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Resonant oscillations of inviscid charged drops

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Moderate-amplitude axisymmetric oscillations of charged inviscid drops held together by surface tension are calculated by a multiple-timescale expansion. The corrections to the drop shape and velocity caused by mode coupling at second order in amplitude are predicted for two-, three- and four-lobed motions of drops with net charge up to the Rayleigh limit $Q_c \equiv 4\pi^{\frac{1}{2}}$. Resonant oscillations between four- and six-lobed motions occur for total charge values near $Q_r \equiv (\frac{32}{3}\pi)^{\frac{1}{2}}$ and are analysed. Both frequency and amplitude modulation of the oscillation are predicted for drop motions starting from general initial deformations.

1. Introduction

The dynamic response of liquid drops held together by surface tension and with electrical charge is important in a great variety of applications. The motion of such drops has been studied in physical systems ranging in size from millimetre raindrops (Brazier-Smith *et al.* 1971; Tsang 1974) and micron-sized spheres produced by fuel atomizers and ink-jet delivery systems (Williams 1973) to the femtometre drops used as models for nuclear fission (Cohen & Swiatecki 1962, 1963; Nix 1972). Rayleigh (1882; also see Hendricks & Schneider 1963) was the first to treat the effect of electrical charge on nearly spherical drops. By an energy stability analysis applied to conducting drops immersed in an insulating medium, Rayleigh calculated the frequencies for small-amplitude oscillations of an inviscid drop and established the amount of charge necessary to fission the drop. The modes of shape oscillation were described by single Legendre polynomials and the levels of charge necessary to disrupt the nth mode were given as

$$\tilde{Q}^{(n)} = 4\pi[\epsilon_m \sigma R^3(n+2)]^{\frac{1}{2}}, \quad n \geqslant 2, \tag{1.1}$$

where σ is the surface tension of the drop, ϵ_m is the permittivity of the medium and R is the radius of the spherical shape. The mode number n corresponds to the number of lobes on the deformed drop. The two-lobed form becomes unstable at the lowest value $Q = Q_c^{(2)}$, which markes the absolute stability limit for nearly spherical drops.

Rayleigh's pioneering work has been the basis for other analyses of drop dynamics which generalize the calculations to include viscosity (Tang & Wong 1974; Hasse 1975) and charge relaxation due to a dielectric drop and external medium (Saville 1974). These studies considered only small-amplitude oscillations and values of charge below the Rayleigh limit. The finite-difference calculations of Alonso (1974) for a slightly viscous drop with volumetrically distributed charge and the finite-element calculations of Basaran *et al.* (1982) for inviscid drops with surface charge are the only studies of nonlinear dynamics for charged drops. Such computations are relatively expensive, so that an exhaustive mapping of drop response as a function of the initial drop shape and the net electrical charge has not been undertaken. Experiments aboard the Space Shuttle are also planned, but must be even more limited in scope because of time restrictions. The use of perturbation analysis to guide both the experiments and further calculations is in order.

In this paper we present asymptotic analysis of the moderate-amplitude axisymmetric oscillations of an inviscid conducting drop with electrical charge suspended in a tenuous insulating medium. The calculations are focused on describing the effect of the amplitude of deformation on the form of the oscillation and on describing the harmonic resonance between the fundamental motion and secondary modes induced by nonlinear interactions. Our analysis is based on the method of multiple timescales as applied to approximating the time-periodic solutions of nonlinear conservative differential equations. In a previous paper (Tsamopoulos & Brown 1983; henceforth referred to as I), we used the Poincaré–Lindstedt method to calculate the dependence of the frequency modulations on amplitude for regular oscillations of an inviscid uncharged drop. The multiple-scale expansions used here are generalizations of the Poincaré–Lindstedt technique which allow for both frequency and amplitude modulations on timescales that differ from the one associated with the fundamental motion. As shown in §3, the two approaches are identical for regular oscillations. The multiple-timescale approach affords systematic treatment of the cases of harmonic resonance.

Harmonic resonance has long been known for two- and three-dimensional inviscid waves. Wilton (1915) first demonstrated secondary harmonic resonance in capillary-gravity waves and computed the special case of waves of permanent form with a phase shift relative to the fundamental frequency. Bretherton (1964) was the first to solve the general equations derived from a multiple-scale expansion which included slow amplitude modulation of the waveform. In a series of papers McGoldrick (1965, 1970*b*, 1972) and his collaborators have thoroughly analysed second- and third-order resonance in water waves including both surface tension and gravity. Both phenomena have been confirmed experimentally (McGoldrick *et al.* 1966; McGoldrick 1970*a*). Phillips (1981) has recently reviewed the rapid developments in the theory of nonlinear wave interactions.

The analysis of nonlinear oscillations and resonance of an isolated drop has advantages over the parallel studies for initially planar waves. The natural periodicity of the drop's surfaces results in a discrete spectrum of the fundamental modes as described by Rayleigh. For planar water waves the spectrum is continuous in the spatial wavenumber and detuning of resonant interactions caused by variation of this wavenumber from the critical value for resonance must be considered in both experiments and calculations. Also, the formulation of the planar-wave problem requires consideration of the variation of this spatial wavelength with the amplitude of the oscillation. The general problem of combined wavelength, amplitude and frequency modulation for two-dimensional waves is, as yet, unsolved. Again, the periodicity of the drop removes the need for incorporating wavelength variation and makes the formulation in §4 the most general for this problem. The major disadvantage of calculations for an oscillating drop over the analysis for planar waves is the more complex algebra generated by the velocity and electrostatic potentials expressed in terms of Legendre polynomials. We have carried out these calculations using the symbolic manipulator MACSYMA (Pavelle, Rothstein & Fitch 1981), which is available on the MIT computer system.

The multiple-timescale expansions valid up to third order in the initial amplitude of the deformation are developed in §3 and their solutions valid away from resonance are presented. In this section we correct a slight algebraic error in the third-order problem made in I and present the emended coefficients for the dependence of oscillation frequency for uncharged drops and bubbles on amplitude. In §4 we analyse second-harmonic resonance for charged drops which occurs at critical values $\tilde{Q} \equiv \tilde{Q}_r$. The detuning caused by a slight variation in the total charge from this value is also considered. Third-order harmonic resonance which occurs for uncharged drops and particular values of $\tilde{Q}$ is discussed briefly in §5. The effect of viscosity on the nonlinear interactions predicted by this inviscid analysis is discussed in §6.

2. Formulation

We consider the irrotational and incompressible motion of an electrically conducting inviscid drop with volume $\tilde{V} = \frac{4}{3}\pi R^3$, density ρ, surface tension σ and net electrical charge Q. The motion of the drop in a tenuous surrounding medium is caused by initially introducing a small axisymmetric deformation. As in I, the surface of the drop during this motion is described by $RF(\theta, t)$, where $F(\theta, t)$ is the dimensionless shape function of the drop and θ is the meridional angle in spherical coordinates. The same scales employed in I are used to define the dimensional velocity potential $(\sigma R/\rho)^{\frac{1}{2}}\phi(r,\theta,t)$, pressure $(2\sigma/R)\ p(r,\theta,t)$ and time $(\rho R^3/\sigma)^{\frac{1}{2}}t$ in terms of their dimensionless counterparts. The inviscid equations of motion and boundary conditions are

$$\nabla^2\phi = 0 \quad (0 \leqslant r \leqslant F(\theta,t), \quad 0 \leqslant \theta \leqslant \pi), \tag{2.1}$$

$$\frac{\partial \phi}{\partial r} = 0 \quad (r = 0, \quad 0 \leqslant \theta \leqslant \pi), \tag{2.2}$$

$$2p + \frac{\partial \phi}{\partial t} + \frac{1}{2}\left[\left(\frac{\partial \phi}{\partial r}\right)^2 + \left(\frac{1}{r}\frac{\partial \phi}{\partial \theta}\right)^2\right] = G(t) \quad (0 \leqslant r \leqslant F(\theta,t), \quad 0 \leqslant \theta \leqslant \pi), \tag{2.3}$$

$$\frac{\partial F}{\partial t} = \frac{\partial \phi}{\partial r} - \frac{1}{r^2}\frac{\partial \phi}{\partial \theta}\frac{\partial F}{\partial \theta} \quad (r = F(\theta,t), \quad 0 \leqslant \theta \leqslant \pi), \tag{2.4}$$

$$\Delta p_0 + 2p + \frac{1}{4\pi}(T^e_{n2} - T^e_{n1}) = -2\mathscr{H} \quad (r = F(\theta,t), \quad 0 \leqslant \theta \leqslant \pi), \tag{2.5}$$

$$\int_0^\pi F^3(\theta,t)\sin\theta\,\mathrm{d}\theta = 2. \tag{2.6}$$

Laplace's equation (2.1) and Bernoulli's equation (2.3) describe the velocity and pressure everywhere in the drop. Equation (2.4) kinematically relates the motion of the surface to the velocity there. The normal-stress balance (2.5) equates the pressure differences caused by capillarity and drop motion to the contributions of the normal electric stress from inside T^e_{n1} and outside T^e_{n2} the drop. The reference pressure difference Δp_0 in (2.5) is determined by the constraint of constant drop volume (2.6). We absorb the constant of integration $G(t)$ into the time derivative $\partial\phi/\partial t$. As explained by Lamb (1932, §227), including this integration constant in (2.3) leads to terms constant in space and proportional to t in the velocity potential, but has no other effect on the solution to (2.1)–(2.6) because only space derivatives of ϕ appear in these equations.

The medium surrounding the drop is assumed to be electrically insulating, and the dimensionless electrostatic potential $V(r,t,\theta)$ and the uniform potential in the drop $V_0(t)$ are both scaled with $(4\pi\epsilon_m/\sigma R)^{-\frac{1}{2}}$, where ϵ_m is the permittivity of the medium.

The electric field is related to the potential as $\boldsymbol{E} = -\nabla V$ and is scaled with $(4\pi\epsilon_m R/\sigma)^{-\frac{1}{2}}$; the dimensionless net charge Q is scaled with $(\sigma 4\pi\epsilon_m R^3)^{-\frac{1}{2}}$. The equations and boundary conditions governing the electrostatic potentials are

$$\nabla^2 V = 0 \quad (F(\theta,t) \leqslant r < \infty, \quad 0 \leqslant \theta \leqslant \pi), \tag{2.7}$$

$$V = 0 \quad (r \rightarrow \infty, \quad 0 \leqslant \theta \leqslant \pi), \tag{2.8}$$

$$-\boldsymbol{n}\cdot\nabla V = 4\pi q(\theta,t) \quad (r = F(\theta,t), \quad 0 \leqslant \theta \leqslant \pi), \tag{2.9}$$

$$\boldsymbol{t}\cdot\nabla V = 0 \quad (r = F(\theta,t), \quad 0 \leqslant \theta \leqslant \pi), \tag{2.10}$$

$$2\pi \int_0^\pi qF(F^2+F_\theta^2)^{\frac{1}{2}} \sin\theta \, \mathrm{d}\theta = Q, \tag{2.11}$$

in which $q(\theta,t)$ is the local surface charge density and $\boldsymbol{n}$ and $\boldsymbol{t}$ are the unit vectors normal and tangential to the drop surface and are defined as

$$\boldsymbol{n} \equiv \frac{F\boldsymbol{e}_r - F_\theta \boldsymbol{e}_\theta}{(F^2+F_\theta^2)^{\frac{1}{2}}}, \quad \boldsymbol{t} \equiv \frac{F\boldsymbol{e}_\theta + F_\theta \boldsymbol{e}_r}{(F^2+F_\theta^2)^{\frac{1}{2}}}. \tag{2.12}$$

In these equations $F_\theta \equiv \partial F/\partial\theta$, and $(\boldsymbol{e}_r, \boldsymbol{e}_\theta)$ are the unit vectors in spherical coordinates. In formulating (2.7)–(2.12) we have assumed that charge is confined to the interface and equilibrates in a time much shorter than that characteristic of the fluid motion. With these assumptions, (2.11) is the charge balance on the interface. Equation (2.11) follows from the general conservation equation for a species on a deforming surface first derived by Bupara (1965; see also Moeckel 1974) when bulk and surface convection are negligible compared with conduction. Then the assumption of electrostatic equilibrium follows if the characteristic time for conduction is much smaller than the time for a typical drop oscillation, or

$$\left(\frac{\rho R^3}{\sigma}\right)^{\frac{1}{2}} \gg \frac{\chi_0}{4\pi\epsilon_m R^2},$$

where χ_0 is the resistivity of the drop. For the case of even distilled water ($\chi_0 = 10^4\ \Omega\ \mathrm{m}^2$, $\rho = 1\ \mathrm{g/cm}^3$, $\sigma = 75\ \mathrm{dyn/cm}$) in air ($\epsilon_m = 8.8 \times 10^{-2}\ \mathrm{F/m}$) this inequality is satisfied by several orders of magnitude when $R = 0.1$ cm. Equation (2.10) guarantees that the tangential component of the electric field is continuous across the interface. It is equivalent to the requirement that the potential be continuous across $F(\theta,t)$, and sets the constant potential V_0.

The electric stress caused by the external electric field is defined as (Stratton 1941)

$$\boldsymbol{T}_2^e \equiv \boldsymbol{E}\boldsymbol{E} - \tfrac{1}{2}|\boldsymbol{E}|^2 \boldsymbol{I}, \tag{2.13}$$

where $\boldsymbol{I}$ is the identity tensor and $|\boldsymbol{E}|$ is the magnitude of $\boldsymbol{E}$. The component of this stress normal to the surface of the drop,

$$T_{n2}^e \equiv \boldsymbol{nn}:\boldsymbol{T}_2^e = \tfrac{1}{2}(\boldsymbol{n}\cdot\boldsymbol{E})^2, \tag{2.14}$$

appears in the normal-stress balance (2.5) and couples together the fluid flow and electrostatic problems. The spatially uniform potential inside the conducting drop forces $\boldsymbol{T}_1^e$ to be zero, hence $T_{n1}^e \equiv 0$.

The dynamical problem for the velocity and electrostatic potentials and the drop shape are solved for motions originating with an initial deformation of the drop. We describe initial deformations which satisfy conservation of mass (2.6) and which have no initial velocity, i.e.

$$\frac{\partial F}{\partial t}(\theta, 0) = 0. \tag{2.15}$$

We define the amplitude of the oscillation ϵ in terms of this initial deformation as

$$F(\theta,0) = 1+\epsilon P_n(\theta)+O(\epsilon^2), \tag{2.16}$$

where $P_n(\theta)$ is the Legendre polynomial of nth order. The forms of the initial condition (2.16) that satisfy the volume integral (2.6) up to $O(\epsilon^3)$ are

$$F(\theta,0) = 1+\epsilon P_2(\theta)-\tfrac{1}{5}\epsilon^2-\tfrac{2}{105}\epsilon^3+\ldots \quad \text{for } n=2, \tag{2.16a}$$

$$F(\theta,0) = 1+\epsilon P_3(\theta)-\tfrac{1}{7}\epsilon^2-0\cdot\epsilon^3+\ldots \quad \text{for } n=3, \tag{2.16b}$$

$$F(\theta,0) = 1+\epsilon P_4(\theta)-\tfrac{1}{9}\epsilon^2-\tfrac{6}{1001}\epsilon^3+\ldots \quad \text{for } n=4. \tag{2.16c}$$

The amplitude ϵ is taken to be a small parameter in the analysis that follows.

3. Perturbation solution away from resonance

We determine the potential fields $(\phi(r,\theta,t),\ V(r,\theta,t))$ and drop shape $F(\theta,t)$ for moderate-amplitude motions by constructing expansions in the initial amplitude of the deformation ϵ. The asymptotic methods couple together the method of multiple timescales for freely oscillating non-dissipative systems and the domain perturbation technique outlined by Joseph (1973) and used in I to account for changes in the drop shape with time and amplitude. Formally, we assume that the dependent variables are functions of three timescales related to the actual time as $T_0 \equiv t$, $T_1 \equiv \epsilon t$ and $T_2 = \frac{1}{2}\epsilon^2 t$. The different timescales are introduced into the field equations by expanding the partial derivative $\partial/\partial t$ as

$$\frac{\partial}{\partial t} \equiv \frac{\partial}{\partial T_0}+\epsilon\frac{\partial}{\partial T_1}+\frac{\epsilon^2}{2}\frac{\partial}{\partial T_2}+O(\epsilon^3). \tag{3.1}$$

The expansion for the domain shape is implemented by transforming the drop shape to the unit sphere using the change of coordinates $r \equiv \eta F(\theta,t)$ and expanding each dependent variable in a Taylor series

$$\begin{bmatrix} F(\theta,t;\epsilon) \\ \phi(r,\theta,t;\epsilon) \\ V(r,\theta,t;\epsilon) \end{bmatrix} = \sum_{k=0}^{\infty} \frac{\epsilon^k}{k!} \begin{bmatrix} F^{[k]}(\theta,T_0,T_1,T_2) \\ \phi^{[k]}(\eta,\theta,T_0,T_1,T_2) \\ V^{[k]}(\eta,\theta,T_0,T_1,T_2) \end{bmatrix}, \tag{3.2}$$

where the superscript $[k]$ denotes the kth total derivative of the quantity with respect to ϵ. As in the development in I, each term in these expansions for the potentials can be written as a sum of a contribution based on the spherical domain $(0 \leqslant \eta \leqslant 1,\ 0 \leqslant \theta \leqslant \pi)$ and terms that account for the deformation of the domain at each order of ϵ. The derivatives evaluated on the spherical domain are denoted by $\phi^{(k)}(\eta,\theta,T_0,T_1,T_2) \equiv \partial\phi^{(k)}/\partial\epsilon^{(k)}$. Because the drop shape $F(\theta,t)$ is independent of the radial coordinate, $F^{[k]}(\theta,T_0,T_1,T_2) \equiv F^{(k)}(\theta,T_0,T_1,T_2)$. Expressions for the total derivatives of a potential up to $\phi^{[2]}(\eta,\theta,T_0,T_1,T_2)$ are given by equation (14) of I.

We anticipate the form of the solution to the drop shape and expand $F^{(k)}(\theta,T_0,T_1,T_2)$ at each order as a series of Legendre polynomials:

$$\begin{aligned} F^{(k)}(\theta,T_0,T_1,T_2) &= \sum_{m=0}^{\infty} F_m^{(k)}(\theta,T_0,T_1,T_2) \\ &= \sum_{m=0}^{\infty} \delta_m^{(k)}(T_0,T_1,T_2)\,P_m(\theta). \end{aligned} \tag{3.3}$$

Using these forms for the corrections to $F(\theta, t)$, the mean curvature of the drop and the unit normal and tangent vectors are conveniently expanded in ϵ; the results valid up to $O(\epsilon^3)$ are given in Appendix A.

The equations governing the zeroth-order contributions from the set (2.1)–(2.16) describe a static charged drop and have the solution

$$\begin{bmatrix} F^{(0)}(\theta, T_0) \\ \phi^{(0)}(\eta, \theta, T_0) \\ V^{(0)}(\eta, \theta, T_0) \end{bmatrix} = \begin{bmatrix} 1 \\ 0 \\ Q/\eta \end{bmatrix}. \tag{3.4}$$

The arbitrary reference potential V_0 inside the drop has been set equal to Q.

The equation set that governs the first-order corrections $(F^{(1)}, \phi^{(1)}, V^{(1)})$ is

$$\nabla^2 \phi^{(1)} = 0 \quad (0 \leqslant \eta < 1, \quad 0 \leqslant \theta \leqslant \pi), \tag{3.5}$$

$$\frac{\partial \phi^{(1)}}{\partial \eta} = 0 \quad (\eta = 0, \quad 0 \leqslant \theta \leqslant \pi), \tag{3.6}$$

$$\frac{\partial \phi^{(1)}}{\partial \eta} = \frac{\partial F^{(1)}}{\partial T_0} \quad (\eta = 1, \quad 0 \leqslant \theta \leqslant \pi), \tag{3.7}$$

$$\frac{\partial \phi^{(1)}}{\partial T_0} - \frac{1}{4\pi} \frac{\partial V^{(0)}}{\partial \eta} \left[\frac{\partial V^{(1)}}{\partial \eta} + F^{(1)} \frac{\partial^2 V^{(0)}}{\partial \eta^2} \right] = - \sum_{n=2}^{\infty} (n-1)(n+2) F_n^{(1)} \quad (\eta = 1, \quad 0 \leqslant \theta \leqslant \pi), \tag{3.8}$$

$$\int_0^\pi F^{(1)} \sin\theta \, \mathrm{d}\theta = 0, \tag{3.9}$$

$$\nabla^2 V^{(1)} = 0 \quad (1 \leqslant \eta \leqslant \infty, \quad 0 \leqslant \theta \leqslant \pi), \tag{3.10}$$

$$V^{(1)} = 0 \quad (\eta \rightarrow \infty, \quad 0 \leqslant \theta \leqslant \pi), \tag{3.11}$$

$$\int_0^\pi \left[\frac{\partial V^{(1)}}{\partial \eta} + F^{(1)} \frac{\partial^2 V^{(0)}}{\partial \eta^2} \right] \sin\theta \, \mathrm{d}\theta = 0 \quad (\eta = 1), \tag{3.12}$$

$$\frac{\partial V^{(1)}}{\partial \theta} + \frac{\partial F^{(1)}}{\partial \theta} \frac{\partial V^{(0)}}{\partial \eta} = 0 \quad (\eta = 1, \quad 0 \leqslant \theta \leqslant \pi), \tag{3.13}$$

$$F^{(1)}(\theta, 0, 0, 0) = P_n(\theta), \tag{3.14}$$

$$\frac{\partial F^{(1)}}{\partial T_0}(\theta, 0, 0, 0) = 0. \tag{3.15}$$

The pressure and normal electric stress have been eliminated from (3.8) by substituting from the first-order forms of Bernoulli's equation and the definition of the electric stress tensor. Also, (3.12) is a combination of the conditions for the jump in the normal component of the electric field at the interface and the condition for conservation of total charge.

The solutions to (3.5)–(3.15) have the form of the linear-oscillation modes described by Rayleigh (1882) cast in the framework of the multiple-scale expansion:

$$F^{(1)}(\theta, T_0, T_1, T_2) = c_n(T_1, T_2) \cos \psi_n P_n(\theta), \tag{3.16a}$$

$$\phi^{(1)}(\eta, \theta, T_0, T_1, T_2) = -c_n(T_1, T_2) \frac{\eta^n \omega_n^{(0)}}{n} \sin \psi_n P_n(\theta), \tag{3.16b}$$

$$V^{(1)}(\eta, \theta, T_0, T_1, T_2) = c_n(T_1, T_2)\, \eta^{-(n+1)} \cos \psi_n P_n(\theta), \tag{3.16c}$$

where $$\psi_n \equiv \omega_n T_0 + h_n(T_1, T_2), \quad \omega_n \equiv \left\{ n(n-1)\left[n+2-\frac{Q^2}{4\pi}\right]\right\}^{\frac{1}{2}}, \tag{3.16d}$$

and $\{c_n(T_1, T_2), h_n(T_1, T_2)\}$ are functions of the slower timescales which are to be determined as part of the second-order problem subject to the initial conditions

$$c_n(0,0) = 1, \quad h_n(0,0) = 0. \tag{3.17}$$

The functions $\{c_n(T_1, T_2)\}$ and $\{h_n(T_1, T_2)\}$ represent the modulation in the slow timescales of the amplitude and frequency of the oscillations, respectively. As noted by Rayleigh, drops with net charge less than $Q_c^{(n)} = [4\pi(n+2)]^{\frac{1}{2}}$ oscillate with stable standing waves, whereas drops with greater charge are unstable and fission. The lowest value of $Q_c^{(n)}$ sets the maximum admissible charge on the drop, corresponds to $n = 2$ and is the Rayleigh stability limit $Q_c = 4\pi^{\frac{1}{2}}$. We consider only drops with net charge below this limit.

The equation set for the second-order terms $(\phi^{(2)}, V^{(2)}, F^{(2)})$ is cumbersome because of the multitude of non-homogeneous terms that are generated by the domain perturbation. Because of the relevance of these equations to other analyses of drop dynamics we list the set in Appendix B. These equations are solved by expanding the two potentials $(\phi^{(2)}, V^{(2)})$ in series of Legendre polynomials and powers of η:

$$\begin{bmatrix} \phi^{(2)}(\eta, \theta, T_0, T_1, T_2) \\ V^{(2)}(\eta, \theta, T_0, T_1, T_2) \end{bmatrix} = \sum_{m=0}^{\infty} P_m(\theta) \begin{bmatrix} \gamma_m(T_0, T_1, T_2)\, \eta^m \\ \beta_m(T_0, T_1, T_2)\, \eta^{-(m+1)} \end{bmatrix}, \tag{3.18}$$

which satisfy the field equations and boundary conditions, except those on the drop surface. The correction to the drop shape is given by (3.3). The displacement condition (B 9) can be integrated directly with respect to θ to give $V^{(2)}$ in terms of $F^{(2)}$ and lower-order quantities. Substituting this result into (B 3) and (B 4) and forming the integrals with $\{\sin\theta P_m(\theta)\}$ yields a sequence of second-order non-homogenous equations for the coefficients $\{\gamma_m(T_0, T_1, T_2)\}$. These are

$$\begin{aligned} \frac{\partial^2 \gamma_m}{\partial T_0^2} + \omega_m^2 \gamma_m = {} & 2\left(\frac{\omega_m^2}{m} + \frac{\omega_n^2}{n}\right)\left(\frac{\partial c_n}{\partial T_1}\cos\psi_n - c_n \sin\psi_n \frac{\partial h}{\partial T_1}\right)\langle P_n, P_m\rangle \\ & - c_n^2 \omega_n \sin 2\psi_n \left[3\omega_n^2 + 4(n^2+n-1) - \frac{Q^2}{4\pi}(n^2+8n-3) \right. \\ & \left. + \frac{Q^2}{4\pi}(m+1)(2n) - (n-1)\frac{\omega_m^2}{m}\right]\langle P_2^2, P_m\rangle \\ & - c_n^2\omega_n \sin 2\psi_n \left[\frac{Q^2}{4\pi} + \frac{\omega_n^2}{n^2} + \frac{\omega_m^2}{nm}\right]\langle P_n^2, P_m\rangle + \frac{Q}{4\pi}(m+1)\frac{\partial K_m}{\partial T_0}\langle 0, P_m\rangle, \end{aligned} \tag{3.19}$$

where $\langle f(\theta), g(\theta)\rangle$ is the inner product of these functions weighted with $\sin\theta$ on the interval $[0, \pi]$, and K_m is an integration constant from (B 9).

The solvability condition for the equations (3.19) eliminates the secular terms and leads to the result

$$\frac{\partial c_n}{\partial T_1} = \frac{\partial h_n}{\partial T_1} = 0, \tag{3.20}$$

or that the modulation functions depend only on T_2. The solutions of (3.19), determined so that the initial conditions (B 10), (B 11) and integral constraints (B 5), (B 8) are satisfied, are written in the form

$$F_k^{(2)}(\theta, T_0, T_1, T_2) = \sum_{j=0}^{8} L_{2kj}(T_0, T_2)\, P_j(\theta), \tag{3.21}$$

where the exact values for the coefficients are tabulated in Appendix B. The corrections for $\phi^{(2)}$ and $V^{(2)}$ can be expressed in a matrix form similar to (3.21) with the elements $\{\eta^j M_{2kj}(T_0, T_2)\}$ and $\{N_{2kj}(T_0, T_2)/\eta^{j+1}\}$ respectively replacing $\{L_{2kj}(T_0, T_2)\}$. The exact forms of these coefficients are available in Tsamopoulos (1984).

The dependence of the frequency and amplitude modulations of the slow timescale T_2 are computed from the solvability conditions of the third-order problem, which is listed in Appendix C. The corrections to the shape and the potentials $(F^{(3)}, \phi^{(3)}, V^{(3)})$ are again expanded in Legendre series, and a set of second-order ordinary differential equations is derived by the same procedure used to solve the second-order problem (for details see Tsamopoulos 1984). The solvability condition for this set dictates that

$$\frac{\partial c_n}{\partial T_2} = 0, \tag{3.22}$$

so that the amplitude of the oscillation is not modulated up to $O(\epsilon^2)$; the $\{c_n\}$ are taken to be unity to satisfy the initial condition (3.17). Solvability also requires that the frequency modulation takes the form

$$h_2(T_2) = -\frac{8}{245}\frac{\sum_{j=0}^{4} A_{2j}\hat{Q}^j}{\omega_2^3\omega_4^2(\omega_4^2-4\omega_2^2)}T_2 \quad \text{for} \quad n = 2, \tag{3.23a}$$

$$h_3(T_2) = -\frac{288}{11011}\frac{\sum_{j=0}^{6} A_{3j}\hat{Q}^j}{\omega_3\omega_2^2\omega_4^2\omega_6^2(\omega_2^2-4\omega_3^2)(\omega_6^2-4\omega_3^2)}T_2 \quad \text{for}\, n = 3, \tag{3.23b}$$

$$h_4(T_2) = -\frac{2488320}{2433431}\frac{\sum_{j=0}^{8} A_{4j}\hat{Q}^j}{\omega_2^2\omega_4^3\omega_6^2\omega_8^2(\omega_2^2-4\omega_4^2)(\omega_6^2-4\omega_4^2)(\omega_8^2-4\omega_4^2)}T_2 \quad \text{for}\, n = 4, \tag{3.23c}$$

where $\hat{Q} \equiv Q^2/4\pi$, and the coefficients $\{A_{ij}\}$ are listed in Appendix C. Because there can be no dependence on the frequency modulation on any odd powers of the amplitude, the results (3.23) are accurate up to $O(\epsilon^4)$. For values of $\hat{Q}$ in the range $0 \leqslant \hat{Q} < 4$, the drop oscillates stably and both $h_2(T_2)$ and $h_3(T_2)$ remain negative. The frequency modulation $h_4(T_2)$ for the four-lobed oscillation is positive in the region $2.3 \leqslant \hat{Q} < \frac{8}{3}$ and is infinite at $\hat{Q} = \frac{8}{3}$. At this value of the net charge the four-lobed motion resonates with six-lobed oscillations and the scalings for the modulation functions derived in this section are no longer valid. The proper analysis close to this resonant point is described in §4.

The frequency modulations given by (3.23) in the limit $\hat{Q} = 0$ differ slightly from the results in I because of the algebraic error mentioned above. The results for the uncharged oscillating bubble given by equations (59)–(61) in I should be corrected to

$$\omega^{(2)} \equiv h_2(T_2) = -\tfrac{23549}{15435}\omega^{(0)} \approx -1.52569\omega^{(0)} \quad \text{for}\, n = 2, \tag{3.24a}$$

$$\omega^{(2)} \equiv h_3(T_2) = -\tfrac{5672825}{2461536}\omega^{(0)} \approx -2.30459\omega^{(0)} \quad \text{for}\, n = 3, \tag{3.24b}$$

$$\omega^{(2)} \equiv h_4(T_2) = -\tfrac{14775015009}{4939864930}\omega^{(0)} \approx -2.99098\omega^{(0)} \quad \text{for}\, n = 4. \tag{3.24c}$$

These corrections improve the agreement between the calculations and the experiments of Trinh, Zwern & Wang (1982) shown in figure 4 of I.

4. Second harmonic resonance

The regular forms of the weakly nonlinear oscillations described in §3 are not valid when the frequencies of the higher harmonics of the fundamental mode are close to integral multiples of its frequency. This does not occur for uncharged drops up to second order in amplitude, but happens for charged drops that satisfy the condition

$$\frac{m(m-1)\,(m+2-Q^2(4\pi)^{-1})}{\omega_n^2} = \text{integer}^2, \quad m \neq n. \tag{4.1}$$

For the two- and three-lobed modes of oscillation, the values of Q required for resonance are above the Rayleigh limit for electrostatic bursting. Resonance is detectable for four-lobed oscillations when the net charge is near $Q_r \equiv (\frac{32}{3}\pi)^{\frac{1}{2}} < Q_c$, where $\omega_6^2 = 4\omega_4^2$. Then the four-lobed fundamental resonates with the six-lobed form, and the two modes exchange energy in a periodic or aperiodic fashion, depending on the initial deformation of the drop.

In the remainder of this section we analyse this second harmonic resonance. The analysis used here parallels the work of McGoldrick (1972) for capillary–gravity waves. We consider drops with net charge slightly different from Q_r, or

$$Q = Q_r + Q^{(1)}\epsilon \equiv Q_r + \left(\frac{3\pi}{8}\right)^{\frac{1}{2}}\lambda\epsilon, \tag{4.2}$$

where $\lambda = O(1)$. The linear oscillation frequencies at these values of Q are given by

$$\omega_4 = 2\sqrt{10}\,(1-\tfrac{3}{20}\lambda\epsilon), \tag{4.3}$$

$$\omega_6 = 4\sqrt{10}\,(1-\tfrac{3}{32}\lambda\epsilon), \tag{4.4}$$

The formulation of the linear problem (3.5)–(3.15) for $n = 4$ is unaffected by the possibility of resonance, and, for convenience in subsequent manipulations, its solution is written in complex form as

$$F^{(1)}(\theta, T_0, T_1) = \{A(T_1)\,\mathrm{e}^{-\mathrm{i}\omega_4 T_0} + \text{c.c.}\}\,P_4(\theta), \tag{4.5a}$$

$$\phi^{(1)}(\gamma, \theta, T_0, T_1) = \{-\tfrac{1}{4}\mathrm{i}\omega_4\,A(T_1)\,\mathrm{e}^{-\mathrm{i}\omega_4 T_0} + \text{c.c.}\}\,P_4(\theta)\,\eta^4, \tag{4.5b}$$

$$V^{(1)}(\eta, \theta, T_0, T_1) = \{A(T_1)\,\mathrm{e}^{-\mathrm{i}\omega_4 T_0} + \text{c.c.}\}\,P_4(\theta)\,\eta^{-5}, \tag{4.5c}$$

where {c.c.} stands for the complex conjugate of the immediately preceding term. We carry our analysis of resonance only to terms $O(\epsilon)$; so to this order of approximation the term $A(T_1)$ will depend on only the single slow timescale T_1.

The forms of the singularities of the second-order coefficients in the regular perturbation when Q is near Q_r (e.g. see the coefficients I_{61} and I_{62} in (B 13)) suggests that the first-order solution be modified to include a second harmonic constituent ($n = 6$). This new mode may not be present initially ($t = 0$), but will be excited through the resonance with the existing fundamental ($n = 4$). We write the more general $O(\epsilon)$ solution as

$$F^{(1)}(\theta, T_0, T_1) = \{A(T_1)\,\mathrm{e}^{-\mathrm{i}\omega_4 T_0} + \text{c.c.}\}\,P_4(\theta) + \{B(T_1)\,\mathrm{e}^{-\mathrm{i}\omega_6 T_0} + \text{c.c.}\}\,P_6(\theta), \tag{4.6a}$$

$$\begin{aligned}\phi^{(1)}(\eta, \theta, T_0, T_1) = {} & \{-\tfrac{1}{4}\mathrm{i}\omega_4\,A(T_1)\,\mathrm{e}^{-\mathrm{i}\omega_4 T_0} + \text{c.c.}\}\,P_4(\theta)\,\eta^4 \\ & + \{-\tfrac{1}{6}\mathrm{i}\omega_6\,B(T_1)\,\mathrm{e}^{-\mathrm{i}\omega_6 T_0} + \text{c.c.}\}\,P_6(\theta)\,\eta^6,\end{aligned} \tag{4.6b}$$

$$\begin{aligned}V^{(1)}(\eta, \theta, T_0, T_1) = {} & \{A(T_1)\,\mathrm{e}^{-\mathrm{i}\omega_4 T_0} + \text{c.c.}\}\,P_4(\theta)\,Q\eta^{-5} \\ & + \{B(T_1)\,\mathrm{e}^{-\mathrm{i}\omega_6 T_0} + \text{c.c.}\}\,P_6(\theta)\,Q\eta^{-7}.\end{aligned} \tag{4.6c}$$

The dependence of the net charge on the amplitude through (4.2) introduces new terms into the non-homogenous parts of (B 4) and (B 9). These terms are, for (B 4),

$$\frac{-Q^{(0)}Q^{(1)}F^{(1)}}{2\pi} \tag{4.7}$$

and, for (B 9),

$$-2Q^{(1)}\frac{\partial F^{(1)}}{\partial \theta}. \tag{4.8}$$

Introducing the generalized solutions (4.6) into the modified form of the second-order problem and following the same procedure outlined in §3 leads to a second-order differential equation like (3.19). Eliminating secular terms from this equation requires setting to zero the coefficients multiplying not only the terms $\exp(\pm i\omega_4 T_0)$ and $\exp(\pm i\omega_6 T_0)$ that resonate exactly, but also those coefficients multiplied by $\exp(\pm 2i\omega_4 T_0)$ and $\exp(\pm i(\omega_6-\omega_4)T_0)$ that nearly resonate because

$$2\omega_4 T_0 \equiv (\omega_6 - N\epsilon)\,T_0 = \omega_6 T_0 - NT_1, \tag{4.9a}$$

$$(\omega_6-\omega_4)\,T_0 \equiv (\omega_4 + N\epsilon)\,T_0 = \omega_4 T_0 + NT_1, \tag{4.9b}$$

where $N \equiv \frac{9}{40}\lambda\sqrt{10}$. Thus the differences between these two frequencies and the fundamental $n = 4$ and $n = 6$ modes differ only in the slow timescale T_1. The solvability conditions that result from removing these secular terms are

$$\alpha_1\left(\frac{dA}{dT_1} - \alpha_3 iA\right) = iA^*B\,e^{-iNT_1}, \tag{4.10a}$$

$$\alpha_2\left(\frac{dB}{dT_1} - \alpha_4 iB\right) = iA^2\,e^{iNT_1}, \tag{4.10b}$$

where $\alpha_1 \equiv \frac{143}{267}\sqrt{\frac{1}{10}}$, $\alpha_2 \equiv \frac{88}{89}\sqrt{\frac{1}{10}}$, $\alpha_3 \equiv \lambda\sqrt{\frac{1}{10}}$ and $\alpha_4 \equiv \frac{9}{4}\lambda\sqrt{\frac{1}{10}}$. Equations similar to (4.10) were first derived by Bretherton (1964) for planar water waves. In §§4.1 and 4.2 we consider separately the cases of exact resonance ($\lambda = 0$) and the detuning caused by a slight variation in Q from Q_r ($\lambda \neq 0$).

4.1. *Exact resonance*

Introducing the substitutions $A(T_1) \equiv r_1(T_1)\,e^{i\theta_1(T_1)}$ and $B(T_1) \equiv r_2(T_1)\,e^{i\theta_2(T_1)}$, where the $\{r_i, \theta_i\}$ are real functions of the slow timescale, reduces (4.10) to

$$\alpha_1\frac{dr_1}{dT_1} = -r_1 r_2 \sin\hat{\theta}, \quad \alpha_2\frac{dr_2}{dT_1} = r_1^2\sin\hat{\theta}, \tag{4.11a,b}$$

$$\alpha_1 r_1\frac{d\theta_1}{dT_1} = r_1 r_2\cos\hat{\theta}, \quad \alpha_2 r_2\frac{d\theta_2}{dT_1} = r_1^2\cos\hat{\theta}, \tag{4.11c,d}$$

where $\hat{\theta} \equiv \theta_2 - 2\theta_1$. Just as for the modulation equations arising for planar water waves (Benney 1962), the first two relations (4.11*a,b*) have an energy-like integral

$$\alpha_1 r_1^2 + \alpha_2 r_2^2 \equiv E, \tag{4.12}$$

where E is proportional to the $O(\epsilon^2)$ energy carried in the system. The functions $r_1(T_1)$, $r_2(T_1)$ and the relative phase $\hat{\theta}(T_1)$ are, from (4.11), related by

$$r_1^2 r_2\cos\hat{\theta} \equiv L, \tag{4.13}$$

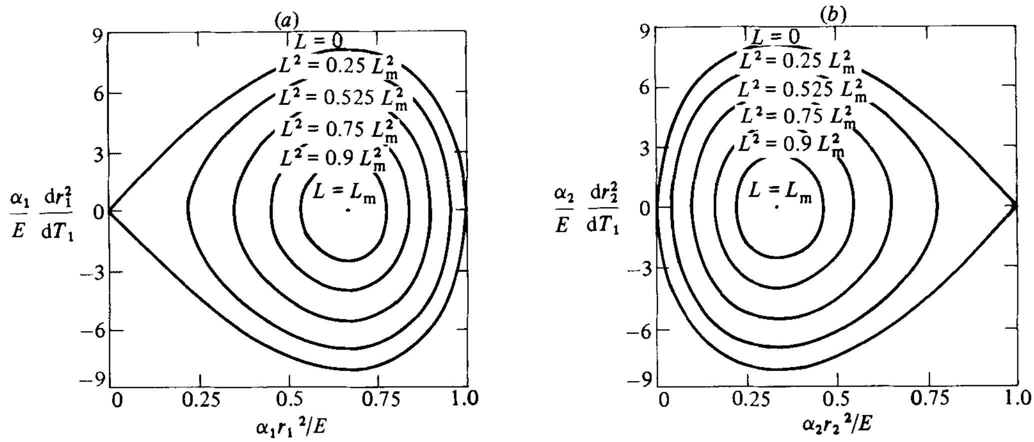

FIGURE 1. Phase-plane plots of the fundamental (*a*) and the resonating mode (*b*) with $A = (\frac{32}{2}\pi)^{\frac{1}{2}}$.

where L is a constant. Using (4.12) and (4.13), the set (4.11) is decoupled into the form

$$\frac{\alpha_1}{2}\frac{\mathrm{d}r_1^2}{\mathrm{d}T_1} = -\frac{1}{\alpha_2^{\frac{1}{2}}}(\alpha_1 r_1^6 + E r_1^4 - \alpha_2 L^2)^{\frac{1}{2}}, \tag{4.14a}$$

$$\frac{\alpha_2}{2}\frac{\mathrm{d}r_2^2}{\mathrm{d}T_1} = \frac{1}{\alpha_1}(\alpha_2^2 r_2^6 - 2E\alpha_2 r_2^4 + E^2 r_2^2 - \alpha_1^2 L^2)^{\frac{1}{2}}, \tag{4.14b}$$

$$\frac{\mathrm{d}\theta_1}{\mathrm{d}T_1} = \alpha_2 r_2^2 \frac{\mathrm{d}\theta_2}{\mathrm{d}T_1} = L. \tag{4.14c}$$

The third-order polynomial in r_1^2 on the right-hand side of (4.14*a*) has three real roots $\{\rho_i\}$ which satisfy the inequalities

$$-E/3\alpha_1 \leqslant \rho_1 \leqslant 0 \leqslant \rho_2 \leqslant 2E/3\alpha_1 \leqslant \rho_3 \leqslant E/\alpha_1.$$

The solutions of (4.14) are expressed as

$$r_1^2(T_1) = \rho_3 + (\rho_2 - \rho_3)\,\mathrm{sn}^2(\tau; k), \tag{4.15a}$$

$$r_2^2(T_1) = \frac{\alpha_1}{\alpha_2}(\rho_1 + \rho_2 - (\rho_2 - \rho_3)\,\mathrm{sn}^2(\tau; k)), \tag{4.15b}$$

$$\theta_1(T_1) = \theta_1(0) + \frac{(\alpha_2/\alpha_1)^{\frac{1}{2}} L}{\rho_3(\rho_3 - \rho_1)^{\frac{1}{2}}}\,\Pi\left(1 - \frac{\rho_2}{\rho_3}; \tau | k\right), \tag{4.15c}$$

$$\theta_2(T_1) = \theta_2(0) + \frac{(\alpha_2/\alpha_1)^{\frac{1}{2}} L}{(\rho_1 + \rho_2)(\rho_3 - \rho_1)^{\frac{1}{2}}}\,\Pi\left(\frac{\rho_2 - \rho_3}{\rho_1 + \rho_2}; \tau | k\right), \tag{4.15d}$$

where sn is Jacobi's elliptic function and Π is the incomplete elliptic integral of the third kind (Abramowitz & Stegun 1964) with $\tau \equiv [(\rho_3 - \rho_1)/(\alpha_1\alpha_2)]^{\frac{1}{2}} T_1$ and $k \equiv (\rho_3 - \rho_2)/(\rho_3 - \rho_1)$. The first-order solution (4*b*) then consists of periodic amplitude modulations between the two largest roots (ρ_2, ρ_3) together with periodic phase modulations of the same period superimposed on a slow linear frequency shift on the slow timescale, which is similar to the $O(\epsilon^2)$ Poincaré correction to the frequency for regular oscillations.

Phase-plane plots for the amplitudes of the two modes are shown in figure 1.

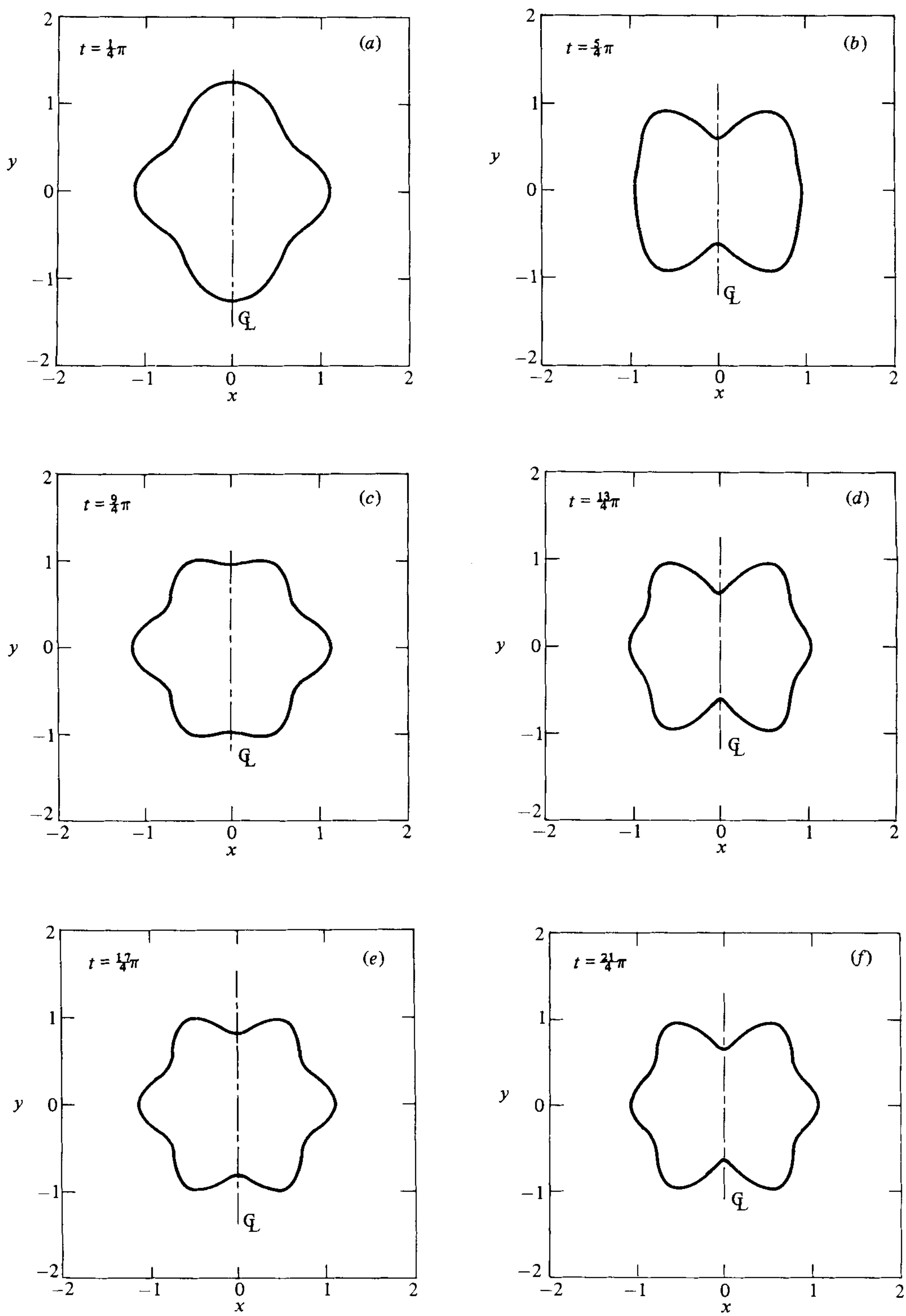

FIGURE 2. Shapes of drops initially perturbed by an $n = 4$ mode with $Q = (\frac{32}{3}\pi)^{\frac{1}{2}}$ and $\epsilon = 0.2$.

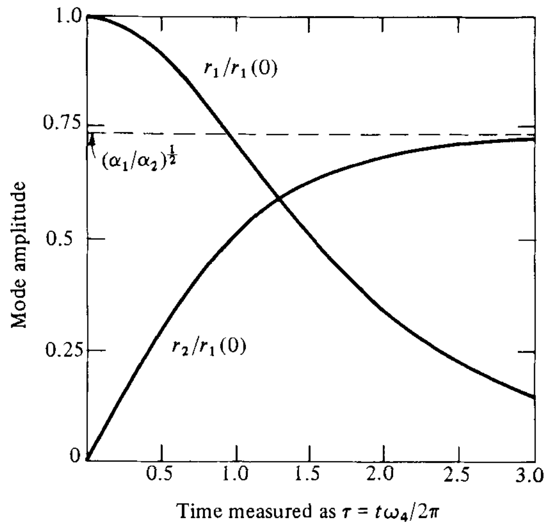

FIGURE 3. Aperiodic modulation of the amplitude of the fundamental and the resonating mode with $Q = (\frac{32}{3}\pi)^{\frac{1}{2}}$ and $\epsilon = 0.2$.

For initial conditions such that $L = 0$, the individual phases of the two interacting modes are constant and the modal amplitudes follow the outermost trajectory in figure 1. Then the initial condition corresponds to purely four-lobed deformation ($r_1(0) = R_1$, $r_2(0) = 0$), and the general solution (4.15) simplifies to

$$r_1(T_1) = R_1 \operatorname{sech} \frac{R_1 T_1}{(\alpha_1 \alpha_2)^{\frac{1}{2}}}, \tag{4.16a}$$

$$r_2(T_1) = R_1 \left(\frac{\alpha_1}{\alpha_2}\right)^{\frac{1}{2}} \tanh \frac{R_1 T_1}{(\alpha_1 \alpha_2)^{\frac{1}{2}}}, \tag{4.16b}$$

$$\theta_2 - 2\theta_1 = \pm\tfrac{1}{2}\pi. \tag{4.16c}$$

At exact resonance a purely four-lobed oscillation of any amplitude cannot persist, but transforms into a six-lobed oscillation within less than three periods of the initially excited mode. Drop shapes for this case are shown in figure 2 for the initial phase conditions $\theta_1(0) = 0$ and $\theta_2(0) = \frac{1}{2}\pi$ with $\epsilon = 0.2$.

The amplitudes of the interacting four- and six-lobed modes are given in figure 3 as functions of time for the same conditions used to calculate the drop shapes in figure 2. When $\epsilon = 0.2$ the $n = 4$ mode has decreased to 71.6 % of its maximum value and the $n = 6$ mode has reached 69.8 % of its maximum value within one oscillation cycle. The corresponding percentages after three cycles are 14.9 % and 98.9 % for the $n = 4$ and $n = 6$ modes respectively. Because of this transient in the $O(\epsilon)$ solution, the complete solution of the second-order problem will initially have terms proportional to the $n = 2, 4, 6$ and 8 Legendre modes, but after about three cycles the shape will have significant components from $n = 10$ and $n = 12$ modes.

The inner trajectories in figure 1 correspond to initial conditions such that $0 < L^2 < 4E^3/27\alpha_1^2\alpha_2 \equiv L_{\mathrm{m}}^2$ for which the disturbances are combinations of four- and six-lobed shapes with continuous modulations of both amplitude and phase. These deformations continuously exchange energy during oscillations at a frequency which is slightly modulated about the mean value. The relative phase $\hat{\theta}$ between these modes always falls between $-\frac{1}{2}\pi < -\hat{\theta}_{\mathrm{m}} \leqslant \hat{\theta} \leqslant \hat{\theta}_{\mathrm{m}} < \frac{1}{2}\pi$, where $\hat{\theta}_{\mathrm{m}} = \cos^{-1}(L/L_{\mathrm{m}})$. This phase is maximum when the amplitudes are undergoing maximum growth ($\alpha_1 r_1^2 = 2\alpha_2 r_2^2$), and is zero when the rate of change of the amplitudes is zero.

The timescale for these resonant oscillations is understood by considering the case $L^2 = \frac{1}{2}L^2_{max}$. If the initial amplitude of the deformation is taken as $\epsilon = 0.2$, one full cycle of the energy exchange between the four- and six-lobed modes occurs in

$$\frac{\omega_4}{\epsilon\pi}\left(\frac{\alpha_1\alpha_2}{\rho_3-\rho_1}\right)^{\frac{1}{2}} \approx \frac{0.8256}{E^{\frac{1}{2}}}$$

cycles for the $n = 4$ fundamental oscillation. This implies that the time for the resonant interaction is inversely proportional to the square root of the total energy input E or the initial amplitude ϵ of the oscillations.

For initial conditions such that $L = L_{max}$, both the amplitude and phase modulations present for $0 < L < L_{max}$ vanish entirely and the timescale of the resonant interaction between the $n = 4$ and $n = 6$ modes has dropped to zero. The trajectories for this initial condition are represented by the single point on each of the phase-plane plots shown in figure 1. The general solution of the modulation equations (4.15) reduces in this case to

$$\tfrac{1}{2}\alpha_1 r_1^2 = \alpha_2 r_2^2 = \tfrac{1}{3}E, \tag{4.17a}$$

$$2\theta_1 = \theta_2 = \pm\left(\frac{2}{\alpha_1\alpha_2}\right)^{\frac{1}{2}} r_1 T_1, \tag{4.17b}$$

where we have assumed $\theta_1(0) = 0$.

4.2. *Oscillations near resonance*

Variations in the oscillation frequencies of the four- and six-lobed motions caused by the dependence of these frequencies on the amplitude and small differences between the true net charge and the value Q_r will detune the resonance. The effect of this detuning on the drop motion is considered by analysing (4.10) with $\lambda \neq 0$. With the substitution

$$W(T_1) \equiv \mathrm{i}A^{*2}B\,\mathrm{e}^{-\mathrm{i}NT_1} \tag{4.18}$$

(4.10) are reduced to

$$\alpha_1\frac{\mathrm{d}}{\mathrm{d}T_1}(AA^*) = -\alpha_2\frac{\mathrm{d}}{\mathrm{d}T_1}(BB^*) = 2\,\mathrm{Re}\,(W), \tag{4.19}$$

so that an integral quantity analogous to E in (4.12) is defined as

$$E = \alpha_1 AA^* + \alpha_2 BB^*, \tag{4.20}$$

and is independent of the detuning parameter λ, which appears only in the coefficients α_3 and α_4. To derive a condition equivalent to (4.13) we introduce a real-valued function

$$Z(T_1) \equiv \alpha_1(R_1^2 - AA^*) \equiv \alpha_2(BB^* - R_2^2), \tag{4.21}$$

where R_1 and R_2 are the moduli of the initial amplitudes of the $n = 4$ and $n = 6$ modes respectively. Substituting (4.18) and (4.21) into the set (4.10) leads to the second integral

$$(2\alpha_3 - \alpha_4 + N)Z - 2L = 2\,\mathrm{Im}\,(W). \tag{4.22}$$

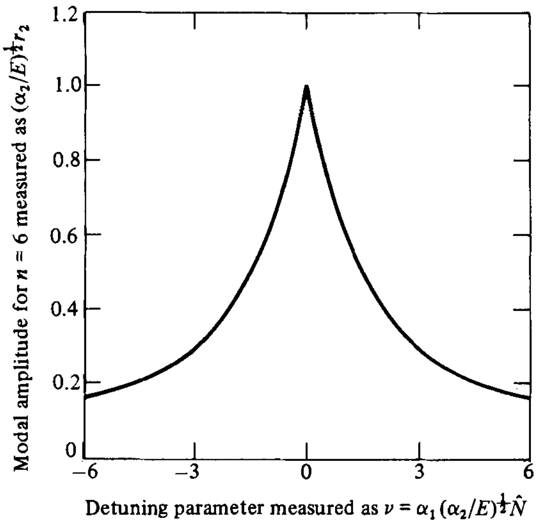

FIGURE 4. Response of the amplitude of the second harmonic near resonance as a function of the detuning parameter $\hat{N}$.

Combining (4.18)–(4.22) gives the general equation

$$\left(\frac{\mathrm{d}Z}{\mathrm{d}T_1}\right)^2 = 4\left\{\left(R_1^2-\frac{Z}{\alpha_1}\right)\left(R_2^2+\frac{Z}{\alpha_2}\right)-[L-\tfrac{1}{2}(2\alpha_3-\alpha_4+N)\,Z]^2\right\}, \tag{4.23}$$

which can be integrated in terms of elliptic integrals.

We consider the solution of (4.23) only in the case when the initial condition is composed of only the fundamental mode ($R_2 = 0$). For this situation it is easily shown that $W(0) = Z(0) = L = 0$, $E = \alpha_1 R_1^2$, and (4.23) reduces to

$$\left(\frac{\mathrm{d}Z}{\mathrm{d}T_1}\right)^2 = 4Z\left\{\frac{1}{\alpha_1^2\alpha_2}(E-Z)^2-\hat{N}^2Z\right\}, \tag{4.24}$$

where $\hat{N} = \frac{1}{2}(2\alpha_3-\alpha_4+N)$; this constant is somewhat different from the one derived for planar waves by McGoldrick (1972) because of the existence of the linear terms in (4.10). The quadratic polynomial in Z in (4.24) has two distinct positive roots, $0 \leqslant \rho_1 \leqslant E \leqslant \rho_2$, if $\hat{N} \neq 0$ or two roots equal to E if $\hat{N} \neq 0$. This latter case corresponds to the situation at exact resonance ($\lambda = 0$) and (4.24) can be easily integrated to yield

$$Z(T_1) = E\tanh^2\left[\frac{2T_1}{\alpha_1}\left(\frac{E}{\alpha_2}\right)^{\frac{1}{2}}\right], \tag{4.25}$$

which when combined with the definition (4.21) can be reduced to a form similar to (4.16). The initial value $Z(0)$ is equal to E and is the maximum value of the four-lobed component of the oscillation.

When detuning occurs $\hat{N}$ is not zero and the general solution of (4.24) is an oscillation of finite period with value between the smaller root of (4.24)

$$\rho_1 = \frac{\alpha_1^2\alpha_2\hat{N}^2}{2}\left\{1+\frac{2E}{\alpha_1^2\alpha_2\hat{N}^2}-\left(1+\frac{4E}{\alpha_1^2\alpha_2\hat{N}^2}\right)^{\frac{1}{2}}\right\} \tag{4.26}$$

and zero. The root ρ_1 is the largest value of Z obtained during the detuned oscillations. The effect of detuning on the energy transfer between the initial fundamental ($n = 4$)

and the second harmonic ($n = 6$) is shown in figure 4 by a plot of the maximum amplitude of the six-lobed component as a function of $\hat{N}$ which measures the variation of Q from Q_r. The amplitude of the $n = 6$ component is scaled with its asymptotic value $(E/\alpha_2)^{\frac{1}{2}}$ for exact resonance. The ability of the resonant mechanism to transfer energy between the fundamental and its second harmonic decreases as $\hat{N}^{-1}$ or λ^{-1}. As this plot indicates there is a band of frequencies and therefore electric charges near Q_r for which the resonant interaction is most effective.

5. Third harmonic resonance

Resonance between the fundamental mode and one of its third-order harmonics occurs for charged drops at particular values of Q. In particular, the fundamental two-lobed motion resonates with its four-lobed harmonic for $Q = 0$, i.e. for uncharged inviscid drops. Also, the $n = 3$ fundamental and its $n = 5$ harmonic resonate at $Q = (\frac{260}{17}\pi)^{\frac{1}{2}}$ and the $n = 4$ fundamental and its $n = 8$ harmonic do so at $Q = (\frac{88}{13}\pi)^{\frac{1}{2}}$. In each case the $O(\epsilon)$ solution $F^{(1)}(T_0, T_1, T_2)$ must be taken to be a combination of the fundamental and resonant harmonic modes and the amplitude modulations (corresponding to $A(T_1, T_2)$ and $B(T_1, T_2)$ in (4.6)) determined from the solvability of the second- and third-order problems. The solvability condition at $O(\epsilon^2)$ will guarantee that the amplitude modulations are only functions of the slowest timescale T_2, and the conditions at $O(\epsilon^3)$ give equations like (4.10) which govern $A(T_2)$ and $B(T_2)$. The derivation of these equations and their solution will be tedious, as suggested by the form of the third-order problem far from resonance given in Appendix C. We have not pursued this work; see McGoldrick (1972) and Nayfeh (1971) for the parallel analysis for capillary–gravity waves. McGoldrick *et al.* (1966) have also observed these third-order interactions experimentally.

The third harmonic resonance for an uncharged drop set into motion from an initial deformation with components of both the $n = 2$ fundamental and $n = 4$ harmonic modes will appear as a continuous and periodic modulation of amplitude for these modes in time with a frequency which is proportional to ϵ^2. For the case of $\epsilon = 0.2$ discussed earlier, this scaling implies that $O(10)$ oscillation cycles of the fundamental frequency will be needed to observe the resonant energy exchange.

6. Discussion

Moderate-amplitude oscillations of inviscid charged drops display an array of nonlinear dynamic phenomena as varied as those that have been observed for nearly planar water waves. Besides a decrease in oscillation frequency with amplitude caused by interactions between the fluid inertia and drop shape, resonant interactions between the fundamental mode and secondary and tertiary harmonics can completely change the pattern of the oscillation. In terms of the classifications used by McGoldrick (1972), these resonances are selective and weak. They are selective in that only particular combinations of the fundamental and its harmonic can resonate at particular values of the electrical charge Q. They are weak because the timescale for the resonant interaction is long when compared with a typical period for the fundamental oscillation.

The analysis presented in §4 for the second harmonic resonance of a four-lobed oscillation for $Q = Q_r$ shows three particular forms for this long-timescale response, depending on the initial deformation of the drop. An aperiodic drop motion is only possible when the initial deformation is composed of the fundamental $n = 4$ mode alone. Small changes in frequency caused by variation in Q from Q_r and oscillation amplitude detune the resonance so that the actual motion described a time-periodic exchange of energy between the $n = 4$ mode and its $n = 6$ harmonic. This type of periodic exchange occurs for more general initial deformations which include both modes and are accompanied by small frequency modulations about the mean.

For either of these periodic and aperiodic motions the oscillation pattern is distinct from the form valid far from resonance. In both cases, the amplitude of the harmonic mode has magnitude similar to the fundamental for the initial part of the motion. This change in pattern should be observable in calculations and experiments so long as data is collected for a period comparable to the long timescale. We feel that the second-harmonic resonance described in §4 will be easily observed, but that some of the cases of tertiary resonance described in §5 will be more difficult. The third type of resonant oscillation is strictly periodic with constant amplitude and phase, but is the most unlikely to be observed because of the precise ratio of initial amplitudes required.

Predictions of experimental observation of these three types of resonance must take into account the effects of the omnipresent viscosity of real drops, which will detune the interactions and cause damping. The quantitative effects of this detuning are difficult to calculate rigorously; however, its qualitative significance can be estimated from simple approximate calculations. Prosperetti (1980) has shown that the damping of small-amplitude oscillations of a viscous drop is governed by an integrodifferential equation which reduces to Lamb's (1932, §305) theory for a drop with small viscosity at short times when the initial disturbance is irrotational. For the resonant interactions predicted by the inviscid analysis to be observed in slightly viscous drops, the timescale for viscous dissipation, or equivalently of vorticity diffusion from the interface, must be much longer than the characteristic time for the inviscid motion:

$$\tau_n \equiv \frac{1}{(n-1)(2n+1)}\frac{R^2\rho}{\mu} \gg \left(\frac{\rho R^3}{\sigma}\right)^{\frac{1}{2}}, \tag{6.1}$$

where n is the primary mode of the oscillation. This condition is satisfied for water drops with n up to eight.

When there are a number of small-amplitude modes of the form given in (4.6), the orthogonality between each component guarantees that, within the order of approximation, each mode decays independently of the others. When the rate of energy transfer between resonantly coupled modes is independent of the viscosity, the amplitude equations for exact second-order resonance are modified to account for viscous damping and give

$$\alpha_1\frac{\mathrm{d}A}{\mathrm{d}T_1} = \mathrm{i}A^*B - \mathrm{i}\frac{\alpha_1}{\tau_4}A, \quad \alpha_2\frac{\mathrm{d}B}{\mathrm{d}T_1} = \mathrm{i}A^2 - \mathrm{i}\frac{\alpha_2}{\tau_6}B. \tag{6.2a,b}$$

The effect of viscosity on the resonant interactions of internal waves (Davis & Acrivos 1967) and capillary waves (McGoldrick 1970) have been derived using similar arguments. McGoldrick analysed the equivalent set of spatial equations in the (A, B) phase plane as a function of initial conditions.

Comparing (6.2) with (4.10) shows that the principal effect of a slight viscosity is to attenuate the two interacting modes at the same decay rates present in the absence of their interaction. Because the timescale for third-order resonance (uncharged drops) is an order of magnitude greater than the one discussed above, it is more comparable to the viscous timescale. As a result, these interactions will be more difficult to observe experimentally.

The experimental systems presently in use rely on acoustic pressure (Jacobi *et al.* 1981) or electric fields (Davis & Ray 1980) to position the drop. The acoustic field forces the drop to oscillate, and opens the possibility of parametrically excited oscillations. Levitation of a charged drop in a d.c. electric field allows the determination of the net charge, but deforms the drop and changes the oscillation frequencies. These deformations have recently been calculated for a static drop (Adornato & Brown 1983), and are small for the full range of values for the field strength and charge accessible before breakup. This suggests that the oscillation frequencies computed for small values of the field by Sample, Raghupathy & Hendricks (1970) are good approximations. Resonant oscillations are detuned even for small changes in these frequencies.

All the calculations presented here are restricted to charge values below the Rayleigh limit (1.1) for breakup of the spherical form. Introducing oscillation will interact with this limit, thus causing the drop to become unstable at lower values of the charge. The variation of the critical value of Q with the amplitude of the oscillation will be considered later.

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Appendix A

The mean curvature $2\mathscr{H}$ and normal tangent vectors are expanded in ϵ as

$$\begin{aligned}-2\mathscr{H} = {} & 2+\epsilon \sum_{i=2}^{\infty} (i-1)(i+2) F_i^{(1)} \\ & +\tfrac{1}{2}\epsilon^2 \left\{ \sum_{j=0}^{\infty} (j-1)(j+2) F_j^{(2)} - 4 \sum_{i=2}^{\infty} (i^2+i-1)(F_i^{(1)})^2 \right\} \\ & +\tfrac{1}{6}\epsilon^3 \left\{ \sum_{l=0}^{\infty} (l-1)(l+2) F_l^{(3)} + 18 \sum_{i=2}^{\infty} (i^2+i-\tfrac{2}{3})(F_i^{(1)})^3 \right. \\ & -6 \sum_{i=2}^{\infty} \sum_{j=0}^{\infty} (i^2+i+j^2+j-2) F_i^{(1)} F_j^{(2)} \\ & \left. +3 \sum_{i=2}^{\infty} (F_{i,\theta}^{(1)})^2 (3F_{i,\theta\theta}^{(1)} + \cot\theta F_{i,\theta}^{(1)}) \right\} \end{aligned} \qquad \text{(A 1)}$$

$$\begin{aligned}\boldsymbol{n} = {} & \{1-\tfrac{1}{2}\epsilon^2 (F_\theta^{(1)})^2 - \tfrac{1}{6}\epsilon^3 [3F_\theta^{(1)} F_\theta^{(2)} - 6F^{(1)} (F_\theta^{(1)})^2]\} \, \boldsymbol{e}_r - \{\epsilon F_\theta^{(1)} + \tfrac{1}{2}\epsilon^2 [F_\theta^{(2)} - 2F^{(1)} F_\theta^{(1)}] \\ & + \tfrac{1}{6}\epsilon^3 [F_\theta^{(3)} - 3F^{(1)} F_\theta^{(2)} - 3(F_\theta^{(1)})^3 - 3F^{(2)} F_\theta^{(1)} + 6(F^{(1)})^2 F_\theta^{(1)}]\} \, \boldsymbol{e}_\theta, \end{aligned} \qquad \text{(A 2)}$$

$$\begin{aligned}\boldsymbol{t} = {} & \{\epsilon F_\theta^{(1)} + \tfrac{1}{2}\epsilon^2 [F_\theta^{(2)} - 2F^{(1)} F_\theta^{(1)}] + \tfrac{1}{6}\epsilon^3 [F_\theta^3 - 3F^{(1)} F_\theta^{(2)} - 3(F_\theta^{(1)})^3 \\ & - 3F^{(2)} F_\theta^{(1)} + 6(F^{(1)})^2 F_\theta^{(1)}]\} \, \boldsymbol{e}_r \\ & + \{1-\tfrac{1}{2}\epsilon^2 (F_\theta^{(1)})^2 - \tfrac{1}{6}\epsilon^3 [3F_\theta^{(1)} F_\theta^{(2)} - 6F^{(1)} (F_\theta^{(1)})^2]\} \, \boldsymbol{e}_\theta. \end{aligned} \qquad \text{(A 3)}$$

Appendix B

The equations that govern the second-order corrections are

$$\nabla^2\phi^{(2)} = 0 \quad (0 \leqslant \eta \leqslant 1, \quad 0 \leqslant \theta \leqslant \pi), \tag{B 1}$$

$$\phi^{(2)}_\eta = 0 \quad (\eta = 0, \quad 0 \leqslant \theta \leqslant \pi), \tag{B 2}$$

$$\phi^{(2)}_\eta - F^{(2)}_{T_0} = -2F^{(1)}\phi^{(1)}_{\eta\eta} + 2F^{(1)}_{T_1} + 2F^{(1)}_\theta \phi^{(1)}_\theta \quad (\eta = 1, \quad 0 \leqslant \theta \leqslant \pi), \tag{B 3}$$

$$\begin{aligned}&\phi^{(2)}_{T_0} + 2F^{(1)}\phi^{(1)}_{T_0\eta} + 2\phi^{(1)}_{T_1} + (\phi^{(1)}_\eta)^2 + (\phi^{(1)}_\theta)^2 - \frac{1}{4\pi}(V^{(1)}_\eta + F^{(1)}V^{(0)}_{\eta\eta})^2 \\ &\quad - \frac{1}{4\pi} V^{(0)}_\eta [V^{(2)}_\eta - (F^{(1)}_\theta)^2 V^{(0)}_\eta + 2F^{(1)}V^{(1)}_{\eta\eta} + F^{(2)}V^{(0)}_{\eta\eta} + (F^{(1)})^2 V^{(0)}_{\eta\eta\eta} - 2F^{(1)}_\theta V^{(1)}_\theta] \\ &= -\sum_{m=0}^{\infty} (m-1)(m+2)F^{(2)} + 4(F^{(1)})^2(n^2+n-1) \quad (\eta = 1, \quad 0 \leqslant \theta \leqslant \pi),\end{aligned} \tag{B 4}$$

$$\int_0^\pi (2(F^{(1)})^2 + F^{(2)}) \sin\theta \, \mathrm{d}\theta = 0, \tag{B 5}$$

$$\nabla^2 V^{(2)} = 0 \quad (1 \leqslant \eta \leqslant \infty, \quad 0 \leqslant \theta \leqslant \pi), \tag{B 6}$$

$$V^{(2)} = 0 \quad (\eta \rightarrow \infty, \quad 0 \leqslant \theta \leqslant \pi), \tag{B 7}$$

$$\begin{aligned}&\int_0^\pi [V^{(2)}_\eta + F^{(2)}V^{(0)}_{\eta\eta} + 2F^{(1)}V^{(1)}_{\eta\eta} + (F^{(1)})^2 V^{(0)}_{\eta\eta\eta} - (F^{(1)}_\theta)^2 V^{(0)}_\eta \\ &\quad - 2F^{(1)}_\theta V^{(1)}_\theta] \sin\theta \, \mathrm{d}\theta + 4\int_0^\pi F^{(1)}(V^{(1)}_\eta + F^{(1)}V^{(0)}_{\eta\eta}) \sin\theta \, \mathrm{d}\theta \\ &\quad - Q^{(0)} \int_0^\pi [2F^{(2)} + 2(F^{(1)})^2 + (F^{(1)}_\theta)^2] \sin\theta \, \mathrm{d}\theta = 0 \quad (\eta = 1),\end{aligned} \tag{B 8}$$

$$\begin{aligned}&V^{(2)}_\theta + 2F^{(1)}(V^{(1)}_{\theta\eta} - V^{(1)}_\theta) + V^{(0)}_\eta(F^{(2)}_\theta - 2F^{(1)}F^{(1)}_\theta) + 2F^{(1)}_\theta(V^{(1)}_\eta + F^{(1)}V^{(0)}_{\eta\eta}) = 0 \\ &\qquad (\eta = 1, \quad 0 \leqslant \theta \leqslant \pi),\end{aligned} \tag{B 9}$$

$$F^{(2)}(\theta, 0, 0, 0) = \begin{cases} -\frac{2}{5} & (n = 2), \\ -\frac{2}{7} & (n = 3), \\ -\frac{2}{9} & (n = 4), \end{cases} \tag{B 10}$$

$$2F^{(1)}_{T_1} + F^{(2)}_{T_0} = 0 \quad (T_0 = T_1 = T_2 = 0). \tag{B 11}$$

The non-zero coefficients in the second-order correction (3.21) to drop shape are

$$\left.\begin{aligned}
L_{220} &= -\tfrac{2}{5}\cos^2\psi_2, \\
L_{222} &= -G_2 \cos(\omega_2 T_0 + \tfrac{1}{21}(G_{21}\cos 2\psi_1 + G_{22}), \\
L_{224} &= -\tfrac{1}{3}G_4 \cos\omega_4 T_0 + \tfrac{48}{35}(G_{41}\cos 2\psi_2 + G_{42}), \\
L_{230} &= -\tfrac{2}{7}\cos^2\psi_3, \\
L_{232} &= -H_2 \cos\omega_2 T_0 + \tfrac{8}{21}(H_{21}\cos 2\psi_3 + H_{22}), \\
L_{234} &= -\tfrac{1}{3}H_4 \cos\omega_4 T_0 + \tfrac{144}{77}(H_{41}\cos 2\psi_3 + H_{42}), \\
L_{236} &= -\tfrac{1}{5}H_6 \cos\omega_6 T_0 + \tfrac{600}{77}(H_{61}\cos 2\psi_3 + H_{62}), \\
L_{240} &= -\tfrac{2}{9}\cos^2\psi_4, \\
L_{242} &= -I_2 \cos\omega_2 T_0 + \tfrac{50}{693}(I_{21}\cos 2\psi_4 + I_{22}), \\
L_{244} &= -\tfrac{1}{3}I_4 \cos\omega_4 T_0 + \tfrac{54}{1001}(I_{41}\cos 2\psi_4 + I_{42}), \\
L_{246} &= -\tfrac{1}{5}I_6 \cos\omega_6 T_0 + \tfrac{10}{11}(I_{61}\cos 2\psi_4 + I_{62}), \\
L_{248} &= -\tfrac{1}{7}I_8 \cos\omega_8 T_0 + \tfrac{15680}{1287}(I_{81}\cos 2\psi_4 + I_{82}),
\end{aligned}\right\} \tag{B 12}$$

where

$$
\left.
\begin{aligned}
&G_{21} = \frac{23\hat{Q}-116}{\omega_2^2}, \quad G_{22} = \frac{132-15\hat{Q}}{\omega_2^2}, \quad G_2 \equiv \tfrac{1}{21}(G_{21}+G_{22}),\\
&G_{41} = -\frac{9\hat{Q}^2+54\hat{Q}-648}{(\omega_4^2-4\omega_2^2)\,\omega_4^2}, \quad G_{42} = \frac{15\hat{Q}^2-258\hat{Q}+1080}{(\omega_4^2-4\omega_2^2)\,\omega_4^2}, \quad G_4 \equiv \tfrac{144}{35}(G_{41}+G_{42}),\\
&H_{21} = \frac{39\hat{Q}^2-380\hat{Q}+896}{(\omega_2^2-4\omega_3^2)\,\omega_2^2}, \quad H_{22} \equiv -\frac{33\hat{Q}^2-652\hat{Q}+2464}{(\omega_2^2-4\omega_3^2)\,\omega_2^2}, \quad H_2 \equiv \tfrac{8}{21}(H_{21}+H_{22}),\\
&H_{41} \equiv \frac{28\hat{Q}^2-470\hat{Q}+1812}{(\omega_4^2-4\omega_3^2)\,\omega_4^2}, \quad H_{42} \equiv -\frac{8\hat{Q}^2-234\hat{Q}+808}{(\omega_4^2-4\omega_3^2)\,\omega_4^2}, \quad H_4 = \tfrac{432}{77}(H_{41}+H_{42}),\\
&H_{61} \equiv \frac{-55\hat{Q}^2+420\hat{Q}+160}{(\omega_6^2-4\omega_3^2)\,\omega_6^2}, \quad H_{62} \equiv \frac{9\hat{Q}^2-284\hat{Q}+2080}{(\omega_6^2-4\omega_3^2)\,\omega_6^2}, \quad H_6 \equiv \tfrac{3000}{77}(H_{61}+H_{62}),\\
&I_{21} = \frac{331\hat{Q}^2-3518\hat{Q}+8776}{(\omega_2^2-4\omega_4^2)\,\omega_2^3}, \quad I_{22} = -\frac{299\hat{Q}^2-8398\hat{Q}+40040}{(\omega_2^2-4\omega_4^2)\,\omega_2^2}, \quad I_2 = \tfrac{50}{693}(I_{21}+I_{22}),\\
&I_{41} = \frac{65\hat{Q}-602}{\omega_4^2}, \quad I_{42} = \frac{3\hat{Q}+618}{\omega_4^2}, \quad I_4 = \tfrac{162}{1001}(I_{41}+I_{42}),\\
&I_{61} = -\frac{15\hat{Q}^2+2510\hat{Q}-21040}{(\omega_6^2-4\omega_4^2)\,\omega_6^2}, \quad I_{62} = -\frac{33\hat{Q}-1918\hat{Q}+4880}{(\omega_6^2-4\omega_4^2)\,\omega_6^2}, \quad I_6 = \tfrac{50}{11}(I_{61}+I_{62}),\\
&I_{81} = -\frac{203\hat{Q}^2-2254\hat{Q}+2240}{(\omega_8^2-4\omega_4^2)\,\omega_8^2}, \quad I_{82} = \frac{13\hat{Q}^2-662\hat{Q}+7480}{(\omega_8^2-4\omega_4^2)\,\omega_8^2}, \quad I_8 = \tfrac{109760}{1287}(I_{81}+I_{82}),
\end{aligned}
\right\} \tag{B 13}
$$

with $\hat{Q} \equiv Q^2/4\pi$.

Appendix C

The equations that govern the third-order problem are

$$\nabla^2\phi^{(3)} = 0 \quad (0 \leqslant \eta \leqslant 1, \quad 0 \leqslant \theta \leqslant \pi), \tag{C 1}$$

$$\phi^{(3)}_{\eta} = 0 \quad (\eta = 0, \quad 0 \leqslant \theta \leqslant \pi), \tag{C 2}$$

$$
\begin{aligned}
\phi^{(3)}_{\eta} - F^{(3)}_{T_0} = {} & -3F^{(2)}\phi^{(1)}_{\eta\eta} - 3F^{(1)}\phi^{(2)}_{\eta\eta} - 3(F^{(1)})^2\,\phi^{(1)}_{\eta\eta\eta} + 3F^{(1)}_{T_2}\\
& + 3F^{(2)}_{T_1} + 3F^{(2)}_{\theta}\,\phi^{(1)}_{\theta} + 3F^{(1)}_{\theta}(\phi^{(2)}_{\theta} + 2F^{(1)}\phi^{(1)}_{\eta\theta} - 4F^{(1)}\phi^{(1)}_{\theta}),
\end{aligned} \tag{C 3}
$$

$$
\begin{aligned}
&\phi^{(3)}_{T_0} + 3F^{(2)}\phi^{(1)}_{T_0\eta} + 3\phi^{(2)}_{T_1} + 3\phi^{(1)}_{T_2} + 3F^{(1)}(\phi^{(2)}_{T_0\eta} + F^{(1)}\phi^{(1)}_{T_0\eta\eta} + 2\phi^{(1)}_{T_1\eta})\\
&\quad + 3\phi^{(1)}_{\eta}\,(\phi^{(2)}_{\eta} + 2F^{(1)}\phi^{(1)}_{\eta\eta}) + 3\phi^{(1)}_{\theta}\,(\phi^{(2)}_{\theta} + 2F^{(1)}\phi^{(1)}_{\theta\eta})\\
&\quad - \frac{3}{4\pi}\,(V^{(1)}_{\eta} + F^{(1)}\,V^{(0)}_{\eta\eta})\,((F^{(1)})^2\,V^{(0)}_{\eta\eta\eta} - V^{(0)}_{\eta}(F^{(1)}_{\theta})^2 + 2F^{(1)}\,V^{(1)}_{\eta\eta}\\
&\quad + F^{(2)}\,V^{(0)}_{\eta\eta} + V^{(2)}_{\eta} - 2F^{(1)}_{\theta}\,V^{(1)}_{\theta})\\
&\quad - \frac{1}{4\pi}\,V^{(0)}_{\eta}\,\{3\,V^{(0)}_{\eta}\,[2F^{(1)}(F^{(1)}_{\theta})^2 - F^{(1)}_{\theta}F^{(2)}_{\theta}] - 3\,V^{(1)}_{\eta}\,(F^{(1)}_{\theta})^2\\
&\quad + (F^{(1)})^2\,[3\,V^{(1)}_{\eta\eta\eta} + F^{(1)}\,V^{(0)}_{\eta\eta\eta\eta}] + 3F^{(1)}[\,V^{(2)}_{\eta\eta} + F^{(2)}\,V^{(0)}_{\eta\eta\eta} - V^{(0)}_{\eta\eta}(F^{(1)}_{\theta})^2]\\
&\quad + F^{(3)}\,V^{(0)}_{\eta\eta} + 3F^{(2)}\,V^{(1)}_{\eta\eta} + V^{(3)}_{\eta} - 3\,V^{(1)}_{\theta}\,(F^{(2)}_{\theta} - 4F^{(1)}F^{(1)}_{\theta})\\
&\qquad\qquad - 3F^{(1)}_{\theta}\,(V^{(2)}_{\theta} + 2F^{(1)}\,V^{(1)}_{\theta\eta})\} = (2\mathscr{H})^{(3)},
\end{aligned} \tag{C 4}
$$

$$\int_0^\pi [6F^{(1)}F^{(2)}+2(F^{(1)})^3+F^{(3)}]\sin\theta\,\mathrm{d}\theta = 0, \tag{C 5}$$

$$\nabla^2 V^{(3)} = 0 \quad (1 \leqslant \eta \leqslant \infty, \quad 0 \leqslant \theta \leqslant \pi), \tag{C 6}$$

$$V^{(3)} = 0 \quad (\eta \to \infty, \quad 0 \leqslant \theta \leqslant \pi), \tag{C 7}$$

$$\begin{aligned}
&\int_0^\pi \{3V_\eta^{0)} F_\theta^{(1)}(2F^{(1)}F_\theta^{(1)} - F_\theta^{(2)}) - 3(F_\theta^{(1)})^2 V_\eta^{(1)} \\
&\quad + (F^{(1)})^2 (F^{(1)} V^{(0)}_{\eta\eta\eta\eta} + 3V^{(1)}_{\eta\eta\eta}) + F^{(3)} V^{(0)}_{\eta\eta} + 3F^{(2)} V^{(1)}_{\eta\eta} + V^{(3)}_\eta \\
&\quad + 3F^{(1)}[V^{(2)}_{\eta\eta} + F^{(2)} V^{(0)}_{\eta\eta\eta} - (F_\theta^{(1)})^2 V^{(0)}_{\eta\eta}] - 3F_\theta^{(1)}(V_\theta^{(2)} + 2F^{(1)} V^{(1)}_{\theta\eta}) \\
&\quad + 3V_\theta^{(1)}(4F^{(1)}F_\theta^{(1)} - F_\theta^{(2)})\}\sin\theta\,\mathrm{d}\theta \\
&\quad + 6\int_0^\pi F^{(1)}[V_\eta^{(2)} + F^{(2)} V^{(0)}_{\eta\eta} + F^{(1)}(2V^{(1)}_{\eta\eta} + F^{(1)} V^{(0)}_{\eta\eta\eta}) - F_\theta^{(1)}(2V_\theta^{(1)} + F_\theta^{(1)} V_\eta^{(0)})]\sin\theta\,\mathrm{d}\theta \\
&\quad + 3\int_0^\pi (V_\eta^{(1)} + F^{(1)} V^{(0)})[2F^{(2)} + 2(F^{(1)})^2 + F_\theta^{(1)})^2]\sin\theta\,\mathrm{d}\theta \\
&\qquad - Q^{(0)}\int_0^\pi [2F^{(3)} + 6F^{(1)}F^{(2)} + 3F_\theta^{(1)} F_\theta^2]\sin\theta\,\mathrm{d}\theta = 0 \quad (\eta = 1),
\end{aligned} \tag{C 8}$$

$$\begin{aligned}
&V_\theta^{(3)} + 3F^{(2)} V^{(1)}_{\theta\eta} + 3F^{(1)}(V^{(2)}_{\theta\eta} + F^{(1)} V^{(1)}_{\theta\eta\eta}) \\
&\quad - 3V_\theta^{(1)}[F^{(2)} - 2(F^{(1)})^2 + (F_\theta^{(1)})^2] \\
&\quad - 3F^{(1)}(V_\theta^{(2)} + 2F^{(1)} V^{(1)}_{\theta\eta}) + 3F_\theta^{(1)}(F^{(2)} V^{(0)}_{\eta\eta} + V_\eta^{(2)}) \\
&\quad + V_\eta^{(0)}[3F^{(1)}(2F^{(1)}F_\theta^{(1)} - F_\theta^{(2)}) - 3F_\theta^{(1)}(F^{(2)} + (F_\theta^{(1)})^2) + F_\theta^{(3)}] \\
&\quad + 3V_\eta^{(1)}(F_\theta^{(2)} - 2F^{(1)}F_\theta^{(1)}) + 3(F^{(1)})^2 F_\theta^{(1)}(V^{(0)}_{\eta\eta\eta} - 2V^{(0)}_{\eta\eta}) \\
&\qquad + 3F^{(1)}(F_\theta^{(2)} V^{(0)}_{\eta\eta} + 2F_\theta^{(1)} V^{(1)}_{\eta\eta}) = 0 \\
&\qquad\qquad (\eta = 1, \quad 0 \leqslant \theta \leqslant \pi),
\end{aligned} \tag{C 9}$$

$$F^{(3)}(\theta,0,0,0) = \begin{cases} -\frac{4}{35} & (\eta = 2), \\ 0 & (\eta = 3), \\ -\frac{36}{1001} & (\eta = 4), \end{cases} \tag{C 10}$$

$$3F^{(1)}_{T_2} + 3F^{(2)}_{T_1} + F^{(3)}_{T_0} = 0 \quad (T_0 = T_1 = T_2 = 0). \tag{C 11}$$

The coefficients in the frequency corrections (3.23) are

	$L = 2$	$L = 3$	$L = 4$
A_{10}	6608528	4214241024000	55735591155609600
A_{11}	−4961440	−4128178176000	−77949491906388480
A_{12}	1419804	1682216124000	45351373912349312
A_{13}	−177168	−362825358328	−14555386948486656
A_{14}	7945	43303979512	2840187292166640
A_{15}	—	−2679419780	−345863703031648
A_{16}	—	66094721	25619763735024
A_{17}	—	—	−1049270108016
A_{18}	—	—	18006768899

REFERENCES

ABRAMOWITZ, M. & STEGUN, I. A. 1964 *Handbook of Mathematical Functions*, chaps 16 and 17. National Bureau of Standards, Washington, D.C.

ADORNATO, P. M. & BROWN, R. A. 1983 Shape and stability of electrostatically levitated drops. *Proc. R. Soc. Lond.* A **389**, 101–117.

ALONSO, C. T. 1974 The dynamics of colliding and oscillating drops. In *Proc. Intl Colloq. on Drops and Bubbles* (ed. D. J. Collins, M. S. Plesset & M. M. Saffren). Jet Propulsion Laboratory.

BASARAN, O. A., AMUNDSON, K. R., PATZEK, T. W., BENNER, R. E. & SCRIVEN, L. E. 1982 Deformation, oscillation and break-up of charged drops in electric field. *Bull. Am. Phys. Soc.* **27**, 1168.

BENNEY, D. J. 1962 Non-linear gravity wave interactions. *J. Fluid Mech.* **14**, 577–584.

BRAZIER-SMITH, P. R., BROOK, M., LATHAM, J., SAUNDERS, C. P. R. & SMITH, M. H. 1971 The vibration of electrified water drops. *Proc. R. Soc. Lond.* A **322**, 523–534.

BRETHERTON, F. P. 1964 Resonant interactions between waves. The case of discrete oscillations. *J. Fluid Mech.* **20**, 457–479.

BUPARA, S. S. 1965 Spontaneous movements of small round bodies in viscous fluids. Ph.D. thesis, University of Minnesota.

COHEN, S. & SWIATECKI, W. J. 1962 The deformation energy of a charged drop. IV: Evidence for a discontinuity in the conventional family of saddle point shapes. *Ann. of Phys.* **19**, 67–164.

COHEN, S. & SWIATECKI, W. J. 1963 The deformation energy of a charged drop. V: Results of electronic computer studies. *Ann. of Phys.* **22**, 406–437.

DAVIS, R. E. & ACRIVOS, A. 1967 The stability of oscillatory internal waves. *J. Fluid Mech.* **30**, 723–736.

DAVIS, E. I. & RAY, A. K. 1980 Single aerosol particle size and mass measurements using an electrodynamic balance. *J. Colloid Interface Sci.* **75**, 566–576.

HASSE, R. W. 1975 Inertia, friction and angular momentum of an oscillating viscous charged liquid drop under surface tension. *Ann. of Phys.* **93**, 68–87.

HENDRICKS, C. D. & SCHNEIDER, J. M. 1963 Stability of a conducting droplet under the influence of surface tension and electrostatic forces. *Am. J. Phys.* **31**, 450–453.

JACOBI, N., CROONQUIST, A. P., ELLEMAN, D. D. & WANG, T. G. 1981 Acoustically induced oscillation and rotation of a large drop in space. In *Proc. 2nd Intl Colloq. on Drops and Bubbles* (ed. D. H. Le Croissette). Jet Propulsion Laboratory.

JOSEPH, D. D. 1973 Domain perturbations: the higher order theory of infinitesimal water waves. *Arch. Rat. Mech. Anal.* **51**, 295–303.

LAMB, H. 1932 *Hydrodynamics*, 6th edn. Cambridge University Press.

MCGOLDRICK, L. F. 1965 Resonant interactions among capillary–gravity waves. *J. Fluid Mech.* **21**, 305–331.

MCGOLDRICK, L. F. 1970*a* An experiment on second-order capillary–gravity resonant wave interactions. *J. Fluid Mech.* **40**, 251–271.

MCGOLDRICK, L. F. 1970*b* On Wilton's ripples: a special case of resonant interactions. *J. Fluid Mech.* **42**, 193–200.

MCGOLDRICK, L. F. 1972 On the rippling of small waves: a harmonic nonlinear nearly resonant interaction. *J. Fluid Mech.* **52**, 725–751.

MCGOLDRICK, L. F., PHILLIPS, O. M., HUNG, N. E. & HODGSON, T. H. 1966 Measurements of third-order resonant wave interactions. *J. Fluid Mech.* **25**, 437–456.

MOECKEL, G. P. 1975 Thermodynamics of an interface. *Arch. Rat. Mech Anal.* **57**, 255–280.

NAYFEH, A. H. 1971 Third-harmonic resonance in the interaction of capillary and gravity waves. *J. Fluid Mech.* **48**, 385–395.

NIX, J. R. 1972 Calculation of fission barriers for heavy and superheavy nuclei. *Ann. Rev. Nucl. Sci.* **22**, 65–120.

PAVELLE, R., ROTHSTEIN, M. & FITCH, J. 1981 Computer algebra. *Sci. Am.* **245**, 136–152.

PHILLIPS, O. M. 1981 Wave interactions – the evolution of an idea. *J. Fluid Mech.* **106**, 215–227.

PROSPERETTI, A. 1980 Free oscillations of drops and bubbles: the initial-value problem. *J. Fluid Mech.* **100**, 333–347.

RAYLEIGH, J. W. S. 1882 On the equilibrium of liquid conducting masses charged with electricity. *Phil. Mag.* **14**, 184–186.

SAMPLE, S. B., RAGHUPATHY, B. & HENDRICKS, C. D. 1970 Quiescent distortion and resonant oscillations of a liquid drop in an electric field. *Intl J. Engng Sci.* **8**, 97–109.

SAVILLE, D. A. 1974 Electrohydrodynamic oscillation and stability of a charged drop. *Phys. Fluids* **17**, 54–60.

STRATTON, J. A. 1941 *Electrodynamic Theory.* McGraw-Hill, New York.

TANG, H. H. K. & WONG, C. Y. 1974 Vibration of a viscous liquid sphere. *J. Phys.* A: *Math. Nucl. Gen.* **7**, 1038–1050.

TRINH, E., ZWERN, A. & WANG, T. G. 1982 an experimental study of small-amplitude drop oscillations in immiscible liquid systems. *J. Fluid Mech.* **115**, 453–474.

TSAMOPOULOS, J. A. 1984 Ph.D. thesis, Massachusetts Institute of Technology.

TSAMOPOULOS, J. A. & BROWN, R. A. 1983 Nonlinear oscillations of inviscid drops and bubbles. *J. Fluid Mech.* **127**, 519–537.

TSANG, C. F. 1974 Similarities and differences between volume-charged (nuclear) drops and charged conducting (rain) drops. In *Proc. Intl Colloq. on Drops and Bubbles* (ed. D. J. Collins, M. S. Plesset & M. M. Saffren). Jet Propulsion Laboratory.

WILLIAMS, A. 1973 Combustion of droplets of fluid fuels: A review. *Combust. Flame* **21**, 1–31.

WILTON, J. R. 1915 On ripples. *Phil. Mag.* **29**, 688–700.