J. Fluid Mech. (2010), *vol.* 654, *pp.* 141–159. © Cambridge University Press 2010
doi:10.1017/S0022112010000480

Modulation equations for strongly nonlinear oscillations of an incompressible viscous drop

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(Received 15 May 2009; revised 22 January 2010; accepted 26 January 2010; first published online 11 May 2010)

Large-amplitude oscillations of incompressible viscous drops are studied at small capillary number. On the long viscous time scale, a formal perturbation scheme is developed to determine original modulation equations. These two ordinary differential equations comprise the averaged condition for conservation of energy and the averaged projection of the Navier–Stokes equations onto the vorticity vector. The modulation equations are applied to the free decay of axisymmetric oblate–prolate spheroid oscillations. On the long time scale, only the modulation equation for energy is required. In this example, the results compare well with linear viscous theory, weakly nonlinear inviscid theory and experimental observations. The new results show that previous experimental observations and numerical simulations are all manifestations of a single-valued relationship between dimensionless decay rate and amplitude. Moreover, if the amplitude of the oscillations does not exceed 30 % of the drop radius, this decay rate may be approximated by a quadratic. The new results also show that, when the amplitude of the oscillations exceeds 20 % of the drop radius, fluid in the inviscid bulk of the drop is undergoing abrupt changes in its acceleration in comparison to the acceleration during small-amplitude deformations.

1. Introduction

The dynamics of an oscillating viscous drop is of fundamental interest, not only because it represents a canonical example of the interplay between convective and free-surface nonlinearities but also because of the practical need to understand the physical mechanisms in important large-scale industrial processes such as multi-phase dispersions. Moreover, the decay of droplet oscillations is one of the experimental techniques by which the viscosity of a fluid may be evaluated.

The large-amplitude oscillations that arise during the recoil of extended filaments are particularly relevant to this study (see Schulkes 1996; Notz & Basaran 2004). When the filaments are quite viscous and/or their initial deformations are not inordinately large, such filaments correspond to highly deformed drops whose responses are either damped oscillations or aperiodic decay. This wide range of motivations has prompted many excellent theoretical and experimental studies of this challenging time-dependent three-dimensional problem.

Rayleigh (1879) described the oscillations of drops in the inviscid linear limit where the flow was irrotational. If the centre of a drop of radius R_∞ is taken as the centre of a spherical coordinate system, then the perturbation in the free surface is given by $S_m(\theta,\phi)\cos(\omega_m t^*)$, where $S_m(\theta,\phi)$ is the spherical harmonic of degree m, ω_m is the corresponding angular frequency, t^* is time, θ is the polar angle and ϕ is the

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azimuthal angle. It was reported that

$$\omega_m^2 = \frac{\sigma}{\rho R_\infty^3} m(m-1)(m+2), \tag{1.1}$$

where ρ is the density, σ is the surface tension and $m \geqslant 2$. In the viscous linear limit, Lamb (1932) showed that if the capillary number is small, then the oscillation amplitude is damped like $\exp(-\lambda_m^* t^*)$, where

$$\lambda_m^* = \frac{\nu}{R_\infty^2}(m-1)(2m+1), \tag{1.2}$$

where μ is the dynamic viscosity and $\nu = \mu/\rho$ is the kinematic viscosity. Chandrasekhar (1961) also studied the free decay of a viscous drop in the linear limit. He showed that if $\omega_m R_\infty^2/\nu$ is larger than a constant κ, then a damped oscillation results. An aperiodic decay is found if it is smaller than κ. The constant κ is essentially a Reynolds number with a velocity scale based on the frequency of oscillations. The dimensionless parameter $\omega_m R_\infty^2/\nu$ is assumed to be large, so modulated oscillations are predicted.

Prosperetti (1977, 1980) studied the initial-value problem of a viscous drop in the linear limit. The initial condition corresponded to zero vorticity distribution and a static deformation. Three phases were identified: an initial phase in which the flow is irrotational, an intermediate phase where vorticity is generated at the free surface that diffuses into the drop and a final phase described by the least-damped normal mode. In the small capillary number limit $\epsilon = \mu/\sqrt{\rho R_\infty \sigma} \ll 1$, the vorticity penetrates to a depth of the order of $\sqrt{\epsilon} R_\infty$ from the drop surface, which is a boundary layer (see Lamb 1932; Lundgren & Mansour 1988). The flow in the bulk of the drop remains irrotational at leading order.

The experimental literature describes either two immiscible liquids or a liquid drop surrounded by a gas. Trinh & Wang (1982) observed the large-amplitude oscillations of drops of silicone oil and carbon tetrachloride immersed in distilled water. A drop with $R_\infty \approx 6.2 \times 10^{-3}$ m, $\rho \approx 10^3$ kg m^{-3}, $\mu \approx 3.2 \times 10^{-3}$ kg m^{-1} s^{-1} and $\sigma \approx 4 \times 10^{-2}$ kg s^{-2} had a capillary number of 6.4×10^{-3}. A decrease in frequency with increasing oscillation amplitude, a longer prolate phase in each oscillation and an exponential decay in oscillation amplitude were all reported. Becker, Hiller & Kowalewski (1991) investigated falling drops of ethanol in a gas. A typical drop with $R_\infty \approx 1.72 \times 10^{-4}$ m, $\rho \approx 803$ kg m^{-3}, $\mu \approx 1.2 \times 10^{-3}$ kg m^{-1} s^{-1} and $\sigma \approx 2.29 \times 10^{-2}$ kg s^{-2} had a capillary number of 2.1×10^{-2}. A least-squares fit was performed to obtain the coefficients in the Fourier–Legendre expansion of the free surface: the viscous decay, coupling and amplitude-dependent frequency of individual modes were observed. Wang, Anilkumar & Lee (1996) studied the large-amplitude oscillations of drops of glycerine/water drop (65/35) from a Space Shuttle-based experiment carried out in microgravity. A drop with $R_\infty \approx 1.01 \times 10^{-2}$ m, $\rho \approx 1.168 \times 10^3$ kg m^{-3}, $\nu \approx 9.2 \times 10^{-6}$ m^2 s^{-1} and $\sigma \approx 6.4 \times 10^{-2}$ kg s^{-2} had a capillary number of 1.2×10^{-2}. These studies, which fit into the small-capillary-number parameter regime, will be the experimental comparisons in this paper.

Over one hundred years after Rayleigh (1879), Tsamopoulos & Brown (1983) extended the linear limit on inviscid drops to moderate-amplitude oscillations by a sophisticated weakly nonlinear analysis. A quadratic decrease in frequency with increasing amplitude and a longer prolate phase were predicted, these being compared against the experiments of Trinh & Wang (1982). We note that all the existing analytical approaches to nonlinear droplet dynamics neglect viscosity (see also Natarajan & Brown 1986).

The theoretical approaches to nonlinear viscous droplet dynamics are restricted to numerical methods. Three techniques are pursued in the literature, namely the boundary integral method applied by Lundgren & Mansour (1988), the finite element method by Basaran (1992) and the mode expansion method by Becker, Hiller & Kowalewski (1994). Patzek *et al.* (1991) have shown that boundary integral methods cannot model droplet oscillations with the viscosity in the range of interest. The finite element and mode expansion methods require long computation times that limit their applicability in solving practical problems. Despite the technical difficulties, all three numerical approaches have found that viscosity has a large effect on mode coupling.

More recent and current studies involving drop oscillations have included the capillary dynamics of coupled spherical-cap droplets (see Theisen *et al.* 2007), the capillary dynamics of a constrained liquid drop (see Bostwick & Steen 2009) and the damped oscillations of pendant drops from a tube (see Suryo & Basaran 2006). This field of research is still of great importance and interest to the scientific community.

The first purpose of this article is to gain new physical insights into the free decay of the large-amplitude oscillations of a viscous drop. The capillary number is small in many cases of interest (see Trinh & Wang 1982; Becker *et al.* 1991; Wang *et al.* 1996), where modulation equations provide a description of the physics on the long viscous time scale and extend the applicability of some numerical approaches from an inviscid to a viscous problem. The second purpose relates to a general open problem: oscillations and oscillatory waves in fluid mechanics are strongly nonlinear. Weakly nonlinear analysis has been widely applied to these problems over four decades. As the leading-order problem is usually nonlinear, it is necessary to construct very sophisticated expansions over several orders to obtain agreement with experimental observations (see e.g. Tsamopoulos & Brown 1983). However, these attempts to capture the leading-order nonlinearity have a limited range of agreement with experiment at best (see Wang *et al.* 1996). Fluid mechanics requires the techniques of strongly nonlinear analysis, but the formal perturbation theory has only recently been developed (see Smith 2007). This new topic faces two immediate challenges: the modulation equations may form an underdetermined system and require experimental validation.

In § 2, formal perturbation theory is applied to the general case of a drop in rotational flow. The leading-order problem is stated, but cannot be solved analytically. This does not prevent us from evaluating the linear differential operator for the first correction and its adjoint. Modulation equations are derived using linearly independent vectors in the null space of the adjoint. In § 3, the free decay of axisymmetric oblate–prolate spheroid oscillations is studied. On the short time scale, a truncated expansion of spherical harmonics is adopted to convert the leading-order problem into a system of nonlinear ordinary differential equations. On the long viscous time scale, a single modulation equation for the energy is necessary and sufficient. The predictions are compared with previous asymptotic results and experimental observations. Finally, § 4 gives a brief discussion of the results.

2. Strongly nonlinear analysis

2.1. *Introduction*

We define R_∞ to be the radius of the spherical drop at steady state, ρ the density and σ the surface tension. We transform to dimensionless variables via

$$r^* = R_\infty r, \qquad t^* = R_\infty\sqrt{\frac{\rho R_\infty}{\sigma}}t, \qquad [u^*, v^*, w^*] = \sqrt{\frac{\sigma}{\rho R_\infty}}[u, v, w],$$

$$p^* = \frac{\sigma}{R_\infty}p, \qquad R^* = R_\infty R,$$

where r^* is the radial coordinate, t^* is time, $\boldsymbol{q}^* = (u^*, v^*, w^*)^{\mathrm{T}}$ is the velocity vector, p^* is pressure and $r^* = R^*$ is the free surface. The three-dimensional continuity and Navier–Stokes equations in spherical polar coordinates become

$$\frac{1}{r^2}\frac{\partial}{\partial r}(r^2 u) + \frac{1}{r\sin(\theta)}\frac{\partial}{\partial \theta}(v\sin(\theta)) + \frac{1}{r\sin(\theta)}\frac{\partial w}{\partial \phi} = 0, \tag{2.1}$$

$$\frac{\partial u}{\partial t} + (\boldsymbol{q}\cdot\nabla)u - \frac{(v^2+w^2)}{r} + \frac{\partial p}{\partial r} = \epsilon\left[\nabla^2 u - \frac{2u}{r^2} - \frac{2}{r^2\sin(\theta)}\frac{\partial}{\partial\theta}(v\sin(\theta)) - \frac{2}{r^2\sin(\theta)}\frac{\partial w}{\partial\phi}\right], \tag{2.2a}$$

$$\frac{\partial v}{\partial t} + (\boldsymbol{q}\cdot\nabla)v + \frac{uv}{r} - \frac{w^2\cot(\theta)}{r} + \frac{1}{r}\frac{\partial p}{\partial\theta} = \epsilon\left[\nabla^2 v + \frac{2}{r^2}\frac{\partial u}{\partial\theta} - \frac{v}{r^2\sin^2(\theta)} - \frac{2\cos(\theta)}{r^2\sin^2(\theta)}\frac{\partial w}{\partial\phi}\right], \tag{2.2b}$$

$$\frac{\partial w}{\partial t} + (\boldsymbol{q}\cdot\nabla)w + \frac{uw}{r} + \frac{vw\cot(\theta)}{r} + \frac{1}{r\sin(\theta)}\frac{\partial p}{\partial\phi} = \epsilon\left[\nabla^2 w + \frac{2}{r^2\sin(\theta)}\frac{\partial u}{\partial\phi} + \frac{2\cos(\theta)}{r^2\sin^2(\theta)}\frac{\partial v}{\partial\phi} - \frac{w}{r^2\sin^2(\theta)}\right], \tag{2.2c}$$

in which ϵ is the capillary number (or the Ohnesorge number) given by

$$0 < \epsilon = \frac{\mu}{\sigma}\sqrt{\frac{\sigma}{\rho R_\infty}} \ll 1,$$

∇ and ∇^2 are the differential operators given by

$$\nabla = \left(\frac{\partial}{\partial r}, \frac{1}{r}\frac{\partial}{\partial\theta}, \frac{1}{r\sin(\theta)}\frac{\partial}{\partial\phi}\right)^{\mathrm{T}},$$

$$\nabla^2 u = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial u}{\partial r}\right) + \frac{1}{r^2\sin(\theta)}\frac{\partial}{\partial\theta}\left(\sin(\theta)\frac{\partial u}{\partial\theta}\right) + \frac{1}{r^2\sin^2(\theta)}\frac{\partial^2 u}{\partial\phi^2}.$$

The boundary conditions on the free surface $r = R$ are

$$u = \frac{\partial R}{\partial t} + \frac{v}{R}\frac{\partial R}{\partial\theta} + \frac{w}{R\sin(\theta)}\frac{\partial R}{\partial\phi}, \tag{2.3}$$

$$\begin{aligned}-\Delta p - p + \epsilon D = \frac{1}{R^2M^3}\Bigg\{&-2R - \frac{3}{R}\left(\frac{\partial R}{\partial\theta}\right)^2 - \frac{3}{R\sin^2(\theta)}\left(\frac{\partial R}{\partial\phi}\right)^2 \\ &+ \cot(\theta)\frac{\partial R}{\partial\theta}\left[1 + \frac{1}{R^2}\left(\frac{\partial R}{\partial\theta}\right)^2 + \frac{2}{R^2\sin^2(\theta)}\left(\frac{\partial R}{\partial\phi}\right)^2\right] - \frac{2}{R^2\sin^2(\theta)}\frac{\partial R}{\partial\theta}\frac{\partial R}{\partial\phi}\frac{\partial^2 R}{\partial\theta\partial\phi} \\ &+ \frac{1}{\sin^2(\theta)}\frac{\partial^2 R}{\partial\phi^2}\left[1 + \frac{1}{R^2}\left(\frac{\partial R}{\partial\theta}\right)^2\right] + \frac{\partial^2 R}{\partial\theta^2}\left[1 + \frac{1}{R^2\sin^2(\theta)}\left(\frac{\partial R}{\partial\phi}\right)^2\right]\Bigg\},\end{aligned} \tag{2.4}$$

$$\begin{aligned}\Bigg(\frac{1}{R\sin(\theta)}\frac{\partial R}{\partial\phi}\left\{2\frac{\partial u}{\partial r} - \frac{1}{R}\frac{\partial R}{\partial\theta}d_{r\theta} - \frac{1}{R\sin(\theta)}\frac{\partial R}{\partial\phi}d_{r\phi}\right\} - \frac{1}{R\sin(\theta)}\frac{\partial R}{\partial\phi}d_{\phi\phi} \\ + d_{r\phi} - \frac{1}{R}\frac{\partial R}{\partial\theta}d_{\theta\phi}\Bigg)\epsilon = 0,\end{aligned} \tag{2.5}$$

$$\left(\frac{1}{R}\frac{\partial R}{\partial \theta}\left\{2\frac{\partial u}{\partial r}-\frac{1}{R}\frac{\partial R}{\partial \theta}d_{r\theta}-\frac{1}{R\sin(\theta)}\frac{\partial R}{\partial \phi}d_{r\phi}\right\}-\frac{1}{R}\frac{\partial R}{\partial \theta}d_{\theta\theta}+d_{r\theta}\right.$$
$$\left.-\frac{1}{R\sin(\theta)}\frac{\partial R}{\partial \phi}d_{\theta\phi}\right)\epsilon=0, \quad (2.6)$$

where $\Delta p=2$ is the static pressure difference across the interface,

$$M=\sqrt{1+\frac{1}{R^2}\left\{\left(\frac{\partial R}{\partial \theta}\right)^2+\frac{1}{\sin^2(\theta)}\left(\frac{\partial R}{\partial \phi}\right)^2\right\}}, \qquad d_{\theta\theta}=2\left[\frac{1}{R}\frac{\partial v}{\partial \theta}+\frac{u}{R}\right],$$

$$d_{\phi\phi}=2\left[\frac{1}{R\sin(\theta)}\frac{\partial w}{\partial \phi}+\frac{u}{R}+\frac{v\cot(\theta)}{R}\right], \qquad d_{r\theta}=\frac{1}{R}\frac{\partial u}{\partial \theta}+\frac{\partial v}{\partial r}-\frac{v}{R},$$

$$d_{r\phi}=\frac{1}{R\sin(\theta)}\frac{\partial u}{\partial \phi}+\frac{\partial w}{\partial r}-\frac{w}{R}, \qquad d_{\theta\phi}=\frac{1}{R\sin(\theta)}\frac{\partial v}{\partial \phi}+\frac{1}{R}\frac{\partial w}{\partial \theta}-\frac{w\cot(\theta)}{R},$$

$$D=\frac{1}{M^2}\left\{2\frac{\partial u}{\partial r}+\frac{1}{R^2}\left(\frac{\partial R}{\partial \theta}\right)^2 d_{\theta\theta}+\frac{1}{R^2\sin^2(\theta)}\left(\frac{\partial R}{\partial \phi}\right)^2 d_{\phi\phi}\right.$$
$$\left.-\frac{2}{R}\frac{\partial R}{\partial \theta}d_{r\theta}-\frac{2}{R\sin(\theta)}\frac{\partial R}{\partial \phi}d_{r\phi}+\frac{2}{R^2\sin(\theta)}\frac{\partial R}{\partial \theta}\frac{\partial R}{\partial \phi}d_{\theta\phi}\right\}.$$

Equation (2.3) represents the kinematic condition, (2.4) the normal stress condition and (2.5)–(2.6) the tangential stress conditions. We assume that the boundary value problem (2.1)–(2.6) with $\epsilon=0$ has time periodic solutions. Moreover, we assume that the initial conditions with $\epsilon=0$ are consistent with an oscillatory solution.

The kinetic energy and vorticity vector are defined to be $E=\boldsymbol{q}\cdot\boldsymbol{q}/2$ and $\boldsymbol{\omega}=(\omega^{(r)},\omega^{(\theta)},\omega^{(\phi)})^{\mathrm{T}}$, respectively. The components of the vorticity vector are given by

$$\omega^{(r)}=\frac{1}{r\sin(\theta)}\left\{\frac{\partial}{\partial \theta}(w\sin(\theta))-\frac{\partial v}{\partial \phi}\right\}, \qquad \omega^{(\theta)}=\frac{1}{r\sin(\theta)}\frac{\partial u}{\partial \phi}-\frac{1}{r}\frac{\partial}{\partial r}(rw),$$
$$\omega^{(\phi)}=\frac{1}{r}\left\{\frac{\partial}{\partial r}(rv)-\frac{\partial u}{\partial \theta}\right\}.$$

2.2. *The leading-order outer problem*

We adopt the usual procedure of introducing a fast time scale t^+ and a slow time scale $\tilde{t}$ with

$$\frac{\mathrm{d}t^+}{\mathrm{d}t}=\omega(\tilde{t}), \qquad \tilde{t}=\epsilon t,$$

where the frequency of oscillation $\omega(\tilde{t})$ needs to be chosen so that, in terms of t^+, the period of oscillation of the leading-order solution is independent of $\tilde{t}$ (see e.g. Kuzmak 1959; Smith 2005). The period on this t^+ scale is then an arbitrary constant that we specify to be 2π without loss of generality. We introduce expansions of the form

$$
\begin{aligned}
\boldsymbol{q} &\sim \boldsymbol{q}_0(t^+, r, \theta, \phi, \tilde{t}) + \epsilon \boldsymbol{q}_1(t^+, r, \theta, \phi, \tilde{t}),\\
p &\sim p_0(t^+, r, \theta, \phi, \tilde{t}) + \epsilon p_1(t^+, r, \theta, \phi, \tilde{t}),\\
E &\sim E_0(t^+, r, \theta, \phi, \tilde{t}) + \epsilon E_1(t^+, r, \theta, \phi, \tilde{t}),\\
\boldsymbol{\omega} &\sim \boldsymbol{\omega}_0(t^+, r, \theta, \phi, \tilde{t}) + \epsilon \boldsymbol{\omega}_1(t^+, r, \theta, \phi, \tilde{t}),\\
R &\sim R_0(t^+, \theta, \phi, \tilde{t}) + \epsilon R_1(t^+, \theta, \phi, \tilde{t}),\\
M &\sim M_0(t^+, \theta, \phi, \tilde{t}) + \epsilon M_1(t^+, \theta, \phi, \tilde{t}),\\
\left[d_{\theta\theta}, d_{\phi\phi}, d_{r\theta}, d_{r\phi}, d_{\theta\phi}\right] &\sim [d_{\theta\theta 0}, d_{\phi\phi 0}, d_{r\theta 0}, d_{r\phi 0}, d_{\theta\phi 0}](t^+, \theta, \phi, \tilde{t}),
\end{aligned}
$$

as $\epsilon \to 0$, where $\boldsymbol{q}_0 = (u_0, v_0, w_0)^{\mathrm{T}}$, $\boldsymbol{q}_1 = (u_1, v_1, w_1)^{\mathrm{T}}$, $\boldsymbol{\omega}_0 = (\omega_0^{(r)}, \omega_0^{(\theta)}, \omega_0^{(\phi)})^{\mathrm{T}}$ and $\boldsymbol{\omega}_1 = (\omega_1^{(r)}, \omega_1^{(\theta)}, \omega_1^{(\phi)})^{\mathrm{T}}$. At leading order, we obtain

$$\bar{L}u_0 - \frac{(v_0^2 + w_0^2)}{r} + \frac{\partial p_0}{\partial r} = 0, \tag{2.7a}$$

$$\bar{L}v_0 + \frac{u_0 v_0}{r} - \frac{w_0^2 \cot(\theta)}{r} + \frac{1}{r}\frac{\partial p_0}{\partial \theta} = 0, \tag{2.7b}$$

$$\bar{L}w_0 + \frac{u_0 w_0}{r} + \frac{v_0 w_0 \cot(\theta)}{r} + \frac{1}{r\sin(\theta)}\frac{\partial p_0}{\partial \phi} = 0, \tag{2.7c}$$

$$\frac{1}{r^2}\frac{\partial}{\partial r}(r^2 u_0) + \frac{1}{r\sin(\theta)}\frac{\partial}{\partial \theta}(v_0 \sin(\theta)) + \frac{1}{r\sin(\theta)}\frac{\partial w_0}{\partial \phi} = 0, \tag{2.7d}$$

with the differential operator

$$\bar{L} = \omega\frac{\partial}{\partial t^+} + u_0\frac{\partial}{\partial r} + \frac{v_0}{r}\frac{\partial}{\partial \theta} + \frac{w_0}{r\sin(\theta)}\frac{\partial}{\partial \phi}$$

and the boundary conditions

$$u_0(t^+ + \Psi, R_0, \theta, \phi, \tilde{t}) = \omega\frac{\partial R_0}{\partial t^+} + \frac{v_0(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})}{R_0}\frac{\partial R_0}{\partial \theta} + \frac{w_0(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})}{R_0\sin(\theta)}\frac{\partial R_0}{\partial \phi}, \tag{2.8a}$$

$$
\begin{aligned}
-\Delta p - p_0(t^+ + \Psi, R_0, \theta, \phi, \tilde{t}) = \frac{1}{R_0^2 M_0^3}\Bigg\{ &-2R_0 - \frac{3}{R_0}\left(\frac{\partial R_0}{\partial \theta}\right)^2 - \frac{3}{R_0\sin^2(\theta)}\left(\frac{\partial R_0}{\partial \phi}\right)^2\\
&+ \cot(\theta)\frac{\partial R_0}{\partial \theta}\left[1 + \frac{1}{R_0^2}\left(\frac{\partial R_0}{\partial \theta}\right)^2 + \frac{2}{R_0^2\sin^2(\theta)}\left(\frac{\partial R_0}{\partial \phi}\right)^2\right] - \frac{2}{R_0^2\sin^2(\theta)}\frac{\partial R_0}{\partial \theta}\frac{\partial R_0}{\partial \phi}\frac{\partial^2 R_0}{\partial \theta \partial \phi}\\
&+ \frac{1}{\sin^2(\theta)}\frac{\partial^2 R_0}{\partial \phi^2}\left[1 + \frac{1}{R_0^2}\left(\frac{\partial R_0}{\partial \theta}\right)^2\right] + \frac{\partial^2 R_0}{\partial \theta^2}\left[1 + \frac{1}{R_0^2\sin^2(\theta)}\left(\frac{\partial R_0}{\partial \phi}\right)^2\right]\Bigg\},
\end{aligned} \tag{2.8b}
$$

$$[u_0, v_0, w_0, p_0](t^+ + \Psi, r, \theta, \phi, \tilde{t}) = [u_0, v_0, w_0, p_0](t^+ + \Psi - 2n\pi, r, \theta, \phi, \tilde{t}), \tag{2.8c}$$

$$R_0(t^+ + \Psi, \theta, \phi, \tilde{t}) = R_0(t^+ + \Psi - 2n\pi, \theta, \phi, \tilde{t}), \tag{2.8d}$$

where n is an integer and $\Psi(\tilde{t})$ is the phase shift. We note that (2.7) and (2.8) are time reversible, that is invariant under $t^+ + \Psi \to -(t^+ + \Psi)$, $u_0 \to -u_0$, $v_0 \to -v_0$ and $w_0 \to -w_0$. We anticipate solutions such that u_0, v_0 and w_0 (p_0 and R_0) are odd (even) about zeros of u_0. Such solutions have not been found in terms of well-known functions, except for very special initial conditions. However, this does not prevent us from determining modulation equations for them.

2.3. *The boundary-layer problem*

In general, the solution of the leading-order outer problem will be inconsistent with the tangential stress conditions (2.5) and (2.6). A boundary layer is required to describe the significance of viscous effects near the free surface. For brevity, we will not discuss it any further, but return to the derivation of the modulation equations.

2.4. *Modulation equations*

In this section, the linear differential operator for the first correction is stated and the adjoint operator is determined (see Smith 2007). Nine linearly independent solutions to the adjoint problem are then deduced. However, only two of these result in physically meaningful modulation equations.

At next order, we have

$$\boldsymbol{L}\boldsymbol{x} = \begin{pmatrix} -\dfrac{\partial u_0}{\partial \tilde{t}} + \Upsilon_1 \\ -\dfrac{\partial v_0}{\partial \tilde{t}} + \Upsilon_2 \\ -\dfrac{\partial w_0}{\partial \tilde{t}} + \Upsilon_3 \\ 0 \end{pmatrix}, \tag{2.9}$$

where

$$\boldsymbol{x} = (u_1, v_1, w_1, p_1)^{\mathrm{T}},$$

$$\Upsilon_1 = \nabla^2 u_0 - \frac{2u_0}{r^2} - \frac{2}{r^2\sin(\theta)}\frac{\partial}{\partial\theta}(v_0\sin(\theta)) - \frac{2}{r^2\sin(\theta)}\frac{\partial w_0}{\partial\phi},$$

$$\Upsilon_2 = \nabla^2 v_0 + \frac{2}{r^2}\frac{\partial u_0}{\partial\theta} - \frac{v_0}{r^2\sin^2(\theta)} - \frac{2\cos(\theta)}{r^2\sin^2(\theta)}\frac{\partial w_0}{\partial\phi},$$

$$\Upsilon_3 = \nabla^2 w_0 + \frac{2}{r^2\sin(\theta)}\frac{\partial u_0}{\partial\phi} + \frac{2\cos(\theta)}{r^2\sin^2(\theta)}\frac{\partial v_0}{\partial\phi} - \frac{w_0}{r^2\sin^2(\theta)},$$

$$\boldsymbol{L} = \begin{pmatrix} \bar{L} + \dfrac{\partial u_0}{\partial r} & \dfrac{1}{r}\dfrac{\partial u_0}{\partial\theta} - \dfrac{2v_0}{r} & \dfrac{1}{r\sin(\theta)}\dfrac{\partial u_0}{\partial\phi} - \dfrac{2w_0}{r} & \dfrac{\partial}{\partial r} \\ \dfrac{\partial v_0}{\partial r} + \dfrac{v_0}{r} & \bar{L} + \dfrac{1}{r}\dfrac{\partial v_0}{\partial\theta} + \dfrac{u_0}{r} & \dfrac{1}{r\sin(\theta)}\dfrac{\partial v_0}{\partial\phi} - \dfrac{2w_0}{r}\cot(\theta) & \dfrac{1}{r}\dfrac{\partial}{\partial\theta} \\ \dfrac{\partial w_0}{\partial r} + \dfrac{w_0}{r} & \dfrac{1}{r}\dfrac{\partial w_0}{\partial\theta} + \dfrac{w_0}{r}\cot(\theta) & L_{33} & \dfrac{1}{r\sin(\theta)}\dfrac{\partial}{\partial\phi} \\ \dfrac{2}{r} + \dfrac{\partial}{\partial r} & \dfrac{\cot(\theta)}{r} + \dfrac{1}{r}\dfrac{\partial}{\partial\theta} & \dfrac{1}{r\sin(\theta)}\dfrac{\partial}{\partial\phi} & 0 \end{pmatrix},$$

$$L_{33} = \bar{L} + \frac{1}{r\sin(\theta)}\frac{\partial w_0}{\partial\phi} + \frac{u_0}{r} + \frac{v_0}{r}\cot(\theta),$$

with the periodicity conditions

$$[u_1, v_1, w_1, p_1](t^+ + \Psi, r, \theta, \phi, \tilde{t}) = [u_1, v_1, w_1, p_1](t^+ + \Psi - 2n\pi, r, \theta, \phi, \tilde{t}). \tag{2.10}$$

Taking great care with the evaluation of the adjoint operator, we obtain

$$
\begin{aligned}
\boldsymbol{r}^{\mathrm{T}} L \boldsymbol{x} - \boldsymbol{x}^{\mathrm{T}} L^{*} \boldsymbol{r} = {} & \omega \frac{\partial}{\partial t^{+}} \{a u_1 + b v_1 + c w_1\} \\
& + \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \left[u_0 \{a u_1 + b v_1 + c w_1\} + a p_1 + d u_1 \right] \right) \\
& + \frac{1}{r \sin(\theta)} \frac{\partial}{\partial \theta} \left(\sin(\theta) \left[v_0 \{a u_1 + b v_1 + c w_1\} + b p_1 + d v_1 \right] \right) \\
& + \frac{1}{r \sin(\theta)} \frac{\partial}{\partial \phi} \left(w_0 \{a u_1 + b v_1 + c w_1\} + c p_1 + d w_1 \right), \qquad (2.11)
\end{aligned}
$$

where

$$
L^{*} = \begin{pmatrix}
-\bar{L} + \dfrac{\partial u_0}{\partial r} & \dfrac{1}{r} \dfrac{\partial}{\partial r}(r v_0) & \dfrac{1}{r} \dfrac{\partial}{\partial r}(r w_0) & -\dfrac{\partial}{\partial r} \\
\dfrac{1}{r} \dfrac{\partial u_0}{\partial \theta} - \dfrac{2 v_0}{r} & -\bar{L} + \dfrac{1}{r} \dfrac{\partial v_0}{\partial \theta} + \dfrac{u_0}{r} & L_{23}^{*} & -\dfrac{1}{r} \dfrac{\partial}{\partial \theta} \\
\dfrac{1}{r \sin(\theta)} \dfrac{\partial u_0}{\partial \phi} - \dfrac{2 w_0}{r} & L_{32}^{*} & L_{33}^{*} & -\dfrac{1}{r \sin(\theta)} \dfrac{\partial}{\partial \phi} \\
-\dfrac{2}{r} - \dfrac{\partial}{\partial r} & -\dfrac{\cot(\theta)}{r} - \dfrac{1}{r} \dfrac{\partial}{\partial \theta} & -\dfrac{1}{r \sin(\theta)} \dfrac{\partial}{\partial \phi} & 0
\end{pmatrix},
$$

$$
L_{23}^{*} = \frac{1}{r} \frac{\partial w_0}{\partial \theta} + \frac{w_0}{r} \cot(\theta), \qquad L_{32}^{*} = \frac{1}{r \sin(\theta)} \frac{\partial v_0}{\partial \phi} - \frac{2 w_0}{r} \cot(\theta),
$$

$$
L_{33}^{*} = -\bar{L} + \frac{1}{r \sin(\theta)} \frac{\partial w_0}{\partial \phi} + \frac{1}{r}(u_0 + v_0 \cot(\theta)), \qquad \boldsymbol{r} = (a, b, c, d)^{\mathrm{T}}.
$$

Equation (2.11) may be integrated to yield

$$
\langle \boldsymbol{r}^{\mathrm{T}} L \boldsymbol{x} - \boldsymbol{x}^{\mathrm{T}} L^{*} \boldsymbol{r} \rangle = I_1 + \int_{t^{+}=-\Psi}^{2\pi - \Psi} I_2 \; \mathrm{d}t^{+},
$$

where

$$
\langle \; \bullet \; \rangle = \int_{t^{+}=-\Psi}^{2\pi-\Psi} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \int_{r=0}^{R_0(t^{+}, \theta, \phi, \tilde{t})} \bullet \; r^2 \sin(\theta) \, \mathrm{d}r \, \mathrm{d}\theta \, \mathrm{d}\phi \, \mathrm{d}t^{+},
$$

$$
\begin{aligned}
I_1 = \Bigg\langle & \omega \frac{\partial}{\partial t^{+}} \{a u_1 + b v_1 + c w_1\} + \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 u_0 \{a u_1 + b v_1 + c w_1\} \right) \\
& + \frac{1}{r \sin(\theta)} \frac{\partial}{\partial \theta} \left(\sin(\theta) v_0 \{a u_1 + b v_1 + c w_1\} \right) + \frac{1}{r \sin(\theta)} \frac{\partial}{\partial \phi} \left(w_0 \{a u_1 + b v_1 + c w_1\} \right) \Bigg\rangle,
\end{aligned}
$$

$$
\begin{aligned}
I_2 = \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \int_{r=0}^{R_0} \Bigg\{ & \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \left[a p_1 + d u_1 \right] \right) + \frac{1}{r \sin(\theta)} \frac{\partial}{\partial \theta} \left(\sin(\theta) \left[b p_1 + d v_1 \right] \right) \\
& + \frac{1}{r \sin(\theta)} \frac{\partial}{\partial \phi} \left(c p_1 + d w_1 \right) \Bigg\} r^2 \sin(\theta) \, \mathrm{d}r \, \mathrm{d}\theta \, \mathrm{d}\phi.
\end{aligned}
$$

Application of the divergence theorem, (2.8*a*), (2.8*d*) and (2.10), shows that $I_1 = 0$, provided

$$
[a, b, c](t^{+} + \Psi, r, \theta, \phi, \tilde{t}) = [a, b, c](t^{+} + \Psi - 2n\pi, r, \theta, \phi, \tilde{t}). \qquad (2.12)
$$

Similarly, the divergence theorem yields

$$I_2 = \int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi}\left(p_1(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\left[a(t^+ + \Psi, R_0, \theta, \phi, \tilde{t}) - \frac{b(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})}{R_0}\frac{\partial R_0}{\partial \theta} - \frac{c(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})}{R_0 \sin\theta}\frac{\partial R_0}{\partial \phi}\right] + d(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\left[\boldsymbol{q}_1(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\cdot \boldsymbol{n}_0\right]\right) R_0^2 \sin(\theta)\, \mathrm{d}\theta\, \mathrm{d}\phi,$$

where

$$\boldsymbol{n}_0 = \left(1, -\frac{1}{R_0}\frac{\partial R_0}{\partial \theta}, -\frac{1}{R_0 \sin\theta}\frac{\partial R_0}{\partial \phi}\right)^{\mathrm{T}}.$$

From this, it follows that if

$$L^* \boldsymbol{r} = \boldsymbol{0}, \tag{2.13}$$

subject to the boundary conditions (2.12), then our linear problem for the first correction can only have a solution if

$$\left\langle a\left[-\frac{\partial u_0}{\partial \tilde{t}} + \Upsilon_1\right] + b\left[-\frac{\partial v_0}{\partial \tilde{t}} + \Upsilon_2\right] + c\left[-\frac{\partial w_0}{\partial \tilde{t}} + \Upsilon_3\right]\right\rangle = \int_{t^+=-\Psi}^{2\pi-\Psi}\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi}\left(p_1(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\left[a(t^+ + \Psi, R_0, \theta, \phi, \tilde{t}) - \frac{b(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})}{R_0}\frac{\partial R_0}{\partial \theta} - \frac{c(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})}{R_0 \sin\theta}\frac{\partial R_0}{\partial \phi}\right] + d(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\left[\boldsymbol{q}_1(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\cdot \boldsymbol{n}_0\right]\right) R_0^2 \sin(\theta)\, \mathrm{d}\theta\, \mathrm{d}\phi\, \mathrm{d}t^+, \tag{2.14}$$

for any $\boldsymbol{r}$.

Nine linearly independent solutions of the adjoint problem (2.13) and (2.12) have been determined:

$$\boldsymbol{r}_1 = (u_0, v_0, w_0, p_0 + E_0)^{\mathrm{T}}, \quad \boldsymbol{r}_2 = (\omega_0^{(r)}, \omega_0^{(\theta)}, \omega_0^{(\phi)}, 0)^{\mathrm{T}}, \quad \boldsymbol{r}_3 = (0, 0, 0, 1)^{\mathrm{T}},$$

$$\boldsymbol{r}_4 = (\sin(\theta)\cos(\phi), \cos(\theta)\cos(\phi), -\sin(\phi), u_0\sin(\theta)\cos(\phi) + v_0\cos(\theta)\cos(\phi) - w_0\sin(\phi))^{\mathrm{T}},$$

$$\boldsymbol{r}_5 = (\sin(\theta)\sin(\phi), \cos(\theta)\sin(\phi), \cos(\phi), u_0\sin(\theta)\sin(\phi) + v_0\cos(\theta)\sin(\phi) + w_0\cos(\phi))^{\mathrm{T}},$$

$$\boldsymbol{r}_6 = (\cos(\theta), -\sin(\theta), 0, u_0\cos(\theta) - v_0\sin(\theta))^{\mathrm{T}},$$

$$\boldsymbol{r}_7 = (0, -r\sin(\phi), -r\cos(\theta)\cos(\phi), -rv_0\sin(\phi) - rw_0\cos(\theta)\cos(\phi))^{\mathrm{T}},$$

$$\boldsymbol{r}_8 = (0, r\cos(\phi), -r\cos(\theta)\sin(\phi), rv_0\cos(\phi) - rw_0\cos(\theta)\sin(\phi))^{\mathrm{T}},$$

$$\boldsymbol{r}_9 = (0, 0, r\sin(\theta), rw_0\sin(\theta))^{\mathrm{T}}.$$

The leading-order vorticity equations, that is

$$\bar{L}\omega_0^{(r)} = \omega_0^{(r)}\frac{\partial u_0}{\partial r} + \frac{\omega_0^{(\theta)}}{r}\frac{\partial u_0}{\partial \theta} + \frac{\omega_0^{(\phi)}}{r\sin(\theta)}\frac{\partial u_0}{\partial \phi},$$

$$\bar{L}\omega_0^{(\theta)} = \omega_0^{(r)}\frac{\partial v_0}{\partial r} + \frac{\omega_0^{(\theta)}}{r}\frac{\partial v_0}{\partial \theta} + \frac{\omega_0^{(\phi)}}{r\sin(\theta)}\frac{\partial v_0}{\partial \phi} + \frac{1}{r}[u_0\omega_0^{(\theta)} - v_0\omega_0^{(r)}],$$

$$\bar{L}\omega_0^{(\phi)} = \omega_0^{(r)}\frac{\partial w_0}{\partial r} + \frac{\omega_0^{(\theta)}}{r}\frac{\partial w_0}{\partial \theta} + \frac{\omega_0^{(\phi)}}{r\sin(\theta)}\frac{\partial w_0}{\partial \phi} + \frac{1}{r}[u_0\omega_0^{(\phi)} - w_0\omega_0^{(r)}] + \frac{\cot(\theta)}{r}[v_0\omega_0^{(\phi)} - w_0\omega_0^{(\theta)}],$$

are useful in the evaluation of $\boldsymbol{r}_2$. The first solution corresponds to conservation of energy, the second to projection of the Navier–Stokes equations onto the vorticity vector and the third to conservation of mass. The fourth, fifth and sixth solutions express conservation of momentum along the x-, y- and z-axes, respectively. The seventh, eighth and ninth solutions correspond to the x-, y- and z-components of angular momentum, respectively. The fourth component of the last six solutions is all invariants on the long time scale.

2.4.1. *Energy modulation equation*

If we substitute the first vector $\boldsymbol{r}_1$ into (2.14), then we obtain our first secularity condition

$$\int_{t^+=-\Psi}^{2\pi-\Psi}\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi} E_0(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\left[\boldsymbol{q}_1(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\cdot\boldsymbol{n}_0\right] \times R_0^2\sin(\theta)\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}t^+ + \left\langle\frac{\partial E_0}{\partial \tilde{t}}\right\rangle = \langle u_0\Upsilon_1 + v_0\Upsilon_2 + w_0\Upsilon_3\rangle$$
$$- \int_{t^+=-\Psi}^{2\pi-\Psi}\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi}\left(p_1(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\left[\boldsymbol{q}_0(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\cdot\boldsymbol{n}_0\right] + p_0(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\left[\boldsymbol{q}_1(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\cdot\boldsymbol{n}_0\right]\right) R_0^2\sin(\theta)\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}t^+. \quad (2.15)$$

Firstly, we simplify the left-hand side of this secularity condition (2.15). We apply the transport theorem as follows:

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\{\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi}\int_{r=0}^{R_0} E_0 r^2\sin(\theta)\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi\right\} = \int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi}\int_{r=0}^{R_0}\left[\frac{\partial E_0}{\partial t} + \nabla\cdot(\boldsymbol{q}E_0)\right] r^2\sin(\theta)\,\mathrm{d}r\,\mathrm{d}\theta\,\mathrm{d}\phi.$$

We expand this equation and integrate over a period to obtain the rate of change of kinetic energy

$$\frac{\mathrm{d}}{\mathrm{d}\tilde{t}}\langle E_0\rangle = \left\langle\frac{\partial E_0}{\partial \tilde{t}} + \frac{1}{r^2}\frac{\partial}{\partial r}(r^2u_1E_0) + \frac{1}{r\sin(\theta)}\frac{\partial}{\partial \theta}(v_1E_0\sin(\theta)) + \frac{1}{r\sin(\theta)}\frac{\partial}{\partial \phi}(w_1E_0)\right\rangle. \quad (2.16)$$

Application of the divergence theorem and (2.16) to the left-hand side of (2.15) and (2.8a) to the right-hand side of (2.15) yields

$$\frac{\mathrm{d}}{\mathrm{d}\tilde{t}}\langle E_0\rangle = \langle u_0\Upsilon_1 + v_0\Upsilon_2 + w_0\Upsilon_3\rangle - \int_{t^+=-\Psi}^{2\pi-\Psi}\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi}\left(p_1(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\omega\frac{\partial R_0}{\partial t^+}\right.$$
$$\left. + p_0(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\left[\boldsymbol{q}_1(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})\cdot\boldsymbol{n}_0\right]\right)R_0^2\sin(\theta)\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}t^+.$$

We incorporate the standard terms for the rate of dissipation of energy (see e.g. Lamb 1932). Moreover, we employ (2.3) to obtain

$$\frac{\mathrm{d}}{\mathrm{d}\tilde{t}}\langle E_0\rangle + \int_{t^+=-\Psi}^{2\pi-\Psi}\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi} p_0\frac{\partial R_0}{\partial\tilde{t}}R_0^2\sin(\theta)\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}t^+$$
$$= -\left\langle|\,\boldsymbol{\omega}_0\,|^2\right\rangle - 2\int_{t^+=-\Psi}^{2\pi-\Psi}\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi}\boldsymbol{n}_0\cdot\{\nabla E_0 - \boldsymbol{q}_0\wedge\boldsymbol{\omega}_0\}\,R_0^2\sin(\theta)\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}t^+$$
$$+\int_{t^+=-\Psi}^{2\pi-\Psi}\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi}\left\{\boldsymbol{n}_0\cdot\left[u_0\left(2\frac{\partial u_0}{\partial r}, d_{r\theta 0}, d_{r\phi 0}\right) + v_0(d_{r\theta 0}, d_{\theta\theta 0}, d_{\theta\phi 0})\right.\right.$$
$$\left. + w_0(d_{r\phi 0}, d_{\theta\phi 0}, d_{\phi\phi 0})\right] - p_1\omega\frac{\partial R_0}{\partial t^+} - p_0\left[\omega\frac{\partial R_1}{\partial t^+} - R_1\frac{\partial u_0}{\partial r} - \frac{v_0 R_1}{R_0^2}\frac{\partial R_0}{\partial\theta}\right.$$
$$+\frac{v_0}{R_0}\frac{\partial R_1}{\partial\theta} + \frac{R_1}{R_0}\frac{\partial v_0}{\partial r}\frac{\partial R_0}{\partial\theta} + \frac{1}{R_0\sin(\theta)}\left(w_0\frac{\partial R_1}{\partial\phi} - \frac{w_0 R_1}{R_0}\frac{\partial R_0}{\partial\phi}\right.$$
$$\left.\left.\left. + R_1\frac{\partial w_0}{\partial r}\frac{\partial R_0}{\partial\phi}\right)\right]\right\}R_0^2\sin(\theta)\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}t^+. \qquad (2.17)$$

The first volume integral and the first surface integral on the right-hand side of (2.17) correspond to the rate of dissipation of energy. The second surface integral on the right-hand side of (2.17) is the rate at which work is being done at the surface by the surrounding matter over the short time scale, which is zero. The surface integral on the left-hand side of (2.17) is the rate of change of potential energy over the long time scale, which may be rewritten as follows:

$$\int_{t^+=-\Psi}^{2\pi-\Psi}\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi} p_0\frac{\partial R_0}{\partial\tilde{t}}R_0^2\sin(\theta)\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}t^+ = \frac{\mathrm{d}}{\mathrm{d}\tilde{t}}V_0,$$

where

$$V_0 = \int_{t^+=-\Psi}^{2\pi-\Psi}\left\{\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi} M_0 R_0^2\sin(\theta)\,\mathrm{d}\theta\,\mathrm{d}\phi\; - 4\pi R_\infty^2\right\}\mathrm{d}t^+.$$

Thus, we have the modulation equation

$$\frac{\mathrm{d}}{\mathrm{d}\tilde{t}}(\langle E_0\rangle + V_0) = -\left\langle|\,\boldsymbol{\omega}_0\,|^2\right\rangle - 2\int_{t^+=-\Psi}^{2\pi-\Psi}\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi}\boldsymbol{n}_0\cdot\{\nabla E_0 - \boldsymbol{q}_0\wedge\boldsymbol{\omega}_0\}$$
$$(t^+ + \Psi, R_0, \theta, \phi, \tilde{t})R_0^2\sin(\theta)\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}t^+. \qquad (2.18)$$

Equation (2.18) is a generalization of the linear theory of Lamb (1932) to the nonlinear problem. Lamb (1932) did not integrate over the period of oscillation. This integration is unnecessary on the left-hand side of (2.18) as the sum of kinetic energy integrated over the volume and potential energy integrated over the surface is independent of the short time scale. However, in the linear theory, Lamb (1932) obtained the mean value of the total dissipation using a solution for irrotational flow. In the absence of an explicit solution, it is necessary to integrate over the period of oscillation to determine the appropriate dissipation on the right-hand side of (2.18).

2.4.2. *Vorticity modulation equation*

If we substitute the second vector $\boldsymbol{r}_2$ into (2.14), then we obtain our second secularity condition

$$\left\langle \boldsymbol{\omega}_0 \cdot \frac{\partial \boldsymbol{q}_0}{\partial \tilde{t}} \right\rangle = \langle \omega_0^{(r)} \Upsilon_1 + \omega_0^{(\theta)} \Upsilon_2 + \omega_0^{(\phi)} \Upsilon_3 \rangle - \int_{t^+=-\Psi}^{2\pi-\Psi} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \{p_1[\boldsymbol{\omega}_0 \cdot \boldsymbol{n}_0]\}(t^+ + \Psi, R_0, \theta, \phi, \tilde{t}) R_0^2 \sin(\theta)\, \mathrm{d}\theta\, \mathrm{d}\phi\, \mathrm{d}t^+. \quad (2.19)$$

The right-hand side of (2.19) may be split into two by integration by parts. The first term is the component of surface force exerted by the surrounding matter in the direction of the vorticity vector and across a surface normal to $\boldsymbol{n}_0$, this being zero. The second term represents viscous dissipation. After a further application of integration by parts, we have the modulation equation

$$\left\langle \boldsymbol{\omega}_0 \cdot \frac{\partial \boldsymbol{q}_0}{\partial \tilde{t}} \right\rangle = \langle \boldsymbol{q}_0 \cdot \nabla^2 \boldsymbol{\omega}_0 \rangle - \int_{t^+=-\Psi}^{2\pi-\Psi} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \{\boldsymbol{n}_0 \cdot [(\boldsymbol{q}_0 \cdot \nabla)\boldsymbol{\omega}_0] + \boldsymbol{q}_0 \cdot [(\boldsymbol{n}_0 \cdot \nabla)\boldsymbol{\omega}_0]\}(t^+ + \Psi, R_0, \theta, \phi, \tilde{t}) R_0^2 \sin(\theta)\, \mathrm{d}\theta\, \mathrm{d}\phi\, \mathrm{d}t^+. \quad (2.20)$$

The left-hand side of (2.20) is the rate of change of momentum projected onto the vorticity vector and the right-hand side is its rate of dissipation. Lamb (1932) did not derive an equation involving the vorticity. His analysis only considered the situation in which the inviscid bulk of the drop was in irrotational flow. A modulation equation involving the vorticity in a rotational bulk flow has not been previously reported in the literature.

3. Axisymmetric oblate–prolate oscillation

3.1. *Introduction*

In this section, we restrict our attention to one of the lowest modes of vibration, namely the axisymmetric oblate–prolate spheroid oscillation. The initial conditions are

$$u(r, \theta, \phi, t = 0) = O(\epsilon),$$
$$v(r, \theta, \phi, t = 0) = O(\epsilon),$$
$$w(r, \theta, \phi, t = 0) = O(\epsilon),$$
$$R(\theta, \phi, t = 0) = \bar{b}_0 + \bar{b}_2 P_2(\xi) + O(\epsilon),$$

as $\epsilon \to 0$, where $\xi = \cos(\theta)$ and $P_l(\xi)$ are the Legendre polynomials. Conservation of mass,

$$\int_{\xi=-1}^{1} \left[\bar{b}_0 + \bar{b}_2 P_2(\xi)\right]^3 \mathrm{d}\xi = 2, \quad (3.1)$$

permits the calculation of $\bar{b}_0$ for a given initial amplitude $\bar{b}_2$. These particular initial conditions are adopted because an approximate analytical solution may be readily calculated to the leading-order outer problem.

3.2. *The leading-order outer solution*

We consider an approximate solution of the leading-order outer problem in the form of a truncated expansion of spherical harmonics. Equations (2.7), (2.8*c*) and (2.8*d*) will be exactly satisfied, but (2.8*a*) and (2.8*b*) will not. The truncated expansion of spherical harmonics is

$$u_0 = \sum_{k=1}^{4} a_{2k}(t^+ + \Psi, \tilde{t}) 2k r^{2k-1} P_{2k}(\xi), \tag{3.2}$$

$$v_0 = -\frac{\sin(\theta)}{r} \sum_{k=1}^{4} a_{2k}(t^+ + \Psi, \tilde{t}) r^{2k} P'_{2k}(\xi), \tag{3.3}$$

$$w_0 = 0, \tag{3.4}$$

$$\begin{aligned} p_0 = {} & G(t^+ + \Psi, \tilde{t}) - \omega(\tilde{t}) \sum_{k=1}^{4} \frac{\partial a_{2k}}{\partial t^+}(t^+ + \Psi, \tilde{t}) r^{2k} P_{2k}(\xi) \\ & - \frac{1}{2}\left(\sum_{k=1}^{4} a_{2k}(t^+ + \Psi, \tilde{t}) 2k r^{2k-1} P_{2k}(\xi)\right)^2 \\ & - \frac{(1-\xi^2)}{2r^2}\left(\sum_{k=1}^{4} a_{2k}(t^+ + \Psi, \tilde{t}) r^{2k} P'_{2k}(\xi)\right)^2, \end{aligned} \tag{3.5}$$

$$R_0 = \sum_{k=0}^{4} b_{2k}(t^+ + \Psi, \tilde{t}) P_{2k}(\xi), \tag{3.6}$$

where the coefficients $a_l(t^+ + \Psi, \tilde{t})$ and $b_l(t^+ + \Psi, \tilde{t})$ must be chosen such that the boundary conditions (2.8*a*) and (2.8*b*) are approximately satisfied. We specify the phase shift Ψ by taking u_0, v_0 and w_0 to be odd (and p_0 and R_0 to be even) about $t^+ + \Psi = n\pi$. It is common practice to assume that b_0 is constant; however, this assumption would result in more equations than unknowns in the strongly nonlinear problem. Only even harmonics are adopted, which is consistent with our choice of initial conditions (see Basaran 1992).

Using the orthogonality properties of Legendre polynomials, we obtain the following autonomous system of ordinary differential equations from (2.8*a*) and (2.8*b*):

$$\begin{aligned} \omega \sum_{k=1}^{4} \frac{\partial a_{2k}}{\partial t^+} \int_{\xi=-1}^{1} P_n(\xi) R_0^{2k} P_{2k}(\xi) \mathrm{d}\xi = {} & -\frac{1}{2} \int_{\xi=-1}^{1} P_n(\xi) \left(\sum_{k=1}^{4} a_{2k} 2k R_0^{2k-1} P_{2k}(\xi)\right)^2 \mathrm{d}\xi \\ & - \frac{1}{2} \int_{\xi=-1}^{1} P_n(\xi)(1-\xi^2) \left(\sum_{k=1}^{4} a_{2k} R_0^{2k-1} P'_{2k}(\xi)\right)^2 \mathrm{d}\xi \\ & + \int_{\xi=-1}^{1} \frac{P_n(\xi)}{R_0^2 M_0^3} \left\{-2R_0 - \frac{3}{R_0}\left(\frac{\partial R_0}{\partial \theta}\right)^2 + \frac{\partial^2 R_0}{\partial \theta^2} + \cot(\theta) \frac{\partial R_0}{\partial \theta} M_0^2\right\} \mathrm{d}\xi, \end{aligned} \tag{3.7}$$

in which $n = 2, 4, 6, 8$ and

$$\begin{aligned} \frac{2\omega}{2n+1} \frac{\partial b_n}{\partial t^+} = {} & \int_{\xi=-1}^{1} P_n(\xi) \sum_{k=1}^{4} a_{2k} 2k R_0^{2k-1} P_{2k}(\xi) \, \mathrm{d}\xi \\ & - \int_{\xi=-1}^{1} P_n(\xi)(1-\xi^2) \left(\sum_{k=1}^{4} a_{2k} R_0^{2k-2} P'_{2k}(\xi)\right) \left(\sum_{k=1}^{4} b_{2k} P'_{2k}(\xi)\right) \mathrm{d}\xi, \end{aligned} \tag{3.8}$$

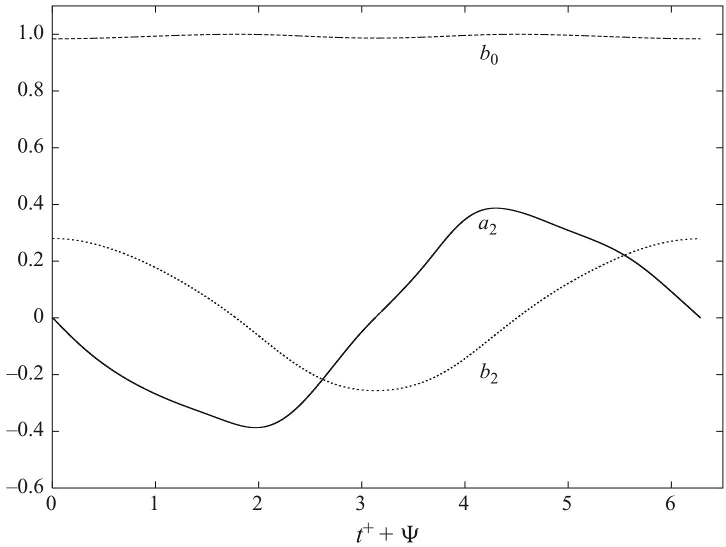

FIGURE 1. Numerical solution of the autonomous system of ordinary differential equations (3.7)–(3.9) over a single period. The solution is periodic in $t^+ + \Psi$ with period 2π and b_0 and b_2 (a_2) are even (odd) about the half period $t^+ + \Psi = n\pi$.

in which $n = 0, 2, 4, 6, 8$. In (3.7) and (3.8), R_0 is given by (3.6). The initial conditions are

$$a_{2k}(0,0) = 0, \quad b_0(0,0) = \bar{b}_0, \quad b_2(0,0) = \bar{b}_2, \quad b_4(0,0) = 0, \quad b_6(0,0) = 0, \quad b_8(0,0) = 0. \quad (3.9)$$

It is not possible to integrate (3.7)–(3.9) analytically, we must proceed numerically. The frequency $\omega(\tilde{t})$ must be selected such that the period is 2π over each cycle. A combined shooting and bisection method may be adopted to determine this frequency. This algorithm is verified against the linear theory (1.1) of Rayleigh (1879).

A typical solution of (3.7)–(3.9) is shown in figure 1 for $\bar{b}_0 = 0.28$, the nonlinear frequency being $\omega \approx 2.705$. In order to validate the truncated expansion of spherical harmonics, the same calculation was performed taking the upper limit of the sum to be eight in the leading-order solution (3.2)–(3.6). Similarly, the calculation was also performed with half of the time step to ensure convergence. A value of $\omega \approx 2.705$ was produced in the former case and $\omega \approx 2.706$ in the latter case. We therefore conclude that the truncated expansion of spherical harmonics and the time step provide suitable resolution at this amplitude of oscillation.

In figure 1, $b_2 < 0$ corresponds to oblate spheroid and $b_2 > 0$ corresponds to prolate spheroid. The modulus of the maximum value of b_2 in the prolate shape is larger than the modulus of the minimum value of b_2 in the oblate shape, that is the variation of b_2 exhibits asymmetry. We note that 43.4 % of the period is spent in the oblate form rather than in the prolate, as noted by previous authors, for example Wang *et al.* (1996). It is clear from figure 1 that the time dependence of a_2, b_0 and b_2 could not be accurately described by a normal-mode analysis with variable frequency.

In figure 2(a), the frequency shift is plotted versus amplitude $b_2(0, \tilde{t})$ along with the prediction of the weakly nonlinear analysis of Tsamopoulos & Brown (1983) and the experimental results of Wang *et al.* (1996). The numerical solution of the

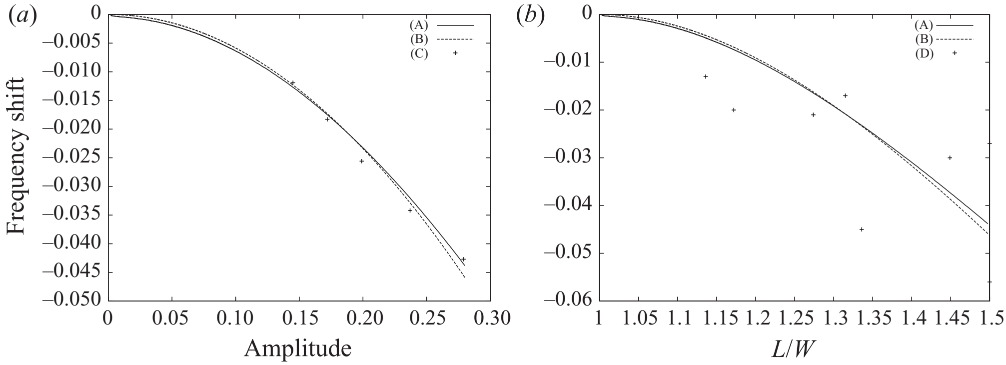

FIGURE 2. Frequency shift versus (*a*) amplitude $b_2(0,\tilde{t})$ and (*b*) aspect ratio L/W: (A) numerical solution of the leading-order problem (3.7)–(3.9), (B) weakly nonlinear analysis of Tsamopoulos & Brown (1983), (C) experimental results of Wang *et al.* (1996) and (D) experimental results of Trinh & Wang (1982).

leading-order outer problem agrees well with Tsamopoulos & Brown (1983); however, the frequency shift is slightly greater at smaller amplitudes and slightly less at larger amplitudes. This trend is also observed in figure 2(*b*), where the frequency shift is plotted versus the ratio of the polar diameter L to equatorial diameter W along with the prediction of Tsamopoulos & Brown (1983) and the experimental results of Trinh & Wang (1982). Our prediction of the amplitude dependence of frequency compares well with the experimental results in figure 2.

3.3. *The modulation equation*

The leading-order initial condition has zero vorticity. Therefore, irrotational flow will persist at leading order in the outer problem, which significantly simplifies the modulation equations (2.18) and (2.20): (2.20) is automatically satisfied and (2.18) becomes

$$\frac{\mathrm{d}}{\mathrm{d}\tilde{t}}(\langle E_0\rangle + V_0) = -2\int_{t^+=-\Psi}^{2\pi-\Psi}\int_{\phi=0}^{2\pi}\int_{\theta=0}^{\pi}\{\boldsymbol{n}_0\cdot\nabla E_0\}\,R_0^2\sin(\theta)\,\mathrm{d}\theta\,\mathrm{d}\phi\,\mathrm{d}t^+. \tag{3.10}$$

As the leading-order solution (3.2)–(3.6) is axisymmetric, the modulation equation (3.10) may be rewritten as

$$\frac{\mathrm{d}}{\mathrm{d}\tilde{t}}(\langle E_0\rangle + V_0) = -4\pi\int_{t^+=-\Psi}^{2\pi-\Psi}\int_{\xi=-1}^{1}\left\{\frac{\partial E_0}{\partial r} - \frac{1}{R_0^2}\frac{\partial R_0}{\partial\theta}\frac{\partial E_0}{\partial\theta}\right\}R_0^2\,\mathrm{d}\xi\,\mathrm{d}t^+. \tag{3.11}$$

The modulation equation (3.11) is solved numerically using the initial condition shown in figure 1, where $\langle E_0\rangle(0) + V_0(0) = 2.32$ and $b_2(0,0) = 0.28$. In figure 3(*a*), the energy and amplitude, $\langle E_0\rangle(\tilde{t}) + V_0(\tilde{t})$ and $b_2(0,\tilde{t})$, are plotted versus the long viscous time scale, $\tilde{t}$, showing an approximately exponential decay. If the oscillation energy and amplitude are damped in time as $\exp(-\lambda\tilde{t})$, then figure 3(*b*) corresponds to the decay rates (λ) in figure 3(*a*). The decay rate varies as a function of the amplitude that has previously been reported by Basaran (1992). Moreover, as the amplitude becomes small, the decay rate approaches the linear theory (1.2) of Lamb (1932). We note that a quadratic approximates the computed free decay rate well for this range of amplitudes.

We seek a comparison with the experimentally observed minima of aspect ratio in figure 1 of Wang *et al.* (1996). The aspect ratio W/L may be approximated using the

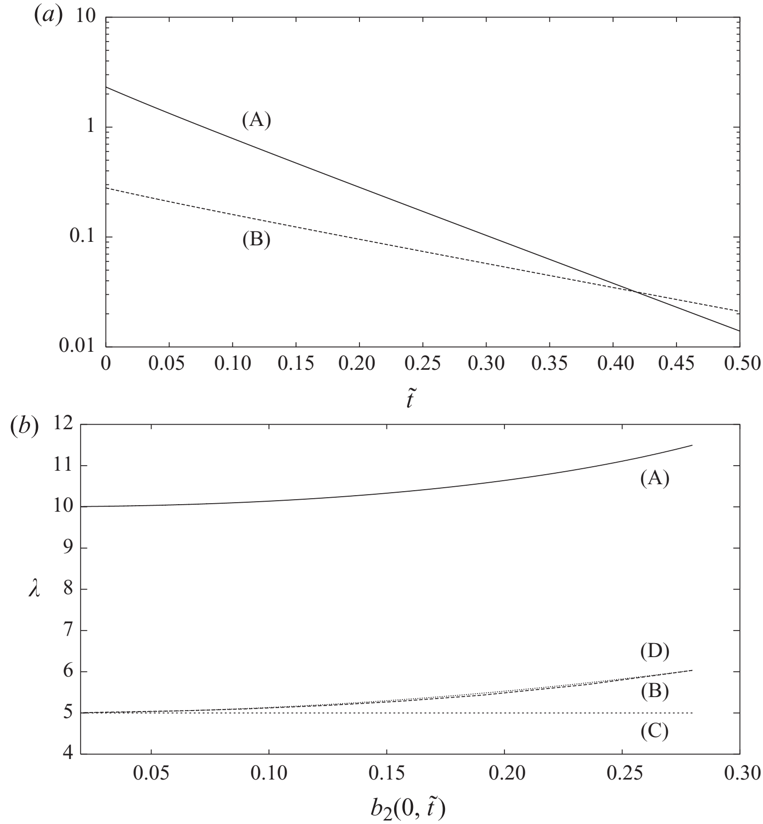

FIGURE 3. (*a*) Numerical solution of the modulation equation (3.11) and (*b*) decay rates versus amplitude $b_2(0, \tilde{t})$: (A) energy $\langle E_0 \rangle(\tilde{t}) + V_0(\tilde{t})$, (B) amplitude $b_2(0, \tilde{t})$, (C) the linear theory (1.2) of Lamb (1932) and (D) the quadratic approximation $5 + 13.2 b_2(0, \tilde{t})^2$.

first two terms in the expansion of Legendre polynomials

$$\frac{W}{L}(0, \tilde{t}) = \frac{b_0(0, \tilde{t}) - \dfrac{b_2(0, \tilde{t})}{2}}{b_0(0, \tilde{t}) + b_2(0, \tilde{t})}.$$

The values of $b_2(0, \tilde{t})$ are determined via the quadratic approximation to the free decay rate and

$$b_2(0, \tilde{t}) = b_2(0, 0) \exp(-\lambda \tilde{t}),$$

where the initial condition $b_2(0, 0)$ is chosen to be consistent with figure 1 of Wang *et al.* (1996). The value of $b_0(0, \tilde{t})$ is obtained by ensuring that mass is conserved (as in (3.1)). The envelope is compared with the experimental minima in figure 4, the agreement being excellent.

In figures 3 and 4 of Becker *et al.* (1991), the Legendre coefficient b_2 is plotted. We seek a comparison with our quadratic approximation to the free decay rate, even though the initial condition is more complicated than that in our calculations (and a satellite droplet merges after 0.4 ms in figure 4). We choose the initial condition $b_2(0, 0)$ to be consistent with Becker *et al.* (1991). The two envelopes are compared with the experimental maxima in figure 5, the agreement being good.

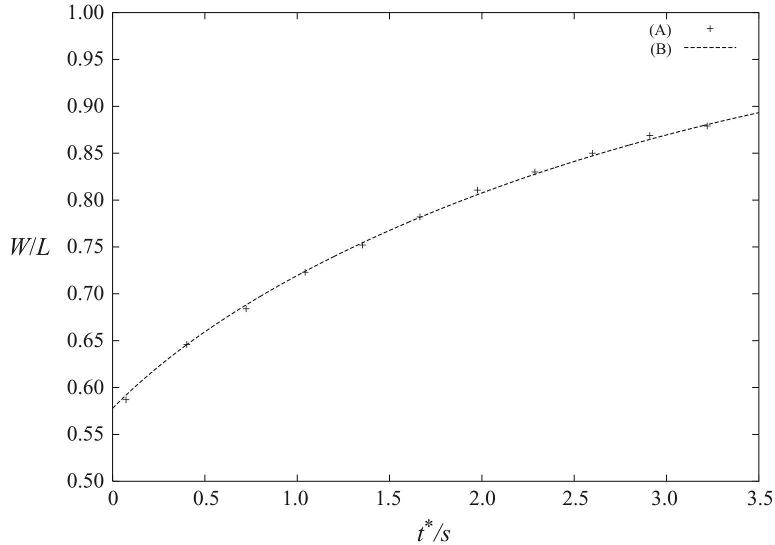

FIGURE 4. Aspect ratio W/L versus time t^*: (A) minima from the experimental results in figure 1 of Wang *et al.* (1996), (B) the envelope from the quadratic decay rate.

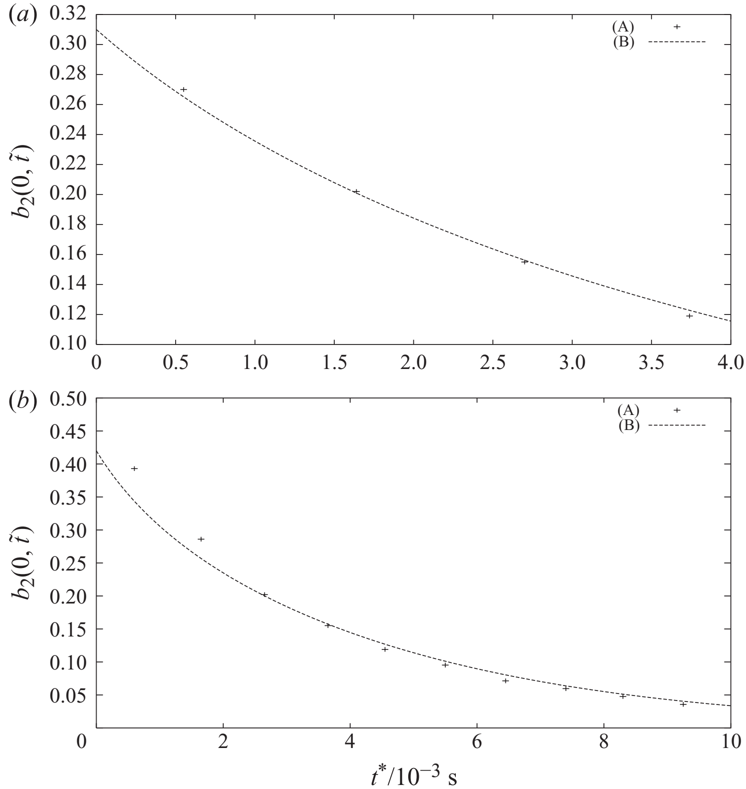

FIGURE 5. Amplitude $b_2(0, \tilde{t})$ versus time t^* for (*a*) figure 3 and (*b*) figure 4 of Becker *et al.* (1991): (A) maxima from the experimental results, (B) the envelope from the quadratic decay rate. We note that, in figure 4 of Becker *et al.* (1991), a satellite droplet merges after 0.4 ms.

The increase in decay rate with amplitude may be understood as follows. At small amplitudes, the drop is approximately spherical and the surface area is a minimum. At large amplitudes, the drop moves from its extreme prolate shape to its extreme oblate, which correspond to greater surface areas than the spherical. The normal derivative of the kinetic energy with respect to $\boldsymbol{n}_0$ integrated over the surface is greater when the surface area is greater. The result is an increase in decay rate.

4. Summary and conclusions

The free decay of large-amplitude oscillations of an incompressible viscous drop is a canonical problem in free-surface flow, the two major nonlinearities of fluid mechanics, convective and free-surface effects, being coupled. In the general case, when the capillary number is order 1, numerical methods are the only viable theoretical approach. However, at small capillary number, the techniques of strongly nonlinear analysis allow two modulation equations to be determined on the long viscous time scale. The first is a generalization of the linear energy equation of Lamb (1932) to the nonlinear problem. The second is the projection of the Navier–Stokes equations onto the vorticity vector, which has not been previously reported in this context. The two modulation equations may form an overdetermined, closed or underdetermined system depending on the number of slowly varying unknowns.

A physical example must be considered to validate and establish the sufficiency of the modulation equations. The axisymmetric oblate–prolate oscillation is considered, which is the most widely studied in experiment and theory. In this case, the second modulation equation is degenerate. We have one modulation equation for the slowly varying oscillation amplitude, this equation proving to be necessary and sufficient. The results for this physical example are verified by comparison with linear viscous theory, weakly nonlinear inviscid theory and experiment.

Trinh & Wang (1982) investigated the decay rate but found it difficult to draw definitive conclusions based on their experiments, although they could perceive a slight increase with larger deformations. Basaran (1992) explained this difficulty in terms of the insensitivity of the decay factor to the deformation during small- and moderate-amplitude oscillations. Basaran (1992) then studied the decay rate at larger amplitudes and different Reynolds numbers. The new result in this paper is to show that the experimental observations of Trinh & Wang (1982) and the numerical results of Basaran (1992) are all manifestations of a single-valued relationship between dimensionless decay rate and amplitude. If the amplitude of the oscillations does not exceed 30 % of the drop radius, then this decay rate may be approximated by the following quadratic:

$$\lambda = 5 + 13.2 b_2(0, \tilde{t})^2.$$

When the amplitude of the oblate–prolate oscillation exceeds approximately 10 % of the drop radius, typical nonlinear phenomena such as the dependence of the oscillation frequency on the amplitude and asymmetry of the amplitude of the free surface are observed. If the amplitude of the oscillations exceeds approximately 20 % of the drop radius, then nonlinear effects become more pronounced. The time dependence of the coefficients in the Fourier–Legendre expansion for the fluid flow shows a zigzag oscillation, this being far from the smooth sinusoidal oscillation that characterizes the linear limit. The fluid in the inviscid bulk of the drop is undergoing abrupt changes in its acceleration in comparison to the acceleration during small-amplitude deformations. The most abrupt change takes place as the free surface passes through the spherical shape.

We note that one property that remains unaltered by the large variation of amplitude is the parity of the oscillations in time. The velocities are odd about the zeros of the radial velocity, while the deformation of the free surface is even. The acceleration of the fluid always changes sign when the free surface is spherical.

This project has benefited from the constructive comments of two anonymous referees.

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