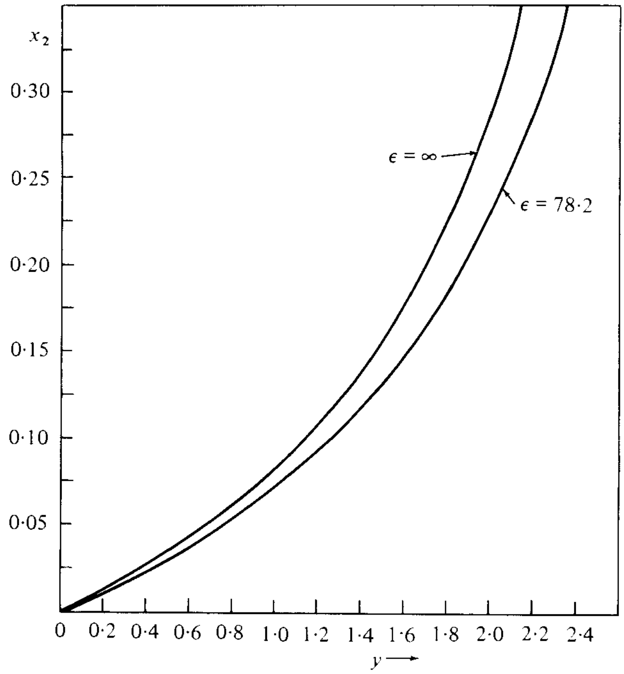

FIGURE 1. Amplitude of the second mode, x_2, as a function of y $(=E^2a/\gamma)$, for $\epsilon = 78{\cdot}2$ and $\epsilon = \infty$.

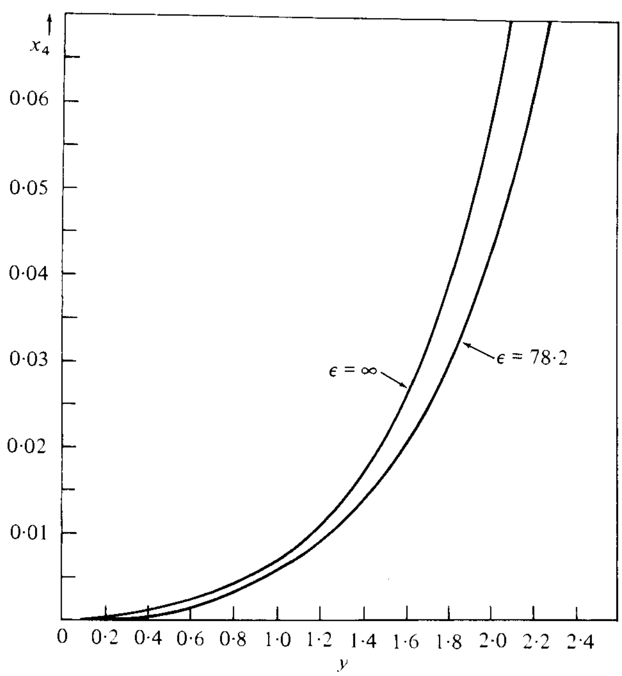

FIGURE 2. Amplitude of the fourth mode, x_4, as a function of y $(=E^2a/\gamma)$, for $\epsilon = 78{\cdot}2$ and $\epsilon = \infty$.

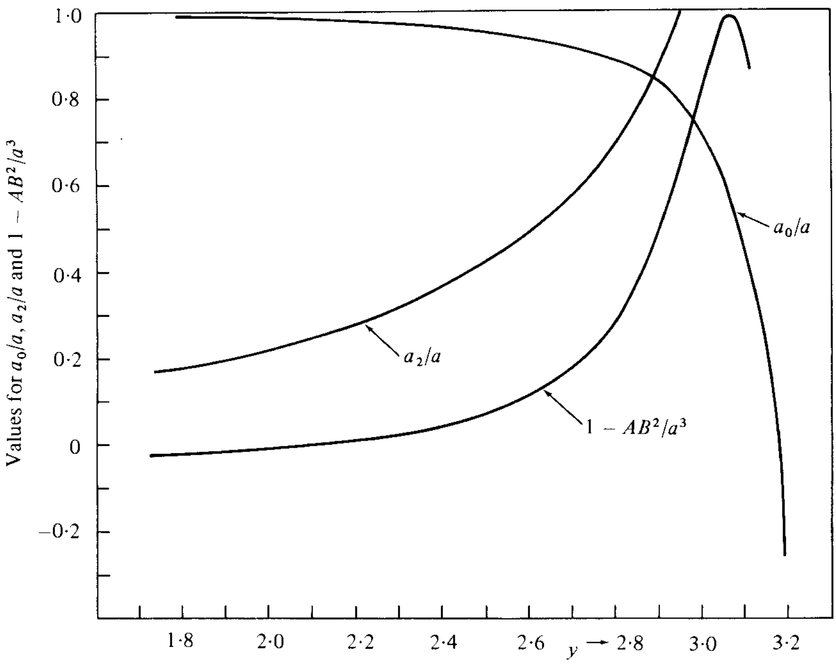

FIGURE 3. The quantities a_0/a, a_2/a, and $1-AB^2/a^3$ obtained from the normal-mode analysis, versus field strength, y.

that the drop distorts into an ellipsoid of revolution. To examine the validity of this assumption, we have calculated from (2.1) the values of the semimajor and semiminor axes, A and B, of the distorted drop. If the drop were truly an ellipsoid, then A and B would satisfy the constraint

$$AB^2 = a^3. \tag{4.11}$$

Therefore the quantity $1-AB^2/a^3$ gives a measure of the deviation of the drop from ellipsoidal shape. The quantities a_2/a, a_0/a, and $1-AB^2/a^3$ are shown in figure 3 for a drop with $\epsilon = \infty$. For small values of y, the latter quantity is quite small, showing that the drop can be well approximated by an ellipse. However, for $y = 2{\cdot}8$, $1-AB^2/a^3 \sim 0{\cdot}28$, and a_0/a is still close to 1 ($\sim 0{\cdot}9$), showing that our expansion procedure is still valid at this value of y. It would seem that the ellipsoidal approximation, when viewed in this light, is not very good and perhaps would indicate the source of the difference in the critical field values obtained here and by other workers.

(*b*) *Dynamical equation for varying electric fields*

The dynamical equation (3.20) is, to our knowledge, a new result in the sense that the losses due to viscosity and the effect of a finite dielectric constant have been included in the analysis. Further, the entire equation of motion was derived in a consistent manner by using Rayleigh's original method. For convenience, we can write (3.20) in the form

$$M_n \ddot{x}_n + M_n \Gamma_n \dot{x}_n + M_n \omega_n^2 x_n = \tfrac{3}{5}\lambda E^2 a \delta_{n,2} + 3\lambda E^2 a[G_{n+2} x_{n+2} + G_n x_{n-2}], \tag{4.12}$$

where $M_n = 4\pi\rho a^3/n(2n+1)$ (note that M_1 is the total mass of the drop), $\Gamma_n = 2\eta(n-1)(2n+1)/\rho a^2$, $\omega_n^2 = \omega_n^2(0) - (2\lambda E^2 a/M_1)n(2n+1)H_n$, and where H_n is given in (3.18) and $\omega_n^2(0)$ is the frequency in the absence of an electric field and damping, as given by (2.10). If the electric field is assumed to be of the form

$$E = E_0 \cos(\omega t), \tag{4.13}$$

then

$$E^2 = \tfrac{1}{2}E_0^2 + \tfrac{1}{2}E_0^2 \cos(2\omega t). \tag{4.14}$$

Thus, the driving force in (4.12) varies at twice the frequency of the electric field, and resonances for small amplitude should occur near $2\omega = \omega_n$.

If we ignore the coupling to higher modes ($G_n = 0$) in (4.12) and replace E^2 by $\frac{1}{2}E_0^2$ in ω_n^2, we have

$$\ddot{x}_2 + \Gamma_2 \dot{x}_2 + \omega_2^2 x_2 = \frac{3\lambda E_0^2 a}{10M_2} + \frac{3\lambda E_0^2 a}{10M_2}\cos(2\omega t). \tag{4.15}$$

where we have used the results of (4.14). The result given in (4.15) is a standard linear equation, and the solution can be written as $x_2(t) = x_2^0 + \xi_2(t)$, where

$$x_2^0 = \frac{3\lambda E_0^2 a}{10M_2\omega_2^2},$$

$$\xi_2(t) = \frac{3\lambda E_0^2 a}{10M_2 R_2}\cos(2\omega t - \theta_2), \tag{4.16}$$

where $R_2 = [(\omega_2^2 - 4\omega^2)^2 + 4\omega^2\Gamma_2^2]^{\frac{1}{2}}$ and $\tan\theta_2 = 2\omega\Gamma_2/(\omega_2^2 - 4\omega^2)$. Also, in the derivation of (4.16), we have ignored terms of higher powers than E_0^2 in the electric field.

Thus, the results given in (4.16) show that, in small electric fields, the equation of motion of the drop in the fundamental mode is similar to that of an ordinary resonant circuit with damping; however, the driving force on the drop has a frequency twice that of the applied electric field. The higher modes are driven by the electric field indirectly by coupling through the lower modes, as shown by (4.12), and approximate solutions can be obtained by a perturbative solution of the equations of motion.

The dynamical equation (4.12) can be used in more complicated situations in which the electric field is of an impulse nature, such as that caused by the passage of charged drops. A number of sources of electrical and mechanical disturbances have been considered by Brook & Lantham (1968) in their study of modulation of radar echo from rainstorms.

A second and perhaps more interesting case of varying fields is the case in which the electric field is an amplitude-modulated high-frequency wave. The carrier may be radar or an infrared laser, and the amplitude modulation can be chosen near a resonant frequency of the drop. Such a field may be represented by

$$E = E_0(1 + m_0\cos(\omega t))\cos(\omega_0 t), \tag{4.17}$$

where $\omega_0 \gg \omega$, m_0 is the modulation index, and the spatial dependence of the wave has been ignored, because we assume that the drop radius is small compared to the wavelength of the carrier, $\lambda_0 = 2\pi c/\omega_0$. We need the square of the field, which is given by

$$E^2 = \tfrac{1}{2}E_0^2[1 + \tfrac{1}{2}m_0^2 + m_0\cos(\omega t) + \tfrac{1}{2}m_0^2\cos(2\omega t)], \tag{4.18}$$

where we have averaged over a time period long compared to a period of oscillation of the carrier. If the modulation index m_0 is small compared to unity, the terms involving m_0^2 can be ignored. Substituting (4.18) into (4.12), we obtain

$$\ddot{x}_n + \Gamma_n \dot{x}_n + \omega_n^2 x_n = \tfrac{3}{10}\lambda E_0^2 a[1 + 2m_0 \cos(\omega t)]\, \delta_{n,2} + \tfrac{3}{2}\lambda E_0^2 a[1 + 2m_0 \cos(\omega t)]\, [G_{n+2} x_{n+2} + G_n x_{n-2}], \quad (4.19)$$

where, as in (4.15), we assume that the term E^2 in ω_n^2 is replaced by $\frac{1}{2}E_0^2$. Also implicit in the equation is the assumption that the dielectric constant, ϵ, wherever it appears, is to be evaluated at the carrier frequency, ω_0. Only in cases of extreme dispersion is this assumption invalid, and such cases would have to be treated quite differently. The solution to (4.19) is the same as in (4.15) for $n = 2$, except that the driving frequency occurring in (4.15) should be reduced by a factor of two.

The time-varying displacement of the drops can be studied (Wortman 1979) by amplitude modulation of a high power laser and observation of the reflected signal of a second low power laser. Some of the analysis given by Brook & Lantham (1968) would also apply to this case, except that the modulation of the reflected signal from the low-power laser would be at the frequency ω of (4.13) rather than a distribution of different frequencies. By observing the reflected signal of the low power laser and sweeping through a range of values of ω, one obtains information regarding the distribution of particles in a fog or cloud.

5. Concluding remarks

We have used the methods of Rayleigh (1879) to derive a consistent theory of the oscillation of liquid drops, including viscosity and electric field effects. In particular, in § 2 we considered the equations of motion for free drops and showed that the results of Lamb (1932) for the decay of drops can be obtained from the Rayleigh formalism. In § 3, the effects of a uniform external field were considered, and we derived equations of motion that coupled together the natural modes of oscillation of the drop. In § 4, we considered the application of the formalism to several problems of interest. For the case of a static applied field, we considered the condition for instability of the drops and showed that the result differed from that obtained previously by other workers. The difference was attributed to the fact that previous work considered the drops as constrained to ellipsoids of revolution, whereas our result was derived with no constraint other than constancy of the drop volume. For the case of an oscillatory applied field, we derived the solution for small amplitude of oscillation and showed that the actual force on the drop oscillated at twice the frequency of the applied field. For the case of an amplitude-modulated wave, we showed that the solution was similar to that for an oscillating field and gave an application of the results to a technique for measuring particle size distributions in fogs or clouds.

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Added note: One of the referees has called to our attention Steven B. Sample's Ph.D. thesis (University of Illinois, 1965, available from University Microfilms, number 65–11,860) in which he addresses the problem of the oscillation of conducting liquid drops by methods similar to those described in the present work. Results for a conducting drop can be obtained from the present results in the limit $\epsilon \to \infty$; however, we have found that the results so obtained disagree with Sample's. The probable reason for this discrepancy is that Sample did not retain all terms to the same order in the drop distortion in a systematic way.

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