

Three-dimensional oscillation characteristics of electrostatically deformed drops

By JAMES Q. FENG† AND KENNETH V. BEARD†

Department of Atmospheric Sciences, University of Illinois at Urbana-Champaign,
Urbana, IL 61801, USA

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A three-dimensional asymptotic analysis of the oscillations of electrically charged drops in an external electric field is carried out by means of the multiple-parameter perturbation method. The mathematical framework allows separate treatments of the quiescent deformation due to the electric field and the oscillatory motions caused by other physical factors. Without oscillations, the solution for the quiescent drop shape exhibits a prolate deformation with a slight asymmetry about the drop's equatorial plane. This axisymmetric quiescent deformation of the equilibrium drop shape is shown to modify the oscillation characteristics of axisymmetric as well as asymmetric modes. The expression of the characteristic frequency modification is derived for the oscillation modes, manifesting fine structure in the frequency spectrum so the degeneracy of Rayleigh's normal modes for charged drops is removed in the presence of an electric field. Physical reasoning indicates that the degeneracy of the oscillation modes is associated with the spherical symmetry of the system, so the removal of the degeneracy may be regarded as a consequence of the symmetry breaking caused by the electric field. In addition, the small-amplitude oscillation mode shapes are also modified as a result of the coupling between the oscillatory motions and the electric field as well as the quiescent deformation.

1. Introduction

An electrically charged conducting drop tends to be spherical due to the action of a uniform surface tension. Rayleigh (1882) first calculated the characteristic frequencies for small-amplitude oscillations of a charged drop about the spherical equilibrium shape and established from an energy stability analysis the amount of charge necessary to induce disruption of the drop surface. According to Rayleigh's analysis, we see that for the drop shape perturbation of each normal mode written in terms of the spherical harmonics $Y_{lk}(\theta, \phi)$ (see Appendix A) of the form

$$e^{i\omega_{lk}t} Y_{lk}(\theta, \phi) \quad (-l \leq k \leq l),$$

the square of the characteristic frequency is determined as

$$\omega_{lk}^2 = \frac{l(l-1)}{\rho R^3} \left[\sigma(l+2) - \frac{Q^{*2}}{(4\pi)^2 \epsilon_m R^3} \right], \quad (1.1)$$

where R is the radius of a sphere having the same volume as the drop, ρ the density, σ the uniform surface tension, Q^* the total charge on the drop and ϵ_m the permittivity of the insulating medium surrounding the drop. (The asterisked

† Also affiliated with Cloud and Precipitation Research, Illinois State Water Survey.

variables denote dimensional quantities hereafter.) Equation (1.1) indicates that ω_{lk} is independent of k , suggesting that there is a $2l+1$ spatial degeneracy associated with one characteristic frequency for the oscillations of a charged drop with the spherically symmetric equilibrium shape, i.e. all normal modes of the spherical harmonics of the same degree l with different rank k have the same characteristic frequency. Thus, in a spherical coordinate system with θ and ϕ denoting the meridional and azimuthal angles, the asymmetric modes ($k \neq 0$) have the same characteristic frequencies as the axisymmetric modes ($k = 0$) of the same l . That is why the abbreviation ω_l is usually seen in the literature rather than the notation ω_{lk} . Intuitively, these sorts of mode degeneracies may be considered as a result of the spherical symmetry of the equilibrium shape about which the oscillatory motions are taking place, because there is no physically preferential direction in defining θ and ϕ in a spherically symmetric system.

When an electrostatic field is applied, however, a non-uniform electric stress distribution will elongate the drop surface in the direction of the electric field, so the equilibrium drop shape becomes approximately prolate (Taylor 1964; Brazier-Smith *et al.* 1971). Moreover, as a charged drop is levitated by applying an electrostatic field parallel to the direction of gravity, the presence of gravity and net electric charge will also destroy the mirror symmetry about the drop's equatorial plane because of the different local hydrostatic pressure and charge concentrations on the upper and lower surfaces (Adornato & Brown 1983). A recent study of axisymmetric drop oscillations (Feng & Beard 1990) reveals some modifications in characteristic frequencies and mode shapes of such electrostatically levitated drops. In this paper, we present generalized three-dimensional features of those modifications for the oscillation characteristics of electrostatically deformed drops.

The behaviour of electrified drops is of interest in a variety of applications and is of fundamental scientific significance. For example, electrostatic levitators have been used to investigate physical properties and size distributions of aerosol particles (O'Konski & Thacher 1953; Davis & Ray 1980). In thunderstorm clouds, natural raindrops are subject to external electric stresses and might also be electrostatically levitated (Pruppacher & Klett 1978; Beard, Feng & Chuang 1989). The oscillation frequencies and mode shapes of such strongly electrified drops can affect the radar return signal and thereby be of use in determining drop-size distributions (Rogers 1984). Also, use of electrostatic levitators has been proposed in the containerless processing of materials such as semi-conductor melts (Carruthers 1974).

Most published works about electrified drops treat charge and external electric field effects separately. In the absence of an external electric field, the equilibrium shape of a charged drop is spherical which greatly simplifies the mathematical treatment. As a consequence, the dynamics and instability of a charged drop without any external fields have been studied extensively (Tsamopoulos & Brown 1984; Tsamopoulos, Akylas & Brown 1985; Natarajan & Brown 1987*a*). Studies of the oscillation behaviour of an uncharged drop in an electric field have been mainly based on simplified models by assuming spheroidal deformations (Rosenkilde 1969; Brazier-Smith *et al.* 1971). In the presence of both electric charge and external field, the drop encounters a net electric force. This net electric force may cause the drop to accelerate or be balanced by some other forces such as the hydrostatic pressure arising from gravity, dynamic drag if the drop is moving in another fluid, etc. For the sake of performing closed-form mathematical analysis without losing the basic features of electrified drops, it seems to be simplest to use the model of electrostatically levitated drops in which an overall force balance is achieved as the

hydrostatic pressure offsets the net electric force (Adornato & Brown 1983; Feng & Beard 1990). Another reason for using the present model is that a comparison can be realized between the theoretical results and experimental observations from electrostatic levitators already in use (e.g. Rhim *et al.* 1987).

In general, the problem of motions in an electrified drop is a complicated free-boundary problem even if the assumptions of inviscid flow and a conducting fluid are used, because of nonlinearities arising from inertia, capillarity and the coupling of the surface kinematics to the velocity field, and also the coupling of the electric field to the changing drop shape. In order to make the problem analytically tractable, an asymptotic approach is used based on the assumptions of both small quiescent deformation in the equilibrium shape and small amplitude of oscillatory motions. Since the quiescent deformation and oscillatory motions are caused by essentially independent physical factors, it is appropriate to involve two small parameters in carrying out the asymptotic expansion about a spherical domain. The previous studies of axisymmetric oscillations of uncharged drops in an electric field (Feng 1990) and of electrostatically levitate drops (Feng & Beard 1990) have already demonstrated the usefulness of the multiple-parameter perturbation technique. Its employment in the present analysis of the more intricate three-dimensional dynamics of electrostatically deformed drops is very helpful in delineating causative roles played by different physical factors in drop behaviour.

2. Problem formulation

In this paper, we consider the irrotational, incompressible motion of an electrically conducting drop in a tenuous insulating medium subjected to an externally applied electrostatic field E_0^* . Under the assumption of electrostatic levitation, the external electrostatic field is placed parallel to the direction of gravity. The mathematical problem is made dimensionless in the same way as that employed in our previous work (Feng & Beard 1990) by scaling the radial coordinate (r_0) with R , time (t) with $(\rho R^3/\sigma)^{1/2}$, velocity potential (Φ) with $(\sigma R/\rho)^{1/2}$, as well as stress terms such as pressure and electric stress with σ/R . The dimensionless surface of the drop is described by $r_0 = F(\theta, \phi, t)$, where θ is the meridional angle measured from the axis parallel to the direction of the external electric field and ϕ is the corresponding azimuthal angle in spherical coordinates.

Thus, the velocity potential is governed by the Laplace equation

$$\nabla^2\Phi = 0 \quad (0 \leq r_0 \leq F(\theta, \phi, t)). \tag{2.1}$$

The condition for a finite radial velocity at the centre of the drop is

$$\frac{\partial\Phi}{\partial r_0} \neq \infty \quad (r_0 = 0). \tag{2.2}$$

The kinematic condition and the normal stress balance at the drop surface are

$$\frac{\partial\Phi}{\partial r_0} = \frac{\partial F}{\partial t} + \frac{1}{r_0^2} \frac{\partial\Phi}{\partial\theta} \frac{\partial F}{\partial\theta} + \frac{1}{r_0^2 \sin^2\theta} \frac{\partial\Phi}{\partial\phi} \frac{\partial F}{\partial\phi} \quad (r_0 = F(\theta, \phi, t)) \tag{2.3}$$

and

$$\nabla_s \cdot \mathbf{n} = \Delta p_0 - \frac{\partial\Phi}{\partial t} - \frac{1}{2} \left[\left(\frac{\partial\Phi}{\partial r_0} \right)^2 + \left(\frac{1}{r_0} \frac{\partial\Phi}{\partial\theta} \right)^2 + \left(\frac{1}{r_0 \sin\theta} \frac{\partial\Phi}{\partial\phi} \right)^2 \right] - B\sigma r_0 \cos\theta + N_E \tag{2.4}$$

$(r_0 = F(\theta, \phi, t)),$

where ∇_s in the term $\nabla_s \cdot \mathbf{n}$ is a surface gradient operator defined in the plane locally tangent to the drop surface (Weatherburn 1927). The unit normal vector of the surface \mathbf{n} is given by

$$\mathbf{n} = \frac{F\mathbf{e}_r - (\partial F/\partial\theta)\mathbf{e}_\theta - (\operatorname{cosec}\theta\partial F/\partial\phi)\mathbf{e}_\phi}{[F^2 + (\partial F/\partial\theta)^2 + (\operatorname{cosec}\theta\partial F/\partial\phi)^2]^{\frac{1}{2}}}. \tag{2.5}$$

The Bond number is defined as $Bo = \rho g R^2/\sigma$ with g denoting the acceleration of gravity. The term N_E is the dimensionless counterpart of the electric stress of the form $\frac{1}{2}\epsilon_m E^{*2}$ (Landau & Lifshitz 1959), where ϵ_m is the permittivity of the medium surrounding the drop in SI units. If the electric potential (V) is scaled with $(\sigma R/\epsilon_m)^{\frac{1}{2}}$, electric field ($E = -\nabla V$) with $[\sigma/(R\epsilon_m)]^{\frac{1}{2}}$ and net electric charge (Q) with $(\epsilon_m \sigma R^3)^{\frac{1}{2}}$, the equation governing the electric potential around a conducting drop is Laplace equation

$$\nabla^2 V = 0 \quad (F(\theta, \phi, t) \leq r_0 < \infty). \tag{2.6}$$

The far-field condition of uniform electric field along the vertical axis is written as

$$V = -E_0 r_0 \cos\theta \quad (r_0 \rightarrow \infty). \tag{2.7}$$

The continuity of the tangential component of the electric field across the interface is guaranteed by

$$\mathbf{n} \times \nabla V = \mathbf{0} \quad (r_0 = F(\theta, \phi, t)). \tag{2.8}$$

The conservation of electric charge in a conducting drop requires

$$\int_0^{2\pi} \int_0^\pi (\mathbf{n} \cdot \nabla V)_{r_0=F} \left[F^2 + \left(\frac{\partial F}{\partial\theta}\right)^2 + \left(\frac{1}{\sin\theta} \frac{\partial F}{\partial\phi}\right)^2 \right]^{\frac{1}{2}} F \sin\theta \, d\theta \, d\phi = -Q. \tag{2.9}$$

The electric stress can be written in terms of the electric potential V as

$$N_E = \frac{1}{2} \left[\left(\frac{\partial V}{\partial r_0}\right)^2 + \left(\frac{1}{r_0} \frac{\partial V}{\partial\theta}\right)^2 + \left(\frac{1}{r_0 \sin\theta} \frac{\partial V}{\partial\phi}\right)^2 \right]_{r_0=F(\theta, \phi, t)}. \tag{2.10}$$

Moreover, the requirement of volume conservation takes the form

$$\int_0^{2\pi} \int_0^\pi F^3(\theta, \phi, t) \sin\theta \, d\theta \, d\phi = 4\pi \tag{2.11}$$

and the constraint that the centre of mass of the drop remains at the origin is

$$\int_0^{2\pi} \int_0^\pi F^4(\theta, \phi, t) Y_{1k}(\theta, \phi) \sin\theta \, d\theta \, d\phi = 0. \tag{2.12}$$

It might be noted that the condition (2.12) also guarantees an overall force balance on the drop.

3. Perturbation approach

In order to make the complicated free-boundary problem (2.1)–(2.12) mathematically tractable, the method of multiple-parameter perturbations (Feng 1990) is used to account for two independent shape deformation characteristics of oscillating

drops with electrostatically deformed equilibrium shapes. Formally, two small parameters ϵ_1 and ϵ_2 are introduced with $\epsilon_1 = E_0$ scaling the externally applied electrostatic field and ϵ_2 scaling the amplitude of the oscillatory motions. Based on the assumption of a nearly spherical drop shape, the expansion of the domain shape is implemented by transforming the complex configuration of the drop shape to the unit sphere using the change of coordinates $r_0 \equiv rF(\theta, \phi, t)$ (cf. Joseph 1967, 1973; Tsamopoulos & Brown 1983) and expanding each dependent variable in a Taylor series. Thus, the nonlinear mathematical problem is transformed into a sequence of linear, inhomogeneous problems at each order of ϵ_1 and ϵ_2 for

$$f^{(n,m)}(r, \theta, \phi, t) \equiv \left[\frac{\partial^{n+m} f(r, \theta, \phi, t; \epsilon_1, \epsilon_2)}{\partial \epsilon_1^n \partial \epsilon_2^m} \right], \quad \epsilon_1 = \epsilon_2 = 0, \tag{3.1}$$

where $f(r, \theta, \phi, t; \epsilon_1, \epsilon_2)$ stands for Φ, V, F , etc. (Feng & Beard 1990). Since the expansions of the governing field equations (2.1) and (2.6) lead to

$$\nabla^2 \Phi^{(n,m)} = 0 \quad (0 \leq r \leq 1) \quad \text{and} \quad \nabla^2 V^{(n,m)} = 0 \quad (1 \leq r < \infty), \tag{3.2}$$

the solutions for the velocity and electric potentials satisfying the natural boundary conditions (2.2) and (2.7) may be written as

$$\begin{bmatrix} \Phi^{(n,m)} \\ V^{(n,m)} \end{bmatrix} = \sum_{l=0}^{\infty} \sum_{k=-l}^l \left[\frac{r^l \beta_{lk}^{(n,m)}}{r^{-l-1} \xi_{lk}^{(n,m)}} \right] Y_{lk}(\theta, \phi) + \begin{bmatrix} 0 \\ -\delta_{n1} \delta_{m0} r \cos \theta \end{bmatrix}, \tag{3.3}$$

where $Y_{lk}(\theta, \phi)$ are the spherical harmonics of the form defined in Appendix A and δ_{nm} denotes the Kronecker delta. For convenience, it is also natural to have the shape function expanded in terms of the spherical harmonics

$$F^{(n,m)} = \sum_{l=0}^{\infty} \sum_{k=-l}^l \alpha_{lk}^{(n,m)} Y_{lk}(\theta, \phi). \tag{3.4}$$

The solution of the equations governing the zeroth-order problem is the base state

$$\begin{bmatrix} F^{(0,0)} \\ \Phi^{(0,0)} \\ V^{(0,0)} \\ \Delta p_0^{(0,0)} \\ B_0^{(0,0)} \end{bmatrix} = \begin{bmatrix} 1 \\ 0 \\ \frac{Q}{4\pi r} \\ 2 - \frac{1}{2} \left(\frac{Q}{4\pi} \right)^2 \\ 0 \end{bmatrix}, \tag{3.5}$$

which, since $\epsilon_1 = \epsilon_2 = 0$, describes an isolated, static spherical drop bearing net charge Q .

Furthermore, to account for the nonlinear dependence of the oscillation frequencies on the deformation amplitudes, a method of multiple timescales is also employed. Formally, the different timescales are introduced into the dynamic equations by expanding the partial derivative with respect to time t as

$$\frac{\partial}{\partial t} \equiv \sum_{n=0}^{\infty} \sum_{m=0}^{\infty} \epsilon_1^n \epsilon_2^m \frac{\partial}{\partial T_{(n,m)}}, \tag{3.6}$$

where the timescales $T_{(n,m)}$ are assumed to be related to the actual time as $T_{(n,m)} \equiv \epsilon_1^n \epsilon_2^m t$.

4. Quiescent drop shapes

Under the influence of an external electric field, the quiescent drop shape is described by the perturbation solutions when the amplitude of oscillatory motions is set to zero ($\epsilon_2 = 0$). Details discussion of the physical mechanism in quiescent deformations can be found in our previous work (Feng & Beard 1990). Hence, only the solution forms written in the notation used in this paper are listed here.

The equations governing the $O(\epsilon_1)$ problem yield the results

$$\begin{bmatrix} F^{(1,0)} \\ V^{(1,0)} \\ \Delta p_0^{(1,0)} \\ Bo^{(1,0)} \end{bmatrix} = \begin{bmatrix} 0 \\ -(r - 1/r^2) \cos \theta \\ 0 \\ \frac{3Q}{4\pi} \end{bmatrix} \tag{4.1}$$

which describe a charged conducting sphere levitated by an externally applied uniform electric field. The overall electric force exactly balances the weight of the sphere, as indicated by the relation $Bo^{(1,0)} = 3Q/(4\pi)$. There is no drop deformation at this order of expansion.

Carrying out the calculations to the $O(\epsilon_1^2)$ problem leads to

$$\begin{bmatrix} F^{(2,0)} \\ V^{(2,0)} \\ \Delta p_0^{(2,0)} \\ Bo^{(2,0)} \end{bmatrix} = \begin{bmatrix} \alpha_{20}^{(2,0)} Y_{20} \\ \frac{Q}{4\pi r^3} \alpha_{20}^{(2,0)} Y_{20} \\ -3 \\ 0 \end{bmatrix}. \tag{4.2}$$

where

$$\alpha_{20}^{(2,0)} = \left(\frac{4\pi}{5}\right)^{\frac{1}{2}} \frac{3}{2[1 - (Q/Q_c^{(2)})^2]} \quad \text{with} \quad Q_c^{(l)^2} = (4\pi)^2(l+2) \tag{4.3}$$

in which $Q_c^{(l)}$ denotes the dimensionless form of Rayleigh’s critical values of charge.

At this level of expansion, the quiescent electrostatical deformation occurs approximately as a prolate spheroid with its major axis along the direction of the applied electric field. As shown in previous works (Adornato & Brown 1983; Feng & Beard 1990), there will appear a third Legendre function in the solution to the $O(\epsilon_1^3)$ problem, indicating that the mirror symmetry of the interface about its equatorial plane is also broken. The quiescent deformation is enhanced as the amount of net charge Q increases, because of the overwhelming effect of the diminished denominators of $\alpha_{20}^{(2,0)}$ and $\alpha_{30}^{(3,0)}$. However, in the absence of the net electric charge Q , the governing equations for the $O(\epsilon_1^3)$ problem render the solutions trivial, i.e. $\alpha_{30}^{(3,0)} = 0$. For the uncharged drop shape further corrections to the above well-known results should be sought from the $O(\epsilon_1^4)$ problem which admit the non-zero coefficients for the shape function

$$\alpha_{00}^{(4,0)} = -\frac{27}{10}(4\pi)^{\frac{1}{2}}, \quad \alpha_{20}^{(4,0)} = \frac{2133}{70}\left(\frac{4\pi}{5}\right)^{\frac{1}{2}}, \quad \alpha_{40}^{(4,0)} = \frac{351}{35}\left(\frac{4\pi}{9}\right)^{\frac{1}{2}}. \tag{4.4}$$

The results of (4.2) and (4.4) yield an expression for the equilibrium shape of an uncharged drop in an electric field

$$F \approx 1 - \epsilon_1^4 \frac{9}{80} + \epsilon_1^2 \left(\frac{3}{4} + \frac{711}{560} \epsilon_1^2\right) \left(\frac{4\pi}{5}\right)^{\frac{1}{2}} Y_{20} + \epsilon_1^4 \frac{117}{280} \left(\frac{4\pi}{9}\right)^{\frac{1}{2}} Y_{40}. \tag{4.5}$$

Actually, the electric field effects on the shape of an uncharged drop were calculated by Taylor (1964) using the spheroidal approximation. His results may be written as

$$F = \frac{(1 - e^2)^{\frac{1}{2}}}{(1 - e^2 \cos^2 \theta)^{\frac{1}{2}}} \cong 1 - \frac{e^4}{45} + \left(\frac{e^2}{3} + \frac{10e^4}{63} \right) \left(\frac{4\pi}{5} \right)^{\frac{1}{2}} Y_{20} + \frac{3e^4}{35} \left(\frac{4\pi}{9} \right)^{\frac{1}{2}} Y_{40}. \tag{4.6}$$

where e^2 denotes the eccentricity of the spheroid. From Taylor's derivation we can obtain the approximate relation

$$e^2 \approx \frac{9}{4} \epsilon_1^2 + \frac{837}{640} \epsilon_1^4. \tag{4.7}$$

Substitution of (4.7) into (4.6) gives the spheroidal result for the shape of an uncharged drop in an electric field

$$F \approx 1 - \epsilon_1^4 \frac{9}{80} + \epsilon_1^2 \left(\frac{3}{4} + \frac{5553}{4480} \epsilon_1^2 \right) \left(\frac{4\pi}{5} \right)^{\frac{1}{2}} Y_{20} + \epsilon_1^4 \frac{343}{560} \left(\frac{4\pi}{9} \right)^{\frac{1}{2}} Y_{40}. \tag{4.8}$$

The comparison of (4.5) and (4.8) shows that to $O(\epsilon_1^2)$ the perturbation and spheroidal models agree perfectly, whereas to $O(\epsilon_1^4)$ there is only a slight difference between the two.

5. Oscillatory motions about the quiescent shape

In this section, we solve the problem of small-amplitude oscillations (first order in ϵ_2) about the equilibrium shape obtained in §4 up to the order of ϵ_1^2 . Solutions to higher-order problems are quite formidable, in spite of the fact that the method of multiple-parameter perturbations provides a systematic way to proceed to any desired order in ϵ_1 and ϵ_2 .

For the $O(\epsilon_2)$ problem, the kinematic condition and the equation of continuity of the tangential electric field lead to

$$l\beta_{ik}^{(0,1)} = \frac{\partial \alpha_{ik}^{(0,1)}}{\partial T_{(0,0)}}, \quad \xi_{ik}^{(0,1)} = \frac{Q}{4\pi} \alpha_{ik}^{(0,1)}. \tag{5.1}$$

Substituting (5.1) into the equation of the normal stress balance leads to a linear oscillator equation for the shape function coefficients $\alpha_{ik}^{(0,1)}$

$$\frac{\partial^2 \alpha_{ik}^{(0,1)}}{\partial T_{(0,0)}^2} + l(l-1)(l+2) \left[1 - \left(\frac{Q}{Q_c^{(l)}} \right)^2 \right] \alpha_{ik}^{(0,1)} = 0 \quad (l \geq 2). \tag{5.2}$$

Thus the solution for the shape function coefficients is found as

$$\alpha_{ik}^{(0,1)} = c_{ik}^{(0,1)} \exp [i\omega_{ik} T_{(0,0)}], \tag{5.3}$$

where $c_{ik}^{(0,1)}$ can be a function of slower timescales such as $T_{(1,0)}$, $T_{(0,1)}$, $T_{(2,0)}$, $T_{(1,1)}$, ..., and

$$\omega_{ik}^2 = l(l-1)(l+2) \left[1 - \left(\frac{Q}{Q_c^{(l)}} \right)^2 \right] \tag{5.4}$$

for $l = 2, 3, 4, \dots$, corresponding to the linear modes of oscillation analysed by Rayleigh (1882). The modes $l = 0$ and $l = 1$ are not included in order to satisfy volume conservation and the condition of fixed centre of mass.

The equation of the normal stress balance also yields

$$\Delta p_0^{\langle 0,1 \rangle} = B_0^{\langle 0,1 \rangle} = 0. \quad (5.5)$$

It is anticipated that if we were carrying out the higher-order expansions in powers of ϵ_2 , the axisymmetric results of Tsamopoulos & Brown (1984) would be recovered for the nonlinear resonant oscillations of inviscid charged drops.

The interactions between the oscillatory motion and the external electric field may appear in the $O(\epsilon_1 \epsilon_2)$ problem solved as follows. By combining the kinematic condition, the equation of continuity of the tangential electric field and the equation of normal stress balance, we obtain an inhomogeneous equation for the shape function coefficients

$$\begin{aligned} \frac{\partial^2 \alpha_{lk}^{\langle 1,1 \rangle}}{\partial T_{(0,0)}^2} + l(l-1)(l+2) \left[1 - \left(\frac{Q}{Q_c^{(l)}} \right)^2 \right] \alpha_{lk}^{\langle 1,1 \rangle} \\ = -2 \frac{\partial^2 \alpha_{lk}^{\langle 0,1 \rangle}}{\partial T_{(1,0)} \partial T_{(0,0)}} + \frac{3Q}{2\pi} [l(l-2) J_{+1}(l-1, k) \alpha_{l-1, k}^{\langle 0,1 \rangle} + l(l-1) J_{-1}(l+1, k) \alpha_{l+1, k}^{\langle 0,1 \rangle}], \end{aligned} \quad (5.6)$$

with the expressions for $J_{-1}(l, k)$ and $J_{+1}(l, k)$ given in Appendix B. The solvability condition, which requires elimination of the secular term in (5.6), leads to

$$\frac{\partial \alpha_{lk}^{\langle 0,1 \rangle}}{\partial T_{(1,0)}} = 0, \quad (5.7)$$

indicating that there is no frequency modification at this level of approximation. Thus the shape coefficients are of the form

$$\alpha_{lk}^{\langle 1,1 \rangle} = A_{-1}^{\langle 1,1 \rangle}(l, k; Q) \alpha_{l-1, k}^{\langle 0,1 \rangle} + A_{+1}^{\langle 1,1 \rangle}(l, k; Q) \alpha_{l+1, k}^{\langle 0,1 \rangle} \quad (l \geq 2), \quad (5.8)$$

with

$$A_{-1}^{\langle 1,1 \rangle}(l, k; Q) = \frac{3Q}{2\pi} \frac{l(l-2) J_{+1}(l-1, k)}{(l-1)(3l+2-Q^2/8\pi^2)},$$

$$A_{+1}^{\langle 1,1 \rangle}(l, k; Q) = -\frac{3Q}{2\pi} \frac{(l-1) J_{-1}(l+1, k)}{(3l+5-Q^2/8\pi^2)}.$$

As discussed in Nayfeh & Mook (1979), the solution to the homogeneous part of (5.6) need not be included because it can always be absorbed in the linear modes (5.3).

From the known shape function coefficients and the kinematic condition as well as the equation of continuity of the tangential electric field, the coefficients for the velocity and electric potentials can be readily determined through the relations

$$l\beta_{lk}^{\langle 1,1 \rangle} = \frac{\partial \alpha_{lk}^{\langle 1,1 \rangle}}{\partial T_{(0,0)}} \quad (5.9)$$

and

$$\xi_{lk}^{\langle 1,1 \rangle} = H_{-1}^{\langle 1,1 \rangle}(l, k; Q) \alpha_{l-1, k}^{\langle 0,1 \rangle} + H_{+1}^{\langle 1,1 \rangle}(l, k; Q) \alpha_{l+1, k}^{\langle 0,1 \rangle}, \quad (5.10)$$

where

$$H_{-1}^{\langle 1,1 \rangle}(l, k; Q) = \frac{Q}{4\pi} A_{-1}^{\langle 1,1 \rangle}(l, k; Q) + 3J_{+1}(l-1, k),$$

$$H_{+1}^{\langle 1,1 \rangle}(l, k; Q) = \frac{Q}{4\pi} A_{+1}^{\langle 1,1 \rangle}(l, k; Q) + 3J_{-1}(l+1, k).$$

At this order of expansion, the modifications to oscillatory mode shapes result from the non-zero Bond number as well as the combination of the external electric

field and the net electric charge, although the equilibrium shape of the drop still exhibits spherical symmetry. Such a phenomenon may be explained as a locally unbalanced electric stress and hydrostatic pressure at the drop surface when it is deformed from the equilibrium spherical shape as oscillatory motions take place.

Furthermore, to satisfy volume conservation and the condition of fixed centre of mass as well as the equation of normal stress balance we should have

$$\alpha_{00}^{\langle 1,1 \rangle} = \alpha_{1k}^{\langle 1,1 \rangle} = \Delta p_0^{\langle 1,1 \rangle} = B_0^{\langle 1,1 \rangle} = 0. \quad (5.11)$$

In order to obtain the first frequency modification, we must extend the analysis to the $O(\epsilon_1^2 \epsilon_2)$ problem. From the equation of conservation of electric charge and the condition of continuity of the tangential electric field on the drop surface we get

$$\xi_{00}^{\langle 2,1 \rangle} = 0$$

and

$$\xi_{lk}^{\langle 2,1 \rangle} = \frac{Q}{4\pi} \alpha_{lk}^{\langle 2,1 \rangle} + H_{-2}^{\langle 2,1 \rangle}(l, k; Q) \alpha_{l-2, k}^{\langle 0,1 \rangle} + H_0^{\langle 2,1 \rangle}(l, k; Q) \alpha_{lk}^{\langle 0,1 \rangle} + H_{+2}^{\langle 2,1 \rangle}(l, k; Q) \alpha_{l+2, k}^{\langle 0,1 \rangle} \quad (l \geq 1), \quad (5.12)$$

with the expressions for $H_{-2}^{\langle 2,1 \rangle}(l, k; Q)$, $H_0^{\langle 2,1 \rangle}(l, k; Q)$ and $H_{+2}^{\langle 2,1 \rangle}(l, k; Q)$ given in Appendix C. The kinematic condition leads to

$$l\beta_{lk}^{\langle 2,1 \rangle} = \frac{\partial \alpha_{lk}^{\langle 2,1 \rangle}}{\partial T_{(0,0)}} + 2 \frac{\partial \alpha_{lk}^{\langle 0,1 \rangle}}{\partial T_{(2,0)}} - B_{-2}^{\langle 2,1 \rangle}(l, k; Q) \frac{\partial \alpha_{l-2, k}^{\langle 0,1 \rangle}}{\partial T_{(0,0)}} - B_0^{\langle 2,1 \rangle}(l, k; Q) \frac{\partial \alpha_{l, k}^{\langle 0,1 \rangle}}{\partial T_{(0,0)}} - B_{+2}^{\langle 2,1 \rangle}(l, k; Q) \frac{\partial \alpha_{l+2, k}^{\langle 0,1 \rangle}}{\partial T_{(0,0)}}, \quad (5.13)$$

with the expressions for $B_{-2}^{\langle 2,1 \rangle}(l, k; Q)$, $B_0^{\langle 2,1 \rangle}(l, k; Q)$ and $B_{+2}^{\langle 2,1 \rangle}(l, k; Q)$ also in Appendix C. Making use of (5.12) and the equation for the normal stress balance yields

$$\begin{aligned} & -\Delta p_0^{\langle 2,1 \rangle} + B_0^{\langle 2,1 \rangle} \left(\frac{4\pi}{3} \right)^{\frac{1}{2}} Y_{10} \\ & + \sum_{l=1}^{\infty} \sum_{k=-l}^l \frac{\partial \beta_{lk}^{\langle 2,1 \rangle}}{\partial T_{(0,0)}} Y_{lk} + \sum_{l=0}^{\infty} \sum_{k=-l}^l (l-1)(l+2) \left[1 - \left(\frac{Q}{Q_c^{(l)}} \right)^2 \right] \alpha_{lk}^{\langle 2,1 \rangle} Y_{lk} \\ & = -2 \sum_{l=2}^{\infty} \sum_{k=-l}^l \frac{1}{l} \frac{\partial^2 \alpha_{lk}^{\langle 0,1 \rangle}}{\partial T_{(0,0)} \partial T_{(2,0)}} Y_{lk} \\ & + \sum_{l=0}^{\infty} \sum_{k=-l}^l [C_{-2}^{\langle 2,1 \rangle}(l, k; Q) \alpha_{l-2, k}^{\langle 0,1 \rangle} + C_0^{\langle 2,1 \rangle}(l, k; Q) \alpha_{lk}^{\langle 0,1 \rangle} + C_{+2}^{\langle 2,1 \rangle}(l, k; Q) \alpha_{l+2, k}^{\langle 0,1 \rangle}] Y_{lk}. \end{aligned} \quad (5.14)$$

where $C_{-2}^{\langle 2,1 \rangle}(l, k; Q)$, $C_0^{\langle 2,1 \rangle}(l, k; Q)$ and $C_{+2}^{\langle 2,1 \rangle}(l, k; Q)$ are explicitly written out in Appendix C. Thus by eliminating $\beta_{lk}^{\langle 2,1 \rangle}$ from (5.13) and (5.14), we obtain an equation for the coefficients of the shape function

$$\begin{aligned} & \frac{\partial^2 \alpha_{lk}^{\langle 2,1 \rangle}}{\partial T_{(0,0)}^2} + l(l-1)(l+2) \left[1 - \left(\frac{Q}{Q_c^{(l)}} \right)^2 \right] \alpha_{lk}^{\langle 2,1 \rangle} \\ & = -4 \frac{\partial^2 \alpha_{lk}^{\langle 0,1 \rangle}}{\partial T_{(0,0)} \partial T_{(2,0)}} + A_{-2}^{\langle 2,1 \rangle}(l, k; Q) \alpha_{l-2, k}^{\langle 0,1 \rangle} + A_0^{\langle 2,1 \rangle}(l, k; Q) \alpha_{lk}^{\langle 0,1 \rangle} + A_{+2}^{\langle 2,1 \rangle}(l, k; Q) \alpha_{l+2, k}^{\langle 0,1 \rangle}, \end{aligned} \quad (5.15)$$

$l \backslash k$	0	1	2	3	4
2	1.382	0.916	-0.482	—	—
3	0.979	0.814	0.321	-0.500	—
4	0.810	0.727	0.479	0.0666	-0.511

TABLE 1. Values of $A_0^{\langle 2,1 \rangle}(l, k; Q)/(4\omega_{lk}^2)$ for $Q = 0$ with various l and k

where

$$A_{-2}^{\langle 2,1 \rangle}(l, k; Q) = lC_{-2}^{\langle 2,1 \rangle}(l, k; Q) - (l-2)(l-3) \left[l - \left(\frac{Q}{4\pi} \right)^2 \right] B_{-2}^{\langle 2,1 \rangle}(l, k; Q),$$

$$A_0^{\langle 2,1 \rangle}(l, k; Q) = lC_0^{\langle 2,1 \rangle}(l, k; Q) - l(l-1) \left[l + 2 - \left(\frac{Q}{4\pi} \right)^2 \right] B_0^{\langle 2,1 \rangle}(l, k; Q),$$

$$A_{+2}^{\langle 2,1 \rangle}(l, k; Q) = lC_{+2}^{\langle 2,1 \rangle}(l, k; Q) - (l+1)(l+2) \left[l + 4 - \left(\frac{Q}{4\pi} \right)^2 \right] B_{+2}^{\langle 2,1 \rangle}(l, k; Q).$$

The solvability condition for (5.15) requires that

$$-4 \frac{\partial^2 \alpha_{lk}^{\langle 0,1 \rangle}}{\partial T_{(0,0)} \partial T_{(2,0)}} + A_0^{\langle 2,1 \rangle}(l, k; Q) \alpha_{lk}^{\langle 0,1 \rangle} = 0. \tag{5.16}$$

Hence at this level of approximation we find

$$\alpha_{lk}^{\langle 0,1 \rangle} = c_{lk}^{\langle 2,1 \rangle} \exp \left\{ i \left[\omega_{lk} T_{(0,0)} - \frac{A_0^{\langle 2,1 \rangle}(l, k; Q)}{4\omega_{lk}} T_{(2,0)} \right] \right\}, \tag{5.17}$$

where $c_{lk}^{\langle 2,1 \rangle}$ could be functions of slower timescales $T_{(0,1)}, T_{(1,1)}, T_{(2,1)}, \dots$. As a result, the characteristic frequencies are modified as

$$\omega_{lk} \left[1 - \frac{A_0^{\langle 2,1 \rangle}(l, k; Q)}{4\omega_{lk}^2} \epsilon_1^2 \right] \tag{5.18}$$

with ω_{lk} being the linear normal mode frequency given by (5.4). For convenient reference, some typical values of the frequency modification factor in (5.18) for $Q = 0$ are listed in table 1. Since the value of $A_0^{\langle 2,1 \rangle}(l, k; Q)$ depends on both l and k , finer structure of the characteristic frequency spectrum can be calculated in the $O(\epsilon_1^2 \epsilon_2)$ problem.

Shown in figure 1 is the normalized frequency $1 - [A_0^{\langle 2,1 \rangle}(l, k; Q)/(4\omega_{lk}^2)] \epsilon_1^2$ as a function of the dimensionless electric field strength $E_0 = \epsilon_1$ when $Q = 0$ for the zonal harmonics (axisymmetric modes $k = 0$) and the sectoral harmonics ($|k| = l$). It is seen that as the external electric field strength increases, all frequencies for the axisymmetric modes are lowered, whereas all sectoral harmonic frequencies are increased. Whether the frequency of a particular mode increases or decreases in the presence of the electric field depends on several factors. The first frequency modification is related to the square of ϵ_1 in (5.18), thus suggesting that the quiescent electrostatic deformation, which also first appears at the level of ϵ_1^2 , could be major cause of the frequency shifts.

From a geometric point of view, the corresponding wavelengths for the zonal harmonics are intimately related to the distance between the upper and lower poles

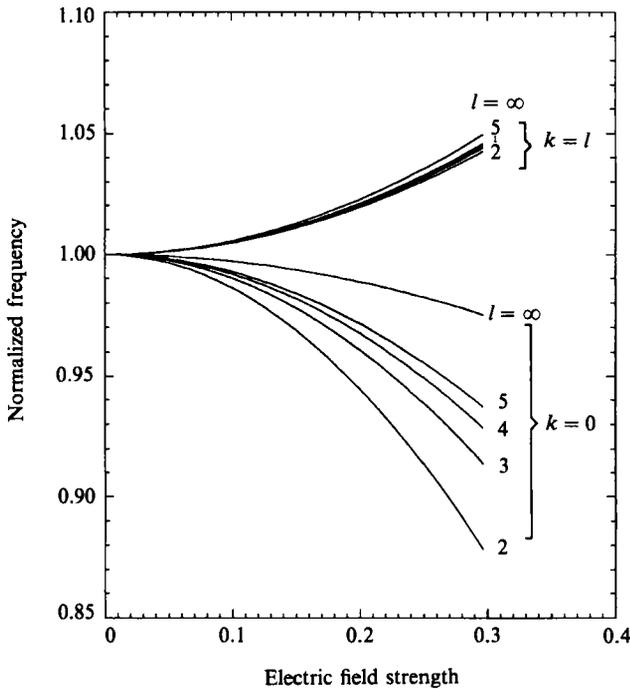


FIGURE 1. The normalized characteristic frequency $1 - [A_0^{(2,1)}(l, k; Q)/(4\omega_k^2)] E_0^2$ of a conducting drop in an electric field with zero net electric charge Q as a function of the dimensionless electric field strength $E_0 = \epsilon_1$ for the zonal and sectoral harmonics.

along a meridian. Physically speaking, increasing the zonal harmonics wavelengths in a prolate spheroid tends to lower the corresponding frequencies. Likewise, the wavelengths for the sectoral harmonics are determined mainly by the size of the equatorial circle, which is contracted for a prolate electrostatical drop so the corresponding frequencies tend to increase. Although the physical problem studied here differs from that of a drop trapped in standing acoustic waves, the geometric consideration in the present analysis seems to be also useful in the explanation of the opposite frequency shifts for axisymmetric drop oscillations when the acoustic driving mode is chosen to be prolate- and oblate-biased (Trinh & Wang 1982). Since the geometric wavelengths for the axisymmetric modes (zonal harmonics) are increased in the prolate-biased drops and decreased in the oblate-biased drops, the corresponding characteristic frequencies should become lower for the former case and higher for the latter case, just as reported by Trinh & Wang for experimental observations of a drop in an acoustic field when it is trapped at a stable position and driven into oscillation with deformed equilibrium shape.

In addition to the geometric effects mentioned above, there is another factor that also obviously affects the characteristic frequencies, viz. a net reduction in the surface restoring force. This is evident from (4.2) which indicates a quiescent decrease in the uniform pressure difference across the interface ($\Delta p_0^{(2,0)} = -3$). Such a softening effect due to the quiescent deformation attempts to cause all characteristic frequencies to decrease. That might be why the net increase of the sectoral harmonic frequencies is relatively insignificant in comparison with the decrease of the zonal harmonic ones.

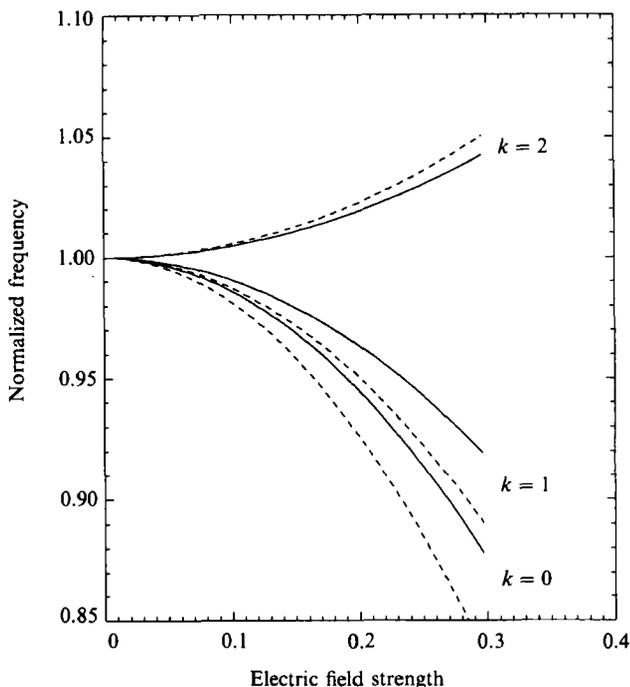


FIGURE 2. The normalized frequencies for two-lobed oscillations ($l = 2$) for the cases $Q = 0$ (solid curves) and $Q = 10$ (dashed curves).

Figures 2, 3 and 4 show the effects of electric charge on the normalized frequency curves for the two-, three- and four-lobed oscillations respectively, with the solid curves for $Q = 0$ and the dashed curves for $Q = 10$. The frequency shifts for the case of $Q = 0$ obtained here for the two-lobed oscillation modes are in general agreement with those of Rosenkilde (1969) who conducted the calculation based on the assumption of spheroidal drop shapes. It is obvious that the net electric charge enhances the frequency shifts for all modes, since it exaggerates the quiescent deformation. The frequency shift enhancements due to the net electric charge, however, differ for the modes of different degree (l) and different rank (k), as can be seen from figure 2, 3 and 4 when the relative difference between the dashed and solid curves for each mode is compared with others.

According to (5.15), the mode shape modifications at this order of expansion can be expressed in the form

$$\alpha_{lk}^{\langle 2,1 \rangle} = \frac{A_{-2}^{\langle 2,1 \rangle}(l, k; Q)}{2[l(3l-4) - (2l-3)(Q/4\pi)^2]} \alpha_{l-2, k}^{\langle 0,1 \rangle} - \frac{A_{+2}^{\langle 2,1 \rangle}(l, k; Q)}{2[(l+2)(3l+2) - (2l+1)(Q/4\pi)^2]} \alpha_{l+2, k}^{\langle 0,1 \rangle} \quad (l \neq 0). \quad (5.19)$$

Since $A_{-2}^{\langle 2,1 \rangle}(l, k; Q)$ and $A_{+2}^{\langle 2,1 \rangle}(l, k; Q)$ in (5.19) do not become zero when $Q = 0$, the mode shape modifications are present even in the absence of net electric charge and gravity, in contrast to the $O(\epsilon_1 \epsilon_2)$ solution. The modification to the shape function in the $O(\epsilon_1^2 \epsilon_2)$ solution is caused mostly by the direct coupling between the oscillatory motions and quiescent electrostatic deformation calculated in the $O(\epsilon_1^2)$ problem, which can exist in the absence of net electric charge Q . There is, however, one thing

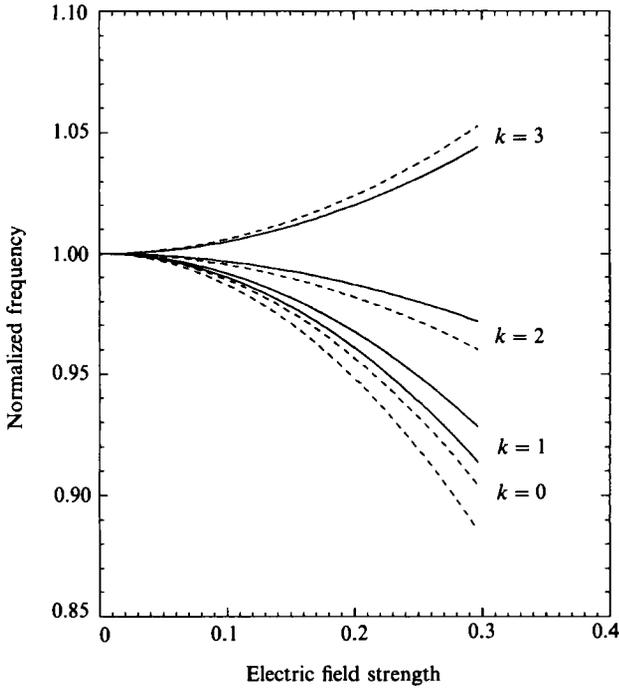


FIGURE 3. The normalized frequencies for three-lobed oscillations ($l = 3$) for the cases $Q = 0$ (solid curves) and $Q = 10$ (dashed curves).

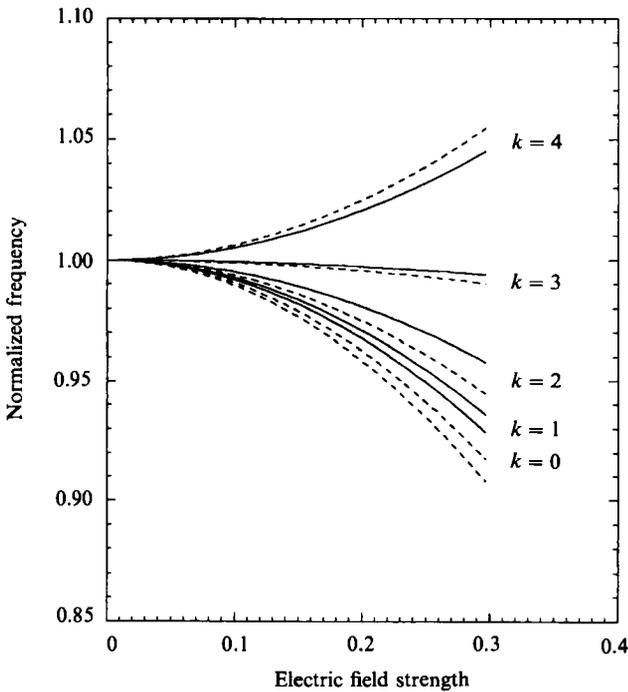


FIGURE 4. The normalized frequencies for four-lobed oscillations ($l = 4$) for the cases $Q = 0$ (solid curves) and $Q = 10$ (dashed curves).

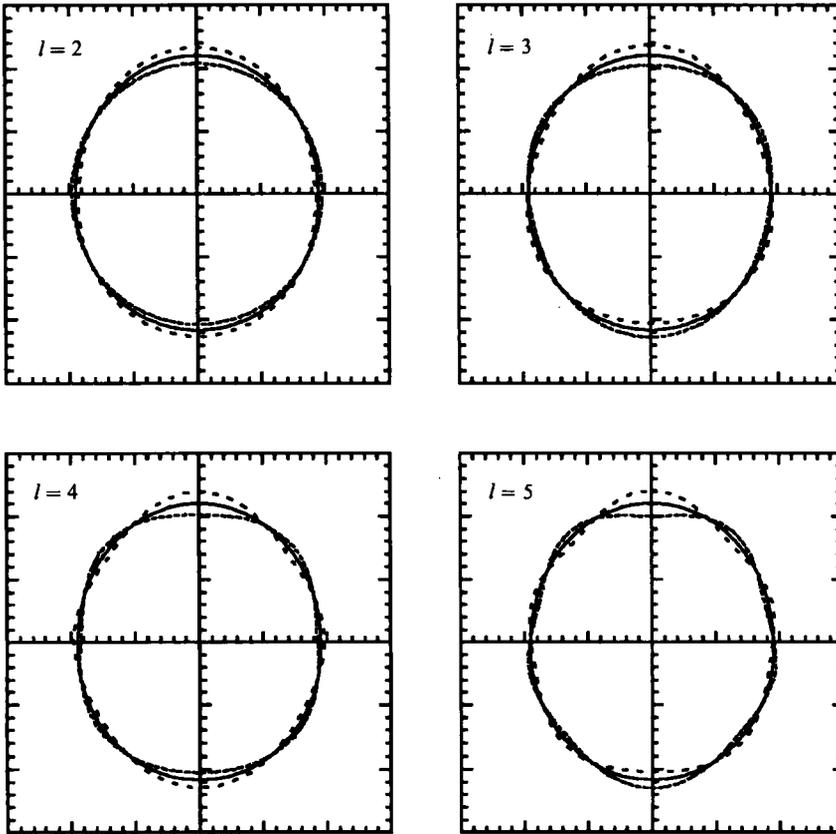


FIGURE 5. Axisymmetric shapes of drop oscillations for $\epsilon_2 = \epsilon_1^2$, $\epsilon_1 = 0.353$ and $Q = 10$. The solid line represents the quiescent drop shape at $t = \frac{1}{4}T_{10}$, the dashed line at $t = 0$, and the dotted line at $t = \frac{1}{2}T_{10}$, with T_{10} being the period of the oscillation mode of degree l and rank $k = 0$.

in common in both (5.8) and (5.19) for the modifications in the oscillation mode shapes; namely there are no modifications in the mode rank (k). This is due to the fact that for each mode the modification uncovered here arises from the coupling between the oscillatory motion and gravity, electric fields and the subsequent quiescent deformation, where the quiescent partners are essentially axisymmetric. Hence, the axisymmetry of the zonal modes is not affected by the shape modifications.

For convenience of illustration, only axisymmetric drop shapes are shown in figure 5 for various oscillatory zonal modes. In order to keep a consistent truncation error for the shape function, a scale relation $\epsilon_2 = \epsilon_1^2$ is used and we retain only the terms up to $O(\epsilon_1^3)$ in the shape function. For each mode, $|c_{lk}^{0,1}| = 1$ is assumed and the parameters are set to $\epsilon_1 = 0.353$ and $Q = 10$ which simulates a water drop of 2.5 mm radius electrostatically levitated in air at the earth's surface. Even when the deformations are not very large, the asymmetric aspect with respect to the equatorial plane can be recognized from figure 5.

6. Concluding remarks

By means of the multiple-parameter perturbation method, fine structure in the characteristic frequency spectrum of the oscillations of electrostatically deformed drops is uncovered through a three-dimensional asymptotic analysis. In the presence of an external electric field, the characteristic frequencies for the modes of the same degree (l) but different rank (k) diverge so the mode degeneracy in Rayleigh's normal modes is removed. This phenomenon may provide some general insight into the relationship of the degenerate characteristics and the symmetries in physical systems. In the present problem, the degeneracy of the oscillation modes is associated with the spherical symmetry of the system where no physical differences can be identified by choosing different coordinate directions. Once this symmetry is destroyed by an external electric field, which obviously gives rise to a specific direction for the system, fine structure in the frequency spectrum emerges whereby the degeneracy disappears. This is similar to the well-known Zeeman effect in quantum mechanics, for which the fine structure in the energy spectrum of hydrogen atoms arises from an external magnetic field that breaks the spherical symmetry.

For the case of drop oscillations, physical considerations suggest that the changes in the geometric lengths of the pole-to-pole meridian and equatorial circle for a prolate electrostatical drop shape are mainly responsible for the lowering of the zonal harmonic frequency and the increase in the sectoral harmonic frequency. Since the mode frequencies of tesseral harmonics are influenced by both the pole-to-pole meridian and the equatorial circle with the significance of each effect depending on the rank of the spherical harmonics k , the mode frequencies associated with $0 < k < l$ lie between those for the zonal and sectoral harmonics. In the presence of net electric charge, the Coulomb repulsion will enhance the quiescent deformation in the equilibrium shape so the frequency shifts become more pronounced. However, the net electric charge seems to affect the frequency shifts differently for the modes of spherical harmonics with different degree l and different rank k .

Although the quiescent deformation of the drop shape is axisymmetric, it affects the oscillation characteristics of both axisymmetric and asymmetric modes and also gives rise to the mode shape modifications. With a net electric charge, the oscillation mode shape modifications can appear in the $O(\epsilon_1 \epsilon_2)$ problem due to the locally unbalanced electric stress and hydrostatic pressure at the drop surface, even though at that level of approximation the equilibrium shape is still spherical and there are no frequency modifications. In the $O(\epsilon_1^2 \epsilon_2)$ problem, however, the mode shape modifications are present even without a net electric charge, because of the quiescent deformation. Corresponding to one characteristic frequency, the oscillation mode consists of spherical harmonics of different degrees (l) but with the same rank (k) in addition to the major component described by Rayleigh's normal mode. The absence of the modifications of the mode rank (k) in (5.8) and (5.19) is a result of the current level of approximations where we have considered only the coupling between the oscillation modes and the quiescent fields such as gravity, external electric field and the subsequent quiescent deformation which are all axisymmetric in nature.

Following the procedure of multiple-parameter perturbations, a further analysis of the effects of the quiescent deformation on the nonlinear resonances seems straightforward. It is, however, beyond the present scope in view of the tedious algebra involved in such a large set of interaction equations. The three-dimensional analysis of nonlinear resonances for a drop free from external fields (Natarajan & Brown 1987*b*) shows that the oscillation mode degeneracy appearing in the small-

amplitude ($O(\epsilon_2)$ solution) plays an important role in causing long-timescale dynamics which may display stochastic behaviour. Without carrying out rigorous calculations for higher-order problems, some general conclusions may be drawn from the present results about the role played by an external field in nonlinear drop resonances. Since the external field tends to destroy the spherical symmetry in the equilibrium drop shape and remove the degeneracy in the small-amplitude oscillation modes, its presence may hinder the nonlinear interactions among the modes of the same degree (l) (or different degree with commensurate Rayleigh frequencies) because the frequencies are no longer the same (or no longer exactly commensurate) even when the oscillation amplitude is small; thereby it may become more difficult to observe the stochastic behaviour of the long-timescale dynamics when the drop is influenced by an external field.

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Appendix A

The basis functions used in this paper are spherical harmonics defined as

$$Y_{lk}(\theta, \phi) = \left[\frac{(2l+1)(l-|k|)!}{4\pi(l+|k|)!} \right]^{\frac{1}{2}} P_{lk}(\cos \theta) e^{ik\phi} \quad (-l \leq k \leq l),$$

with $P_{lk}(\theta)$ being the associated Legendre polynomials written as

$$P_{lk}(x) = \frac{(1-x^2)^{|k|/2}}{2^l l!} \frac{d^{l+|k|}}{dx^{l+|k|}} (1-x^2)^l.$$

The $Y_{lk}(\theta, \phi)$ satisfy the orthogonality conditions

$$\int_0^{2\pi} \int_0^\pi Y_{lk}(\theta, \phi) Y_{l'k'}^*(\theta, \phi) \sin \theta \, d\theta \, d\phi = \delta_{ll'} \delta_{kk'},$$

where the asterisk denotes the complex conjugate and δ_{nm} stands for the Kronecker delta.

Appendix B

From the well-known properties of the associate Legendre polynomials, some recurrence formulae often used in this paper for the spherical harmonics are obtained as follows.

$$xY_{lk} = J_{-1}(l, k) Y_{l-1, k} + J_{+1}(l, k) Y_{l+1, k}$$

with

$$J_{-1}(l, k) = \frac{(l+|k|)}{(2l+1)} \left[\frac{(2l+1)(l-|k|)}{(2l-1)(l+|k|)} \right]^{\frac{1}{2}},$$

$$J_{+1}(l, k) = \frac{(l-|k|+1)}{(2l+1)} \left[\frac{(2l+1)(l+|k|+1)}{(2l+3)(l-|k|+1)} \right]^{\frac{1}{2}}.$$

Hence,

$$x^2 Y_{lk} = I_{-2}(l, k) Y_{l-2, k} + I_{+2}(l, k) Y_{l+2, k} + I_0(l, k) Y_{lk},$$

where

$$I_{-2}(l, k) = \frac{(l+|k|)(l+|k|-1) \left[(2l+1)(l-|k|)(l-|k|-1) \right]^{\frac{1}{2}}}{(2l-1)(2l+1) \left[(2l-3)(l+|k|)(l+|k|-1) \right]^{\frac{1}{2}}},$$

$$I_0(l, k) = \frac{2l^2 + 2l - 2k^2 - 1}{(2l-1)(2l+3)},$$

$$I_{+2}(l, k) = \frac{(l-|k|+1)(l-|k|+2) \left[(2l+1)(l+|k|+1)(l+|k|+2) \right]^{\frac{1}{2}}}{(2l+1)(2l+3) \left[(2l+5)(l-|k|+1)(l-|k|+2) \right]^{\frac{1}{2}}}.$$

Moreover, we have

$$(1-x^2) \frac{dY_{lk}}{dx} = (l+1) J_{-1}(l, k) Y_{l-1, k} - l J_{+1}(l, k) Y_{l+1, k};$$

thereby

$$x(1-x^2) \frac{dY_{lk}}{dx} = (l+1) I_{-2}(l, k) Y_{l-2, k} - I_{+2}(l, k) Y_{l+2, k}$$

$$+ [(l+1) J_{-1}(l, k) J_{+1}(l-1, k) - l J_{+1}(l, k) J_{-1}(l+1, k)] Y_{lk}$$

and

$$(l+1) J_{-1}(l, k) J_{+1}(l-1, k) - l J_{+1}(l, k) J_{-1}(l+1, k) = \frac{l^2 + l - 3k^2}{(2l-1)(2l+3)}.$$

Appendix C

The coefficients in (5.12) are derived from the condition of continuity of the tangential electric field on the drop surface as

$$H_{-2}^{(2,1)}(l, k; Q) = 6J_{+1}(l-1, k) A_{-1}^{(1,1)}(l-1, k; Q) + \frac{3Q}{8\pi} \left(\frac{5}{4\pi} \right)^{\frac{1}{2}} \alpha_{20}^{(2,0)} I_{+2}(l-2, k),$$

$$H_0^{(2,1)}(l, k; Q) = 6J_{-1}(l+1, k) A_{-1}^{(1,1)}(l+1, k; Q) + 6J_{+1}(l-1, k) A_{+1}^{(1,1)}(l-1, k; Q) + \frac{3Q}{8\pi} \left(\frac{5}{4\pi} \right)^{\frac{1}{2}} \alpha_{20}^{(2,0)} (l+2) \left[I_0(l, k) - \frac{1}{3} \right],$$

$$H_{+2}^{(2,1)}(l, k; Q) = 6J_{-1}(l+1, k) A_{+1}^{(1,1)}(l+1, k; Q) + \frac{3Q}{8\pi} \left(\frac{5}{4\pi} \right)^{\frac{1}{2}} \alpha_{20}^{(2,0)} (l+4) I_{-2}(l+2, k),$$

with the expressions for $I_{-2}(l, k)$, $I_0(l, k)$ and $I_{+2}(l, k)$ given in Appendix B. The coefficients in (5.13) are derived from the kinematic condition as

$$B_{-2}^{(2,1)}(l, k; Q) = \frac{3}{2} \left(\frac{5}{4\pi} \right)^{\frac{1}{2}} \alpha_{20}^{(2,0)} (l-1) I_{+2}(l-2, k),$$

$$B_0^{(2,1)}(l, k; Q) = \frac{3}{2} \left(\frac{5}{4\pi} \right)^{\frac{1}{2}} \alpha_{20}^{(2,0)} \left[(l+1) I_0(l, k) - \frac{(l-1)}{3} - \frac{2(l+|k|)(l-|k|)}{l(2l-1)} \right],$$

$$B_{+2}^{(2,1)}(l, k; Q) = \frac{3}{2} \left(\frac{5}{4\pi} \right)^{\frac{1}{2}} \alpha_{20}^{(2,0)} \left[(l+1) - \frac{2(l+3)}{(l+2)} \right] I_{-2}(l+2, k),$$

where the Q dependence comes from the fact that $\alpha_{20}^{(2,0)}$ is a function of charge Q . The coefficients in (5.14) are derived from the equation for the normal stress balance as

$$\begin{aligned}
 C_{-2}^{(2,1)}(l, k; Q) &= \frac{3}{2} \left(\frac{5}{4\pi} \right)^{\frac{1}{2}} \alpha_{20}^{(2,0)} \left\{ 2(l^2 - 3l + 6) \right. \\
 &\quad \left. + (l-2)(l-3) \left[l - \left(\frac{Q}{4\pi} \right)^2 \right] \right\} I_{+2}(l-2, k) + E_{-2}^{(2,1)}(l, k; Q), \\
 C_0^{(2,1)}(l, k; Q) &= \frac{3}{2} \left(\frac{5}{4\pi} \right)^{\frac{1}{2}} \alpha_{20}^{(2,0)} \left\{ 2(l^2 + l + 4) \right. \\
 &\quad \left. + l(l-1) \left[l + 2 - \left(\frac{Q}{4\pi} \right)^2 \right] \right\} \left[I_0(l, k) - \frac{1}{3} \right] + E_0^{(2,1)}(l, k; Q), \\
 C_{+2}^{(2,1)}(l, k; Q) &= \frac{3}{2} \left(\frac{5}{4\pi} \right)^{\frac{1}{2}} \alpha_{20}^{(2,0)} \left\{ 2(l^2 + 5l + 10) \right. \\
 &\quad \left. + (l+1)(l+2) \left[l + 4 - \left(\frac{Q}{4\pi} \right)^2 \right] \right\} I_{-2}(l+2, k) + E_{+2}^{(2,1)}(l, k; Q),
 \end{aligned}$$

with $E_{-2}^{(2,1)}(l, k; Q)$, $E_0^{(2,1)}(l, k; Q)$ and $E_{+2}^{(2,1)}(l, k; Q)$ denoting the contribution from $N_E - B o r_0 \cos \theta$:

$$\begin{aligned}
 E_{-2}^{(2,1)}(l, k; Q) &= \frac{Q}{4\pi} (l+1) H_{-2}^{(2,1)}(l, k; Q) \\
 &\quad + 6J_{+1}(l-1, k) \left[l H_{-1}^{(1,1)}(l-1, k; Q) - \frac{5Q}{4\pi} A_{-1}^{(1,1)}(l-1, k; Q) \right] \\
 &\quad - \left\{ \frac{3}{2} \left(\frac{Q}{4\pi} \right)^2 \left(\frac{5}{4\pi} \right)^{\frac{1}{2}} \alpha_{20}^{(2,0)} [(l-1)^2 + 8 + 2(l-2)] + 36 \right\} I_{+2}(l-2, k), \\
 E_0^{(2,1)}(l, k; Q) &= \frac{Q}{4\pi} (l+1) H_0^{(2,1)}(l, k; Q) \\
 &\quad + 6J_{+1}(l-1, k) \left[l H_{+1}^{(1,1)}(l-1, k; Q) - \frac{5Q}{4\pi} A_{+1}^{(1,1)}(l-1, k; Q) \right] \\
 &\quad + 6J_{-1}(l+1, k) \left[(l+2) H_{-1}^{(1,1)}(l+1, k; Q) - \frac{5Q}{4\pi} A_{-1}^{(1,1)}(l+1, k; Q) \right] \\
 &\quad - \frac{3}{2} \left(\frac{Q}{4\pi} \right)^2 \left(\frac{5}{4\pi} \right)^{\frac{1}{2}} \alpha_{20}^{(2,0)} [(l+1)^2 + 8] \left[I_0(l, k) - \frac{1}{3} \right] - 36 I_0(l, k) \\
 &\quad + 3 \left(\frac{Q}{4\pi} \right)^2 \left(\frac{5}{4\pi} \right)^{\frac{1}{2}} \alpha_{20}^{(2,0)} \frac{l^2 + l - 3k^2}{(2l-1)(2l+3)}, \\
 E_{+2}^{(2,1)}(l, k; Q) &= \frac{Q}{4\pi} (l+1) H_{+2}^{(2,1)}(l, k; Q) \\
 &\quad + 6J_{-1}(l+1, k) \left[(l+2) H_{+1}^{(1,1)}(l+1, k; Q) - \frac{5Q}{4\pi} A_{+1}^{(1,1)}(l+1, k; Q) \right] \\
 &\quad - \left\{ \frac{3}{2} \left(\frac{Q}{4\pi} \right)^2 \left(\frac{5}{4\pi} \right)^{\frac{1}{2}} \alpha_{20}^{(2,0)} [(l+3)^2 + 8 - 2(l+3)] + 36 \right\} I_{-2}(l+2, k).
 \end{aligned}$$

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