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Stokes flow past bubbles and drops partially coated with thin films. Part 2. Thin films with internal circulation – a perturbation solution

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In the present study we examine the steady axisymmetric creeping flow due to the motion of a liquid drop or a bubble which is partially covered by a thin immiscible fluid layer or film. The analysis is based on the assumption that surface-tension forces are large compared with viscous forces which deform the drop, and that the circulation in the film is weak. The latter assumption is satisfied provided that the film-fluid viscosity is not too small. A perturbation scheme based on the thinness of the fluid layer is used to construct the solution.

One of the principal results is an expression for the drag force on the complex drop. We also find that the extent to which the drop or bubble is covered by the film has a maximum value depending on the magnitude of the driving force on the film. In addition, we find the rather interesting result that when the ratio of the primary drop viscosity and bulk fluid viscosity is greater than $\frac{1}{2}$, the circulation within the film may have a double-cell structure.

1. Introduction

Heat- and mass-transfer processes involving drops and bubbles have been and continue to be an important area of research often requiring an understanding of the associated hydrodynamics. The recent development of direct-contact heat and mass exchangers has resulted in the need to study the fluid mechanics of two-fluid drops. The laboratory observations of Mori (1978) show the wide range of two-fluid drop configurations which are encountered in these processes. Here we study the motion of one of these two-fluid drop configurations. Namely, we examine the uniform translation of a two-fluid drop comprised of a primary drop, which is partially surrounded by a thin layer of a second immiscible fluid, i.e. a fluid film (figure 1). Attention will be restricted to the case when the film covers the rear of the primary drop and the motion is axisymmetric. This seems to be the most frequently observed situation. The analysis is also applicable to cases in which the primary-fluid region is a gas bubble. This corresponds to the situation in which the primary-fluid viscosity is very small. In this paper we use the term 'drop' to refer to both bubbles and drops.

The following simplifying assumptions have been made in the analysis. First, the fluids are assumed to be incompressible and the inertial effects are neglected in each

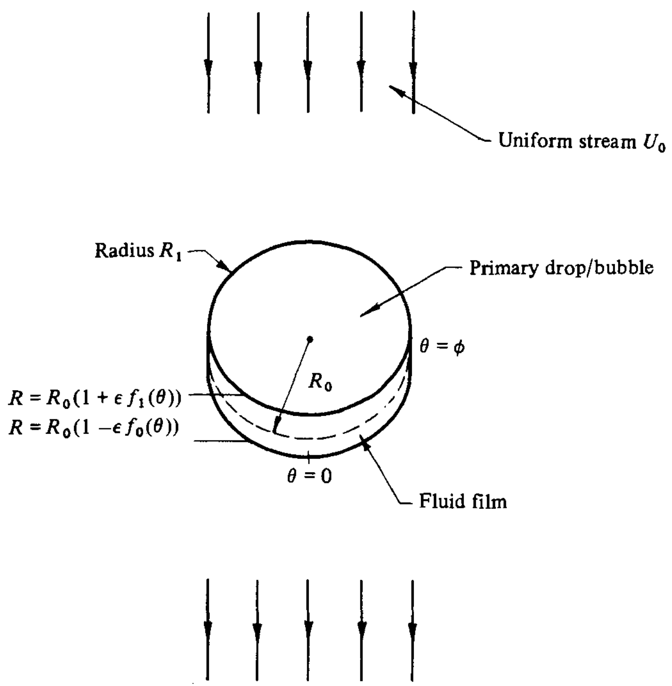

FIGURE 1. Fluid-film geometry.

of the three flow regions; the outer bulk fluid, the interior primary-drop fluid, and the fluid film. Consequently, the Stokes equations describe the fluid motion in each region.

Surface-tension forces of the three fluid–fluid interfaces are assumed to be large compared with the viscous forces which deform the drop, and the surface tensions are constant, i.e. the effects of surface-active agents are ignored. Therefore, to leading order the drop consists of a spherical interface of radius R_1 at the front of the drop separating the bulk fluid and primary-drop fluid ($\phi \leqslant \theta \leqslant \pi$), and two nearly spherical surfaces at the rear bounding the fluid film ($0 \leqslant \theta \leqslant \phi$); $R = R_0(1+\epsilon f_1(\theta))$ and $R = R_0(1-\epsilon f_0(\theta))$. Here ϵ is the thinness parameter given by the ratio of the characteristic film thickness t_0 and the primary-drop radius R_0, i.e. $\epsilon = t_0/R_0$. In mathematical terms, the assumption that the surface-tension forces are large amounts to assuming that for each interface there is a capillary number of the form $\mu U_0/\sigma$ which is small, where U_0 is the free-stream velocity, and μ and σ are the viscosity and surface tension appropriate for each interface. The specific conditions will be made precise in §2. Note, however, that for arbitrary values of the three surface tensions the two radii R_0 and R_1 would not be equal, and therefore the leading-order shape of the drop consists of two spherical caps of different radii. In particular, for the surface-tension-dominated problem considered here there is a pressure jump equal to $2\sigma/R$ for each of the fluid–fluid interfaces, where σ is the surface tension and R the surface radius. Consequently, with the outer bulk-fluid pressure taken to be zero, the pressure within the primary drop evaluated across the two fluid-film interfaces is equal to $2(\hat{\sigma}_0+\sigma_0)/R_0$, where $\hat{\sigma}_0$ and σ_0 are the surface tensions of the inner ($R = R_0(1-\epsilon f_0)$) and outer ($R = R_0(1+\epsilon f_1)$) fluid-film interfaces respectively. Similarly, evaluating the primary-drop pressure across the front interface between bulk and primary-drop fluids having surface tension σ_1 and radius R_1 gives $2\sigma_1/R_1$. Since the calculated primary-drop pressure must be the same in each case, we must have $R_1/R_0 = \sigma_1/(\hat{\sigma}_0+\sigma_0)$. Therefore, if $\sigma_1 \approx \hat{\sigma}_0+\sigma_0$ then $R_1 \approx R_0$, and the complex drop is nearly spherical. In fact, the experimental literature indicates that the special case $R_0 \approx R_1$ is often observed (Mori 1978), at least for liquid–gas drops for which there is literature available. We will limit our study to this special situation, in which case the analysis is considerably simplified; however, we still expect the qualitative behaviour of the solution to be useful when this is not the case.

Note that in the present problem the condition for an approximately spherical interface is that the capillary numbers must be small. This is in contrast with the classical problem of a drop without a film, where the distortion from sphericity is proportional to the Weber number (Taylor & Acrivos 1964). Owing to the unique symmetry in the classical drop problem, the jump in the viscous normal stress at the drop interface exactly cancels with the hydrostatic pressure. Consequently, deviations from the spherical shape are due to inertial effects, which are measured by the Weber number. In the present problem the geometry is considerably complicated by the presence of the film, and the viscous stresses do not cancel with the hydrostatic pressure. Consequently, we find that distortions from sphericity are capillary-number dependent.

We make a further assumption that the mechanism driving circulation within the film is sufficiently weak, so that the film-fluid velocity is small compared with the free-stream velocity. The fluid velocity within the film is readily estimated from a consideration of the continuity of shear stress at the fluid-film interfaces. The shear stress at the outer film interface will be of order $\mu U_0/R_0$ and that at the inner interface will be of order $\hat{\mu} U_0/R_0$, where μ and $\hat{\mu}$ are the viscosities of the bulk fluid and primary-drop fluid respectively. The shear stress within the film will be of order $\mu_f u_0/t_0$, where t_0 is the characteristic film thickness, u_0 is the characteristic film-fluid velocity and μ_f is the film-fluid viscosity. Consequently, stress continuity gives the magnitude of the film-fluid velocity as $u_0/U_0 = O(\epsilon\mu/\mu_f, \epsilon\hat{\mu}/\mu_f)$. Therefore, since $\epsilon \ll 1$, u_0 will be small provided that the viscosities of the bulk fluid and primary-drop fluid are not too large, i.e. the driving force on the film is sufficiently weak. Note that the film-fluid velocity is generally small because the fluid layer is thin and the restriction on the viscosities is not very severe.

The solution is constructed by expanding the velocity and pressure fields in terms of the thinness parameter ϵ. The analysis closely follows that considered by Johnson (1981) for the problem of a thin film on a solid sphere. Since the film-fluid velocity has been assumed to be small, the leading-order problem corresponds to the Stokes flow past a drop having a stagnant can over that portion of the drop covered by the film. This leading-order problem has been recently solved in connection with drops having a stagnant cap of surfactant. The reader is referred to Sadhal & Johnson (1983) for a detailed discussion of this problem and related work. At second order, motion in the film is driven by the leading-order shear stresses exerted at the fluid-film interfaces by the motion of the primary-drop fluid and bulk fluid. Finally, the circulation in the film and the film's shape result in a second-order correction to the primary-drop and the bulk-fluid flow fields. We note that the approximations made for the film which are based on the thinness of the fluid layer are analogous to those of classical lubrication theory.

2. Formulation

Since inertial effects are neglected, the outer bulk-fluid velocity U and pressure P, and the primary-drop fluid velocity $\hat{U}$ and pressure $\hat{P}$ satisfy the Stokes equations

$$\left.\begin{aligned} \mu\nabla^2 \boldsymbol{U} = \nabla P, \quad \nabla\cdot\boldsymbol{U} = 0, \\ \hat{\mu}\nabla^2\hat{\boldsymbol{U}} = \nabla\hat{P}, \quad \nabla\cdot\hat{\boldsymbol{U}} = 0. \end{aligned}\right\} \tag{1}$$

Inherent in the approximation of negligible inertial effects is the assumption that the Reynolds numbers $Re = \rho U_0 R_0/\mu$ and $Re = \hat{\rho} U_0 R_0/\hat{\mu}$ are small (ρ and $\hat{\rho}$ are the densities of the bulk fluid and primary-drop fluid respectively). Similarly in the fluid film the velocity $\boldsymbol{u}$ and pressure p are assumed to satisfy the Stokes equations

$$\mu_{\mathrm{f}}\nabla^2\boldsymbol{u} = \nabla p, \quad \nabla\cdot\boldsymbol{u} = 0. \tag{2}$$

The parameter, which must be small for the Stokes-flow approximation to be justified, has been shown by Johnson (1981) to be $\epsilon Re_{\mathrm{f}} = \epsilon\rho_{\mathrm{f}} u_0 t_0/\mu_{\mathrm{f}}$, where ρ_{f} is the density of the film fluid.

In the following, dimensionless quantities will be used. Velocity and spatial coordinates will be non-dimensionalized by U_0 and R_0 respectively. Stress and pressure will be non-dimensionalized in the bulk fluid, primary-drop fluid and film fluid by $\mu U_0/R_0$, $\hat{\mu} U_0/R_0$ and $\mu_{\mathrm{f}} U_0/R_0$ respectively.

The problem is formulated using spherical polar coordinates (R, θ, Φ), where θ is the angle between a field point and the symmetry axis, which is taken to be the x-axis with unit vector $\boldsymbol{e}_x$. The components of the velocity field in spherical coordinates for the axisymmetric flow considered here are $\boldsymbol{U} = (U, V, 0)$, $\hat{\boldsymbol{U}} = (\hat{U}, \hat{V}, 0)$ and $\boldsymbol{u} = (u, v, 0)$.

The boundary conditions for the outer bulk fluid far from the drop are

$$\boldsymbol{U} \to \boldsymbol{e}_x, \quad P \to 0 \quad \text{as} \quad R \to \infty. \tag{3}$$

At each of the fluid–fluid interfaces we require: (i) continuity of the tangential component of velocity, (ii) no fluid flux across the interface, (iii) continuity of the shear stress, and (iv) the jump in the normal stress equals the product of the surface tension and the sum of the principal curvatures of the surface.

Therefore the boundary conditions at outer film interface $R = 1 + \epsilon f_1(\theta)$ are

$$\left.\begin{aligned} & v - V + \epsilon\frac{\partial f_1}{\partial\theta}(u - U) + O(\epsilon^2) \approx 0, \\ & u - \epsilon\frac{\partial f_1}{\partial\theta} v + O(\epsilon^2) \approx 0, \\ & U - \epsilon\frac{\partial f_1}{\partial\theta} V + O(\epsilon^2) \approx 0, \\ & \tau_{R\theta} - \frac{\mu}{\mu_{\mathrm{f}}} T_{R\theta} + O(\epsilon) \approx 0, \\ & \beta_1\left[\tau_{RR} - \frac{\mu}{\mu_{\mathrm{f}}} T_{RR} + O(\epsilon)\right] \approx -\kappa_1, \end{aligned}\right\} \tag{4}$$

where the components of stress in spherical coordinates have been introduced (τ_{RR} and $\tau_{R\theta}$ for the film fluid; T_{RR} and $T_{R\theta}$ for the bulk fluid; and $\hat{T}_{RR}$ and $\hat{T}_{R\theta}$ for the primary-drop fluid). As discussed in §1, the capillary number $\beta_1 = \mu_{\mathrm{f}} U_0/\sigma_0$ is assumed to be small. The curvature κ_1 is given in terms of the fluid-film profile function $f_1(\theta)$ by

$$\kappa_1 \approx 2-\epsilon\left(\frac{\partial^2 f_1}{\partial\theta^2}+\cot\theta\frac{\partial f_1}{\partial\theta}+2f_1\right)+O(\epsilon^2). \tag{5}$$

Similarly, for the inner film interface $R = 1-\epsilon f_0(\theta)$ we have

$$\left.\begin{aligned}
&v-\hat{V}-\epsilon\frac{\partial f_0}{\partial\theta}(u-\hat{U})+O(\epsilon^2)\approx 0,\\
&u+\epsilon\frac{\partial f_0}{\partial\theta}v+O(\epsilon^2)\approx 0,\\
&\hat{U}+\epsilon\frac{\partial f_0}{\partial\theta}\hat{V}+O(\epsilon^2)\approx 0,\\
&\tau_{R\theta}-\frac{\hat{\mu}}{\mu_f}\hat{T}_{R\theta}+O(\epsilon)\approx 0,\\
&\beta_0\left[\tau_{RR}-\frac{\hat{\mu}}{\mu_f}\hat{T}_{RR}+O(\epsilon)\right]\approx\kappa_0,
\end{aligned}\right\} \tag{6}$$

where the capillary number $\beta_0 = \mu_f U_0/\hat{\sigma}_0$ is also assumed to be small, and the curvature κ_0 is given by

$$\kappa_0 \approx 2+\epsilon\left(\frac{\partial^2 f_0}{\partial\theta^2}+\cot\theta\frac{\partial f_0}{\partial\theta}+2f_0\right)+O(\epsilon^2). \tag{7}$$

Lastly, the boundary conditions at the front interface are applied at $R = 1$ after assuming $R_1 \approx R_0$ $(\sigma_1 \approx \sigma_0+\hat{\sigma}_0)$ as discussed earlier, and are given by

$$\left.\begin{aligned}
&V=\hat{V},\quad U=\hat{U}=0,\\
&T_{R\theta}=\frac{\hat{\mu}}{\mu}\hat{T}_{R\theta},\quad \beta\left[\frac{\mu}{\hat{\mu}}T_{RR}-\hat{T}_{RR}\right]\approx 2,
\end{aligned}\right\} \tag{8}$$

where the capillary number $\beta = \hat{\mu}U_0/\sigma_1$ and $(\mu/\hat{\mu})\beta$ are assumed to be small. The last boundary condition assumes that the interface is spherical and will only be satisfied approximately. An assumed spherical shape will satisfy this condition with an error $O(\beta, (\mu/\hat{\mu})\beta)$. We will see that this deviation from the spherical shape is small compared with the deviations of the fluid-film interfaces.

As discussed in §1, we anticipate that for a thin fluid film a leading-order flow in the outer bulk and primary-drop fluids drives motion within the film, which in turn leads to modifications of the flow in the outer fluid and primary drop. Hence we assume

$$\left.\begin{aligned}
&\boldsymbol{U}=\boldsymbol{U}^{(0)}+\epsilon\boldsymbol{U}^{(1)}+\dots,\quad P=P^{(0)}+\epsilon P^{(1)}+\dots,\\
&\hat{\boldsymbol{U}}=\hat{\boldsymbol{U}}^{(0)}+\epsilon\hat{\boldsymbol{U}}^{(1)}+\dots,\quad \hat{P}=\frac{2}{\beta}+\hat{P}^{(0)}+\epsilon\hat{P}^{(1)}+\dots.
\end{aligned}\right\} \tag{9}$$

The large constant pressure term $2/\beta$ in $\hat{P}$ is necessary so that the drop is nearly spherical to leading order (see (8)). At each order in ϵ the velocity and pressure fields satisfy the Stokes equations.

For the film fluid we assume that the velocity field is given by

$$\left.\begin{aligned}
&v=\epsilon v^{(1)}+\epsilon^2 v^{(2)}+\dots,\\
&u=\epsilon^2 u^{(1)}+\epsilon^3 u^{(2)}+\dots.
\end{aligned}\right\} \tag{10}$$

Note that the radial component of velocity u is nearly perpendicular to the fluid-film interfaces and is assumed to be small compared with the tangential component v. This is because the surface tension forces are large compared with the deforming effect of the stresses on the fluid–fluid interfaces, and therefore the slope of the interface will be small. A conequence of this slope being small is the fact that the velocity perpendicular to the interface will be small in comparison with the tangential component. Note, however, that there will generally be a small region of non-uniformity near the three-fluid contact line $\theta = \phi$. This is because the interface slopes or contact angles at $\theta = \phi$ are material properties which would be specified for given fluids and may not be small (see Dussan V. 1979). The characteristics of this region of non-uniformity are very similar to those discussed by Johnson (1981) for the analogous problem of a thin film on a solid sphere, and the reader is referred to that paper for a complete discussion. The point to make here is that the details of the fluid motion in this small region near the contact line does not affect the leading-order solution being considered here.

For the fluid-film pressure we take

$$p = \frac{2}{\beta_1} + \frac{1}{\epsilon} p^{(1)} + p^{(2)} + \dots. \tag{11}$$

As before, the constant pressure term $2/\beta_1$ is required so that the interface is nearly spherical (see (4) and (5)). The next term, i.e. $p^{(1)}/\epsilon$, is the pressure term required to balance the viscous-stress term in the equation of motion. This pressure term is large and of order ϵ^{-1} owing to the fact that the film is thin and the radial derivatives are large compared with the tangential ones. This is essentially one of the standard results of classical lubrication theory.

For the thin film we introduce the film variable $\xi = (R-1)/\epsilon$, and the leading-order governing equations for the fluid motion within the film become

$$\frac{\partial^2 v}{\partial \xi^2} = \frac{\partial p^{(1)}}{\partial \theta}, \quad \frac{\partial p^{(1)}}{\partial \xi} = 0, \tag{12}$$

$$\frac{\partial u^{(1)}}{\partial \xi} + \frac{1}{\sin\theta} \frac{\partial}{\partial \theta} (\sin\theta v^{(1)}) = 0. \tag{13}$$

Here we see the balance between the pressure and the viscous stress.

The boundary conditions at the two fluid-film interfaces are further simplified by expanding the variables associated with the bulk and primary-drop fluids (U, T_{RR}, $T_{R\theta}$, $\hat{U}$, $\hat{T}_{RR}$ and $\hat{T}_{R\theta}$) in a Taylor series about $R = 1$. By doing this for the conditions at $R = 1 + \epsilon f_1(\theta)$ ($\xi = f_1$) we obtain

$$v - V + \epsilon\left[\frac{\partial f_1}{\partial \theta}(u-U) - f_1 \frac{\partial V}{\partial R}\right] \approx 0, \quad u - \epsilon \frac{\partial f_1}{\partial \theta} v \approx 0,$$

$$U + \epsilon\left[f_1 \frac{\partial U}{\partial R} - \frac{\partial f_1}{\partial \theta} V\right] \approx 0, \quad \tau_{R\theta} - \frac{\mu}{\mu_f} T_{R\theta} \approx 0, \quad \beta_1\left[\tau_{RR} - \frac{\mu}{\mu_f} T_{RR}\right] \approx -\kappa_1.$$

Here it is understood that the bulk-fluid variables are evaluated at $R = 1$ and the film-fluid variables are evaluated at $\xi = f_1(\theta)$.

By substituting the perturbation expansions for the velocity and pressure fields we finally arrive at the conditions at $R = 1 + \epsilon f_1(\theta)$ as

$$V^{(0)} - \epsilon\left[v^{(1)} - V^{(1)} - f_1 \frac{\partial V^{(0)}}{\partial R} - \frac{\partial f_1}{\partial \theta} U^{(0)}\right] \approx 0, \tag{14}$$

$$u^{(1)} - \frac{\partial f_1}{\partial \theta} v^{(1)} \approx 0, \tag{15}$$

$$U^{(0)} + \epsilon\left[U^{(1)} + f_1 \frac{\partial U^{(0)}}{\partial R} - \frac{\partial f_1}{\partial \theta} V^{(0)}\right] \approx 0, \tag{16}$$

$$\frac{\partial v^{(1)}}{\partial \xi} \approx \frac{\mu}{\mu_f} T^{(0)}_{R\theta} = \frac{\mu}{\mu_f}\left(\frac{\partial V^{(0)}}{\partial R} - \frac{V^{(0)}}{R} + \frac{1}{R}\frac{\partial U^{(0)}}{\partial \theta}\right), \tag{17}$$

$$-\frac{\beta_1}{\epsilon^2}\left[p^{(1)} + O\left(\epsilon, \epsilon\frac{\mu}{\mu_f}, \epsilon\frac{\mu}{\mu_f}\frac{\rho g R_0^2}{\mu U_0}\right)\right] \approx \frac{\partial^2 f_1}{\partial \theta^2} + \cot\theta \frac{\partial f_1}{\partial \theta} + 2f_1. \tag{18}$$

In obtaining these equations we have used the expansions for the stresses

$$\tau_{R\theta} \approx \frac{\partial v^{(1)}}{\partial \xi} + O(\epsilon),$$

$$\tau_{RR} \approx -\frac{2}{\beta_1} - \frac{1}{\epsilon} p^{(1)} + O(1),$$

and we include the magnitude of the hydrostatic pressure $\rho g R_0$ in (18). A few points concerning the hydrostatic pressure will be made shortly.

From the last boundary condition (18), note that we now have a specific restriction on the capillary number β_1, namely $\beta_1 = O(\epsilon^2)$. Note also that the error term $\epsilon\mu/\mu_f$ is small, since this is equal to the magnitude of the film velocity, which has been assumed small.

Owing to the large fluid-film pressure $p^{(1)}/\epsilon$, variations of the fluid-film interfaces from spherical are large compared with the variation of the front interface ($\phi \leqslant \theta \leqslant \pi$). In particular, from (18) we see that the variation in the fluid-film interface $\epsilon f_1(\theta)$ is of order β_1/ϵ. As discussed earlier, the variation of the front interface from spherical is $O(\beta, (\mu/\hat{\mu})\beta)$. Since $\beta = \epsilon(\hat{\mu}/\mu_f)(\beta_1/\epsilon)(1+\hat{\sigma}_0/\sigma_0)^{-1}$, we can see that β is small compared with the film-interface variation β_1/ϵ since $\epsilon\hat{\mu}/\mu_f$ has been assumed small. Similarly $(\mu/\hat{\mu})\beta$ is small since $\epsilon\mu/\mu_f$ has been assumed small. Consequently, it is sufficient to assume that the front interface is spherical, with the associated error being of higher order than that considered here.

Furthermore, since the problem realistically pertains to a drop moving in a gravity field, we should point out that in the normal-stress condition (18) we have neglected the hydrostatic pressure compared with the large pressure $p^{(1)}/\epsilon$ generated in the film. This assumption implies the following weak restriction on the validity of the present solution. Since we are assuming that the hydrostatic pressure is small compared with the pressure in the film, which is very large, we require $\rho g R_0 \ll \epsilon^{-1}\mu_f U_0/R_0$ or $\rho g R_0^2/\mu_f U_0 \ll \epsilon^{-1}$. Now, the magnitude of the drop velocity is readily estimated from the fact that the viscous-drag force balances the buoyancy force and drop weight, i.e. $\mu U_0 R_0 = O[\rho g R_0^3(1-\hat{\rho}/\rho)]$, the contribution of the film to the weight being negligible since the film has a small volume. Therefore, after substituting for the magnitude of the velocity, our condition becomes $O(1-\hat{\rho}/\rho) \gg \epsilon\mu/\mu_f \ll 1$. In physical terms this simply says that the density difference between the drop and bulk must be large enough (simply larger than some small number) so that the resulting drop velocity is not too small. More specifically, the drop velocity must be sufficiently large so that the corresponding viscous stresses produce a film pressure that is large compared to the hydrostatic pressure. Consequently, the present analysis does not consider the case when the drop density very nearly equals the density of the bulk fluid, in which case the drop moves very slowly. This restriction is relatively weak by virtue of the fact that the film pressure generally dominates the other stresses because the film is thin and therefore the film pressure is $O(1/\epsilon)$.

The analogous boundary conditions at the interface $R = 1-\epsilon f_0(\theta)$ are obtained from the above equations (14)–(18) by interchanging U, V, $T_{R\theta}$, β_1, μ and f_1 with $\hat{U}$, $\hat{V}$, $\hat{T}_{R\theta}$, $-\beta_0$, $\hat{\mu}$ and $-f_0$. Note that to obtain the conditions we also use the approximation $\hat{T}_{RR} \approx -\hat{P}+O(1) \approx -2/\beta+O(1)$, where $\beta = \hat{\mu}U_0/(\hat{\sigma}_0+\sigma_0)$, and neglect the hydrostatic pressure. The normal-stress condition in this case also gives a restriction similar to that obtained before, namely $\beta_0 = O(\epsilon^2)$. Furthermore, it is easy to show that the deviation of the inner film interface $\epsilon f_0(\theta)$ from spherical is also generally large compared with that of the front interface ($\phi \leqslant \theta \leqslant \pi$).

Finally, from the boundary conditions and governing equations we obtain the following hierarchy of problems.

At leading-order we have

$$\left.\begin{aligned} \nabla^2 U^{(0)} &= \nabla P^{(0)}, \quad \nabla\cdot U^{(0)} = 0, \\ \nabla^2 \hat{U}^{(0)} &= \nabla \hat{P}^{(0)}, \quad \nabla\cdot \hat{U}^{(0)} = 0, \end{aligned}\right\} \tag{19}$$

with the boundary conditions

$$U^{(0)} = V^{(0)} = \hat{U}^{(0)} = \hat{V}^{(0)} = 0 \quad (R = 1, 0 \leqslant \theta \leqslant \phi),$$

$$\left.\begin{aligned} &U^{(0)} = \hat{U}^{(0)} = 0, \\ &V^{(0)} = \hat{V}^{(0)}, \\ &T^{(0)}_{R\theta} = \frac{\hat{\mu}}{\mu}\hat{T}^{(0)}_{R\theta} \end{aligned}\right\} \quad (R = 1, \phi \leqslant \theta \leqslant \pi). \tag{20}$$

Note that the normal-stress condition has been approximately satisfied by the pressure with an error $O(\beta, (\mu/\hat{\mu})\beta)$. As mentioned in §1, this problem corresponds to a spherical drop with a stagnant cap for $0 \leqslant \theta \leqslant \phi$.

The leading-order fluid-film problem is

$$\frac{\partial^2 u^{(1)}}{\partial \xi^2} = \frac{\partial p^{(1)}}{\partial \theta}, \quad \frac{\partial p^{(1)}}{\partial \xi} = 0, \tag{21}$$

$$\frac{\partial u^{(1)}}{\partial \xi} + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}(\sin\theta v^{(1)}) = 0, \tag{22}$$

with boundary conditions

$$\frac{\partial v^{(1)}}{\partial \xi} = \frac{\mu}{\mu_f} T^{(0)}_{R\theta}(1,\theta), \tag{23}$$

$$u^{(1)} - \frac{\partial f_1}{\partial \theta} v^{(1)} = 0 \qquad (\xi = f_1(\theta)), \tag{24}$$

$$\frac{\partial v^{(1)}}{\partial \xi} = \frac{\hat{\mu}}{\mu_f} \hat{T}^{(0)}_{R\theta}(1,\theta), \tag{25}$$

$$u^{(1)} + \frac{\partial f_0}{\partial \theta} v^{(1)} = 0 \qquad (\xi = -f_0(\theta)). \tag{26}$$

The film profile functions are determined from

$$\frac{\partial^2 f_1}{\partial \theta^2}+\cot\theta\frac{\partial f_1}{\partial \theta}+2f_1 = -\frac{\beta_1}{\epsilon^2}p^{(1)}, \tag{27}$$

$$\frac{\partial^2 f_0}{\partial \theta^2}+\cot\theta\frac{\partial f_0}{\partial \theta}+2f_0 = -\frac{\beta_0}{\epsilon^2}p^{(1)}, \tag{28}$$

with boundary conditions which will be discussed shortly.

Lastly, the first correction to the motions of the bulk fluid and primary-drop fluid are determined by

$$\left.\begin{aligned}\nabla^2 \boldsymbol{U}^{(1)} &= \nabla P^{(1)}, \quad \nabla\cdot \boldsymbol{U}^{(1)} = 0,\\ \nabla^2 \hat{\boldsymbol{U}}^{(1)} &= \nabla \hat{P}^{(1)}, \quad \nabla\cdot \hat{\boldsymbol{U}}^{(1)} = 0,\end{aligned}\right\} \tag{29}$$

with the boundary conditions

$$\left.\begin{aligned}V^{(1)} &= v^{(1)}-f_1\frac{\partial V^{(0)}}{\partial R},\\ U^{(1)} &= 0\end{aligned}\right\} \quad (R = 1, \xi = f_1(\theta), 0 \leqslant \theta \leqslant \phi), \tag{30}$$

$$\left.\begin{aligned}\hat{V}^{(1)} &= v^{(1)}+f_0\frac{\partial \hat{V}^{(0)}}{\partial R},\\ \hat{U}^{(1)} &= 0\end{aligned}\right\} \quad (R = 1, \xi = -f_0(\theta), 0 \leqslant \theta \leqslant \phi), \tag{31}$$

$$\left.\begin{aligned}U^{(1)} &= \hat{U}^{(1)} = 0,\\ V^{(1)} &= \hat{V}^{(1)},\\ T^{(1)}_{R\theta} &= \frac{\hat{\mu}}{\mu}\hat{T}^{(1)}_{R\theta}\end{aligned}\right\} \quad (R = 1, \phi \leqslant \theta \leqslant \pi), \tag{32}$$

where $T^{(1)}_{R\theta}$ and $\hat{T}^{(1)}_{R\theta}$ are obtained from the corresponding leading-order expressions (17) by substituting $\boldsymbol{U}^{(1)}$ and $\hat{\boldsymbol{U}}^{(1)}$ for $\boldsymbol{U}^{(0)}$ and $\hat{\boldsymbol{U}}^{(0)}$ respectively. Note that in obtaining the boundary conditions for $0 \leqslant \theta \leqslant \phi$ we have used the leading-order conditions on $R = 1$, $\boldsymbol{U}^{(0)} = \hat{\boldsymbol{U}}^{(0)} = 0$, and the fact that on $R = 1$, $\partial U^{(0)}/\partial R = \partial \hat{U}^{(0)}/\partial R = 0$. The latter result is readily found from the continuity equation. Also, far from the drop

$$\boldsymbol{U}^{(1)}, P^{(1)} \to 0 \quad \text{as} \quad R \to \infty. \tag{33}$$

It is convenient to define the non-dimensional film thickness $t(\theta) = f_0(\theta)+f_1(\theta)$, and then to combine the governing equations for f_0 and f_1 by adding them to give

$$\frac{\partial^2 t}{\partial \theta^2}+\cot\theta\frac{\partial t}{\partial \theta}+2t = -\frac{\beta_0+\beta_1}{\epsilon^2}p^{(1)}. \tag{34}$$

From the equations for f_0, f_1 and t it is easy to see that f_0 and f_1 are related to t by

$$f_0(\theta) = \frac{\sigma_0}{\sigma_0+\hat{\sigma}_0}t(\theta), \quad f_1(\theta) = \frac{\hat{\sigma}_0}{\sigma_0+\hat{\sigma}_0}t(\theta). \tag{35}$$

Taking the non-dimensional thickness $t(\theta)$ to be equal to unity at the rear of the sphere $\theta = 0$, using symmetry at $\theta = 0$, and requiring the film to vanish at the three-fluid contact line $\theta = \phi$, gives the boundary conditions $t(0) = 1$, $\partial t(0)/\partial\theta = 0$ and $t(\phi) = 0$. We will see that, although (34) is of second order, three boundary conditions are necessary because the pressure $p^{(1)}$ contains an additive constant which will be determined by one of these conditions. Furthermore, the condition $t(\phi) = 0$ is complete when the contact line position is specified. In the appendix the position of the contact line is determined in terms of the surface tensions and contact angles from the condition of global force equilibrium on the film.

The final point to make here is that the volume of fluid in the film is assumed to be known. Therefore, for a given volume the characteristic film thickness t_0 or equivalently $\epsilon = t_0/R_0$ is determined by the relation

$$\text{film volume} \approx 2\pi R_0^3 \epsilon \int_0^{\phi} t(\theta) \sin\theta \, d\theta \equiv 2\pi R_0^3 \epsilon \gamma(\phi). \tag{36}$$

3. Solution

3.1. *The leading-order primary-drop and bulk-fluid motion*

As already discussed, the leading-order motion of the primary-drop and bulk fluid governed by (19) and (20) is analogous to the problem of a drop having a stagnant cap of surfactant. Recently Sadhal & Johnson (1983) obtained an exact solution for this problem using Collin's (1961) method to solve the dual-series equations which arise from the mixed boundary conditions due to the stagnant cap. In that solution the stream function $\Psi^{(0)}$ and $\hat{\Psi}^{(0)}$ are given by

$$\Psi^{(0)} = \left(R^2 - \frac{1}{R}\right) \sin\theta \, T_1^{-1}(\cos\theta) + \sum_{k=1}^{\infty} C_k^*(R^{2-k} - R^{-k}) \sin\theta \, T_k^{-1}(\cos\theta), \tag{37}$$

$$\hat{\Psi}^{(0)} = \tfrac{3}{2} R^2(R^2 - 1) \sin\theta \, T_1^{-1}(\cos\theta) + \sum_{k=1}^{\infty} C_k^*(R^{k+3} - R^{k+1}) \sin\theta \, T_k^{-1}(\cos\theta), \tag{38}$$

where T_k^{-1} denotes the associated Legendre functions

$$\sin\theta \, T_k^{-1}(\cos\theta) = \int_{\cos\theta}^{1} P_k(x) \, dx, \tag{39}$$

and

$$\left.\begin{aligned} C_1^* &= -\frac{\mu}{4\pi(\mu+\hat{\mu})}[2\phi + \sin\phi - \sin 2\phi - \tfrac{1}{3}\sin 3\phi] - \frac{1}{2}\frac{2\mu + 3\hat{\mu}}{\mu + \hat{\mu}}, \\ C_k^* &= \frac{\mu}{4\pi(\mu+\hat{\mu})}\left\{\sin(k+2)\phi - \sin k\phi + \sin(k+1)\phi - \sin(k-1)\phi \right. \\ &\qquad \left. - 2\left[\frac{\sin(k+2)\phi}{k+2} + \frac{\sin(k-1)\phi}{k-1}\right]\right\} \quad (k \geqslant 2), \end{aligned}\right\} \tag{40}$$

where $P_k(x)$ is the Legendre polynomial. The corresponding velocity fields are given by

$$\left.\begin{aligned} U^{(0)} &= \frac{1}{R^2 \sin\theta}\frac{\partial \Psi^{(0)}}{\partial\theta}, \quad V^{(0)} = \frac{-1}{R\sin\theta}\frac{\partial \Psi^{(0)}}{\partial R}, \\ \hat{U}^{(0)} &= \frac{1}{R^2 \sin\theta}\frac{\partial \hat{\Psi}^{(0)}}{\partial\theta}, \quad \hat{V}^{(0)} = \frac{-1}{R\sin\theta}\frac{\partial \hat{\Psi}^{(0)}}{\partial R}. \end{aligned}\right\} \tag{41}$$

Prior to this solution only approximate numerical solutions were available. The pressure fields are easily obtained, but are not presented here since they will not be used in the subsequent analysis.

3.2. *The leading-order fluid-film solution*

From (21), (23) and (25) we see that the pressure is a function only of θ, and we find

$$v^{(1)} = \tfrac{1}{2}G(\theta)\,\xi(\xi-2f_1)+T(\theta)\,\xi+B(\theta), \tag{42}$$

where

$$G(\theta) = \frac{\partial p^{(1)}}{\partial\theta} = \frac{T(\theta)+\hat{T}(\theta)}{t(\theta)}, \tag{43}$$

$$T(\theta) = \frac{\mu}{\mu_f}T^{(0)}_{R\theta}(1,\theta), \tag{44}$$

$$\hat{T}(\theta) = -\frac{\hat{\mu}}{\mu_f}\hat{T}^{(0)}_{R\theta}(1,\theta). \tag{45}$$

$T^{(0)}_{R\theta}, \hat{T}^{(0)}_{R\theta}$ are easily determined from the leading-order solution (37) and (38). We obtain $B(\theta)$ from the physical constraint that for steady flow the net volume flux through a section of the fluid film must be zero, i.e.

$$\int_{-f_0}^{f_1} v^{(1)}(\xi)\,\mathrm{d}\xi = 0, \tag{46}$$

giving

$$B(\theta) = \tfrac{1}{2}T(f_0-f_1)+\tfrac{1}{3}G(f_1^2-f_0f_1-\tfrac{1}{2}f_0^2).$$

The condition (46) is simply an integral form of the continuity equation, which is easily obtained from (22).

From the continuity equation (22) and the boundary conditions (24) and (26) on $u^{(1)}$ we find

$$u^{(1)} = -\tfrac{1}{6}(G'+G\cot\theta)\,[\xi^3+f_0^3-3f_1(\xi^2-f_0^2)] -\tfrac{1}{2}(\xi^2-f_0^2)\,(T'+T\cot\theta-Gf_1')-(B'+B\cot\theta)\,(\xi+f_0)-\tfrac{1}{3}f_0'(\hat{T}-\tfrac{1}{2}T)\,t, \tag{47}$$

where a prime denotes differentiation with respect to ξ. From the velocity field $(\epsilon^2u^{(1)}, \epsilon v^{(1)})$ the stream function ψ for the fluid film is found to be

$$\psi = -\epsilon^2\sin\theta\{\tfrac{1}{6}G[\xi^2(\xi-3f_1)+f_0^2(f_0+3f_1)]+\tfrac{1}{2}T(\xi^2-f_0^2)+B(\xi+f_0)\}. \tag{48}$$

At this point the film solution is determined in terms of the film thickness $t(\theta)$ and the profile functions $f_0(\theta)$ and $f_1(\theta)$. These are determined from (34) and (35), where the pressure $p^{(1)}$ is given by (43) as

$$p^{(1)} = p^{(1)}(0)+\int_0^\theta \frac{T(\theta')+\hat{T}(\theta')}{t(\theta')}\mathrm{d}\theta'. \tag{49}$$

In §4 the film thickness $t(\theta)$ will be computed numerically for a variety of flow conditions, and the additive constant pressure term $p^{(1)}(0)$ is found as part of the solution.

3.3. *The second-order solution in the primary drop and bulk fluid*

The solution to the second-order Stokes-flow problem (29) with boundary conditions (30)–(33) is obtained using the method given by Sadhal & Johnson (1983). The stream functions $\Psi^{(1)}$ and $\hat{\Psi}^{(1)}$ are introduced, and the corresponding velocity field is given by (41), replacing the superscript zero with a one. The Stokes equations become

$$\mathrm{L}^2_{-1}(\Psi^{(1)}) = 0, \quad \mathrm{L}^2_{-1}(\hat{\Psi}^{(1)}) = 0 \tag{50}$$

where

$$\mathrm{L}_{-1} = \frac{\partial^2}{\partial R^2} + \frac{\sin\theta}{R^2}\frac{\partial}{\partial\theta}\left(\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\right).$$

The boundary conditions (30)–(33) are given below.

(I) At the outer fluid–film interface, $R = 1$, $\xi = f_1$, $0 \leqslant \theta \leqslant \phi$:

(a) the tangential velocity is specified

$$\frac{-1}{R\sin\theta}\frac{\partial\Psi^{(1)}}{\partial R} = v^{(1)}_{\cdot} - f_1\frac{\partial V^{(0)}}{\partial R} \equiv q(\theta). \tag{51}$$

(b) The radial velocity vanishes

$$\Psi^{(1)} = 0. \tag{52}$$

(II) At the inner fluid-film interface, $R = 1$, $\xi = -f_0$, $0 \leqslant \theta \leqslant \phi$:

(a) the tangential velocity is specified

$$\frac{-1}{R\sin\theta}\frac{\partial\hat{\Psi}^{(1)}}{\partial R} = v^{(1)} + f_0\frac{\partial \hat{V}^{(0)}}{\partial R} \equiv \hat{q}(\theta). \tag{53}$$

(b) The radial velocity vanishes

$$\hat{\Psi}^{(1)} = 0. \tag{54}$$

(III) At the front interface, $R = 1$, $\phi \leqslant \theta \leqslant \pi$:

(a) the radial velocity vanishes

$$\Psi^{(1)} = \hat{\Psi}^{(1)} = 0. \tag{55}$$

(b) The tangential velocity is continuous

$$\frac{\partial\Psi^{(1)}}{\partial R} = \frac{\partial\hat{\Psi}^{(1)}}{\partial R}. \tag{56}$$

(c) The shear stress is continuous

$$\frac{\partial}{\partial R}\left(\frac{1}{R^2}\frac{\partial\Psi^{(1)}}{\partial R}\right) = \frac{\hat{\mu}}{\mu}\frac{\partial}{\partial R}\left(\frac{1}{R^2}\frac{\partial\hat{\Psi}^{(1)}}{\partial R}\right). \tag{57}$$

(IV) Far from the drop

$$\Psi^{(1)} \rightarrow 0 \quad \text{as} \quad R \rightarrow \infty. \tag{58}$$

The general solution of (50) satisfying condition (52), (54), (55), (58) and the requirement that the motion inside the drop be bounded is

$$\left.\begin{aligned}\Psi^{(1)} &= \sum_{k=1}^{\infty} D_k(R^{2-k} - R^{-k})\sin\theta\, T_k^{-1}(\cos\theta),\\ \hat{\Psi}^{(1)} &= \sum_{k=1}^{\infty} \hat{D}_k(R^{k+3} - R^{k+1})\sin\theta\, T_k^{-1}(\cos\theta),\end{aligned}\right\} \tag{59}$$

where as before $T_k^{-1}(\cos\theta)$ denotes the associated Legendre functions (39). The remaining four boundary conditions (51), (53), (56) and (57) may be recast into the following two sets of Legendre-series equations:

$$\left.\begin{aligned}\sum_{k=1}^{\infty} G_k T_k^{-1}(\cos\theta) &= -\tfrac{1}{2}[\hat{\mu}\hat{q}(\theta)+\mu q(\theta)] \equiv Q(\theta) \quad (0 \leqslant \theta \leqslant \phi),\\ \sum_{k=1}^{\infty} (2k+1)\, G_k T_k^{-1}(\cos\theta) &= 0 \quad (\phi \leqslant \theta \leqslant \pi),\end{aligned}\right\} \tag{60}$$

where $G_k = \hat{\mu}\hat{D}_k + \mu D_k$; and

$$\sum_{k=1}^{\infty} H_k T_k^{-1}(\cos\theta) = \begin{cases} \tfrac{1}{2}[q(\theta)-\hat{q}(\theta)] & (0 \leqslant \theta \leqslant \phi),\\ 0 & (\phi \leqslant \theta \leqslant \pi),\end{cases} \tag{61}$$

where $H_k = \hat{D}_k - D_k$.

Following Sneddon (1966), the exact solution of the first set of dual-series equations (60) is

$$G_k = \left(\frac{2}{\pi}\right)^{\frac{1}{2}} \frac{\Gamma(2+k)}{\Gamma(k+\frac{1}{2})} \int_0^{\phi} Q^*(u) \cos^3 \tfrac{1}{2}u\, P_k^{(\frac{1}{2},\frac{3}{2})}(\cos u)\, \mathrm{d}u, \tag{62}$$

$$Q^*(u) = \frac{\mathrm{d}}{\mathrm{d}u} \int_0^u \frac{\tan\frac{1}{2}\theta \sin\theta\, Q(\theta)}{(\cos\theta - \cos u)^{\frac{1}{2}}}\, \mathrm{d}\theta, \tag{63}$$

where $Q(\theta)$ is defined in (60), Γ denotes the Gamma function, and $P_k^{(\frac{1}{2},\frac{3}{2})}(\cos u)$ is the Jacobi function, which may be expressed as

$$P_k^{(\frac{1}{2},\frac{3}{2})}(\cos u) = -2\frac{\Gamma(k+\frac{1}{2})}{\Gamma(k+2)\,\Gamma(\frac{1}{2})} \frac{1}{\sin u} \frac{\mathrm{d}}{\mathrm{d}u}\left[\frac{\cos(k+\frac{1}{2})\,u}{\cos\frac{1}{2}u}\right]. \tag{64}$$

Consequently, substituting (64) into (62) and integrating by parts gives

$$\begin{aligned} G_k = {} & \frac{\surd 2}{\pi}\left[(k+\tfrac{1}{2})\cos\tfrac{1}{2}\phi \frac{\sin(k+\frac{1}{2})\,\phi}{\sin\frac{1}{2}\phi} - \tfrac{1}{2}\cos(k+\tfrac{1}{2})\,\phi\right] K(\phi) \\ & -\frac{\surd 2}{\pi}\int_0^{\phi} (k+\tfrac{1}{2}) \frac{\cos\frac{1}{2}u}{\sin^2\frac{1}{2}u}[(k+\tfrac{1}{2})\sin\tfrac{1}{2}u \cos(k+\tfrac{1}{2})\,u \\ & -\tfrac{1}{2}\sin(k+\tfrac{1}{2})\,u\cos\tfrac{1}{2}u]\, K(u)\,\mathrm{d}u, \end{aligned} \tag{65a}$$

where

$$K(u) = \int_0^u \frac{\tan\frac{1}{2}\theta \sin\theta\, Q(\theta)}{(\cos\theta - \cos u)^{\frac{1}{2}}}\, \mathrm{d}\theta. \tag{65b}$$

The second set of equations (61) are readily solved using the orthogonality principle for the associated Legendre functions T_k^{-1}. Taking $\eta = \cos\theta$, we find

$$\begin{aligned} H_k &= \frac{\dfrac{1}{2}\displaystyle\int_{\cos\phi}^{1} (q-\hat{q})\, T_k^{-1}(\eta)\, \mathrm{d}\eta}{\displaystyle\int_{-1}^{1} [T_k^{-1}(\eta)]^2\, \mathrm{d}\eta} \\ &= \tfrac{1}{4}k(k+1)(2k+1)\int_{\cos\phi}^{1} (q-\hat{q})\, T_k^{-1}(\eta)\, \mathrm{d}\eta. \end{aligned} \tag{66}$$

Finally, we have from the relations between G_k, H_k and D_k, $\hat{D}_k$,

$$D_k = \frac{G_k - \hat{\mu} H_k}{\mu + \hat{\mu}}, \quad \hat{D}_k = \frac{G_k + \mu H_k}{\mu + \hat{\mu}}. \tag{67}$$

3.4. *The drag force*

The drag force on the two-fluid drop is easily shown to be

$$\begin{aligned} \text{drag} &= -4\pi\mu U_0 R_0[C_1^* + \epsilon D_1] \\ &= -4\pi\mu U_0 R_0\left[C_1^* + \epsilon \frac{G_1 - \hat{\mu} H_1}{\mu + \hat{\mu}}\right], \end{aligned} \tag{68}$$

where C_1^*, G_1 and H_1 are given by (40), (65) and (66) respectively. This is the well known result that the drag force is proportional to the Stokeslet strength (the Stokeslet being the solution of the Stokes equations corresponding to a point force in unbounded fluid).

Now G_1 can be simplified as follows:

$$\begin{aligned} G_1 &= \frac{\sqrt{2}}{\pi}[\tfrac{3}{2}\cot\tfrac{1}{2}\phi \sin\tfrac{3}{2}\phi - \tfrac{1}{2}\cos\tfrac{3}{2}\phi]\,K(\phi) \\ &\quad - \frac{3}{\sqrt{2}\pi}\int_0^\phi \cot\tfrac{1}{2}u[\tfrac{3}{2}\cos\tfrac{3}{2}u - \tfrac{1}{2}\sin\tfrac{3}{2}u\cot\tfrac{1}{2}u]\,K(u)\,\mathrm{d}u \\ &= \frac{2\sqrt{2}}{\pi}\left\{2\cos^3\tfrac{1}{2}\phi K(\phi) + 3\int_0^\phi \cos^2\tfrac{1}{2}u \sin\tfrac{1}{2}u K(u)\,\mathrm{d}u\right\}, \end{aligned}$$

After substituting for $K(u)$ from (65*b*) and noting that the order of integration in the last term may be changed, we have

$$\begin{aligned} G_1 &= \frac{2\sqrt{2}}{\pi}\left\{2\cos^3\tfrac{1}{2}\phi K(\phi) + 3\int_0^\phi\left[\int_\theta^\phi \frac{\cos^2\frac{1}{2}u\sin\frac{1}{2}u}{(\cos\theta-\cos u)^{\frac{1}{2}}}\,\mathrm{d}u\right]\tan\tfrac{1}{2}\theta\sin\theta\,Q(\theta)\,\mathrm{d}\theta\right\}, \\ &= \frac{6}{\pi}\int_0^\phi \tan\tfrac{1}{2}\theta\sin\theta Q(\theta)\left\{\cos^2\tfrac{1}{2}\theta\cos^{-1}\left(\frac{\cos\frac{1}{2}\phi}{\cos\frac{1}{2}\theta}\right)\right. \\ &\quad \left. + \sqrt{\tfrac{1}{2}}\cos\tfrac{1}{2}\phi\,(\cos\theta-\cos\phi)^{\frac{1}{2}} + \frac{2\sqrt{2}\cos^3\frac{1}{2}\phi}{3(\cos\theta-\cos\phi)^{\frac{1}{2}}}\right\}\mathrm{d}\theta, \end{aligned} \tag{69}$$

where $Q(\theta) = -\frac{1}{2}(\hat{\mu}\hat{q} + \mu q)$, with q and $\hat{q}$ given by (51) and (53). For H_1 we find

$$H_1 = \frac{3}{4}\int_0^\phi [q(\theta) - \hat{q}(\theta)]\sin^2\theta\,\mathrm{d}\theta. \tag{70}$$

In order to evaluate G_1 and H_1 we use the following expressions for $q(\theta)$ and $\hat{q}(\theta)$, which are determined from (51) and (53):

$$q(\theta) = \tfrac{1}{3}[T(\theta) - \tfrac{1}{2}\hat{T}(\theta)]\,t(\theta) - \frac{\mu_f}{\mu}f_1(\theta)\,T(\theta), \tag{71}$$

$$\hat{q}(\theta) = \tfrac{1}{3}[\hat{T}(\theta) - \tfrac{1}{2}T(\theta)]\,t(\theta) - \frac{\mu_f}{\hat{\mu}}f_0(\theta)\,\hat{T}(\theta), \tag{72}$$

where

$$T(\theta) = \frac{\mu}{\mu_f}T_{R\theta}^{(0)}(1,\theta),$$

$$\hat{T}(\theta) = -\frac{\hat{\mu}}{\mu_f}\hat{T}_{R\theta}^{(0)}(1,\theta).$$

Using the leading-order solution, $T_{R\theta}^{(0)}(1,\theta)$ and $\hat{T}_{R\theta}^{(0)}(1,\theta)$ are found to be

$$T_{R\theta}^{(0)}(1,\theta) = \left[R\frac{\partial}{\partial R}\left(\frac{V^{(0)}}{R}\right)\right]_{R=1}$$
$$= 2\sum_{k=1}^{\infty}(2k+1)\,C_k T_k^{-1} - \frac{3\hat{\mu}}{\mu+\hat{\mu}}T_1^{-1},$$
$$\hat{T}_{R\theta}^{(0)}(1,\theta) = \left[R\frac{\partial}{\partial R}\left(\frac{\hat{V}^{(0)}}{R}\right)\right]_{R=1}$$
$$= -2\sum_{k=1}^{\infty}(2k+1)\,C_k T_k^{-1} - \frac{3\mu}{\mu+\hat{\mu}}T_1^{-1},$$

where

$$C_1 = C_1^* + \frac{1}{2}\frac{2\mu+3\hat{\mu}}{\mu+\hat{\mu}}, \quad C_k = C_k^*.$$

The C_k^* are given by (40). Furthermore, from the results of Sadhal & Johnson (1983), we have

$$\sum_{k=1}^{\infty} C_k(2k+1)\,T_k^{-1} = \frac{3}{4}\frac{\mu}{\mu+\hat{\mu}}g(\theta,\phi) \quad (0 \leqslant \theta \leqslant \phi), \tag{73a}$$

where

$$g(\theta,\phi) = -\frac{2}{\pi}\tan\tfrac{1}{2}\theta\left\{(1+\cos\theta)\left[\sin^{-1}\left(\frac{\cos\theta-\cos\phi}{1+\cos\theta}\right)^{\frac{1}{2}} + \frac{(\cos\theta-\cos\phi)^{\frac{1}{2}}(1+\cos\phi)^{\frac{1}{2}}}{1+\cos\theta}\right] + \frac{2(1+\cos\phi)^{\frac{3}{2}}}{3(\cos\theta-\cos\phi)^{\frac{1}{2}}}\right\}. \tag{73b}$$

Using (73) and $T_{-1}^{1}(\cos\theta) = \frac{1}{2}\sin\theta$, we have for $0 \leqslant \theta \leqslant \phi$,

$$\left.\begin{aligned} T_{R\theta}^{(0)}(1,\theta) &= \frac{3}{2}\frac{\mu}{\mu+\hat{\mu}}\left[g(\theta,\phi) - \frac{\hat{\mu}}{\mu}\sin\theta\right], \\ \hat{T}_{R\theta}^{(0)}(1,\theta) &= -\frac{3}{2}\frac{\mu}{\mu+\hat{\mu}}[g(\theta,\phi)+\sin\theta]. \end{aligned}\right\} \tag{74}$$

Note that it is easily shown that $T_{R\theta}^{(0)}(1,\theta) \leqslant 0$ and $\hat{T}_{R\theta}^{(0)}(1,\theta) \geqslant 0$, i.e. the shear stresses exerted on the fluid film are towards the rear of the drop.

Finally, after substituting q and $\hat{q}$, (71) and (72), into G_1 and H_1 and using the relations for f_0 and f_1 in terms of t, (35), we find

$$\text{drag} = 4\pi\mu U_0 R_0\left\{d_0 + \epsilon\left[\left(\hat{\Omega} - \frac{1}{3}\frac{\mu}{\mu_f}\right)d_1 + \frac{1}{6}\frac{\mu}{\mu_f}d_2 + \left(\Omega - \frac{1}{3}\frac{\hat{\mu}}{\mu_f}\right)d_3\right]\right\}, \tag{75}$$

where

$$d_0 = -C_1^*,$$
$$d_1 = \frac{-1}{\mu+\hat{\mu}}\{\mu I[\phi;S(\theta)] + \hat{\mu}J[\phi;S(\theta)]\},$$
$$d_2 = \frac{\hat{\mu}}{\mu+\hat{\mu}}\left\{I[\phi;\hat{S}(\theta)] + \frac{\hat{\mu}}{\mu}J[\phi;\hat{S}(\theta)] - I[\phi;S(\theta)] + J[\phi;S(\theta)]\right\},$$
$$d_3 = \frac{\hat{\mu}}{\mu+\hat{\mu}}\{I[\phi;\hat{S}(\theta)] - J[\phi;\hat{S}(\theta)]\},$$

and we define

$$S(\theta) = T^{(0)}_{R\theta}(1,\theta)\,t(\theta), \quad \hat{S}(\theta) = \hat{T}^{(0)}_{R\theta}(1,\theta)\,t(\theta),$$

$$I[\phi;F(\theta)] = \frac{3}{\pi}\int_0^{\phi} F(\theta)\,(1-\cos\theta)\left\{\cos^2\tfrac{1}{2}\theta\cos^{-1}\left(\frac{\cos\frac{1}{2}\phi}{\cos\frac{1}{2}\theta}\right)\right.$$

$$\left. + \sqrt{\tfrac{1}{2}}\cos\tfrac{1}{2}\phi\,(\cos\theta-\cos\phi)^{\frac{1}{2}} + \frac{2\sqrt{2}}{3}\frac{\cos^3\frac{1}{2}\phi}{(\cos\theta-\cos\phi)^{\frac{1}{2}}}\right\}\mathrm{d}\theta,$$

$$J[\phi;F(\theta)] = \frac{3}{4}\int_0^{\phi} F(\theta)\sin^2\theta\,\mathrm{d}\theta,$$

$$\hat{\Omega} = \frac{\hat{\sigma}_0}{\sigma_0+\hat{\sigma}_0}, \quad \Omega = \frac{\sigma_0}{\sigma_0+\hat{\sigma}_0}.$$

A discussion of the leading-order drag term d_0 has been given by Sadhal & Johnson (1983), and will not be repeated. At second-order the constants d_1, d_2 and d_3 are found to be positive. Consequently, a physical interpretation of each of the drag-correction terms can be made as follows. In the term $(\hat{\Omega}-\frac{1}{3}\mu/\mu_f)\,d_1$ the first part involving $\hat{\Omega}$ represents a drag increase due to the increased size of the drop associated with the presence of the film, and the second part $-\frac{1}{3}\mu/\mu_f$ is a drag-reducing effect due to the lubricating nature of the film, i.e. the small slip-velocity at the film interface decreases the drag. Note that if $\sigma_0 \gg \hat{\sigma}_0$ then $\hat{\Omega}$ is small and there is little drag increase from the first term, i.e. the film does not significantly increase the size of the drop. This is because when $\sigma_0 \gg \hat{\sigma}_0$ the outer film interface is much stiffer than the inner interface and most of the film deformation due to the pressure within the film occurs at the inner interface. A similar interpretation may be made for the term $(\Omega-\frac{1}{3}\hat{\mu}/\mu_f)\,d_3$. The first part Ω is a drag increase due to the fact that the presence of the film, i.e. its finite thickness, restricts the flow inside the drop. When $\hat{\sigma}_0 \gg \sigma_0$ this effect is small. The second part is a drag-reducing lubrication effect of the inner film interface. The remaining term $\frac{1}{6}(\mu/\mu_f)\,d_2$ always produces a drag increase. This term can be attributed to the coupling effect between inner and outer film interfaces. For example, motion of the film fluid at the outer film interface due to the shear stress there is felt at the inner interface owing to mass conservation in the film. In particular, since $T^{(0)}_{R\theta}(1,\theta) \leqslant 0$ film fluid near the outer interface is driven towards the rear of the drop, naturally feeding a return flow near the inner interface. This reduces the slip velocity at the inner interface and thereby increases the drag. A similar argument may be made for the inner film interface where $\hat{T}^{(0)}_{R\theta}$ also tends to drive film fluid towards the rear of the drop, and hence ultimately reduces the slip velocity at the outer interface. Note that the total drag correction is a net increase when $\mu/\mu_f \leqslant 3\hat{\Omega}$ and $\hat{\mu}/\mu_f \leqslant 3\Omega$.

4. Results and discussion

In this section we examine the characteristics of the fluid film and the drag-force correction terms for a variety of flow conditions. We begin by considering the solution of (34) for the film thickness $t(\theta)$. This is most easily done by considering the equation obtained from (34) after differentiating once with respect to θ. We find, after using the expression for $\partial p^{(1)}/\partial\theta$,

$$\frac{\partial^3 t}{\partial\theta^3} + \cot\theta\frac{\partial^2 t}{\partial\theta^2} + (2-\csc^2\theta)\frac{\partial t}{\partial\theta} = -\frac{\beta_0+\beta_1}{\epsilon^2}\frac{T(\theta)+\hat{T}(\theta)}{t(\theta)} = -\alpha\frac{g(\theta,\phi)}{t(\theta)}, \tag{76}$$

where

$$\alpha = \frac{3}{2}\frac{\mu}{\mu_f}\frac{(\beta_0+\beta_1)}{\epsilon^2}, \tag{77}$$

and $g(\theta, \phi)$ is given by (73*b*). The boundary conditions for (76) are as before,

$$t(0) = 1, \quad \frac{\partial t}{\partial \theta}(0) = 0, \quad t(\phi) = 0, \tag{78}$$

and the additional condition

$$\frac{\partial^2 t}{\partial \theta^2}(0) = -1 - \frac{1}{2}\frac{\beta_0+\beta_1}{\epsilon^2}p^{(1)}(0),$$

obtained from the original second-order equation (34). Here we see that (76) is of the third order and we have four boundary conditions, one of which essentially determines the unknown constant $p^{(1)}(0)$.

From (76) and (77) note the somewhat-surprising result that the film thickness depends on the bulk-fluid viscosity μ but is independent of the primary-drop viscosity $\hat{\mu}$. This can be explained as follows. The primary-drop fluid affects the film thickness through the action of its shear stress at the inner film interface. Furthermore, the viscous stresses in the primary drop are a consequence of the motion induced in the drop by the contact between the bulk fluid and primary drop fluid at the front interface of the drop. In particular, the shear stress is continuous at the front of the drop, and therefore the magnitude of the shear stresses inside the drop must be equal to the magnitude of the stresses in the bulk fluid. In other words, it is the viscous stresses in the bulk fluid which drive the overall motion. Consequently, it is the magnitude of these stresses and therefore the viscosity of the bulk fluid μ which appears in the coefficient α (76). The magnitude of the driving force on the film is characterized by the coefficient α and will be referred to as the driving-force parameter.

Equation (76) with boundary conditions (78) was solved for specified values of the driving-force parameter α using the shooting method with a fourth-order Runge–Kutta scheme. The quantity $p^{(1)}(0)$ was varied until $t(\phi) = 0$ was satisfied for a given value of ϕ. Recall that ϕ is determined in the appendix by (A 1) and (A 2) for specified values of the contact angles and surface tensions.

One feature of the solution found is that for a given value of α there corresponds a maximum value of ϕ for which $t(\phi) = 0$ can be satisfied. This result is shown in figure 2, where ϕ_{max} is plotted versus α for α between 0.01 and 100.0. As can be expected the maximum extent of the drop which may be covered by the film, i.e. ϕ_{max}, monotonically decreases as the driving force or α increases. Note that for a given value of α any value of ϕ less than ϕ_{max} is a possible steady-state solution, and for each of these solutions there corresponds a specific value of $p^{(1)}(0)$. The specific values of $p^{(1)}(0)$ are of little practical value and therefore will not be presented. The general character of $t(\theta)$ is that it monotonically decreases from unity at $\theta = 0$ to zero at $\theta = \phi$.

In figure 3 the volume factor $\gamma(\phi)$ defined in (36) is plotted versus the contact-line position ϕ for a few values of α. The last point on each curve corresponds to $\phi = \phi_{max}$. As pointed out earlier, for a given film volume this information determines the characteristic film thickness t_0.

The drag-force correction factors d_1, d_2 and d_3 are presented in figures 4, 5 and 6 for the viscosity ratios $\hat{\mu}/\mu = 0$, 1.0 and 100.0 respectively. The results are plotted

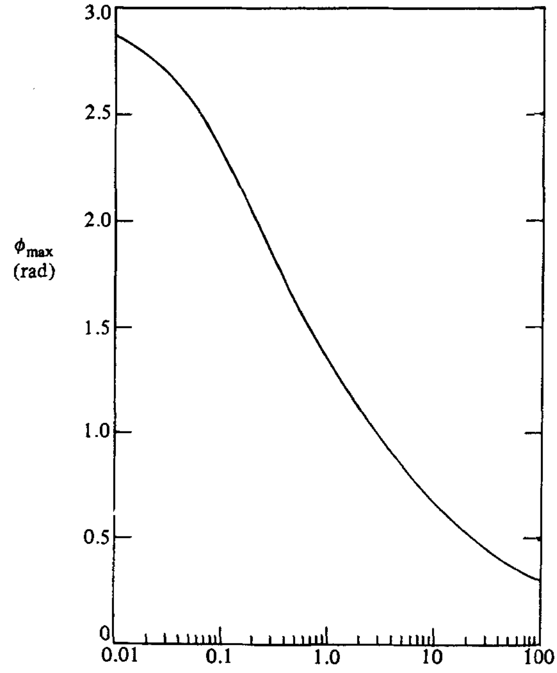

FIGURE 2. The maximum value of ϕ as a function of the driving-force parameter α.

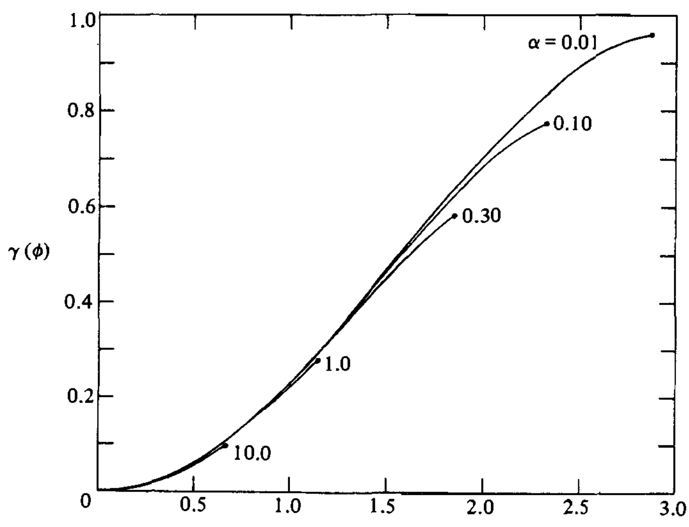

FIGURE 3. The volume factor γ as a function of the contact-line position ϕ and the driving-force parameter α. The largest value of ϕ for each α is ϕ_{max}.

versus ϕ for a range of values of the driving-force parameter. The viscosity ratio $\hat{\mu}/\mu = 0$ corresponds to a primary drop that is a bubble, and in this case $d_2 = d_3 \equiv 0$. The viscosity ratio $\hat{\mu}/\mu = 100.0$ would be representative of a primary drop comprised of oil moving in water or a primry drop slightly more viscous than water moving in air. In order to interpret the results recall that d_1 is due to the outer film interface, d_2 is due to the interaction effect, and d_3 is due to the inner film interface.

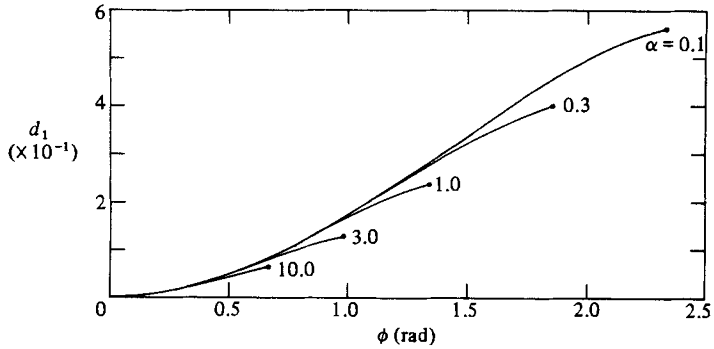

FIGURE 4. The drag-correction factor d_1 as a function of ϕ for a viscosity ratio $\hat{\mu}/\mu = 0$, i.e. a bubble. In this case $d_2 = d_3 = 0$. Five values of the driving-force parameter α are shown, and the largest value of ϕ for each α is ϕ_{max}.

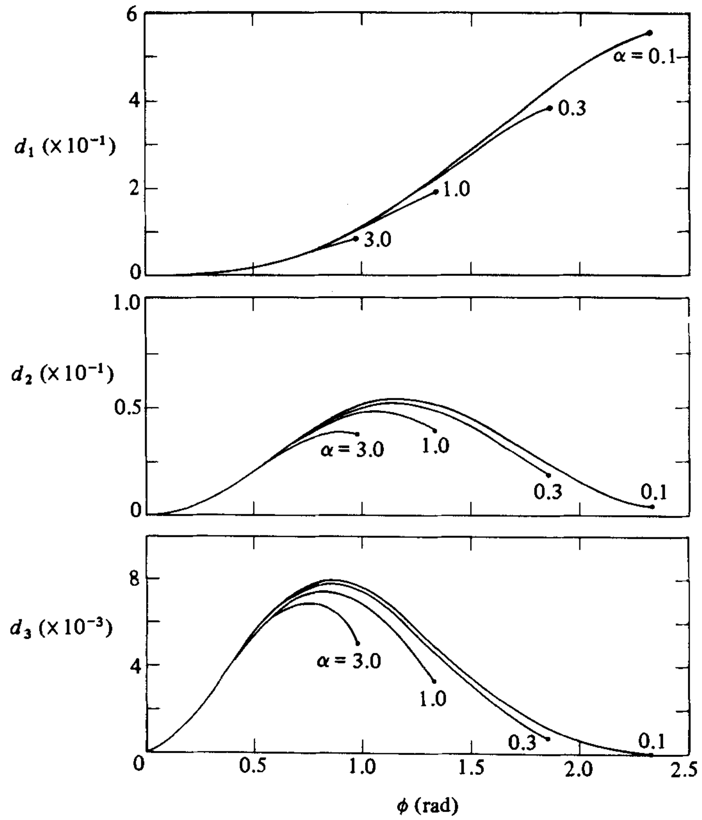

FIGURE 5. The drag-correction factors d_1, d_2 and d_3 as functions of ϕ for a viscosity ratio $\hat{\mu}/\mu = 1.0$. Four values of the driving-force parameter α are shown, and the largest value of ϕ for each α is ϕ_{max}.

The general trends of the three curves can be accounted for in the following way. The term d_1 monotonically increases as ϕ increases simply because the effect of the outer film interface increases as the extent of the drop covered by the film increases. In contrast, d_2 and d_3 have distinct maxima. For small ϕ the extent of the film is small so that d_2 and d_3 are small. As ϕ increases, d_2 and d_3 increase to a maximum value and then decrease. This is because both of these terms are associated with

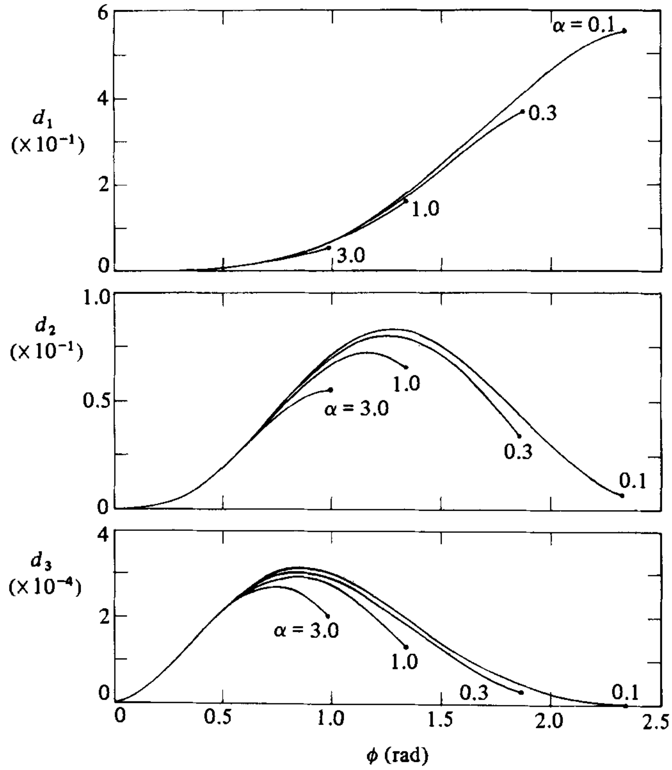

FIGURE 6. The drag-correction factors d_1, d_2 and d_3 as functions of ϕ for a viscosity ratio $\hat{\mu}/\mu = 100$. Four values of the driving-force parameter α are shown, and the largest value of ϕ for each α is ϕ_{max}.

motion in the primary drop, and this motion begins to weaken beyond a critical value of ϕ. This weakening of the primary drop motion is partially due to the fact that as ϕ increases beyond $\frac{1}{2}\pi$ the contact area between the bulk and primary-drop fluids where the motion is being driven is decreasing. However, d_2 and d_3 actually begin decreasing before $\phi = \frac{1}{2}\pi$ owing to the fact that as ϕ approaches $\frac{1}{2}\pi$ from below, the film begins to cover that portion of the primary drop where the shear stress in the bulk fluid driving motion in the drop is the largest.

Another noticeable feature is that d_3 is very small. Consequently, the terms in (75) involving d_1 and d_2 would generally be the dominant effect except when $\hat{\mu}$ is large, since d_3 is multiplied by $\hat{\mu}$ in the expression for the drag.

The circulation within the film has the interesting feature that for $\hat{\mu}/\mu > \frac{1}{2}$ the flow pattern consists of a double-cell structure. Examples of this are shown in figure 7. This criterion for a double-cell structure can be established by examining whether or not there are stagnation points on the inner and outer film interfaces, i.e. the streamlines $\xi = -f_0(\theta)$ and $\xi = f_1(\theta)$. This is accomplished by requiring the tangential velocity to vanish, i.e.

$$v^{(1)}(\xi = -f_0) = \tfrac{1}{3}(\hat{T} - \tfrac{1}{2}T)\, t = 0,$$

$$v^{(1)}(\xi = f_1) = \tfrac{1}{3}(T - \tfrac{1}{2}\hat{T})\, t = 0.$$

After substituting for $T(\theta)$ and $\hat{T}(\theta)$ we find that $v^{(1)}$ vanishes on $\xi = -f_0$ at the point θ that satisfies

$$-\frac{\sin\theta}{g(\theta,\phi)} = \frac{2\hat{\mu}/\mu - 1}{3\hat{\mu}/\mu}, \tag{79}$$

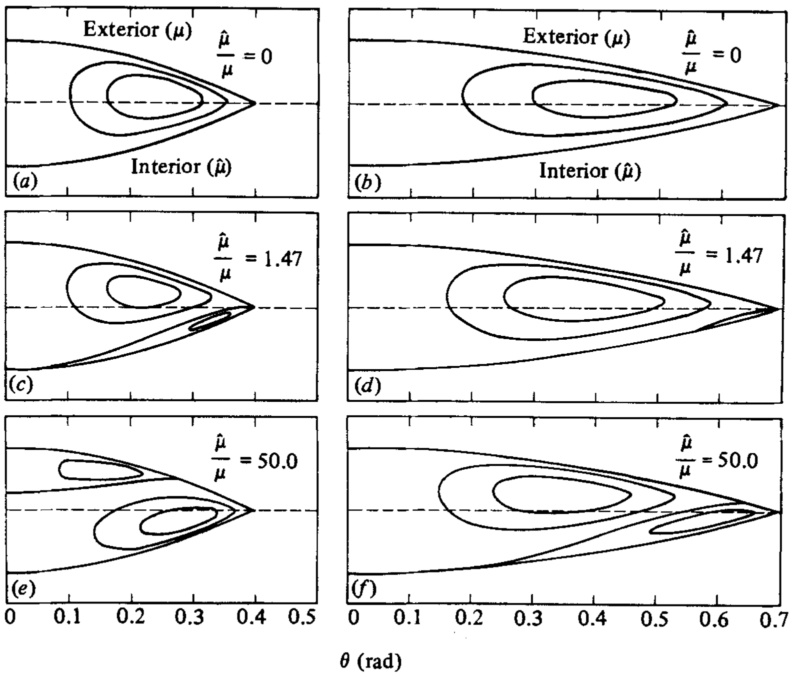

FIGURE 7. Typical fluid-film profiles. Three viscosity ratios are shown $\hat{\mu}/\mu = 0$, 1.47, 50.0. The driving-force parameter $\alpha = 1.0$, and $\Omega = \hat{\Omega} = \frac{1}{2}$. The pressure parameter $\epsilon^{-2}(\beta_0+\beta_1)\,p(0)$ is equal to 50.0 in (*a*), (*c*) and (*e*), and is equal to 15.5 in (*b*), (*d*) and (*f*).

and $v^{(1)}$ vanishes on $\xi = f_1$ when

$$-\frac{\sin\theta}{g(\theta,\phi)} = \frac{\hat{\mu}/\mu - 2}{3\hat{\mu}/\mu}, \tag{80}$$

where $g(\theta,\phi)$ is given by (73*b*). For a specified value of ϕ between 0 and π, the function $-\sin\theta/g(\theta,\phi)$ has the following properties: $0 \leqslant -\sin\theta/g(\theta,\phi) \leqslant 1$, is zero at the contact line $\theta = \phi$, and monotonically increases as θ decreases from $\theta = \phi$ to $\theta = 0$. Consequently a solution of (79) with θ less than ϕ is only possible when the right-hand side is greater than zero, i.e. $\hat{\mu}/\mu > \frac{1}{2}$. When $\hat{\mu}/\mu = \frac{1}{2}$ the solution is $\theta = \phi$. Similarly (80) can only be satisfied for $\hat{\mu}/\mu > 2$. For $\hat{\mu}/\mu$ slightly larger than $\frac{1}{2}$ but smaller than 2 there would be a stagnation point on the inner film interface between 0 and ϕ, and the stagnation point on the outer interface would be at the contact line $\theta = \phi$. An example of this is shown in figures 7 (*c*, *d*). For $\hat{\mu}/\mu$ greater than 2 the stagnation point on the outer interface moves away from $\theta = \phi$ as in figure 7 (*f*). As $\hat{\mu}/\mu \to \infty$ the right-hand sides of (79) and (80) tend to $\frac{2}{3}$ and $\frac{1}{3}$ respectively, and the two stagnation points would be at their furthest distance from the contact line. The stagnation point on the inner interface may in fact move all the way to the rear as in figure 7 (*e*). This possibility exists when ϕ is less than or equal to 0.6689 rad or 38.33°. This maximum value of ϕ is the value when the stagnation point furthest from the contact line ($\hat{\mu}/\mu = \infty$) first reaches the rear of the drop $\theta = 0$, i.e. the value of ϕ for which $[-\sin\theta/g(\theta,\phi)]_{\theta=0} = \frac{2}{3}$. Furthermore, the outer-interface stagnation point may also move all the way to rear, in which case we have a single cell with circulation opposite to that when $\hat{\mu}/\mu = 0$. In this case the circulation in the film is being controlled by the shear at the inner film interface. The largest value of ϕ when this is possible is determined from $[-\sin\theta/g(\theta,\phi)]_{\theta=0} = \frac{1}{3}$, and is found to be $\phi = 0.2930$ rad or 16.79°.

Lastly, note that another film configuration which cannot be ruled out in the present problem is that of a film entirely surrounding the primary drop. This case would correspond to a solution of (76) with boundary conditions $t(0) = 1$ and $\partial t/\partial\theta(0) = \partial t/\partial\theta(\pi) = 0$. Mori (1978) suggests that this is a possible steady-state case, but the experimental evidence has not yet verified his claim. Preliminary numerical calculations conducted here seem to support Mori. However, numerically determining the precise range of parameters when this solution is possible is a somewhat tedious and costly calculation, which at present does not seem particularly valuable.

In the area of direct contact heat- and mass-exchangers, the results from the present study are of fundamental value in the development of a successful model. A common situation of practical interest is the case of a vapor bubble growing from a liquid film in an immisible liquid. For the thermodynamical analysis of this case it is important to know the flow field, the film thickness, and the extent to which the bubble is covered by the film.

The analysis may also be employed to indirectly measure contact angles. It is quite clear that for a given set of fluids and a fixed volume there is a unique angle ϕ for a particular contact angle. Since the angle ϕ is more easily measured than the contact angle, this may be a useful method for determining the contact angle.

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Appendix

The position of the contact line $\theta = \phi$ is most easily determined from a consideration of global force equilibrium on the film. That is, the forces exerted on the film by the surrounding fluids are balanced by the surface tension forces at the contact line. In dimensional quantities this is

$$\int_{\hat{S}} \hat{\boldsymbol{f}}\,\mathrm{d}S + \int_S \boldsymbol{f}\,\mathrm{d}S + \int_C (\hat{\sigma}_0\hat{\boldsymbol{\nu}} + \sigma_0\boldsymbol{\nu})\,\mathrm{d}s = 0,$$

where $\hat{S}$ and S are the inner and outer fluid-film interfaces, C is the curve formed by the contact line $\theta = \phi$, $\hat{\boldsymbol{f}}$ and $\boldsymbol{f}$ are the forces per unit area exerted on the film at the inner and outer interfaces by the surrounding fluid (primary-drop and bulk fluid), and $\hat{\boldsymbol{\nu}}$ and $\boldsymbol{\nu}$ are the unit vectors tangent to the surfaces $\hat{S}$ and S and perpendicular to the curve C respectively (see figure 8). Note that the contact angles denoted by $\hat{\Phi}_0$ and Φ_0 are the angles between $\hat{\boldsymbol{\nu}}$ and $\boldsymbol{\nu}$ and a tangent to the sphere $R = 1$ respectively. These angles are material properties, which we assume are known quantities.

To leading-order the large pressure $\hat{P} \approx 2/\beta$ within the drop dominates the surface-force terms ($\beta = O(\epsilon^2\mu/\mu_f) \ll 1$). Clearly the viscous stresses which are $O(1)$ are negligible. Furthermore, the hydrostatic stresses are also small compared with β^{-1} since they are $O(\rho g R_0^2/\hat{\mu}U_0,\ \hat{\rho} g R_0^2/\hat{\mu}U_0)$ and are much less than $O(\mu_f/\epsilon\hat{\mu}, (\hat{\rho}/\rho)\,\mu_f/\epsilon\hat{\mu})$ based on the restriction made earlier that $1 - \hat{\rho}/\rho \gg \epsilon\mu/\mu_f$. Since the pressure $\hat{P} \approx 2/\beta$ and the surface tensions are constant, the integrals are easily computed, giving

$$\frac{\hat{\mu}U_0}{R}\frac{2}{\beta}\pi R^2 \sin^2\phi \approx 2\pi R_0 \sin\phi\{\hat{\sigma}_0 \sin(\phi - \hat{\Phi}_0) + \sigma_0 \sin(\phi + \Phi_0)\}.$$

Recalling that $\beta = \hat{\mu}U_0/\sigma_1$, we find

$$\tan\phi = \frac{\sigma_0 \sin\Phi_0 - \hat{\sigma}_0 \sin\hat{\Phi}_0}{\sigma_1 - \sigma_0\cos\Phi_0 - \hat{\sigma}_0\cos\hat{\Phi}_0}, \qquad \text{(A 1)}$$

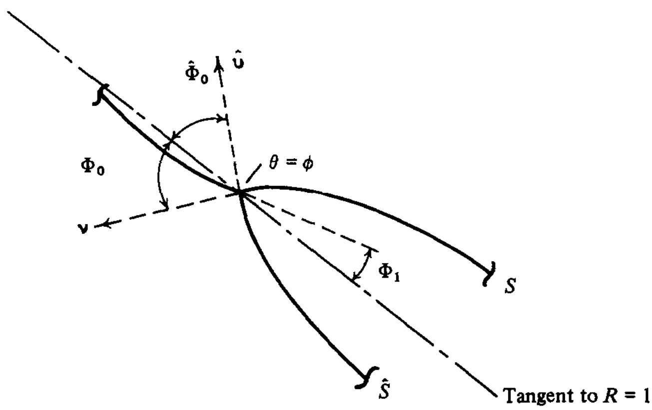

FIGURE 8. The film geometry near the contact line $\theta = \phi$.

where in the problem considered here $\sigma_1 = \sigma_0 + \hat{\sigma}_0$. Similarly we can determine the position of the contact line by considering force equilibrium of the front interface $\phi \leqslant \theta \leqslant \pi$. Given the contact angle Φ_1 of the front interface at $\theta = \phi$ and the surface-tension σ_1 force equilbrium gives

$$\tan\phi = \frac{\sin\Phi_1}{1-\cos\Phi_1}. \tag{A 2}$$

Note that the relation between Φ_1 and Φ_0, $\hat{\Phi}_0$, σ_0, $\hat{\sigma}_0$ and σ_1 obtained by equating (A 1) to (A 2) is consistent with the fact that the resultant of the surface-tension forces acting on the three-fluid contact line must be zero, i.e.

$$\sigma_0 \sin\Phi_0 - \hat{\sigma}_0 \sin\hat{\Phi}_0 = \sigma_1 \sin\Phi_1, \tag{A 3}$$

$$\sigma_0 \cos\Phi_0 + \hat{\sigma}_0 \cos\hat{\Phi}_0 = \sigma_1 \cos\Phi_1 = \sigma_1(1+\cos\Phi_1 - 1). \tag{A 4}$$

Dividing (A 3) by $\sigma_1(1-\cos\Phi_1)$ and using (A 4) gives an expression equivalent to that obtained by equating (A 1) and (A 2). Also note that the restriction $\sigma_1 = \sigma_0 + \hat{\sigma}_0$ is automatically satisfied in the special case when all of the contact angles are small.

We should point out here that the contact angles need not be small. The reader might expect that since the analysis is concerned with small interface deformations that this would require the contact angles Φ_0, $\hat{\Phi}_0$ and Φ_1 to be small. However, as was discussed earlier, the present analysis does not provide a uniformly valid solution to the problem. In fact, an accurate description for the shapes of the interfaces close to the contact line would require an inner solution to be constructed which would admit large interface slopes and have a specified slope (contact angle) at the contact line. This inner solution, however, does not significantly influence the outer solution found here, which is valid everywhere except very close to the contact line. The passive nature of the inner solution is evidenced by the fact that the solution to (34) (the outer solution) is able to satisfy one of the inner boundary conditions, i.e. $t(\phi) = 0$. This situation is a frequent occurrence in asymptotic analysis and additional discussion of this point is given by Johnson (1981).

Lastly, one might anticipate a singularity at the contact line in the viscous stresses exerted on the film by the inner and outer fluids. However, this is found to be a square-root singularity, and consequently its contribution to the force balance considered here is negligible.

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