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VIBRATIONAL FREQUENCY OF A NON-CONDUCTING CHARGED LIQUID DROP*

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ABSTRACT

We consider the vibration of a non-conducting liquid drop endowed with a surface charge, in supplement to the well-known case of a conducting liquid drop studied by Lord Rayleigh in the last century. It is assumed that there is no charge conduction except by hydrodynamical transport due to the flow motion. It is found that the surface flow is of such a form that the charge again maintains an electrostatic equipotential at the surface of the drop at all instants of the vibrational motion, and the same Rayleigh result is obtained. Thus, the Rayleigh result is applicable to more general classes of liquid drops, irrespective of the conductivity of the liquid in these two limits.

INTRODUCTION

The vibrational frequency of a charged liquid drop under the restoring force of its own surface tension was first derived by Lord Rayleigh (1) about a hundred years ago. The case considered was, in Lord Rayleigh's own words, for "liquid conducting masses charged with electricity". Implicit in the assumption of a conducting mass was that the dielectric relaxation time was much shorter than the mechanical vibrational period so that the charge could always redistribute itself to maintain an equipotential surface at the surface of the drop (see, for example, the re-derivation of Lord Rayleigh's result by Hendricks and Schneider (2)). Recent tests of Rayleigh's relation for a charged water drop indicated good agreement with experimental measurements (3,4).

It is of interest to study the influence of the conductivity of the liquid on the vibrational frequency. In the case of water, conductivity increases with the degree of salinity; one wishes to know whether the vibrational frequency may depend on the degree of purity of water under consideration. As

*Research sponsored by the U. S. Atomic Energy Commission under contract with Union Carbide Corporation.

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the vibrational frequencies have been used to estimate the drop size and degree of electrification of water drops in the atmosphere by means of the backscattering effect on radar (5), such a question is of practical interest. On the other hand, there are also insulating liquids which can be readily charged (6). For these liquids, the dielectric relaxation time can be large compared to the mechanical vibrational period. One would be interested to see if the Rayleigh relation needs modification which may have some bearing on the electrohydrodynamical spraying process studied by many authors (6,7,8).

We consider a non-viscous liquid drop endowed with a surface charge. The charge is assumed to be uniformly distributed when the drop assumes its equilibrium spherical shape. We study the case in which the charge element is not free to move except to follow the motion of the surface element in which it resides. We shall see that this motion of the charge in the liquid drop results again in an equipotential surface at the surface of the drop at any instant of the vibrational motion, even though the charge is not free and moves by hydrodynamical transport only. Thus, surprisingly enough, the vibrational frequency of a charged liquid drop turns out to be independent of the conductivity of the liquid in these two limits!

HYDRODYNAMICAL FLOW AND VIBRATIONAL FREQUENCY

The starting point of the present discussion is the Euler equation for a fluid element in the interior

$$\frac{d\vec{u}}{dt} = -\nabla\left(\frac{\delta p}{\rho_M}\right) \tag{1}$$

where $\vec{u}$ is the velocity characterizing the motion of the fluid element, ρ_M the mass density, δp the deviation of pressure from the equilibrium value. We linearize the Euler equation by assuming the amplitude ϵ_o to be small. A normal mode is specified by describing the sharp surface as

$$r = R_o + \epsilon_o e^{i\omega t} Y_{\ell m}(\theta,\phi) \tag{2}$$

where R_o is the radius of the liquid drop in equilibrium and $Y_{\ell m}(\theta,\phi)$ is a spherical harmonic and ω gives the frequency of the vibrational motion. We further assume that the mass is incompressible and the density is uniform. Thus, from the equation of continuity, we have

$$\nabla\cdot\vec{u} = 0. \tag{3}$$

This condition, together with the solenoidal nature of the restoring force vector leads to the important implication that $\vec{u}$ is purely poloidal (Chandrasekhar (9)) (which means that $\vec{u}$ can be written in the form $\vec{u} = \nabla \times [\nabla \times (\Phi/r)\vec{r}]$, where Φ is a scalar function). From this, the mathematical analysis can be considerably simplified. In spherical coordinates, the various components of $\vec{u}$ (for the poloidal solution) are:

$$u_r = e^{i\omega t} \frac{\ell(\ell+1)}{r^2} U(r)\, Y_{\ell m}(\theta,\phi) \tag{4a}$$

$$u_\theta = e^{i\omega t} \frac{1}{r} \frac{dU(r)}{dr} \frac{\partial Y_{\ell m}(\theta,\phi)}{\partial \theta} \tag{4b}$$

$$u_\phi = e^{i\omega t} \frac{1}{r\sin\theta} \frac{dU(r)}{dr} \frac{\partial Y_{\ell m}}{\partial \phi} \tag{4c}$$

where U(r) is some function to be determined by the boundary conditions.

Taking the divergence of Eq. (1), we have, from Eq. (3),

$$\nabla^2 \left(\frac{\delta p}{\rho_M}\right) = 0. \tag{5}$$

A solution to this Laplace equation satisfying the boundary condition at the origin is

$$\delta p/\rho_M = \varepsilon_o e^{i\omega t}(\ell+1)\, \Pi_o r^\ell Y_{\ell m}(\theta,\phi) \tag{6}$$

where Π_o is a constant to be determined.

Replacing each side of Eq. (1) by its defining function as in Eqs. (4) and (6), we have

$$i\omega\, U(r) = -\varepsilon_o\, \Pi_o\, r^{\ell+1}. \tag{7}$$

Thus, the radial component of the velocity is

$$u_r = -\frac{\varepsilon_o e^{i\omega t}}{i\omega}\, \Pi_o\, \ell(\ell+1) r^{\ell-1}\, Y_{\ell m}(\theta,\phi). \tag{8}$$

There are two boundary conditions to be satisfied. First, from the requirement of consistency between the radial component of the velocity (Eq. (8)) and the form of the boundary (Eq. (2)), we have

$$\omega^2 = \ell(\ell+1)\, \Pi_o R_o^{\ell-1}. \tag{9}$$

The second boundary condition requires the balance of pressure at the boundary as arising from surface tension and the electrostatic stress. For the latter quantity, it is necessary to know how the charge redistributes itself. In a conducting liquid, the charge is free to move to maintain an equipotential at the surface of the drop. This was the case considered by Lord Rayleigh. On the opposite extreme, suppose the conductivity of the liquid is such that the charge cannot move freely, then, a surface charge element must follow the motion of the surface element in which it resides. What will be the instantaneous charge distribution?

It is easy to follow the hydrodynamics of any fluid element. From Eqs. (4), (7) and (9), we know that a fluid element at (r_o,θ_o,ϕ_o) is mapped into (r,θ,ϕ) by

$$\begin{cases} r = r_o + \varepsilon_o e^{i\omega t} \dfrac{r^{\ell-1}}{R_o^{\ell-1}} Y_{\ell m}(\theta,\phi) & (10a) \\ \theta = \theta_o + \varepsilon_o e^{i\omega t} \dfrac{r^{\ell-2}}{R_o^{\ell-1}} \dfrac{\partial Y_{\ell m}(\theta,\phi)}{\partial\theta} & (10b) \\ \phi = \phi_o + \varepsilon_o e^{i\omega t} \dfrac{r^{\ell-2}}{R_o^{\ell-1}} \dfrac{\partial Y_{\ell m}(\theta,\phi)}{\sin\theta\,\partial\phi}. & (10c) \end{cases}$$

A surface element at point (R_o,θ_o,ϕ_o) goes into another surface element at point (R,θ,ϕ) as follows

$$\begin{cases} R = R_o + \varepsilon_o e^{i\omega t} Y_{\ell m}(\theta,\phi) & (11a) \\ \theta = \theta_o + \varepsilon_o e^{i\omega t} (\partial Y_{\ell m}/\partial\theta)/\ell R_o & (11b) \\ \phi = \phi_o + \varepsilon_o e^{i\omega t} (\partial Y_{\ell m}/\partial\phi)/\ell R_o \sin\theta. & (11c) \end{cases}$$

On assumption of no conduction except by mechanical transport via the hydrodynamical flow, the surface charge density ρ_Q is related to the uniform equilibrium density $\rho_Q^{(o)}$ by

$$\rho_Q = \rho_Q^{(o)} \frac{R_o^2}{R^2} \frac{\sin\theta_o}{\sin\theta} \frac{D(\theta_o,\phi_o)}{D(\theta,\phi)} \qquad (12)$$

where $D(\theta_o,\phi_o)/D(\theta,\phi)$ is the Jacobian of the transformation. From Eqs. (11a), (11b) and (11c), we obtain the surface charge distribution at any instant of the vibrational motion

$$\rho_Q = \rho_Q^{(o)} [1 + \varepsilon_o e^{i\omega t}(\ell-1) Y_{\ell m}(\theta,\phi)/R_o]. \qquad (13)$$

This charge distribution is exactly the same if the liquid is a conducting liquid (see Hendricks, et al. (2)). One therefore obtains the important result that whether the liquid is conducting or not, the hydrodynamical transport carries the charge so that the surface of the liquid drop is also a surface of electrostatic equipotential as a result of the transport. In fact, a surface charge in the form of Eq. (13) gives rise to a potential

$$V_c(r) = \frac{kQ}{R_o} \text{ for } r \leq R_o + \varepsilon_o e^{i\omega t} Y_{\ell m} \qquad (14)$$

and $$V_c(r) = \frac{kQ}{r} + kQ\,\varepsilon_o e^{i\omega t}\frac{R_o^{\ell-1}}{r^{\ell+1}}Y_{\ell m} \text{ for } r \geq R_o + \varepsilon_o e^{i\omega t}Y_{\ell m} \tag{15}$$

where k is the Coulomb coupling constant (it equals $(4\pi\varepsilon_o)^{-1}$ in the usual convention) and Q is the total charge. The electric field as approached from the outside is

$$E_r = \frac{kQ}{R_o^2}\left[1 + (\ell-1)\frac{\varepsilon_o e^{i\omega t}}{R_o}Y_{\ell m}\right]. \tag{16}$$

The increment in pressure at the surface due to electrostatic interaction is

$$\frac{1}{2}\rho_Q E_r - \frac{kQ^2}{8\pi R_o^4} = (\ell-1)\frac{kQ^2}{4\pi R^5}\varepsilon_o e^{i\omega t}Y_{\ell m}. \tag{17}$$

Combining the contribution from electrostatic interaction and surface tension (10), we have

$$\left(\frac{\delta p}{\rho_M}\right)_{R_o+\varepsilon_o e^{i\omega t}Y_{\ell m}} = \left[(\ell-1)(\ell+2)\frac{T}{\rho_M R^2} - (\ell-1)\frac{kQ^2}{4\pi\rho_M R^5}\right]\varepsilon_o e^{i\omega t}Y_{\ell m}. \tag{18}$$

From Eqs. (18), (5) and (9), we get the vibrational frequency

$$\omega^2 = \frac{\ell(\ell-1)(\ell+2)T}{\rho_M R^3}\left(1 - \frac{kQ^2}{(\ell+2)4\pi TR^3}\right) \tag{19}.$$

which is just the Rayleigh result.

DISCUSSIONS

We have shown that in the vibration of a charged non-viscous liquid drop, the hydrodynamical transport of the surface charge results again in an equipotential surface at the surface of the drop at any instant. Thus, if the liquid is non-conducting in the sense of having a large dielectric relaxation time, the behavior of the surface charge is the same as in a conducting liquid. One concludes that Rayleigh's result is more general than it was formulated.

Our discussion has been limited to the inviscid case for which a potential flow is possible. The fact that an electrostatic potential is again maintained is probably intimately related to the potential flow of the fluid. Some differences in the vibrational behavior due to differences in conductivity may be expected in the vibration of a <u>viscous</u> charged liquid drop for which the flow becomes rotational and the vorticities reside mostly at the surface. In a

conducting liquid, a charge element is free to move to the surface even though the surface fluid element may flow inside the surface. For a non-conducting liquid, the motion of the charge toward the surface may not be fast enough, and the electrostatic stress is modified as a consequence. As a viscous conducting charged liquid drop follows the Chandrasekhar equation (9,11) based on an instantaneous surface charge redistribution, the behavior of a viscous non-conducting charged liquid drop will deviate from the Chandrasekhar equation. This may be checked experimentally.

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