Shape oscillations of a viscoelastic drop

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Small-amplitude axisymmetric shape deformations of a viscoelastic liquid drop in microgravity are theoretically analyzed. Using the Jeffreys constitutive equation for linear viscoelasticity, the characteristic equation for the frequency and decay factor of the shape oscillations is derived. Asymptotic analysis of this equation is performed in the low- and high-viscosity limits and for the cases of small, moderate, and large elasticities. Elastic effects are shown to give rise to a type of shape oscillation that does not depend on the surface tension. The existence of such oscillations is confirmed by numerical solution of the characteristic equation in various regimes. A method for determining the viscoelastic properties of highly viscous liquids based upon experimental measurements of the frequency and damping rate of such shape oscillations is suggested.

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I. INTRODUCTION

Many natural and industrial processes involve shape deformations of liquid drops. Examples include cell division in biology, containerless materials processing in space, impacts between stellar objects, spraying and atomization, evaluating radar cross sections of rain clouds, and indirect measurement of rheological parameters. An example of the latter is as follows: In microgravity, an incompressible liquid drop assumes a spherical shape at equilibrium. This shape can be perturbed by external means. When the external perturbation is removed, the drop eventually returns to its original spherical form. Depending upon the bulk properties of the liquid and the surface parameters this process may take the form of underdamped oscillations about or overdamped aperiodic decay toward the spherical shape. Experimental measurements of the frequency and damping rate of shape oscillations through the acoustic levitation technique would thus enable the physical properties of the liquid to be inferred [1–3].

The study of shape oscillations of liquid drops began with the work of Lord Kelvin [4] where the frequency of inviscid shape oscillations was determined. Lamb [5] developed approximate expressions for the damping rate of weakly viscous liquid drop oscillations. Reid [6] analyzed a viscous liquid drop in a vacuum or low density gas and derived the characteristic equation for the frequency and damping rate of the shape oscillations; this was subsequently solved numerically by Chandrasekar [7]. Miller and Scriven [8] extended Reid's results by including intrinsic surface rheological properties and considering a drop immersed in another immiscible fluid. Further refinements were made by Prosperetti [9] and Marston [10].

In recent years the emphasis has shifted to investigating surface viscoelastic effects on the shape oscillations of weakly viscous drops. In particular, Lu and Apfel [2] considered the case of a purely viscous liquid drop oscillating in another fluid with and without surfactants. Approximate analytical solutions for free-oscillation frequency and damping rate were derived and numerically analyzed by Tian, Holt, and Apfel [11] by introducing surface compositional elasticity and surface dilatational and shear viscosity. Apfel *et al.* [12] demonstrated experimentally the important role of surfactants in liquid drop oscillations under microgravity (see also Ref. [13]). On the numerical side, Lundgren and Mansour [14] first implemented the boundary integral method for numerical simulations of clean axisymmetric drops. The same method was used by Feng and Su [15] to simulate a liquid drop in an acoustic field and by Rush and Nadim [16] for a weakly viscous two-dimensional drop.

To our knowledge, the role of bulk viscoelasticity on the axisymmetric shape oscillations of a liquid drop has not been investigated previously, although transient deformation of a viscoelastic drop in a steady uniaxial extensional flow of a Newtonian liquid has been considered [17]. As we will show here, viscoelasticity of the liquid appears to have a strong influence on the shape oscillations of liquid drops and ignoring this factor when dealing with polymeric and biological liquids may introduce large errors. Moreover, it appears to be feasible to infer the elastic parameters of the liquid from experimental measurements of the frequency and damping rate of shape oscillations.

We present here a complete analysis of small-amplitude axisymmetric shape deformations of viscoelastic liquid drops in microgravity assuming the Jeffreys constitutive equation for linear viscoelasticity. Since during small-amplitude shape oscillations the liquid is subject to small strains, linear viscoelasticity should represent a valid model. We derive the characteristic equation for the frequency and decay factor of the shape oscillations and analyze it asymptotically in the cases of small and large Reynolds number (high- and low-viscosity limits). When the Reynolds number is large, the liquid drop undergoes shape oscillations due to surface tension, i.e., elasticity has a minor effect on the drop dynamics. A decrease in the Reynolds number results in the disappearance of the oscillations. However, upon adding elasticity in that limit (once a critical value of the relaxation time is exceeded), a shape oscillation is seen to emerge, determined solely by the viscous and elastic stresses. This allows an alternative means of estimating the viscoelastic properties of

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liquids based on measuring the frequency and damping rate of liquid drop shape oscillations. We show that a further increase in the relaxation time and viscosity leads to the occurrence of additional shape oscillation modes, so that a large number of such modes exist for incompressible elastic solid balls with a small shear modulus. Numerical solution of the characteristic equation confirms the results of the asymptotic analysis.

II. BASIC EQUATIONS AND BOUNDARY CONDITIONS

Consider a spherical viscoelastic liquid drop of radius R surrounded by a vacuumlike medium. It is assumed that the Bond number $B=gR^2(\rho_l-\rho_m)/\sigma$ is much less than unity, the liquid is incompressible and isothermal, and the drop undergoes small-amplitude deformations. Here g is the acceleration of gravity, ρ_l and ρ_m are the densities of the liquid and the medium respectively, and σ is the surface tension. The continuity equation for the liquid takes the form

$$\nabla\cdot\mathbf{v}=0, \tag{1}$$

and upon neglect of the gravitational force and nonlinear terms the momentum equation reduces to

$$\rho_l\frac{\partial\mathbf{v}}{\partial t}=-\nabla p+\nabla\cdot\tau. \tag{2}$$

The deviatoric stress tensor τ allows for viscoelasticity of the liquid in the form of the linear Jeffreys constitutive equation [18]:

$$\tau+\lambda_1\frac{\partial\tau}{\partial t}=2\mu\left(\dot{\gamma}+\lambda_2\frac{\partial\dot{\gamma}}{\partial t}\right),\quad \dot{\gamma}=\frac{1}{2}(\nabla\mathbf{v}+\nabla\mathbf{v}^\dagger) \tag{3}$$

where $\dot{\gamma}$ is the rate-of-strain tensor, $\mathbf{v}$ is the velocity vector, μ is the shear viscosity, and λ_1 and λ_2 represent the ''relaxation'' and ''retardation'' times.

Equations (1)–(3) need to be supplemented by boundary conditions at the drop surface (s). Denote the outward unit normal and velocity vectors at the surface by $\mathbf{n}$ and $\mathbf{v}_s$ and note that by assumption the stress tensor in the external medium is negligible. The kinematic and dynamic boundary conditions at the surface are then given by

$$\mathbf{v}|_s=\mathbf{v}_s,\quad (p\mathbf{n}-\mathbf{n}\cdot\boldsymbol{\tau})_s=\sigma(\nabla_s\cdot\mathbf{n})\mathbf{n} \tag{4}$$

where $\nabla_s\cdot\mathbf{n}$ is the total surface curvature of the drop, with $\nabla_s\equiv\nabla-\mathbf{nn}\cdot\nabla$.

Consider a spherical coordinate system (r,Θ,φ), introduce a small parameter ε that measures the amplitude of the drop deformation, and assume the shape oscillations to be axisymmetric. For a pure mode, the surface profile of the drop r_s can be expressed in terms of the Legendre polynomial $P_n(\cos\Theta)$

$$r_s=R[1+\varepsilon C_nP_n(\cos\Theta)\exp(-\alpha_nt)], \tag{5}$$

where C_n and $\alpha_n=\delta+i\omega$ are unknown parameters. Obviously, the real part δ of α_n is the amplification or decay factor (positive values of δ correspond to damping) and its imaginary part ω is the angular frequency of shape oscillations

$$\delta=\mathrm{Real}\{\alpha_n\},\quad \omega=\mathrm{Im}\{\alpha_n\}. \tag{6}$$

The general solution for p, $\mathbf{v}$, and τ can then be expanded as

$$p(r,\Theta,t)=\frac{2\sigma}{R}+\varepsilon\rho_l\omega_L^2R^2\,a_n(r)P_n(\cos\Theta)\exp(-\alpha_nt), \tag{7a}$$

$$v_r(r,\Theta,t)=\varepsilon\omega_LR\,b_n(r)P_n(\cos\Theta)\exp(-\alpha_nt), \tag{7b}$$

$$v_\Theta(r,\Theta,t)=\varepsilon\omega_LR\,g_n(r)\frac{dP_n(\cos\Theta)}{d\Theta}\exp(-\alpha_nt), \tag{7c}$$

$$\tau(\mathbf{r},t)=\tau^{(n)}(\mathbf{r})\exp(-\alpha_nt),\quad \dot{\gamma}(\mathbf{r},t)=\dot{\gamma}^{(n)}(\mathbf{r})\exp(-\alpha_nt), \tag{7d}$$

where $\mathbf{r}$ is the position vector and ω_L is the Lamb frequency [5], which for a drop in vacuum is given by

$$\omega_L=\sqrt{\frac{\sigma n(n-1)(n+2)}{\rho_lR^3}}. \tag{8}$$

Substituting Eq. (7d) into the constitutive equation (3) shows that

$$\tau^{(n)}=2\mu_{\mathrm{eff}}\dot{\gamma}^{(n)},\quad \mu_{\mathrm{eff}}=\mu\left(\frac{1-\alpha_n\lambda_2}{1-\alpha_n\lambda_1}\right). \tag{9}$$

We thus see that for an exponential time dependence, viscoelasticity of the liquid can be incorporated into axisymmetric shape oscillations of the drop as a modification of the shear viscosity. It is clear that the momentum equation (2) simply becomes the linearized Navier-Stokes equation in which the shear viscosity μ has been replaced by the effective viscosity μ_{eff}

$$\rho_l\frac{\partial\mathbf{v}}{\partial t}=-\nabla p+\mu_{\mathrm{eff}}\nabla^2\mathbf{v}. \tag{10}$$

When Eqs. (7a)–(7c) are substituted into Eqs. (1) and (10) we obtain

$$a_n(r)=A_n\left(\frac{r}{R}\right)^n, \tag{11a}$$

$$b_n(r)=nA_n\left(\frac{\omega_L}{\alpha_n}\right)\left(\frac{r}{R}\right)^{n-1}+B_n\left(\frac{R}{r}\right)j_n(kr), \tag{11b}$$

$$g_n(r)=A_n\left(\frac{\omega_L}{\alpha_n}\right)\left(\frac{r}{R}\right)^{n-1}+\frac{B_n}{n}\left(\frac{R}{r}\right)\left[j_n(kr)-\frac{(kr)j_{n+1}(kr)}{n+1}\right], \tag{11c}$$

where A_n, B_n are unknown coefficients; j_n is the spherical Bessel function of order n, and $k=\sqrt{\alpha_n\rho_l/\mu_{\mathrm{eff}}}$. The constants A_n, B_n, C_n, and α_n can be found from the boundary conditions (4) that take the forms

$$v_r\Big|_{r=r_s}=\frac{\partial r_s}{\partial t},\quad (p-\tau_{rr})_{r=r_s}=\sigma(\boldsymbol{\nabla}_s\cdot\mathbf{n}),\quad \tau_{r\Theta}\Big|_{r=r_s}=0, \tag{12}$$

with

$$(\boldsymbol{\nabla}_s\cdot\mathbf{n})_{r=r_s}=\frac{1}{R}[2+\varepsilon(n-1)(n+2)C_nP_n(\cos\Theta)\times\exp(-\alpha_n t)].$$

III. DECAY FACTOR AND OSCILLATION FREQUENCY

Upon defining the dimensionless variables

$$\tilde{p}=\frac{p}{\rho_l R^2\omega_L^2},\quad \tilde{\mathbf{v}}=\frac{\mathbf{v}}{R\omega_L},\quad \tilde{t}=\omega_L t,\quad \tilde{r}=\frac{r}{R},$$

and substituting Eqs. (5), (7), and (11) into Eq. (12) we obtain a linear system of algebraic equations in A_n, B_n, C_n (at order ε)

$$nA_n+\frac{\alpha_n j_n(z)}{\omega_L}B_n+\frac{\alpha_n^2}{\omega_L^2}C_n=0, \tag{13a}$$

$$\left[\frac{2n(n-1)}{z^2}-1\right]A_n+\frac{2\alpha_n}{\omega_L z^2}[(n-1)j_n(z)-zj_{n+1}(z)]B_n+\frac{C_n}{n}=0, \tag{13b}$$

$$2(n-1)A_n+\frac{\alpha_n}{\omega_L n(n+1)}[(2n^2-2-z^2)j_n(z)+2zj_{n+1}(z)]B_n=0, \tag{13c}$$

where

$$z=kR=R\sqrt{\frac{\rho_l\alpha_n(1-\lambda_1\alpha_n)}{\mu(1-\lambda_2\alpha_n)}}. \tag{14}$$

Nontrivial solutions of system (13) exist only if its coefficient determinant is zero,

$$2(n-1)(2n+1)-\left(1+\frac{\omega_L^2}{\alpha_n^2}\right)z^2+2Q(z)\times\left[\left(1+\frac{\omega_L^2}{\alpha_n^2}\right)-\frac{2n(n-1)(n+2)}{z^2}\right]=0, \tag{15}$$

where

$$Q(z)=\frac{zj_{n+1}(z)}{j_n(z)}. \tag{16}$$

This characteristic equation for α_n provides a way of evaluating the decay factor δ and the angular frequency ω of free oscillations of a viscoelastic liquid drop. As seen from Eq. (14), the variable z is a more complicated function of α_n compared with the purely viscous case. However, the equation retains its form. Actually, rearrangement of Eq. (15) gives the result

$$\left(\frac{\omega_L}{\alpha_n}\right)^2=\frac{2(n^2-1)}{z^2-2zW(z)}-1+\frac{2n(n-1)}{z^2}\left[1-\frac{(n+1)W(z)}{z/2-W(z)}\right], \tag{17}$$

with $W(z)=j_{n+1}(z)/j_n(z)=Q(z)/z$, which was obtained by Reid [6] for surface oscillations of a viscous liquid drop (see also [7,8]).

Due to the presence of spherical Bessel functions in Eq. (15) it is impossible to calculate α_n at finite values of the shear viscosity μ analytically. Straightforward analysis shows that there are an infinite number of roots of Eq. (15), depending critically on the values of the relaxation and retardation times λ_1 and λ_2, as well as the surface tension σ. In the purely viscous case most of the roots are real and represent various aperiodic modes of decay in the drop shape oscillations. These real roots correspond to the poles of $Q(z)$ [which occur at the zeros of $j_n(z)$] because in the neighborhood of each of these poles the left-hand side of Eq. (15) has a zero [7]. However, the asymptotic behavior of α_n for $z\to 0$ and $z\to\infty$ can be readily established.

Before proceeding with the asymptotic analysis, it is best to choose dimensionless parameters that are appropriate for the limiting cases to be studied. These parameters include the Reynolds number Re, and the relaxation and retardation Deborah numbers De_1 and De_2:

$$\mathrm{Re}=\frac{\rho_l\omega_L R^2}{\mu},\quad \mathrm{De}_1=\omega_L\lambda_1,\quad \mathrm{De}_2=\omega_L\lambda_2. \tag{18}$$

Equation (15) then becomes

$$\left(1+\frac{1}{x^2}\right)z^2-2Q(z)\left[\left(1+\frac{1}{x^2}\right)-\frac{2n(n-1)(n+2)}{z^2}\right]=2(n-1)(2n+1) \tag{19}$$

with

$$z^2=\frac{\mathrm{Re}\,x(1-\mathrm{De}_1 x)}{1-\mathrm{De}_2 x},\quad x=\frac{\alpha_n}{\omega_L}. \tag{20}$$

It is clear that the cases $z\to 0$ and $z\to\infty$ correspond to the high viscosity $\mathrm{Re}\to 0$, and low viscosity $\mathrm{Re}\to\infty$, limits, respectively.

A. High-viscosity limit

For small z, $Q(z)$ can be expanded in powers of z [19]:

$$Q(z)|_{z\to 0}=\frac{z^2}{2n+3}\left[1+\frac{z^2}{(2n+3)(2n+5)}\right]+O(z^6), \tag{21}$$

and Eq. (19) reduces to

$$z^2\left[1+E_n+\frac{1}{x^2}\right]=\frac{2(n-1)(2n^2+4n+3)}{2n+1},$$

$$E_n=\frac{4n(n-1)(n+2)}{(2n+1)(2n+3)(2n+5)}. \tag{22}$$

1. Purely viscous liquid drop

A deformed drop of a highly viscous liquid returns to a spherical shape aperiodically, i.e., there exists a critical value of viscosity μ_c, such that if $\mu>\mu_c$, no shape oscillations occur. This is supported by the numerical solution of Eq. (17) presented by Chandrasekar [7] (see also the next section).

In the high-viscosity limit there are only two possible modes of aperiodic decay. Indeed, in a Newtonian fluid ($\mathrm{De}_1=\mathrm{De}_2=0$) Eq. (22) becomes quadratic in x. Both roots of this equation are real. The first root is proportional to Re and therefore determines the extremely slow decay. It is easy to show by going back to dimensional variables that this mode exists due to a nonzero surface tension (σ or $\omega_L>0$) [7]:

$$\delta_1=x_1\omega_L=\frac{(2n+1)\rho_l R^2\omega_L^2}{2(n-1)(2n^2+4n+3)\mu}$$
$$=\frac{n(n+2)(2n+1)\sigma}{2(2n^2+4n+3)\mu R}. \tag{23}$$

In contrast, the second root gives a decay factor that increases without bound as $\mathrm{Re}\to 0$. The existence of this mode is explained solely by the action of viscous forces on the drop,

$$\delta_2=x_2\omega_L=\frac{2(n-1)(2n^2+4n+3)\mu}{(2n+1)(1+E_n)\rho_l R^2}. \tag{24}$$

It should be noted that the asymptotic expansion (21) holds as $z\to 0$ but not for $z=R\sqrt{\delta_2\rho_l/\mu}\sim O(1)$. Even so, the power series for $Q(z)$ remains convergent and terms of order z^6 (and higher) are rather small such that the high-viscosity root of Eq. (15), corresponding to a very rapid decay, is approximately equal to Eq. (24). For example, in the case of quadrupole deformations ($n=2$), numerical analysis (of the type carried out in Sec. IV) for a typical water drop shows the difference between these roots to be less than a few percent.

2. Viscoelastic liquid drop

The presence of elastic components in the stress tensor has a significant effect on the deformations of a highly viscous drop. In particular, such a drop can undergo shape oscillations rather than just an aperiodic decay. This follows from Eq. (22) that is now cubic

$$\mathrm{De}_1\,\mathrm{Re}\,x^3-(\mathrm{Re}+F_n\,\mathrm{De}_2)x^2+\left(\frac{\mathrm{Re}\,\mathrm{De}_1}{1+E_n}+F_n\right)x$$
$$-\frac{\mathrm{Re}}{1+E_n}=0. \tag{25}$$

This equation has complex roots under the condition that

$$\frac{A_4}{\mathrm{Re}^4}+\frac{A_3}{\mathrm{Re}^3}+\frac{A_2}{\mathrm{Re}^2}+\frac{A_1}{\mathrm{Re}}+A_0>0, \tag{26}$$

where

$$F_n=\frac{2(n-1)(2n^2+4n+3)}{(2n+1)(1+E_n)},$$

$$A_0=\frac{4}{1+E_n}\left[\frac{\mathrm{De}_1^2}{1+E_n}+1\right]^2,$$

$$A_1=\frac{4F_n}{1+E_n}\left[\frac{\mathrm{De}_1^2}{1+E_n}(3\,\mathrm{De}_1-5\,\mathrm{De}_2)-5\,\mathrm{De}_1+3\,\mathrm{De}_2\right],$$

$$A_2=F_n^2\left[-\frac{\mathrm{De}_1^2\,\mathrm{De}_2^2}{(1+E_n)^2}+\frac{2}{1+E_n}\right.$$
$$\left.\times(6\,\mathrm{De}_1^2-11\,\mathrm{De}_1\,\mathrm{De}_2+6\,\mathrm{De}_2^2)-1\right],$$

$$A_3=2F_n^3\left[\frac{\mathrm{De}_2^2}{1+E_n}(2\,\mathrm{De}_2-\mathrm{De}_1)+2\,\mathrm{De}_1-\mathrm{De}_2\right],$$

$$A_4=-F_n^4\,\mathrm{De}_2^2.$$

From Eq. (26) it can be seen that shape oscillations occur for a wide range of relaxation Deborah numbers De_1 but only for small values of the retardation Deborah number De_2. As $\mathrm{Re}\to 0$ the dominant term in this condition is the A_4 term. It is negative and the condition (26) would not be satisfied unless $\mathrm{De}_2^2<4\,\mathrm{Re}\,\mathrm{De}_1/F_n$. We see that shape oscillations of high-viscosity liquid drops appear due to the presence of relaxation terms in the constitutive equation but can be suppressed by the retardation terms. We therefore restrict our attention to liquids with small retardation times.

To obtain asymptotic solutions of Eq. (25), since Re is small, we assume solutions of the form

$$x=\mathrm{Re}^{\nu}[x^{(0)}+\mathrm{Re}^{\varsigma}x^{(1)}+\mathrm{Re}^{2\varsigma}x^{(2)}+\cdots] \tag{27}$$

where ν and ς are to be determined so that the expansion (27) is uniformly valid for $\mathrm{Re}\to 0$.

One obvious possibility is $\nu=\varsigma=1$. We then get a solution that describes a small aperiodic decay of viscoelastic liquid drop deformations by

$$x_1=\frac{\mathrm{Re}}{F_n(1+E_n)}\left[1+\frac{\mathrm{Re}(\mathrm{De}_2-\mathrm{De}_1)}{F_n(1+E_n)}\right]+O(\mathrm{Re}^3). \tag{28}$$

Note that the leading term of Eq. (28) agrees with the purely viscous solution (23). Since the retardation time must always be less than the relaxation one ($\mathrm{De}_2<\mathrm{De}_1$) [18] the decay factor $\delta_1=\omega_L x_1$ decreases with adding elasticity, i.e., a viscoelastic liquid drop returns to its spherical shape more slowly than a purely viscous one.

The other choice of ν and ς that gives the next two solutions is governed by the relative sizes of De_1 and Re. One should choose $\nu=-1$, $\varsigma=1$ for small De_1 and De_2, i.e., in the case of small elasticity

$$\mathrm{De}_1=\xi\,\mathrm{Re},\quad \mathrm{De}_2=\chi\,\mathrm{Re}\quad \text{with}\quad \xi,\chi=O(1).$$

The solution takes the forms

$$x_{2,3}=\frac{(1+\chi F_n)(1\mp X)}{2\xi\,\mathrm{Re}}+\frac{\xi\,\mathrm{Re}[2-(1+\chi F_n)(1\mp X)]}{(1+E_n)[(1+\chi F_n)^2(1\mp X)-4\xi F_n]}+O(\mathrm{Re}^2),\tag{29}$$

where

$$X=\sqrt{1-4\xi F_n(1+\chi F_n)^{-2}},\tag{30}$$

and become complex when $4\xi F_n>(1+\chi F_n)^2$. There therefore exists a critical relaxation time

$$\lambda_{1c}=\frac{(2n+1)(1+E_n)\rho R^2}{8(n-1)(2n^2+4n+3)\mu}\times\left[1+\frac{2(n-1)(2n^2+4n+3)\lambda_2\mu}{(2n+1)(1+E_n)\rho R^2}\right]^2,\tag{31}$$

such that if $\lambda_1>\lambda_{1c}$ and μ is large a viscoelastic liquid drop undergoes shape oscillations. As indicated above the retardation components of the stress tensor hinder the occurrence of oscillations. Increasing λ_2 results in a significant rise in λ_{1c} when the liquid viscosity is large.

The resulting shape oscillations subsist solely on the forces given by the stress tensor (3) and not on surface tension. To show this, we go to dimensional variables, $\alpha_{n2}=\omega_L x_2$, $\alpha_{n3}=\omega_L x_3$. For simplicity, neglect the second term of Eq. (29), assume $\lambda_1=\lambda_{1c}+\Lambda_1$, $\lambda_2=0$ and recall that $\xi=\mu\lambda_1/(\rho_l R^2)$ and $\chi=\mu\lambda_2/(\rho_l R^2)=0$. The angular frequency of shape oscillations then does not depend on ω_L

$$\omega_s=-\mathrm{Im}\{\alpha_{n2}\}=\mathrm{Im}\{\alpha_{n3}\}=\sqrt{\frac{2(n-1)(2n^2+4n+3)\mu\Lambda_1}{(2n+1)(1+E_n)\rho_l R^2\lambda_{1c}^2}},\tag{32}$$

i.e., the oscillations take place even with no surface tension, $\sigma=\omega_L=0$. Thus, elasticity of the liquid leads to another kind of shape oscillation governed only by the viscous and elastic stresses in the drop.

The validity of Eq. (29) is further supported by the fact that the second solution x_2 agrees with the mode of rapid aperiodic decay (24) in the limit $\lambda_1,\lambda_2\to 0$. The third root x_3 comes into existence due to the relaxation components in the stress tensor. When $\lambda_1,\lambda_2\to 0$

$$x_3=\frac{1+\chi F_n}{\xi\,\mathrm{Re}}-\frac{F_n(1-\chi F_n)}{\mathrm{Re}}+o(\xi,\chi).$$

In the purely viscous case x_3 and the decay factor $\delta_3=\omega_L\,\mathrm{Real}(x_3)$ go to infinity, i.e., there is no contribution of this mode to the drop deformation. The inclusion of the relaxation components leads to finite values of x_3. For a small λ_1 we once again have a mode of aperiodic decay. When λ_1 is increased this mode plays an increasingly important part in the drop deformation, because the greater the λ_1, the smaller the decay factor δ_3. But as soon as λ_1 reaches its critical value λ_{1c} the modes of aperiodic decay given by x_2 and x_3 are transformed to a mode of decaying shape oscillations with the frequency (32).

Let us now consider the case of moderate elasticity,

$$\mathrm{De}_2=\chi\,\mathrm{Re},\quad \mathrm{Re}\ll\mathrm{De}_1\ll 1/\mathrm{Re},\quad \chi=O(1).$$

To obtain the complex solutions of Eq. (25) we now take $\nu=-1/2$, $\varsigma=1/2$ and substitute Eq. (27) into Eq. (25). The solutions are in the form

$$x_{2,3}=\frac{1+F_n\chi}{2\,\mathrm{De}_1}+\frac{\mathrm{Re}}{2}\left[\frac{\chi}{(1+E_n)}-\frac{(1+F_n\chi)^3}{4F_n\,\mathrm{De}_1}\right]\mp i\sqrt{\frac{F_n}{\mathrm{Re}\,\mathrm{De}_1}}\left\{1-\frac{\mathrm{Re}}{2F_n}\left[\frac{(1+F_n\chi)^2}{4\,\mathrm{De}_1}+\frac{\mathrm{De}_1}{1+E_n}\right]\right\}+O(\mathrm{Re}^{3/2}).\tag{33}$$

They describe a mode of decaying shape oscillations with the angular frequency

$$\omega_s\approx\sqrt{\frac{2(n-1)(2n^2+4n+3)\mu}{(2n+1)(1+E_n)\rho_l R^2\lambda_1}}\tag{34}$$

and the decay factor

$$\delta_s\approx\frac{1}{2\lambda_1}\left[1+\frac{2(n-1)(2n^2+4n+3)\mu\lambda_2}{(2n+1)(1+E_n)\rho_l R^2}\right].\tag{35}$$

It is significant that the frequency decreases with increasing relaxation and retardation times as seen from Eq. (33). But Eq. (32), which is valid for the relaxation times close to λ_{1c}, exhibits an opposite dependence on frequency. Hence, with increasing λ_1 from λ_{1c} the frequency rises, attaining a maximum value at the relaxation time

$$\lambda_{1m}\approx 3\lambda_{1c},\tag{36}$$

and then falls off. The value λ_{1m} was found from the extremum condition

$$\left.\frac{d\,\mathrm{Im}(x_3)}{d\lambda_1}\right|_{\lambda_1=\lambda_{1m}}=0.$$

The maximal value of the frequency ω_{sm} is found to be

$$\omega_{sm}=\frac{4(n-1)(2n^2+4n+3)\mu}{\sqrt{3}(2n+1)(1+E_n)\rho_l R^2}\times\left[1+\frac{2(n-1)(2n^2+4n+3)\lambda_2\mu}{(2n+1)(1+E_n)\rho R^2}\right]^{-1}. \quad (37)$$

Finally, the high elasticity case

$$\lambda_1=\mu/G, \quad \lambda_2=0 \quad \text{with} \quad G=O(1)$$

is suitable for describing an incompressible elastic solid ''ball'' in a vacuumlike medium because, upon neglecting small terms, the stress tensor is then equivalent to

$$\tau=2G\gamma, \quad (38)$$

where γ is the strain tensor.

The pattern of shape oscillations becomes very complex for large λ_1 (now $\mathrm{De}_1 \gg 1/\mathrm{Re}$). From Eq. (20) it follows that if

$$x>\frac{1}{\mathrm{De}_1}, \quad (39)$$

the argument z is imaginary. This means that the root of Eq. (22) satisfying the condition (39) becomes complex valued. Moreover, in the case of high elasticity the argument z is not small and the high-viscosity asymptotics do not apply. The characteristic equation (15), as indicated above, has an infinite number of real solutions in the purely viscous case. They take values from near zero, given by the slowly decaying mode (23), to infinity. Adding elasticity to the stress tensor causes some of these roots, corresponding to the highest modes of aperiodic decay, to become complex valued. High-frequency modes of shape oscillation come into existence. When the relaxation time is small, all these modes have very large decay factors. They die out far in advance of the normal mode given by the solution (33) and cannot influence the drop deformation. Increasing the relaxation time results in additional complex-valued roots being generated from the real ones, as seen from Eq. (39). Their decay factors are small compared to the ''first'' high-frequency modes because the greater the frequency of a mode, the faster its damping. Finally, in the case of high elasticity, we find a large number of weakly decaying modes of oscillation.

B. Low-viscosity limit

Suppose now that the shear viscosity of the liquid is small so that Re and hence z tend to infinity. The asymptotic form of the characteristic equation is then readily available from Eq. (17). When $z\to\infty$ the function $W(z)$ remains finite everywhere apart from its poles. It is easy to verify that the poles cannot be solutions of Eq. (17) in this limit. Thus we can assume $W(z)$ to be negligibly small compared to z. Equation (17) then reduces to the form

$$\left(1+\frac{1}{x^2}\right)z^2=2(n-1)(2n+1) \quad (40)$$

with z and x given in Eq. (20).

The reduction in the shear viscosity brings into existence shape oscillations in purely viscous liquid drops. In the limit $\mathrm{Re}\to\infty$, as seen from Eq. (40) where De_1 and De_2 are set to zero, the surface of the drop oscillates with the angular frequency $\omega=\omega_L\,\mathrm{Im}(x)$,

$$\omega=\omega_L[1+O(\mathrm{Re}^{-2})]$$

and the decay factor $\delta=\omega_L\,\mathrm{Real}(x)$

$$\delta=\frac{(n-1)(2n+1)\mu}{2\rho_l R^2}.$$

This result was first obtained by Lamb [5] and investigated in detail by Chandrasekar [7]. Obviously, there exists only one mode of surface oscillation and no modes of aperiodic decay in the low-viscosity limit.

Here again elasticity leads to a cubic equation for x,

$$\mathrm{De}_1\,\mathrm{Re}\,x^3-[\mathrm{Re}+G_n\,\mathrm{De}_2]x^2+[G_n+\mathrm{Re}\,\mathrm{De}_1]x-\mathrm{Re}=0, \quad (41)$$

where $G_n=2(n-1)(2n+1)$. Once again, complex roots occur under the condition (26) but with

$$A_0=4[\mathrm{De}_1^2+1]^2,$$

$$A_1=4G_n[\mathrm{De}_1^2(3\,\mathrm{De}_1-5\,\mathrm{De}_2)-5\,\mathrm{De}_1+3\,\mathrm{De}_2],$$

$$A_2=G_n^2[-\mathrm{De}_1^2\,\mathrm{De}_2^2+2(6\,\mathrm{De}_1^2-11\,\mathrm{De}_1\,\mathrm{De}_2+6\,\mathrm{De}_2^2)-1],$$

$$A_3=2G_n^3[\mathrm{De}_2^2(2\,\mathrm{De}_2-\mathrm{De}_1)+2\,\mathrm{De}_1-\mathrm{De}_2],$$

$$A_4=-G_n^4\,\mathrm{De}_2^2.$$

Now $1/\mathrm{Re}\to 0$ and the major contribution to this condition comes from the terms with A_0 and A_1 that are positive for small De_1 and De_2. This is not surprising because a purely viscous liquid drop already undergoes shape oscillations when viscosity is low. The question of interest here is to find when these oscillations do not occur. Increasing De_2 does not accomplish this. The condition for complex roots (i.e., oscillation) is satisfied even when $\mathrm{De}_2=\mathrm{De}_1\to\infty$ because the dominant term in Eq. (26) is the A_0 term (as $1/\mathrm{Re}\to 0$) and even if De_1 and De_2 get large, that term still remains dominant. Hence elasticity has a minor effect on shape oscillations in the low-viscosity limit. Nevertheless, a mode of aperiodic decay appears that is due to viscoelastic properties of the liquid. In order to understand this better, consider the asymptotic solution of Eq. (41). The solution can again be sought in the form (27) with ς less than zero.

Let us restrict our attention to the case of moderate elasticity, so that $\mathrm{De}_1,\ \mathrm{De}_2=O(1)$. In order to obtain all the solutions we suppose $\nu=0$ and $\varsigma=-1$ in Eq. (27). We then obtain an aperiodic decay solution of the form

$$x_1=\frac{1}{\mathrm{De}_1}+\frac{G_n(\mathrm{De}_2-\mathrm{De}_1)}{\mathrm{Re}\,\mathrm{De}_1(1+\mathrm{De}_1^2)}+O(\mathrm{Re}^{-2}) \quad (42)$$

and the oscillatory solutions

$$x_{2,3}=\frac{G_n(1+\mathrm{De}_1\,\mathrm{De}_2)}{2\,\mathrm{Re}(1+\mathrm{De}_1^2)}\pm i\left[1+\frac{G_n(\mathrm{De}_1-\mathrm{De}_2)}{2\,\mathrm{Re}(1+\mathrm{De}_1^2)}\right]+O(\mathrm{Re}^{-2}). \tag{43}$$

The x_1 gives a mode of aperiodic decay that is damped out very fast in the limit $\mathrm{De}_1\to 0$ and, as discussed above, makes no contribution in the subsequent drop deformation in this limit. For the other roots, elasticity lowers the decay factor and slightly enhances the frequency of shape oscillations. In dimensional form

$$\omega=\omega_L\,\mathrm{Im}(x_2)=\omega_L+\omega_{el},$$

$$\omega_{el}=\frac{(n-1)(2n+1)\mu\omega_L(\lambda_1-\lambda_2)}{\rho_l R^2(1+\omega_L^2\lambda_1^2)}. \tag{44}$$

It is noteworthy that the correction to the frequency due to elasticity ω_{el} is at its maximum when $\mathrm{De}_1=1$, i.e., $\lambda_1=1/\omega_L$. In contrast to the high-viscosity limit, there are no shape oscillations when $\sigma=\omega_L=0$, i.e., low-viscosity drops oscillate due to surface tension only.

IV. QUADRUPOLE OSCILLATIONS: NUMERICAL ANALYSIS

In the case of quadrupole deformations ($n=2$), the numerical solution of the characteristic equation (15) has been found using Maple. The first case to be investigated is a water drop of the radius $R=0.1$ mm in zero gravity. A polymer is assumed to be dissolved in the water at low enough concentration that the surface tension, density and shear viscosity are not affected: $\rho_l=10^3\ \mathrm{kg\,m^{-3}}$, $\sigma=0.073\ \mathrm{kg\,s^{-2}}$, $\mu=0.001\ \mathrm{kg\,m^{-1}\,s^{-1}}$, but the drop begins to take on viscoelastic properties. This addresses the question of how elasticity influences the drop deformation. In this case the Lamb frequency ω_L, given by Eq. (8), is $\omega_L\approx 24\,166\ \mathrm{s^{-1}}$. The Reynolds number $\mathrm{Re}=\rho_l R^2\omega_L/\mu$ is much more than unity, $\mathrm{Re}\approx 241$, and one would expect the occurrence of shape oscillations even without elasticity (low-viscosity limit). Figure 1(a) shows the change in the nondimensional frequency of shape oscillations with increasing the relaxation Deborah number $\mathrm{De}_1=\omega_L\lambda_1$ (De_2 is taken to be zero).

The frequency grows with increasing De_1, attains a maximal value at $\mathrm{De}_1=1$, and then slowly falls off. This agrees with the low-viscosity asymptotic formula (43). There is a small difference between theoretical and numerical results that disappears if terms of order 1/Re are accounted for in Eq. (43). The dependence of the decay factor on the relaxation Deborah number is illustrated in Fig. 1(b). We have good agreement between asymptotic and numerical calculations: the decay factor decreases with increasing De_1. The small discrepancy is again due to the neglect of terms of order 1/Re in Eq. (43).

As discussed earlier, highly viscous liquid drops regain their original spherical shapes without oscillation. By this it is meant that there exists a critical value of viscosity μ_c such that if $\mu>\mu_c$ shape oscillations do not occur. Numerical analysis validates this observation. As displayed in Fig. 2 the frequency vanishes at $\mu=\mu_c\approx 0.0655\ \mathrm{kg\,m^{-1}\,s^{-1}}$. Here the density, surface tension, and radius of the drop are identical with those in Fig. 1 but elasticity has not been taken into account ($\mathrm{De}_1=0$). The critical Reynolds number is then

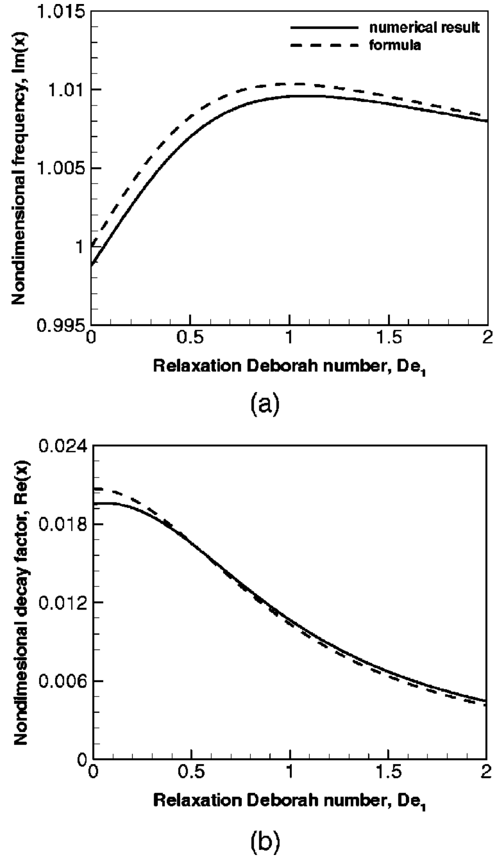

FIG. 1. Quadrupole shape oscillations of a drop comprised of water and a low-fraction polymer material: (a) frequency and (b) decay factor versus the relaxation Deborah number. The solid line is the numerical solution of Eq. (15); the dashed line corresponds to the asymptotic solution (43). Parameters: $R=0.1$ mm, $\rho_l=10^3\ \mathrm{kg\,m^{-3}}$, $\sigma=0.073\ \mathrm{kg\,s^{-2}}$, $\mu=0.001\ \mathrm{kg\,m^{-1}\,s^{-1}}$, $\mathrm{De}_2=0$.

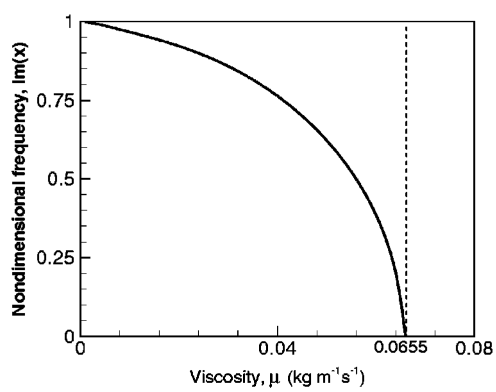

FIG. 2. Quadropule shape oscillations of a purely viscous liquid drop: frequency versus shear viscosity. Parameters: $R=0.1$ mm, $\rho_l=10^3\ \mathrm{kg\,m^{-3}}$, $\sigma=0.073\ \mathrm{kg\,s^{-2}}$, $\mathrm{De}_1=\mathrm{De}_2=0$.

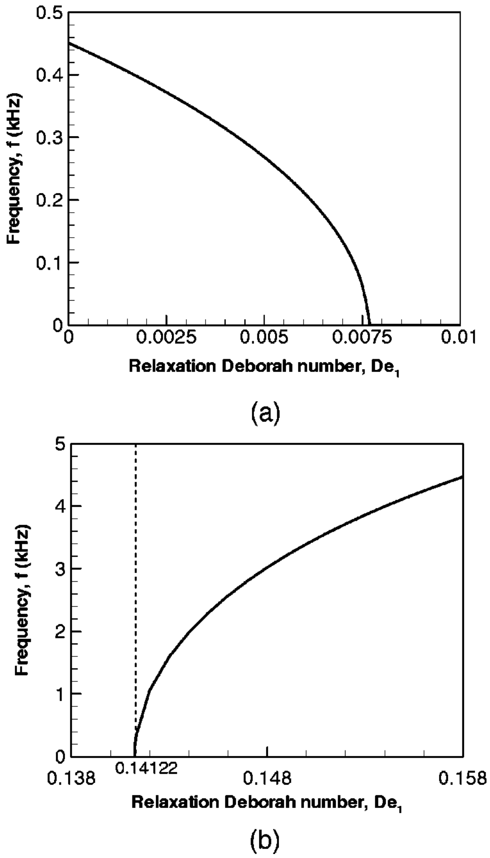

FIG. 3. Variation of frequency of drop shape oscillation versus the relaxation Deborah number: $\mathrm{Re}=3.72$, the remaining parameters are identical with those in Fig. 1.

$\mathrm{Re}=\mathrm{Re}_c\approx 3.69$. Hence, any purely viscous liquid drop with the Reynolds number $\mathrm{Re}<3.69$ does not undergo shape oscillations.

Elasticity of the liquid leads to a reduction in this critical Reynolds number because oscillations disappear even with $\mathrm{Re}=3.72>\mathrm{Re}_c$ [Fig. 3(a)]. What is more important, however, is that a further increase in De_1 to a value above $\mathrm{De}_{1c}\approx 0.14122$ causes a nonzero frequency to appear again [Fig. 3(b)]. A shape oscillation depending on the stress but not on surface tension is generated. The critical relaxation Deborah number De_{1c}, as illustrated in Fig. 4(a), decreases with a reduction in the Reynolds number. Increasing De_1 beyond De_{1c} results in a fast rise of frequency, especially at low Reynolds numbers, to the maximal value $f_m=\omega_m/(2\pi)$ followed by a decrease in frequency with additional increase of De_1. Such an elasticity dependence of frequency is consistent with the results of asymptotic analysis in the case of high viscosity. Figure 4(b) shows how the decay factor depends on elasticity. It decreases monotonically with increasing De_1. Formula (31) approximates the critical relaxation time $\lambda_{1c}=\mathrm{De}_{1c}/\omega_L$ for various values of viscosity very well (Fig. 5). Thus, Eq. (31) gives the actual minimal value of relaxation time needed for ‘‘elastic’’ shape oscillations in most viscoelastic liquid drops.

Figure 6 demonstrates the hinderance due to the retardation time of the occurrence of ‘‘elastic’’ shape oscillations.

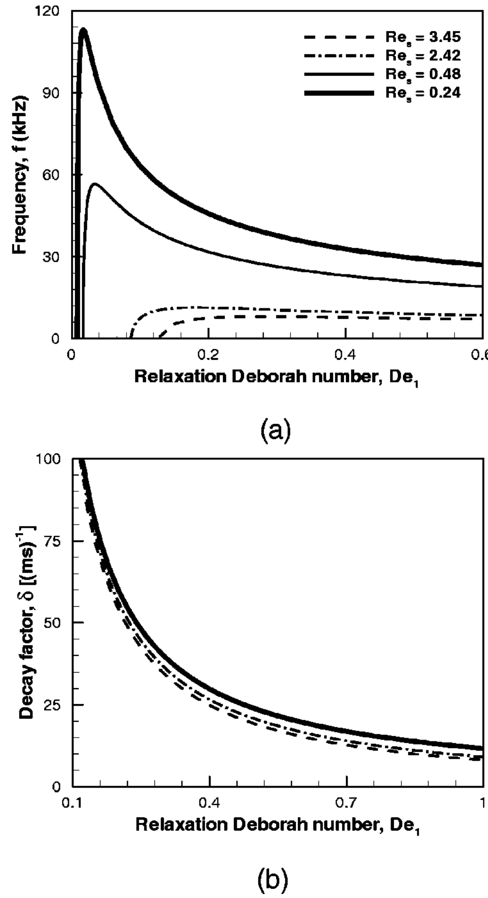

FIG. 4. Variation of frequency and decay factor of drop shape oscillation versus the relaxation Deborah number for different values of shear viscosity ($\mathrm{De}_2=0$).

An increase in the retardation Deborah number De_2 leads to a rise of the critical relaxation Deborah number De_{1c} and a decrease in the frequency of shape oscillation. This is fully in accord with the results of asymptotic analysis.

In the case of moderate elasticity $[\lambda_1=\mathrm{De}_1/\omega_L=O(1/\omega_L)]$ the dependence of frequency and decay factor

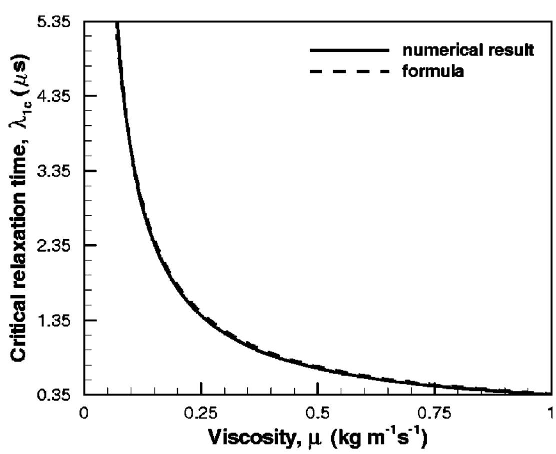

FIG. 5. Comparison between the numerical solution and the approximate formula (31) of the critical relaxation time versus viscosity ($R=0.1$ mm, $\rho_l=10^3$ $\mathrm{kg\,m^{-3}}$, $\sigma=0.073$ $\mathrm{kg\,s^{-2}}$, $\mathrm{De}_2=0$).

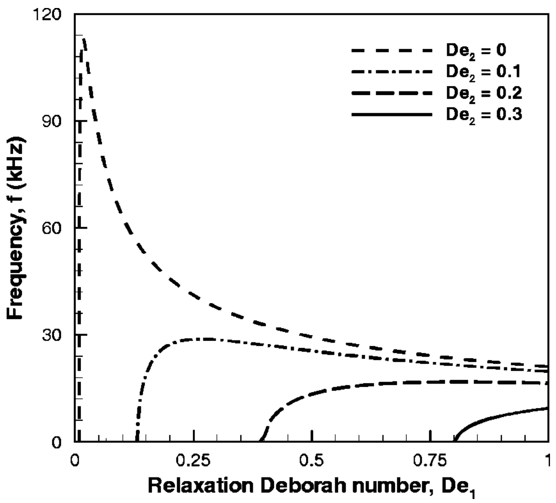

FIG. 6. Variation of the frequency of drop shape oscillation versus the relaxation Deborah number for different retardation Deborah numbers $(\mathrm{Re}=0.24)$.

of viscoelastic drop oscillation on the relaxation time is realistically described by the asymptotic formulas (34) and (35). Numerical analysis verifies the statement that both the frequency and the decay factor decrease with increasing relaxation time (Fig. 7). But there is some quantitative disagreement between the numerical solutions and the asymptotic results at high values of the relaxation time. Actually, increasing the relaxation time leads to an increase in the argument z in the characteristic equation (15) and the high-viscosity approximation ceases to be valid.

As indicated above, when λ_1 and μ tend to infinity, we have a model of an elastic solid sphere instead of a viscoelastic liquid. A large number of shape oscillation modes exist for elastic solid balls. Figure 8 shows decay factors (ordinate) and frequencies (abscissa) for the first 14 modes obtained from the numerical solution of Eq. (15) at $\mu = 100\ \mathrm{kg\,m^{-1}\,s^{-1}}$ and $\lambda_1 = 10$ s, i.e., $G=\mu/\lambda_1 = 10$ Pa. The density, radius, and surface tension are identical with those in Figs. 1 and 2. The first mode has the frequency $\omega_s = \omega_L\,\mathrm{Im}(x) \gtrsim 1/\lambda_1$, i.e., all these modes satisfy the condition (39). The modes are damped out almost simultaneously but slowly, apart from one mode (number 6) that has a much smaller decay factor and can be referred to as the normal mode of oscillation. It is easy to check that the frequency of this mode is almost equal to the Lamb frequency, i.e., it is well approximated by the low-viscosity solution (43). Actually, the relaxation Deborah number is now $\mathrm{De}_1 = \omega_L \lambda_1 \gg \mathrm{Re}$ and the argument z in Eq. (15) is very large and corresponds to the low-viscosity limit. Elastic solid balls therefore undergo high-frequency shape oscillations initially. For long times, however, the remaining dominant angular frequency is simply the Lamb frequency determined by surface tension.

V. CONCLUSION

The characteristic equation determining the frequency and decay factor for shape oscillations of a viscoelastic liquid drop has been derived and investigated analytically and numerically. Asymptotic solutions of the equation obtained in the high-viscosity (low Reynolds number) limit have shown the occurrence of a different kind of shape oscillation once the relaxation time exceeds a critical value. This critical value decreases with increasing viscosity, i.e., even small elasticity would enable a highly viscous liquid drop to undergo shape oscillations.

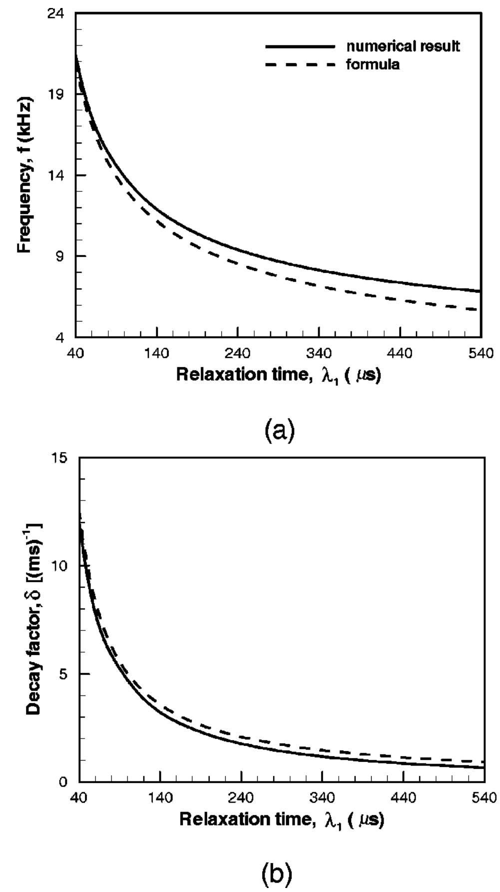

FIG. 7. Comparison between the numerical and asymptotic solutions (34), (35) for the (a) frequency and (b) decay factor of drop shape oscillation as a function of the relaxation Deborah number $(\mathrm{Re}=0.24,\ \mathrm{De}_2=0)$.

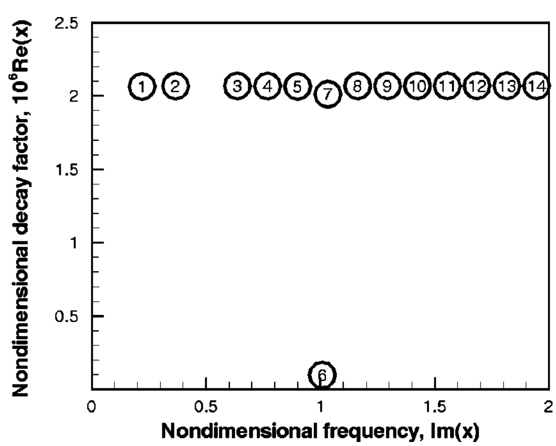

FIG. 8. The first 14 modes of elastic solid ball shape oscillation $(G=10\ \mathrm{Pa})$.

From an experimental point of view, the regime that provides the most promising prospect for measuring the viscoelastic properties is the one of high viscosity ($\mathrm{Re} \ll 1$) and moderate elasticity ($\mathrm{Re} \ll \mathrm{De} \ll 1/\mathrm{Re}$). Suppose such a viscoelastic liquid drop is levitated acoustically and its quadrupole mode ($n=2$) of oscillation is excited, as is routinely done with Newtonian drops and foams. The radius of the drop R, the liquid density ρ_l and the shear viscosity μ are easy to measure independently. Upon measuring the frequency and decay factor of drop surface oscillations from the experiments, one would be able to calculate the relaxation and retardation times λ_1 and λ_2. As seen from Eqs. (34) and (35), these would be given by

$$\lambda_1 \approx \frac{2394}{347} \frac{\mu}{\rho_l R^2 \omega_s^2}, \quad \lambda_2 \approx \frac{2\,\delta_s}{\omega_s^2} - \frac{347}{2394} \frac{\rho_l R^2}{\mu}, \quad (45)$$

under the assumptions that $\rho_l R^2/\mu \ll 1/\omega_L$, $\lambda_1 \gg \rho_l R^2/\mu$, $\lambda_2 \ll \lambda_1$, and $n=2$.

An increase in the relaxation time and viscosity was shown to lead to the occurrence of additional shape oscillation modes, so that a large number of such modes exist for incompressible elastic solid ''balls.'' Nevertheless (and somewhat surprisingly), the dominant angular frequency for shape oscillations of such an elastic ball at long times turns out to be the Lamb frequency that is determined by surface tension.

ACKNOWLEDGMENTS

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