

The oscillations of a fluid droplet immersed in another fluid

By C. A. MILLER AND L. E. SCRIVEN

Department of Chemical Engineering, University of Minnesota,
Minneapolis, Minnesota 55455

(Received 31 May 1967 and in revised form 11 December 1967)

From an analysis of small oscillations of a viscous fluid droplet immersed in another viscous fluid a general dispersion equation is derived by which frequency and rate of damping of oscillations can be calculated for arbitrary values of droplet size, physical properties of the fluids, and interfacial viscosity and elasticity coefficients. The equation is studied for two distinct extremes of interfacial characteristics: (i) a free interface between the two fluids in which only a constant, uniform interfacial tension acts; (ii) an 'inextensible' interface between the two fluids, that is, a highly condensed film or membrane which, to first order, cannot be locally expanded or contracted. Results obtained are compared with those previously published for various special cases.

When the viscosities of both fluids are low, the primary contribution to the rate of damping of oscillations is generally the viscous dissipation in a boundary layer near the interface, in both the free and inextensible interface situations. For this reason inviscid velocity profiles, which do not account for the boundary-layer flow, do not lead to good approximations to the damping rate. The two exceptions in which the approximation based on inviscid profiles is adequate occur when the interface is free and either the interior or exterior fluid is a gas of negligible density and viscosity.

1. Introduction

A droplet of one fluid which is immersed in another fluid assumes a static shape that is nearly spherical when gravitational effects are small in comparison with interfacial tension effects as, for example, when the droplet is small, the two fluid densities are about equal, or interfacial tension is high. If such a droplet is deformed slightly by some external force which is applied and then removed, the droplet will return to its former spherical shape. Depending on the physical properties of the fluids and the composition of the interface this process may involve either a series of oscillations about the spherical shape with continuously decreasing amplitude or else an aperiodic direct return to the spherical shape.

This type of motion of fluid droplets occurs in a wide variety of physical systems. Many mass transfer operations involve small droplets of one fluid immersed in another fluid because such an arrangement provides a relatively large interfacial area for a given volume of the dispersed phase. Emulsions and fluid inclusions in biological cells are other examples. Sometimes the droplets may have mem-

branes or highly condensed films of surface active material at the interface as in cellular organelles and some emulsions. Forces causing initial deformation might be, for example, shear forces resulting from flow or transient gradients in interfacial tension caused by local variations in interfacial concentration or temperature.

In this paper we present a general analysis of small oscillations of a droplet of one viscous fluid immersed in another. The analysis considers an interface with viscoelastic properties but, surprisingly, even for the case of the free interface no general analysis has heretofore been published, to the best of our knowledge. However, solutions do exist for several special cases of the free-interface problem. An expression for oscillation frequency when both fluids are inviscid may be found in Lamb (1932). An analysis applicable to a droplet of a viscous fluid oscillating in a vacuum or low density gas has been performed by Reid (1960) and is conveniently summarized in the book by Chandrasekhar (1961). Lamb (1932) had previously developed approximate expressions for the rate of damping of oscillations for such a droplet when its viscosity is small and for a cavity or bubble of low density gas oscillating in a liquid of low viscosity. In obtaining these expressions he used the velocity fields found for small oscillations of inviscid fluids to estimate the rate of viscous dissipation when viscosity is small but not zero. Valentine, Sather & Heideger (1965) used the same method to obtain an expression for the damping rate when both interior and exterior fluids are liquids of low viscosity. It is established below (§5) that their expression substantially underestimates the damping rate because the inviscid solution, which permits slip to occur at the interface, cannot account for a boundary-layer flow near the interface. This boundary-layer flow is, except in very small droplets, the primary source of viscous dissipation when both fluids are liquids, even when the viscosities are very low.

Various models to describe the rheology of interfaces have been proposed. Such features as interfacial viscosity and elasticity, resistance to bending, and diffusion of surface-active material between interface and bulk fluids have been incorporated in one or more models; these have been adequately described by Oldroyd (1955), Scriven (1960), Goodrich (1961), Levich (1962), Eliassen (1963), Hansen & Mann (1964), and others. In order to simplify the analysis we restrict our consideration to an interface having only viscous and elastic properties. A limiting case of such an interface which can serve as a first approximation for droplets covered by membranes or highly condensed films is the 'inextensible' interface first considered by Lamb (1932). This amounts to an interfacial film sufficiently condensed that no local expansion or contraction may occur. Benjamin (1962) developed an approximate expression relating the rate of decay of oscillations to oscillation frequency for a cavity separated by an inextensible interface from a fluid of low viscosity. We show below that his equation and others for inextensible interfaces can be obtained from the general analysis when the coefficient of either dilatational elasticity or dilatational viscosity becomes very large.

2. Method of analysis. Basic equations

The two bulk fluids are taken as isothermal, incompressible, and Newtonian. It is presumed that the ratio $g_L R^2 \Delta\rho/\gamma$ is sufficiently small that gravitational forces are negligible (g_L is the local acceleration of gravity, R the droplet radius, $\Delta\rho$ the difference in fluid densities and γ the interfacial tension). Moreover, the ratio of radial displacement of the interface, B , to the wavelength along the interface, $2\pi R/l$, say, is supposed to be sufficiently small that the nonlinear term in the Navier-Stokes equation can be neglected (compare Levich 1962). The equation of motion then becomes

$$\frac{\partial \mathbf{v}}{\partial t} = -\frac{1}{\rho} \nabla p + \nu \nabla^2 \mathbf{v}. \quad (1)$$

Taking the curl of this equation gives an equation governing the vorticity field. Its radial component is

$$\left(\frac{\partial}{\partial t} - \nu \nabla^2 \right) (rz) = 0, \quad (2)$$

where z is the radial component of vorticity. Similarly an equation in the radial component of velocity is obtained by taking the radial component of the curl of the vorticity equation

$$\nabla^2 \left(\frac{\partial}{\partial t} - \nu \nabla^2 \right) (rw) = 0, \quad (3)$$

where w is the radial component of velocity. † We suppose that w and z may be expanded in terms of spherical harmonics; each term of the respective expansions has the form

$$rw_{lm} = e^{-\beta_I t} W_{lm}(r) Y_l^m(\boldsymbol{\omega}), \quad (4)$$

$$rz_{lm} = e^{-\beta_I t} Z_{lm}(r) Y_l^m(\boldsymbol{\omega}), \quad (5)$$

where $\boldsymbol{\omega}$ is the position vector of a point on the unit sphere. Note that the real part β_R of β is the amplification or decay factor, a positive β_R corresponding to the latter. The imaginary part β_I is the angular frequency of oscillation. That β depends on l but not m will be seen from the dispersion equation to be derived. As a matter of convenience we shall drop the subscripts l and m except where they are essential for clarity. When (4) and (5) are substituted into (2) and (3) and the resulting differential equations in W and Z are solved, we find

$$W(r) = a_1 r^l + a_2 r^{-l-1} + a_3 \left(\frac{\pi}{2\omega r} \right)^{\frac{1}{2}} \mathcal{Z}_{l+\frac{1}{2}}^{(1)}(\omega r) + a_4 \left(\frac{\pi}{2\omega r} \right)^{\frac{1}{2}} \mathcal{Z}_{l+\frac{1}{2}}^{(2)}(\omega r), \quad (6)$$

$$Z(r) = b_1 \left(\frac{\pi}{2\omega r} \right)^{\frac{1}{2}} \mathcal{Z}_{l+\frac{1}{2}}^{(1)}(\omega r) + b_2 \left(\frac{\pi}{2\omega r} \right)^{\frac{1}{2}} \mathcal{Z}_{l+\frac{1}{2}}^{(2)}(\omega r), \quad (7)$$

where $\omega^2 = \beta/\nu$ and $\mathcal{Z}_{l+\frac{1}{2}}^{(1)}(\omega r)$ and $\mathcal{Z}_{l+\frac{1}{2}}^{(2)}(\omega r)$ are an appropriate pair of independent half-integral-order Bessel functions.

† See Chandrasekhar (1961, pp. 220–222) for a more detailed derivation of these equations.

For an inviscid fluid (3) reduces to the requirement that rw satisfy Laplace's equation; in this instance $W(r)$ is given by the first two terms of (6), the 'inviscid part' of the solution. The two terms involving Bessel functions are the 'viscous part' of the solution. Similarly the entire solution given by (7) can be called the viscous solution for $Z(r)$, since the latter vanishes identically in the inviscid case.

The problem is now to evaluate the time factor β and the constants a_j and b_j of (6) and (7) from the boundary conditions. Once this has been done the problem is completely solved inasmuch as the vector velocity field can be constructed from the scalar w and z fields as has been shown by Sani (1963)

$$\mathbf{v} = \mathbf{e}_r w + \frac{r^2}{l(l+1)} [\nabla_{II} \mathcal{D}w - \mathbf{e}_r \times \nabla_{II} z], \quad (8)$$

where \mathbf{e}_r is the unit vector in the radial direction, ∇_{II} is the surface gradient operator

$$\nabla - \mathbf{e}_r \frac{\partial}{\partial r} \quad \text{and} \quad \mathcal{D} \equiv \frac{1}{r^2} \frac{\partial}{\partial r} r^2.$$

The pressure field may also be constructed from the radial velocity field by combining the divergence of (1) with its radial component.

If $p_{tm} = P_{tm}(r) e^{-\beta t} Y_l^m(\boldsymbol{\omega})$, we find using this procedure that $P(r)$ is given by

$$P(r) = \frac{\mu}{l(l+1)} \frac{\partial}{\partial r} \left[r \left(\frac{\beta}{\nu} + \frac{1}{r^2} \frac{\partial}{\partial r} r^2 \frac{\partial}{\partial r} - \frac{l(l+1)}{r^2} \right) W \right], \quad (9)$$

$P(r)$ is the increment in pressure from its static value due to fluid motion.

3. Boundary conditions

We use the subscript i for quantities associated with the inner fluid and the subscript o for those associated with the outer fluid. Four obvious boundary conditions are that W_i and Z_i must be finite at $r = 0$ and that W_o and Z_o must remain finite as $r \rightarrow \infty$. Applying these conditions gives

$$W_i(r) = a_1 r^l + a_3 \left(\frac{\pi}{2\omega_i r} \right)^{\frac{1}{2}} J_{l+\frac{1}{2}}(\omega_i r), \quad (10)$$

$$W_o(r) = a_2 r^{-l-1} + a_4 \left(\frac{\pi}{2\omega_o r} \right)^{\frac{1}{2}} H_{l+\frac{1}{2}}^{(1)}(\omega_o r), \quad (11)$$

$$Z_i(r) = b_1 \left(\frac{\pi}{2\omega_i r} \right)^{\frac{1}{2}} J_{l+\frac{1}{2}}(\omega_i r), \quad (12)$$

$$Z_o(r) = b_2 \left(\frac{\pi}{2\omega_o r} \right)^{\frac{1}{2}} H_{l+\frac{1}{2}}^{(1)}(\omega_o r), \quad (13)$$

where $J_{l+\frac{1}{2}}$ and $H_{l+\frac{1}{2}}^{(1)}$ are half-integral-order Bessel and Hankel functions of the first kind, respectively.

In addition, velocity must be continuous at the interface and the kinematic condition, which requires radial velocity of the interface itself to equal radial velocity of both fluids at the interface, must be satisfied. The radial displacement

B of the interface from the initial spherical configuration of radius R can be expanded in a series which has terms of the form

$$B_{lm} = B_{o_{lm}} e^{-\beta_l t} Y_l^m(\boldsymbol{\omega}). \quad (14)$$

Then application of the boundary conditions just adduced gives four scalar equations;

(a) kinematic condition

$$\beta B_o + a_1 R^{l-1} + a_3 \left(\frac{\pi}{2\omega_i R^3} \right)^{\frac{1}{2}} J_{l+\frac{1}{2}}(\omega_i R) = 0; \quad (15)$$

(b) continuity of radial velocity

$$a_1 R^l + a_3 \left(\frac{\pi}{2\omega_i R} \right)^{\frac{1}{2}} J_{l+\frac{1}{2}}(\omega_i R) - a_2 R^{-l-1} - a_4 \left(\frac{\pi}{2\omega_o R} \right)^{\frac{1}{2}} H_{l+\frac{1}{2}}^{(1)}(\omega_o R) = 0. \quad (16)$$

Without the Bessel-function terms these two equations are boundary conditions which can be satisfied when both fluids are inviscid. However, the equations requiring continuity of tangential velocity, which are considered next, cannot, in general, be satisfied by inviscid fluids. In fact the occurrence of slip at solid-liquid and liquid-liquid interfaces is a characteristic feature of inviscid solutions.

Because tangential velocity is a two-dimensional vector, requiring its continuity must involve two scalar equations. Although a pair of equations could be obtained by taking components with respect to some co-ordinate system on the interface, it proves far more convenient to work instead with the scalar equations obtained by taking the surface divergence and the radial component of the surface curl of the continuity condition in the vector form, $\mathbf{v}_{II_i} = \mathbf{v}_{II_o}$. The advantage of this procedure results from the fact that the surface divergence equation contains only the radial velocities and not the radial vorticities while the surface curl equation contains the radial vorticities alone. The equations are as follows:

(c) surface divergence

$$a_1 (1-l) R^{l-2} + a_3 \left(\frac{\pi}{2\omega_i R^5} \right)^{\frac{1}{2}} [(1-l) J_{l+\frac{1}{2}}(\omega_i R) + \omega_i R J_{l+\frac{3}{2}}(\omega_i R)] \\ - a_2 (l+2) R^{-l-3} - a_4 \left(\frac{\pi}{2\omega_o R^5} \right)^{\frac{1}{2}} [(1-l) H_{l+\frac{1}{2}}^{(1)}(\omega_o R) + \omega_o R H_{l+\frac{3}{2}}^{(1)}(\omega_o R)] = 0; \quad (17)$$

(d) radial component of surface curl

$$b_1 \left(\frac{\pi}{2\omega_o R} \right)^{\frac{1}{2}} J_{l+\frac{1}{2}}(\omega_i R) - b_2 \left(\frac{\pi}{2\omega_o R} \right)^{\frac{1}{2}} H_{l+\frac{1}{2}}^{(1)}(\omega_o R) = 0. \quad (18)$$

The final boundary condition is the balance of forces at the interface. It again proves convenient to replace the vector boundary condition by an equivalent set of three scalar equations. These are the radial component, the surface divergence and the radial component of the surface curl of the vector force balance. These three equations have been worked out by Bupara (1964), for the case of an interface having viscous properties (Scriven 1960). Forces stemming from elastic behaviour are easily included because they depend on interfacial strain in the

same manner that viscous forces depend on the interfacial rate of strain. The equations incorporating both are:

(a) radial component

$$\begin{aligned}
 & -B_0\beta^{*2} \frac{\Gamma}{l(l+1)} + a_1 R^{l-1} \left[\frac{\rho_i \beta}{l} - \frac{2(l-1)(\mu_i - D)}{R^2} \right] + a_3 \left(\frac{\pi}{2\omega_i R^3} \right)^{\frac{1}{2}} \left[\frac{2(\mu_i - D)}{R^2} \right] \\
 & [(1-l)J_{l+\frac{1}{2}}(\omega_i R) + \omega_i R J_{l+\frac{3}{2}}(\omega_i R)] + a_2 R^{-l-2} \left[\frac{\rho_o \beta}{l+1} - \frac{2\mu_o(l+2)}{R^2} \right] \\
 & - a_4 \left(\frac{\pi}{2\omega_o R^3} \right)^{\frac{1}{2}} \frac{2\mu_o}{R^2} [(1-l)H_{l+\frac{1}{2}}^{(1)}(\omega_o R) + \omega_o R H_{l+\frac{3}{2}}^{(1)}(\omega_o R)] = 0. \quad (19)
 \end{aligned}$$

In this equation β^* is the frequency of oscillation for two inviscid fluids and is inserted merely to simplify the form of this equation and results to be obtained subsequently. It is given by (cf. Lamb 1932)

$$\beta^* = \left(\frac{\gamma l(l+1)(l-1)(l+2)}{R^3 \Gamma} \right)^{\frac{1}{2}}. \quad (20)$$

Other quantities introduced in these equations are Γ , which is given by $p_0 l + \rho_i(l+1)$, the interfacial tension γ , the coefficient of interfacial dilatational viscosity κ , the coefficient of interfacial dilatational elasticity Λ , and D , which is $(\kappa/R) - (\Lambda/\beta R)$;

(b) surface divergence

$$\begin{aligned}
 & a_1 R^{l-1}(l^2-1)[2\mu_i + S(l+2) + Dl] + a_3 \left(\frac{\pi}{2\omega_i R^3} \right)^{\frac{1}{2}} [\mu_i(2(l^2-1)J_{l+\frac{1}{2}}(\omega_i R) \\
 & - \omega_i^2 R^2 J_{l+\frac{1}{2}}(\omega_i R) + 2\omega_i R J_{l+\frac{3}{2}}(\omega_i R)) + S(l+2)(l-1)((l+1)J_{l+\frac{1}{2}}(\omega_i R) \\
 & - \omega_i R J_{l+\frac{3}{2}}(\omega_i R)) - Dl(l+1)((1-l)J_{l+\frac{1}{2}}(\omega_i R) + \omega_i R J_{l+\frac{3}{2}}(\omega_i R))] \\
 & - a_2 R^{-l-2} 2l(l+2)\mu_o - a_4 \left(\frac{\pi}{2\omega_o R^3} \right)^{\frac{1}{2}} [2(l^2-1)H_{l+\frac{1}{2}}^{(1)}(\omega_o R) - \omega_o^2 R^2 H_{l+\frac{1}{2}}^{(1)}(\omega_o R) \\
 & + 2\omega_o R H_{l+\frac{3}{2}}^{(1)}(\omega_o R)] = 0, \quad (21)
 \end{aligned}$$

here ϵ is the coefficient of interfacial shear viscosity, M is the coefficient of interfacial shear elasticity, and S is given by $(\epsilon/R) - (M/\beta R)$;

(c) radial component of surface curl

$$\begin{aligned}
 & b_1 \left(\frac{\pi}{2\omega_i R} \right)^{\frac{1}{2}} \{ [(l-1)\mu_i + (l-1)(l+2)S] J_{l+\frac{1}{2}}(\omega_i R) - \mu_i \omega_i R J_{l+\frac{3}{2}}(\omega_i R) \} \\
 & - b_2 \left(\frac{\pi}{2\omega_o R} \right)^{\frac{1}{2}} \mu_o [(l-1)H_{l+\frac{1}{2}}^{(1)}(\omega_o R) - \omega_o R H_{l+\frac{3}{2}}^{(1)}(\omega_o R)] = 0. \quad (22)
 \end{aligned}$$

4. Evaluation of decay factor and oscillation frequency

Equations (15), (16), (17), (18), (19), (21) and (22) are seven linear homogeneous equations in the seven unknowns $B_o, a_1, a_2, a_3, a_4, b_1$ and b_2 . Hence non-trivial solutions exist only if the determinant of coefficients is zero. The constants b_1 and b_2 appear only in (18) and (22) and the other five unknowns appear only in the other five equations; therefore the determinant of coefficients may be

written as the product of two smaller determinants. The following definitions simplify the form of the determinants:

$$\begin{aligned}
 Q_{l+\frac{1}{2}}^J &\equiv \frac{J_{l+\frac{1}{2}}(\omega_i R)}{J_{l+\frac{1}{2}}(\omega_o R)}, & Q_{l+\frac{1}{2}}^H &\equiv \frac{H_{l+\frac{1}{2}}^{(1)}(\omega_o R)}{H_{l+\frac{1}{2}}^{(1)}(\omega_o R)}, \\
 L_1 &\equiv \frac{\beta\rho_i}{l} - \frac{2(l-1)(\mu_i - D)}{R^2}, \\
 L_2 &\equiv (l^2 - 1)[2\mu_i + S(l+2) + lD], \\
 L_3 &\equiv \frac{2(\mu_i - D)}{R} \omega_i Q_{l+\frac{1}{2}}^J - \frac{\beta\rho_i}{l}, \\
 L_4 &\equiv \mu_i[-\omega_i^2 R^2 + 2\omega_i R Q_{l+\frac{1}{2}}^J] - S(l+2)(l-1)\omega_i R Q_{l+\frac{1}{2}}^J - Dl(l+1)\omega_i R Q_{l+\frac{1}{2}}^J, \\
 L_5 &\equiv -\frac{\beta\rho_o}{l+1} + \frac{2\mu_o}{R^2} C(H), \\
 C(H) &\equiv 2l+1 - \omega_o R Q_{l+\frac{1}{2}}^H, \\
 F(H) &\equiv 2(2l+1) + \omega_o^2 R^2 - 2\omega_o R Q_{l+\frac{1}{2}}^H.
 \end{aligned}$$

With these definitions we have

$$\begin{vmatrix}
 1 & & & & 1 & & & & & & \\
 (l-1)(\mu_i + S(l+2)) - \mu_i \omega_i R Q_{l+\frac{1}{2}}^J & & & & \mu_o(l-1 - \omega_o R Q_{l+\frac{1}{2}}^H) & & & & & & \\
 \beta & 1 & 0 & & 0 & & & & 0 & & \\
 0 & 1 & 0 & & -1 & & & & 0 & & \\
 0 & 1-l & \omega_i R Q_{l+\frac{1}{2}}^J & & -(l+2) & & & & C(H) & & \\
 \frac{-\beta^* \Gamma}{l(l+1)} & L_1 & L_3 & & \frac{\rho_o \beta}{l+1} - \frac{2\mu_o(l+2)}{R^2} & & & & L_5 & & \\
 0 & L_2 & L_4 & & -2\mu_o l(l+2) & & & & \mu_o F(H) & &
 \end{vmatrix} = 0. \quad (23)$$

There are two solutions to (23) corresponding to two types of wave motion which may occur in and around a droplet. In one solution the smaller of the two determinants vanishes while the larger one does not. The former condition determines β ; the latter implies there is no radial displacement of the interface and no radial component of velocity at any point inside or outside the droplet. There is, however, a radial component of vorticity and hence, according to (8), there are tangential velocities. Such motion might be called a shear wave or purely rotational wave. It can be shown that waves of this type always decay without oscillation, i.e. $\beta_I = 0$. Scriven & Sternling (1964) proved this result for the case of a single fluid having a plane interface, and Bupara (1964) extended their proof to include the case of two fluids separated by a spherical interface, the situation under study here. We are concerned with surface waves and so consider the aperiodically damped, purely rotational motions no further.

The other solution of (23) is the more familiar surface wave in which radial velocities and radial displacement of the interface do occur. However, because the smaller determinant of (23) is not zero, the only solution for the b_j 's is the trivial one and there is no radial component of vorticity. The equation obtained by setting the larger determinant equal to zero is an implicit equation which can

be solved numerically to obtain values of decay factor β_R and oscillation frequency β_I for any mode number l , in terms of droplet radius R , inviscid oscillation frequency β^* , physical properties of the two fluids, and coefficients of interfacial elasticity and viscosity.

5. Behaviour when the interface is free

If the droplet is separated from the exterior fluid by a relatively clean, simple interface the coefficients of interfacial viscosity and elasticity are zero and the dispersion equation for the surface-wave solution becomes

$$\begin{vmatrix} \beta & 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & -1 & 0 \\ 0 & 1-l & \omega_i R Q_{l+\frac{1}{2}}^J & -(l+2) & C(H) \\ -\beta^* \Gamma & \beta \rho_i & \frac{2(l-1)\mu_i}{R^2} & \frac{2\mu_i \omega_i}{R} Q_{l+\frac{1}{2}}^J - \frac{\beta \rho_i}{l} & \frac{\beta \rho_o}{l+1} - \frac{2\mu_o(l+2)}{R^2} & \frac{-\beta \rho_o}{l+1} + \frac{2\mu_o}{R^2} C(H) \\ 0 & 2\mu_i(l^2-1) & \mu_i G(J) & -2\mu_o l(l+2) & \mu_o F(H) \end{vmatrix} = 0, \quad (24)$$

where $G(J) \equiv -\omega_i^2 R^2 + 2\omega_i R Q_{l+\frac{1}{2}}^J$.

This equation is still sufficiently complex that solution by numerical methods is usually required. Although we have done no numerical work, it does appear that systematic investigation of the variation of β with fluid properties would be of considerable interest. A full tabulation of computed values would have more than routine use, for it would bring out basic features of the physical behaviour of oscillating droplets, features such as the transition between oscillatory and aperiodic decay, the number and character of solutions for β in various ranges of physical properties, and bounds on the values of β_R and β_I for different solutions. If one may judge from recent discussion of oscillations of two fluids separated by a plane interface, determining all of the significant features of droplet oscillation is scarcely likely to be a trivial matter. The work of Willson (1965) has shown, for example, that, contrary to the belief previously held by some workers, there are important aspects of wave propagation on and instability of plane interfaces which are not revealed by study of the special case of two fluids having equal kinematic viscosities. In particular, Willson established that if the two viscosities are equal but one fluid is of much greater density, there is a range of wavelengths in which only one mode of aperiodic decay exists although two modes are found when the fluids have equal kinematic viscosities. The behaviour of β for other relationships between properties of the two fluids has evidently not yet been studied. But just as detailed study of the analogous plane problem has been fruitful so should further scrutiny of the general equation for droplet oscillation.

There are several special cases in which (24) becomes considerably simplified and it is instructive to consider these. As indicated below, the results for inviscid fluids, for a liquid droplet in gas, and some of those for a cavity are already well known. The remainder, notably those applicable when both fluids are liquids (§§5D and E), appear not to have been published previously. All the results, both old and new, that are presented here are comprehensively summarized in figure 1.

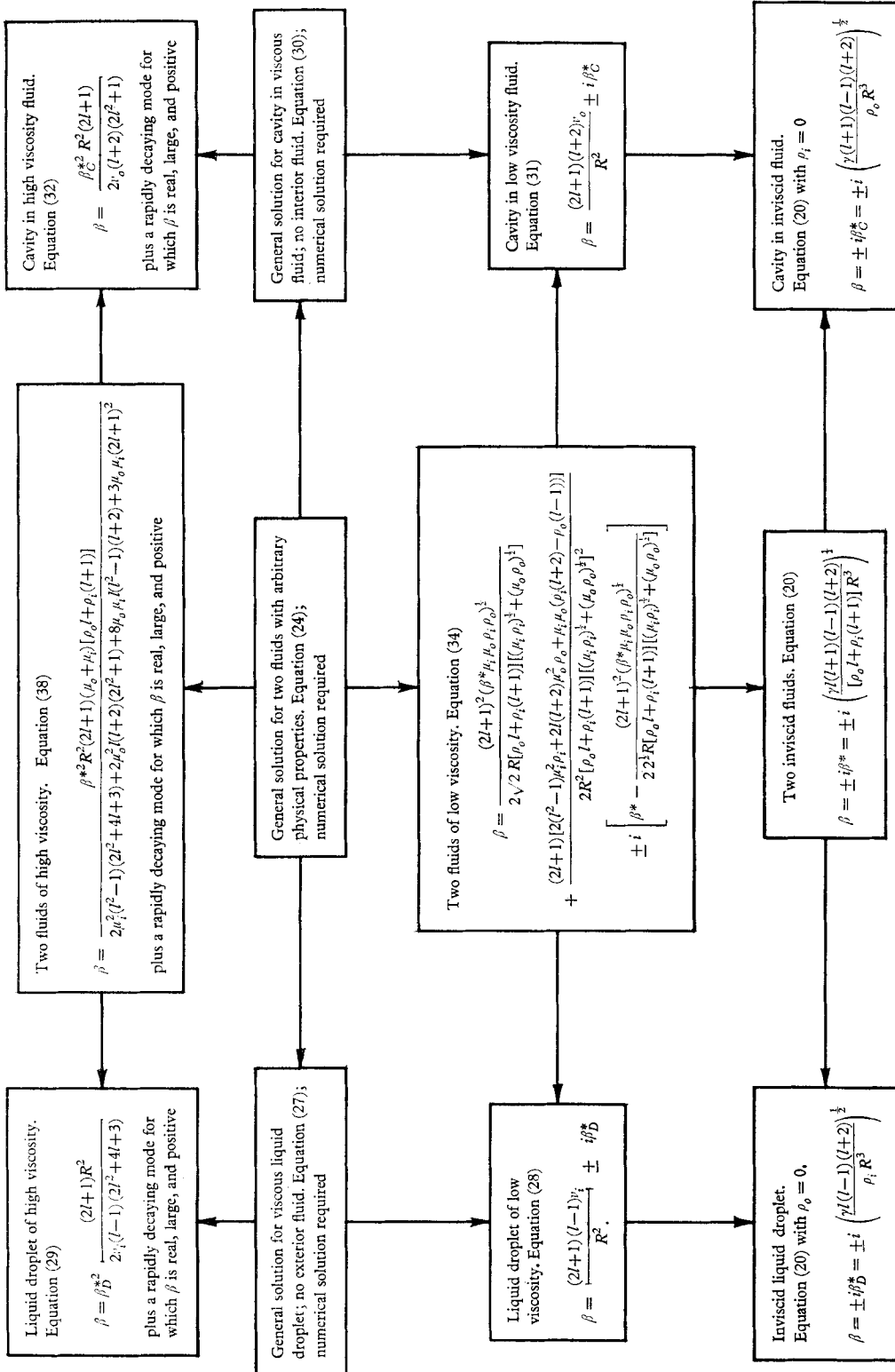


FIGURE 1. Summary of equations for time factor β for free interface analysis. The real part of β is the decay factor; the imaginary part of β is the angular frequency of oscillation.

(A) *Inviscid fluids*

For inviscid fluids only the inviscid part of the radial velocity solution (6) appears. Also, as previously noted, equation (17), the requirement of continuity of tangential velocity at the interface cannot be satisfied. These features of the case of inviscid fluids result in elimination of the third and fifth columns and the third row of the determinant in (24). The fifth row, which amounts to requiring continuity of shear stress at the interface, clearly vanishes as well for inviscid fluids. On expanding the remaining terms of the determinant, we find

$$\beta = \pm i\beta^*. \quad (25)$$

Therefore, as would be expected, there is neither amplification nor decay and the frequency of oscillation is β^* as defined by (20). Indeed this is how β^* would be found if Lamb's result has not been previously substituted to simplify the form of (19).

Calculating velocities of the interior and exterior fluids by means of (8) makes obvious the slip which occurs at the interface. We find that the tangential velocities of the two fluids are in precisely opposite directions and that, except at points where $\nabla_{II} Y_l^m = 0$ (in which case both tangential velocities are zero), the magnitudes of the tangential velocities are related by

$$\frac{|\mathbf{v}_{II_i}|}{|\mathbf{v}_{II_o}|} = \frac{l+1}{l}. \quad (26)$$

It can be shown (see Lamb 1932; Benjamin 1962) that results obtained for fluids separated by a spherical interface should approach those found for the corresponding plane interface problems when droplet radius R and the mode number l (order of spherical harmonic) simultaneously become very large. The ratio l/R is maintained constant while passing to the limit, at which it is identified with the wave-number k of simple oscillations of a plane interface. In this light it is clear from (26) that in the limit the tangential velocities become approximately equal in magnitude and opposite in direction; this is in agreement with the result found for oscillation of two inviscid fluids separated by a plane interface (see Lamb 1932).

(B) *Oscillation of a viscous liquid droplet in a vacuum or gas of negligible density and viscosity*

If the exterior fluid is so rarefied that its effect on motion of the droplet is negligible, the determinant in (24) is considerably simplified. Expansion and rearrangement give

$$\frac{\beta^{*2}}{\beta^2} = \frac{2(l^2-1)}{\omega_i^2 R^2 - 2\omega_i R Q_{l+\frac{1}{2}}^J} - 1 + \frac{2l(l-1)}{\omega_i^2 R^2} \left[1 - \frac{(l+1) Q_{l+\frac{1}{2}}^J}{\frac{1}{2}\omega_i R - Q_{l+\frac{1}{2}}^J} \right], \quad (27)$$

which is equivalent to the result of Reid (1960). A good discussion of this problem is given by Chandrasekhar (1961), who has computed some values of β using (27). His discussion also includes consideration of the following limiting cases.

(a) *Low viscosity*

In the limit as $\nu_i \rightarrow 0$, (27) reduces to

$$\beta = \frac{\nu_i(l-1)(2l+1)}{R^2} \pm i\beta_D^* \quad (28)$$

where β_D^* is the frequency of oscillation of an inviscid liquid droplet obtained by setting ρ_0 equal to zero in (20). The decay factor of (28) was obtained by Lamb (1932), who estimated the rate of viscous dissipation using the velocity distribution for inviscid oscillations. When (28) is reduced to a plane interface formula in the manner described above, a decay factor of $2k^2\nu_i$ is obtained, which agrees with the result given by Lamb (1932).

(b) *High viscosity*

As viscosity increases a point is reached beyond which oscillations no longer occur and a deformed droplet returns to the spherical shape aperiodically. Actually it turns out that there are two possible modes of aperiodic decay. One of these has a decay factor which increases without bound as $\nu_i \rightarrow \infty$, corresponding to a very rapid decay; the other has a decay factor which becomes very small, corresponding to a very slow decay. The latter mode is described by the following equation

$$\beta = \beta_D^{*2} \frac{(2l+1)R^2}{2(l-1)(2l^2+4l+3)\nu_i} \quad (29)$$

A similar result, i.e. one rapidly decaying aperiodic mode and one slowly decaying aperiodic mode, is found in the high-viscosity limit for the plane interface problem. The smaller decay factor is precisely that obtained by reduction of (29) to a plane interface formula (cf. Lamb 1932; Wehausen & Laitone 1960).

(C) *Oscillation of a cavity or bubble of gas with negligible density and viscosity in a viscous liquid*

This special case is very similar to that of the viscous liquid droplet just discussed, except that now the interior fluid is considered to be so rarefied that it has a negligible effect on the outer fluid. In this case (24) can be reduced to

$$\frac{\beta^{*2}}{\beta^2} = \frac{l+2}{\omega_o^2 R^2} \left[\frac{(2l+1)\omega_o^2 R^2 - 2(l-1)(l+1)(2l+1 - \omega_o R Q_{l+\frac{1}{2}}^H)}{2l+1 - \omega_o R Q_{l+\frac{1}{2}}^H + \omega_o^2 R^2/2} \right] - 1. \quad (30)$$

Oscillation frequency and decay factor must be obtained by numerical solution of an awkwardly transcendental equation which is quite similar to that for a droplet and can be treated by the same techniques (Chandrasekhar 1961). Limiting cases are as follows.

(a) *Low viscosity.* As $\nu_o \rightarrow 0$, $|\omega_o| = (|\beta|/\nu_o)^{\frac{1}{2}} \rightarrow \infty$; neglecting terms higher than first order in $(\omega_o R)^{-2}$, we obtain a quadratic equation in β which has the solutions

$$\beta = \frac{(2l+1)(l+2)\nu_o}{R^2} \pm i\beta_C^* \quad (31)$$

where β_C^* , the oscillation frequency for a cavity in an inviscid fluid, may be obtained by setting $\rho_i = 0$ in (20). With regard to the sign of the imaginary part of β we note that choosing the Hankel function of the first, rather than the second, kind in (11) depends on maintaining $\omega_I \geq 0$. For decaying oscillations it is easily shown that if $\beta_I \geq 0$, the proper sign of ω_I is obtained by taking the square root of β having positive real part, i.e. $\omega_R \geq 0$. Likewise if $\beta_I \leq 0$, the root with $\omega_R \leq 0$ must be chosen.

The decay factor of (31) agrees with that obtained by Lamb (1932) using the same method as that used in deriving his result given above in (28) for the low viscosity liquid droplet. It also reduces to the decay factor given above for a low viscosity liquid pool.

(b) *High viscosity*. In the limit $\nu_o \rightarrow \infty$, $\omega_o \equiv (|\beta|/\nu_o)^{\frac{1}{2}} \rightarrow 0$, (30) yields

$$\beta = \beta_C^{*2} \frac{(2l+1)R^2}{2(l+2)(2l^2+1)\nu_o}. \quad (32)$$

This describes a slowly decaying aperiodic mode; (30) also implies the existence of a second, rapidly decaying aperiodic mode. The high-viscosity situation is thus the same as in the liquid droplet and liquid pool. As was found for the corresponding liquid droplet result, (29), equation (32) in the limit as $R \rightarrow \infty$, $l \rightarrow \infty$, with the ratio l/R constant, reduces to the formula for the slowly decaying aperiodic mode in a liquid pool of high viscosity.

(D) Oscillation of two viscous fluids having small viscosities

For both the oscillating liquid droplet and the oscillating cavity just considered it is found that in the low-viscosity limit the oscillation frequency approaches the inviscid frequency and the decay factor approaches that given by Lamb's analysis employing the inviscid velocity profiles. Hence we might expect that two fluids of low viscosity would, in a similar fashion, oscillate with approximately the inviscid frequency and with the decay factor derived by Valentine *et al.* (1965) using Lamb's method. Were this so the expression for β would be†

$$\beta = \frac{(2l+1)}{R^2\Gamma} [(l^2-1)\mu_i + l(l+2)\mu_o] \pm i\beta^*. \quad (33)$$

In fact, when the determinant in (24) is expanded and the low viscosity limit is considered with terms retained to the same order as in the low viscosity results for droplet and cavity, the following expression is obtained:

$$\beta = \frac{(2l+1)^2(\beta^*\mu_i\mu_o\rho_i\rho_o)^{\frac{1}{2}}}{2\sqrt{2R\Gamma}[(\mu_i\rho_i)^{\frac{1}{2}} + (\mu_o\rho_o)^{\frac{1}{2}}]} + \frac{(2l+1)[2(l^2-1)\mu_i^2\rho_i + 2l(l+2)\mu_o^2\rho_o + \mu_o\mu_i(\rho_i(l+2) - \rho_o(l-1))]}{2R^2\Gamma[(\mu_i\rho_i)^{\frac{1}{2}} + (\mu_o\rho_o)^{\frac{1}{2}}]^2} \pm i \left(\beta^* - \frac{(2l+1)^2(\beta^*\mu_i\mu_o\rho_i\rho_o)^{\frac{1}{2}}}{2\sqrt{2R\Gamma}[(\mu_i\rho_i)^{\frac{1}{2}} + (\mu_o\rho_o)^{\frac{1}{2}}]} \right). \quad (34)$$

† Actually this is a slight generalization of their result since they were interested only in the case $\rho_i = \rho_o$.

The first term of the decay factor in (34) is proportional to the square root of the product of frequency and viscosity, the same behaviour that is found in oscillating boundary layers (see Landau & Lifshitz 1959). This term therefore represents at least approximately the rate of decay of oscillations due to viscous dissipation in a boundary layer lying on either side of the interface. On the other hand, the second term of the decay factor in (34) and the decay factors derived by Lamb's method, (28), (31), (33), are all independent of frequency and directly proportional to viscosity. These expressions represent approximately the rate of decay due to viscous dissipation outside the boundary layer in the regions where the velocity distribution is very close to that of the inviscid solution.

In the droplet and cavity cases we have situations where one of the two fluids is sufficiently rarefied as to have negligible effect on flow of the other fluid. Therefore no appreciable boundary-layer flow arises and the inviscid velocity profiles provide a good approximation. This conclusion accords with limiting behaviour of (34), for in both special cases the first or boundary-layer term of the decay factor vanishes and the second or potential-flow term reduces to the corresponding expression obtained by Lamb's method, (28) for the droplet and (31) for the cavity. However, when both fluids are liquids, their interaction at the interface as expressed by the requirements of no slip and continuity of tangential stress, results in a boundary-layer flow which becomes the dominant factor in the rate of viscous dissipation. Very small droplets are the exception; the effect of size is discussed further below. Because the inviscid solution is incapable of accounting for the boundary-layer flow, use of Lamb's method for the two-liquid problem yields a result, (33), which is of limited value: it substantially underestimates the damping rate.

An equation for the rate of decay of oscillations of two viscous fluids of low viscosity separated by a (nearly) plane interface can be obtained from (34) by passing to the limit in the manner described previously. The result agrees with that obtained for the plane interface situation by Harrison† (1908) and is

$$\beta = \frac{2k(\beta_P^* \mu_i \mu_o \rho_i \rho_o)^{\frac{1}{2}}}{2^{\frac{1}{2}}(\rho_o + \rho_i)[(\mu_o \rho_o)^{\frac{1}{2}} + (\mu_i \rho_i)^{\frac{1}{2}}]} + \frac{2k^2(\mu_i^2 \rho_i + \mu_o^2 \rho_o)}{(\rho_o + \rho_i)[(\mu_o \rho_o)^{\frac{1}{2}} + (\mu_i \rho_i)^{\frac{1}{2}}]^2} \pm i \left(\beta_P^* - \frac{2k(\beta_P^* \mu_i \mu_o \rho_i \rho_o)^{\frac{1}{2}}}{2^{\frac{1}{2}}(\rho_o + \rho_i)[(\mu_o \rho_o)^{\frac{1}{2}} + (\mu_i \rho_i)^{\frac{1}{2}}]} \right), \quad (35)$$

where

$$\beta_P^* \equiv \left[\frac{gk(\rho_i - \rho_o) + \gamma k^3}{\rho_o + \rho_i} \right]^{\frac{1}{2}}$$

is the frequency of oscillation of two inviscid fluids with a plane interface. Harrison, we might remark, was aware that the no-slip boundary condition is the key factor causing this equation to differ significantly in form from that for waves on a liquid surface unaffected by the presence of a second phase.

It is instructive to determine the relative magnitudes of the two terms of the frequency and decay factor in (34) for a representative example in which both fluids are liquids. For simplicity we suppose that the densities and viscosities

† During the course of this work we also verified Harrison's result directly by repeating his analysis of the plane interface situation.

of the two liquids are equal. Then the ratio of the two contributions to the decay factor is given by

$$R_D = \frac{\text{second term (rate of viscous dissipation far from interface)}}{\text{first term (rate of dissipation in boundary layer)}} \\ = \frac{\sqrt{2} \left(\frac{\nu^2 \rho}{R\gamma} \right)^{\frac{1}{4}} (4l^2 + 4l - 1)}{(2l + 1)^{\frac{3}{2}} [l(l + 1)(l - 1)(l + 2)]^{\frac{1}{2}}}. \quad (36)$$

The ratio of the two contributions to the frequency is given by

$$R_F = \frac{\text{frequency correction}}{\text{inviscid frequency}} = \frac{1}{4\sqrt{2}} \left(\frac{\nu^2 \rho}{R\gamma} \right)^{\frac{1}{4}} \frac{(2l + 1)^{\frac{3}{2}}}{[l(l + 1)(l - 1)(l + 2)]^{\frac{1}{2}}}. \quad (37)$$

To obtain numerical values of these ratios we further suppose that the values of μ and ρ are those of water, that interfacial tension is 40 dynes/cm, a reasonable value for an oil-water interface with no adsorbed surface-active material, that

Droplet diameter (cm)	Frequency calculated for $l = 2$ (β^* (s ⁻¹))	Frequency (s ⁻¹) (measured)	Reciprocal decay factor (s) for $l = 2$			
			Equation (33)	Equation (34) with calculated frequency	Equation (34) with measured frequency	Measured
0.98	28.3	37.8	2.3	1.0	0.9	0.7
0.78	42.0	63.0	1.5	0.6	0.5	0.2
0.78	36.4	56.8	1.5	0.7	0.6	0.6
1.13	25.7	47.3	3.1	1.2	0.9	0.4
1.13	25.1	44.0	3.1	1.2	0.9	0.4
1.13	25.1	37.8	3.1	1.2	1.0	0.3

TABLE 1. Comparison of decay times calculated by equations (33) and (34) for experimental data of Valentine, Sather & Heideger (1965) on droplets of a CCl_4 - C_6H_6 mixture oscillating in water

droplet radius is 0.1 cm, and that $l = 2$.[†] With these values we find $R_D = 0.16$, which demonstrates that under these conditions the boundary-layer effect is dominant in determining the decay factor. We also calculate $R_F = 0.04$, which indicates that the inviscid frequency is still a good approximation. It is noteworthy that for given values of l and fluid properties, both R_D and R_F increase as droplet radius decreases. Thus it appears that for sufficiently small droplets (34) no longer provides a good approximation to β and numerical solution of (24) would be required.

We have used (34) to calculate values of decay factor from the experimental data of Valentine *et al.* (1965) on droplets of a mixture of carbon tetrachloride and benzene oscillating in water. These droplets were about one centimeter in diameter and so we would expect the boundary-layer effect to be dominant. As shown in table 1 agreement with experimentally measured decay factors is considerably improved over that obtained by use of (33), which does not account

[†] This value of l is of course chosen because lower values of l correspond to motions which are not oscillations (uniform droplet compression or expansion and droplet translation) and because oscillations for higher values of l decay more rapidly.

for the boundary-layer effect. The remaining discrepancy may, as suggested by Valentine *et al.*, be due to the fact that the assumption of small amplitude, on which the analysis rests, was violated by the conditions of the experiment.

(E) *Motion of two fluids having large viscosities*

To determine the behaviour of the dispersion equation (24) in the limit as $\nu_o \rightarrow \infty$ and $\nu_i \rightarrow \infty$ would require considerable effort that can be avoided by using the approach to the slowly decaying aperiodic mode advocated by Lamb (1932) and employed by him in the liquid droplet case. This approach rests on the hypothesis that $\beta/\nu \rightarrow 0$ as $\nu \rightarrow \infty$ and therefore yields no information about the existence of a second, rapidly decaying aperiodic mode. When $\beta/\nu \rightarrow 0$, (3) can be replaced by the biharmonic equation $\nabla^4(rw) = 0$, and (6) goes over to $W(r) = a_1 r^l + a_2 r^{-l-1} + a_3 r^{l+2} + a_4 r^{-l+1}$.† The kinematic, continuity of velocity, and force-balance boundary conditions must still be satisfied but their detailed form is of course altered from (15)–(17), (19) and (21) by the change in form of W . The dispersion equation obtained by essentially the same route as (24) is

$$\beta = \frac{\beta^{*2} R^2 (2l+1) (\mu_o + \mu_i) \Gamma}{2\mu_i^2 (l^2 - 1) (2l^2 + 4l + 3) + 2\mu_o^2 l(l+2) (2l^2 + 1) + 8\mu_o \mu_i l(l^2 - 1) (l+2) + 3\mu_o \mu_i (2l+1)^2}. \quad (38)$$

It is easily seen that this equation reduces to (29) and (32) in the cases of a liquid droplet and of a cavity, respectively. For two highly viscous fluids separated by a plane interface (38) reduces to

$$\beta = \frac{\beta_F^{*2} (\rho_o + \rho_i)}{2k^2 (\mu_o + \mu_i)}, \quad (39)$$

which agrees with the result given by Wehausen & Laitone (1960).

6. Oscillations when the interface is inextensible

Equation (23) is applicable to an oscillating droplet with an interface having arbitrary viscous and elastic properties. We wish to simplify this equation to obtain an approximate expression for β when the interface consists of a membrane or highly condensed film of surface-active material. It seems reasonable to suppose, following Lamb (1932) and Benjamin (1962), that such an interface is essentially ‘inextensible’, i.e. to first order no local expansion or contraction can occur. This restriction can be imposed simply by letting $D \{ \equiv (\kappa/R) - (\Lambda/\beta R) \}$, which is a combined coefficient of surface dilatational elasticity and viscosity, become very large in magnitude. This amounts to supposing that a very small expansion or contraction produces a very large restoring force preventing further motion. Then if the large determinant of (23) is expanded and all terms except

† This functional dependence satisfies the biharmonic equation. It is entirely consistent with the facts that as $\omega \rightarrow 0$,

$$a_3 (\pi/2\omega r)^{\frac{1}{2}} J_{l+\frac{1}{2}}(\omega r) \rightarrow a'_3 \omega^l [r^l - \omega^2 r^{l+2}/2(2l+3)] \text{ in (10)}$$

and $a_4 (\pi/2\omega r)^{\frac{1}{2}} H_{l+\frac{1}{2}}^{(1)}(\omega r) \rightarrow a'_4 \omega^{-l-1} [r^{-l-1} + \omega^2 r^{-l+1}/2(2l-1)] \text{ in (11).}$

those involving D are neglected, an approximate dispersion equation for an oscillating droplet with an inextensible interface is obtained. If both bulk fluids have low viscosities, a procedure similar to that used for the free-interface problem yields

$$\beta = \frac{\beta^{*\frac{1}{2}}[(\mu_o\rho_o)^{\frac{1}{2}}(l+2)^2 + (\mu_i\rho_i)^{\frac{1}{2}}(l-1)^2]}{2\sqrt{2}R\Gamma} \pm i \left[\beta^* - \frac{\beta^{*\frac{1}{2}}[(\mu_o\rho_o)^{\frac{1}{2}}(l+2)^2 + (\mu_i\rho_i)^{\frac{1}{2}}(l-1)^2]}{2\sqrt{2}R\Gamma} \right]. \quad (40)$$

This equation shows that, as was found for the two-fluid free-interface situation (in (34)), the primary cause of damping of oscillations is viscous dissipation in a boundary layer near the interface. In view of the fact that the dispersion equation used to obtain (40) is itself approximate, we have not continued the expansion for the low viscosity case to include terms in the decay factor proportional to higher powers of the fluid viscosities.

When the fluids have equal densities and viscosities, comparison of (40) with the corresponding free-interface result given by (34) reveals that

$$\frac{\text{inextensible interface decay factor}}{\text{free interface decay factor}} = \frac{2(2l^2 + 2l + 5)}{(2l + 1)^2}. \quad (41)$$

This ratio obviously decreases toward unity as l increases. Its maximum value, at $l = 2$, is only 1.36. We see therefore that when both fluids are liquids of low viscosity, the rate of damping with an inextensible interface is not substantially greater than that when the interface is free.

If the exterior fluid is a gas of low density and viscosity, (40) reduces to the formula for a liquid droplet of low viscosity covered by an inextensible film

$$\beta = \frac{(\beta_D^* \nu_i)^{\frac{1}{2}}(l-1)^2}{2\sqrt{2}R(l+1)} \pm i \left(\beta_D^* - \frac{(\beta_D^* \nu_i)^{\frac{1}{2}}(l-1)^2}{2\sqrt{2}R(l+1)} \right). \quad (42)$$

Similarly, if the interior fluid is a gas of low density and viscosity (40) yields a formula for a cavity bounded by an inextensible film

$$\beta = \frac{(\beta_C^* \nu_o)^{\frac{1}{2}}(l+2)^2}{2\sqrt{2}Rl} \pm i \left(\beta_C^* - \frac{(\beta_C^* \nu_o)^{\frac{1}{2}}(l+2)^2}{2\sqrt{2}Rl} \right). \quad (43)$$

These equations indicate that a boundary-layer flow exists near the interface when the interface is inextensible, in contrast to the corresponding free interface results ((28) and (31)) where no appreciable boundary-layer flow exists in the low viscosity limit. Because of the boundary-layer flow the rate of damping is substantially greater for the inextensible interface situation.

Equation (43) is in agreement with the approximate expression derived by Benjamin (1962), who estimated the rate of dissipation on the basis of boundary-layer theory. It is of interest to note that in Benjamin's derivation interfacial shear elasticity and viscosity do not appear at all, while in the foregoing derivation exactly the same result is obtained by requiring only that the magnitude of $S\{ = (\epsilon/R) - (M/\beta R) \}$, the combined coefficient of shear elasticity and viscosity, be small in comparison with the magnitude of D , the combined coefficient of

dilatational elasticity and viscosity. This requirement ensures that dissipation of energy due to shearing deformation of the interface itself is small in comparison with dissipation due to the boundary-layer flow in the underlying bulk liquid.

Using the same procedure as in the preceding section, we may reduce the above equations to equations for the corresponding plane interface situations. For two fluids of low viscosity separated by a plane inextensible interface, (40) reduces to

$$\beta = \frac{k(\beta_P^*)^{\frac{1}{2}}[(\mu_o\rho_o)^{\frac{1}{2}} + (\mu_i\rho_i)^{\frac{1}{2}}]}{2\sqrt{2}(\rho_o + \rho_i)} \pm i \left(\beta_P^* - \frac{k(\beta_P^*)^{\frac{1}{2}}[(\mu_o\rho_o)^{\frac{1}{2}} + (\mu_i\rho_i)^{\frac{1}{2}}]}{2\sqrt{2}(\rho_o + \rho_i)} \right). \quad (44)$$

We have confirmed this result, which does not appear to have been published previously, by direct analysis using the differential equations and boundary-conditions applicable to a plane interface. From (42) and (43) we obtain an equation valid when one of the fluids is a gas of low density and viscosity

$$\beta = \frac{k(\beta_P^* \nu)^{\frac{1}{2}}}{2\sqrt{2}} \pm i \left(\beta_P^* - \frac{k(\beta_P^* \nu)^{\frac{1}{2}}}{2\sqrt{2}} \right). \quad (45)$$

This equation agrees with the result given by Lamb (1932).

There is one significant difference between the droplet and plane interface situations. When the interface is deformed about a plane, the interfacial visco-elastic coefficients S and D always appear in the combination $S + D$. Therefore (44) and (45) are applicable if either or both of S and D become very large. However, in the case of a droplet a dispersion relation differing from that which led to (40) would be found if S were supposed to be very large and D finite. If both S and D become very large, that is, if the interface has a very large resistance to both expansion and shear deformation, there can be no motion at all; this can be seen directly in the force-balance equations. Indeed, the radial-component equation (19) has only one term involving S or D , which expressed in terms of interfacial velocity is

$$\frac{D}{R^2} \left[\nabla_{II} \cdot \mathbf{v}_{II} + \frac{2}{R} v_r \right].$$

If D increases without bound, the expression in brackets, which represents the rate of local expansion of the interface (cf. Weatherburn 1927, 1930), must vanish. The surface divergence equation (21) has two terms in S and D , namely

$$D \nabla_{II}^2 \left[\nabla_{II} \cdot \mathbf{v}_{II} + \frac{2}{R} v_r \right] + S \left[\left(\nabla_{II}^2 + \frac{2}{R^2} \right) (\nabla_{II} \cdot \mathbf{v}_{II}) \right].$$

In view of the radial equation both expressions in brackets must vanish in the limit as S and D simultaneously approach infinite magnitude. It is readily shown that both vanish only when there is no motion whatsoever in or of the interface.

This difference between the plane interface and droplet results may be explained as follows. Because a plane has zero curvature, the rate of local expansion is given simply by $\nabla_{II} \cdot \mathbf{v}_{II}$. Hence elements of the interface can be given small displacements normal to the interface without producing any local expansion, to first order. Moreover, such displacements involve no tangential motion and so shear deformation is absent. We see therefore that very small amplitude oscilla-

tions can occur even when S and D are both very large. In contrast, the local rate of expansion for a nearly spherical interface is $(\nabla_{II} \cdot \mathbf{v}_{II} + (2v_r/R))$; from this expression we see that any radial motion of the droplet interface must produce expansion unless there is a compensating tangential motion. But the tangential motion would involve shear deformation, and therefore no motion can occur when S and D are both large.

7. Summary and conclusions

A general dispersion equation has been derived which is applicable to oscillations of a droplet of one viscous fluid immersed in another viscous fluid, even when the interface between the two possesses viscous and elastic properties of its own. Equations have been obtained for frequency and rate of damping of oscillations for a number of special cases when the interface is free and when it is inextensible.

The analysis shows that if both fluids are liquids of low viscosity, as they are in many instances of practical importance, the primary contribution to the rate of damping of oscillations is, except for very small droplets, from viscous dissipation in a boundary layer near the interface. Owing to this boundary-layer flow the velocity distribution in the region near the interface differs significantly from the velocity distribution obtained by assuming both fluids to be inviscid. The latter, in common with most solutions for inviscid fluids, actually involves slip at the interface. Therefore even if both liquid viscosities are very low, the velocity distribution found for the inviscid case is not a good basis for deriving approximate expressions for the rate of damping. On the other hand, if either the interior or exterior fluid is a gas of low density and viscosity, there is no appreciable boundary-layer flow, provided the interface is free, and a good approximation to the damping rate may be obtained by using the velocity distribution of the inviscid case. But when the interface is inextensible, a boundary-layer flow occurs even in these single-fluid situations.

The authors are indebted to Mr H. K. Rhee for his interest and assistance in some of this study. The work was supported in part by a National Science Foundation Graduate Fellowship awarded to C.A.M. and in part by a National Science Foundation Research Grant to L.E.S.

REFERENCES

- BENJAMIN, T. B. 1962 Non-spherical motions of cavities. In Davies, R. 1964 (ed.), *Cavitation in Real Liquids*, Proceedings of 1962 Symposium. Amsterdam: Elsevier, pp. 164–180.
- BUPARA, S. S. 1964 Spontaneous Movements of Small Round Bodies in Viscous Fluids, Ph.D. Thesis, Department of Chemical Engineering, University of Minnesota.
- CHANDRASEKHAR, S. 1961 *Hydrodynamic and Hydromagnetic Stability*, p. 466–477. Oxford: Clarendon Press.
- ELIASSEN, J. D. 1963 Interfacial Mechanics. Ph.D. Thesis, Department of Chemical Engineering, University of Minnesota.
- GOODRICH, F. C. 1961 Mathematical theory of capillarity. *Proc. Roy. Soc. A* **260**, 481–509.

- HANSEN, R. S. & MANN, J. A. 1964 Propagation characteristics of capillary ripples. I. The theory of velocity dispersion and amplitude attenuation of plane capillary waves on viscoelastic films. *J. Appl. Phys.* **35**, 152–158.
- HARRISON, W. J. 1908 The influence of viscosity on the oscillations of superposed fluids. *Proc. Lond. Math. Soc.* Second Series, **6**, 396–405.
- LAMB, H. 1932 *Hydrodynamics*, sixth edition. Cambridge University Press. Reprinted by Dover, New York, 1945.
- LANDAU, L. D. & LIFSHITZ, E. J. 1959 *Fluid Mechanics*. London: Pergamon Press.
- LEVICH, V. G. 1962 *Physicochemical Hydrodynamics*. Englewood Cliffs, N.J. Prentice-Hall.
- OLDROYD, J. G. 1955 The effect of interfacial stabilizing films on the elastic and viscous properties of emulsions. *Proc. Roy. Soc. A* **232**, 567–577.
- REID, W. H. 1960 The oscillations of a viscous liquid drop. *Quart. Appl. Math.* **18**, 86–89.
- SANI, R. L. 1963 Convective Instability. Ph.D. Thesis, Department of Chemical Engineering, University of Minnesota.
- SCRIVEN, L. E. 1960 Dynamics of a fluid interface. Equations of motion for Newtonian surface fluids. *Chem. Engng. Sci.* **12**, 98–108.
- SCRIVEN, L. E. & STERNLING, C. V. 1964 On cellular convection driven by surface tension gradients: Effects of mean surface tension and surface viscosity. *J. Fluid Mech.* **19**, 321–340.
- VALENTINE, R. S., SATHER, N. F. & HEIDEGGER, W. J. 1965 The motion of drops in viscous media. *Chem. Engng. Sci.* **20**, 719–728.
- WEATHERBURN, C. E. 1927 *Differential Geometry of Three Dimensions*, vol. 1. Cambridge University Press.
- WEATHERBURN, C. E. 1930 *Differential Geometry of Three Dimensions*, vol. 2. Cambridge University Press.
- WEHAUSEN, J. V. & LAITONE, E. V. 1960 Surface Waves, in *Handbuch der Physik*, pp. 446–778. Vol. IX. Berlin: Springer Verlag.
- WILLSON, A. J. 1965 On the stability of two superposed fluids. *Proc. Camb. Phil. Soc.* **61**, 595–607.