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The Stokes force on a droplet in an unbounded fluid medium due to capillary effects

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The Stokes force on a fluid droplet is obtained when the droplet is placed in an unbounded fluid medium and motion ensues due to an arbitrary interfacial-tension gradient on the droplet surface. The force, derived here for a spherical droplet, is proportional to the integral of the interfacial-tension gradient over the droplet surface. It may be calculated without solving the complete governing equations from a knowledge of this integral and the ratio of the viscosity of the droplet phase to that of the continuous phase, as shown in the principal result displayed in (29).

When the interfacial-tension gradients are caused by temperature or concentration variations, the result for the force may be further specialized when convective transport effects are negligible. In this case, it is possible to express the force in terms of the gradient of the undisturbed temperature (or concentration) field evaluated at the location of the droplet centre in a form analogous to Faxén's force law.

1. Introduction

The variation of the interfacial tension along a fluid–fluid interface will result in a tangential stress discontinuity across the interface. The normal consequence is the motion of the fluids present on either side of the interface. Since the interfacial tension depends on the temperature, species concentration, and the electrical charge density at the interface, a gradient in any one of these entities can cause fluid motion.

When a droplet of one fluid is placed in a second fluid, and conditions exist which produce a non-uniform interfacial-tension distribution on the droplet surface, the droplet will experience a hydrodynamic force. It is useful to be able to calculate this force without recourse to the full solution of the governing equations, if possible. This is the quest of the present article.

It is worthwhile to point out that some results are available for the direct calculation of the hydrodynamic force on a *spherical* object placed in an unbounded fluid under conditions of Stokes flow. For instance, Faxén's (1924) relations permit the calculation of the force and torque on a rigid sphere placed in an unbounded arbitrary Stokes flow. For a rigid object, of course, interfacial tension is not a relevant quantity. In the case of a spherical *fluid* droplet of radius a, translating at a velocity $\boldsymbol{U}$ in an unbounded arbitrary Stokes flow $\boldsymbol{u}_\infty(\boldsymbol{r})$ under conditions when the interfacial tension is uniform, Hetsroni & Haber (1970) obtained similar results using a full spherical harmonics solution (here, $\boldsymbol{r}$ is the position vector). Later Hetsroni, Wacholder & Haber (1971) employed the reciprocal theorem of Lorentz (Happel & Brenner 1965) together with a spherical harmonics solution to obtain the same result. In this case, the hydrodynamic force is given by

$$\boldsymbol{F} = 2\pi\mu a\,\frac{2+3\alpha}{1+\alpha}\left[(\boldsymbol{u}_\infty)_0 - \boldsymbol{U} + \frac{\alpha a^2}{2(2+3\alpha)}\,(\nabla^2\boldsymbol{u}_\infty)_0\right]. \tag{1}$$

Here, μ is the viscosity of the continuous phase, and α is the ratio of the viscosity of the droplet phase to that of the continuous phase. The subscript 0 refers to evaluation at the location of the droplet centre. The same equation, excluding translation, was derived more recently by Rallison (1978) who used an elegant alternative approach. It may be noted that (1) contains the rigid-sphere case as the asymptotic limit, $\alpha \rightarrow \infty$.

Returning to the influence of interfacial-tension gradients on droplets, Young, Goldstein & Block (1959) pointed out that a fluid droplet placed in a thermal gradient environment will experience a force due to capillarity. The interfacial tension will typically be a minimum at the warm pole of the droplet, and will increase toward the cooler pole. The resulting tangential stress will drag the neighbouring continuous-phase fluid toward the cooler pole, and the reaction on the droplet will therefore be in the opposite direction. If the droplet is free to move, it will migrate toward warmer regions as a consequence. Young *et al.* demonstrated the phenomenon by applying a downward thermal gradient on gas bubbles introduced into a vertical liquid column. They were able to arrest the buoyant rise of the bubbles, and move them downward by applying a sufficiently large thermal gradient. In fact, under conditions of Stokes flow, a gas bubble approximately 20 μm in diameter will experience the same force due to a 1 K/mm temperature gradient as due to buoyancy in a Dow Corning DC200 silicone oil, a result independent of the viscosity of the fluid. At sizes down to 1–5 μm, capillarity can dominate over both gravity and Brownian motion in governing the migration of bubbles and droplets.

In addition to performing experiments, Young *et al.* also solved the theoretical problem of a fluid droplet placed in an unbounded fluid possessing a *uniform* thermal gradient in the undisturbed state. They assumed negligible convective transport of momentum and energy and constant physical properties, and therefore solved the Stokes and Laplace's equations for the velocity and temperature fields respectively. In the tangential-stress balance, the interfacial tension was assumed linear in temperature. The spherical shape is preserved under the above assumptions. From results given by Young *et al.*, one can obtain the hydrodynamic force exerted on a stationary droplet due to capillarity-induced motion in the surrounding fluid (after correcting some minor typographical errors in their paper),

$$\boldsymbol{F} = -\frac{4\pi a^2 \sigma'}{(1+\alpha)(2+\beta)} \nabla T_\infty. \tag{2}$$

Here, $\sigma' = \mathrm{d}\sigma/\mathrm{d}T$ is the gradient of the interfacial tension with temperature, a is the droplet radius, and β is the ratio of the thermal conductivity of the droplet phase to that of the continuous phase. α has been defined earlier. Note that the result presumes that ∇T_∞ is a constant vector field. It will be shown later that (2) is a special case of the more general results derived in the present work.

Additional literature on capillarity-induced migration of droplets and bubbles is discussed elsewhere (Subramanian 1981, 1983) and will not be reviewed here.

In what follows, constant physical properties are assumed with the exception of the interfacial tension σ. This quantity is permitted to vary with position on the droplet surface.

The symbols employed for the droplet phase are distinguished from those for the continuous phase by a caret. Initially, the Reynolds number for the flow in the continuous phase is assumed negligible. In the latter part of the development, it will become necessary to require this in the droplet phase as well. Thus, the velocity fields $\boldsymbol{v}(\boldsymbol{r})$ and $\hat{\boldsymbol{v}}(\boldsymbol{r})$ both satisfy Stokes equations. The symbols $\pi(\boldsymbol{r})$ and $\hat{\pi}(\boldsymbol{r})$ are used for the corresponding stress tensor fields, in which, by convention, the pressure is taken to be the *hydrodynamic* pressure (Happel & Brenner 1965). One exception is made to this convention, and is noted in the text where it appears.

2. A variant of the Lorentz reciprocal theorem

In order to arrive at the required result, it is necessary to use the Lorentz reciprocal theorem. According to this theorem, if $(\boldsymbol{v}_1, \boldsymbol{\pi}_1)$ and $(\boldsymbol{v}_2, \boldsymbol{\pi}_2)$ are the velocity and the hydrodynamic stress fields corresponding to any two Stokes flows of the same fluid, and a surface S completely encloses an arbitrary region R *occupied by the fluid*,

$$\int_S \mathbf{d}\boldsymbol{S}\cdot\boldsymbol{\pi}_2\cdot\boldsymbol{v}_1 = \int_S \mathbf{d}\boldsymbol{S}\cdot\boldsymbol{\pi}_1\cdot\boldsymbol{v}_2. \tag{3}$$

Here, $\mathbf{d}\boldsymbol{S}$ is the usual directed area element oriented in the direction of the normal pointing outward from the region R.

If the region R is taken to be bounded by the surface of the droplet and an arbitrarily large spherical boundary in the continuous-phase fluid, a useful version of the above result may be obtained. In an unbounded quiescent fluid, at large distances r from the droplet, the velocity field $\boldsymbol{v}$ is $O(1/r)$ while $\boldsymbol{\pi}$ is $O(1/r^2)$. Brenner (1963) used this fact to show that (3) reduces to

$$\int_{S_\mathrm{d}} \mathbf{d}\boldsymbol{S}\cdot\boldsymbol{\pi}_2\cdot\boldsymbol{v}_1 = \int_{S_\mathrm{d}} \mathbf{d}\boldsymbol{S}\cdot\boldsymbol{\pi}_1\cdot\boldsymbol{v}_2 \tag{4}$$

in this situation. Here, S_d is the surface of the droplet and $\mathbf{d}\boldsymbol{S}$ is redefined so that it points *into the continuous-phase fluid*. This result is independent of the boundary conditions used at the surface S_d, and is equally valid for a rigid body or a fluid droplet, stationary, or in quasi-static motion. The only condition imposed in addition to creeping motion is that the fluid at infinity be quiescent.

The next step is to obtain a version of the reciprocal theorem which is useful here. For this, consider a *fluid* droplet held fixed in an unbounded continuous-phase fluid which is quiescent at infinity. Let two Stokes fields $(\boldsymbol{v}_1, \boldsymbol{\pi}_1)$ and $(\boldsymbol{v}_2, \boldsymbol{\pi}_2)$ be induced by capillary effects resulting from interfacial-tension gradients $\boldsymbol{\nabla}_\mathrm{s}\,\sigma_1(\boldsymbol{r})$ and $\boldsymbol{\nabla}_\mathrm{s}\,\sigma_2(\boldsymbol{r})$ respectively (on the droplet surface S_d). Here $\boldsymbol{\nabla}_\mathrm{s}$ is the surface gradient operator. In this situation, since the fluid at infinity is quiescent, (4) is applicable.

The stress balance at the droplet interface in each problem reads[†]

$$\boldsymbol{n}\cdot\boldsymbol{\pi} = \boldsymbol{n}\cdot\hat{\boldsymbol{\pi}} - \boldsymbol{\nabla}_\mathrm{s}\,\sigma + 2\sigma H\boldsymbol{n} \quad \text{on } S_\mathrm{d}. \tag{5}$$

Here, $\boldsymbol{n}$ is the unit normal and H the mean curvature at any point on S_d.

The velocity field $\boldsymbol{v}$ is tangent to the surface S_d everywhere on it. It is also continuous across it (that is, $\boldsymbol{v} = \hat{\boldsymbol{v}}$ on S_d). These facts make it possible to obtain the following result from (5),

$$\mathbf{d}\boldsymbol{S}\cdot\boldsymbol{\pi}_2\cdot\boldsymbol{v}_1 = -\boldsymbol{\nabla}_\mathrm{s}\,\sigma_2\cdot\boldsymbol{v}_1\,\mathrm{d}S + \mathbf{d}\boldsymbol{S}\cdot\hat{\boldsymbol{\pi}}_2\cdot\hat{\boldsymbol{v}}_1 \quad \text{on } S_\mathrm{d}. \tag{6}$$

Here, $\mathrm{d}S = |\mathbf{d}\boldsymbol{S}|$. Substitution of (6), and an analogous result wherein the subscripts 1 and 2 are interchanged, into (4) leads to

$$\int_{S_\mathrm{d}} \boldsymbol{\nabla}_\mathrm{s}\,\sigma_2\cdot\boldsymbol{v}_1\,\mathrm{d}S - \int_{S_\mathrm{d}} \mathbf{d}\boldsymbol{S}\cdot\hat{\boldsymbol{\pi}}_2\cdot\hat{\boldsymbol{v}}_1 = \int_{S_\mathrm{d}} \boldsymbol{\nabla}_\mathrm{s}\,\sigma_1\cdot\boldsymbol{v}_2\,\mathrm{d}S - \int_{S_\mathrm{d}} \mathbf{d}\boldsymbol{S}\cdot\hat{\boldsymbol{\pi}}_1\cdot\hat{\boldsymbol{v}}_2. \tag{7}$$

[†] Strictly, $\boldsymbol{\pi}$ and $\hat{\boldsymbol{\pi}}$ in (5) stand for the complete stress tensors, including hydrostatic contributions in the presence of body forces. However, this distinction becomes irrelevant in the transition to (6).

However, S_{d} completely encloses the droplet phase, and the Lorentz reciprocal theorem may be directly applied to the *droplet phase* to obtain the equality of the second members of the left- and right-hand sides of (7). Thus, the following analogue of the reciprocal theorem is obtained for capillary problems,

$$\int_{S_{\mathrm{d}}} \nabla_{\mathrm{s}}\sigma_2 \cdot \boldsymbol{v}_1 \, \mathrm{d}S = \int_{S_{\mathrm{d}}} \nabla_{\mathrm{s}}\sigma_1 \cdot \boldsymbol{v}_2 \, \mathrm{d}S. \tag{8}$$

Note that, up to this point, the shape of the droplet has been permitted to be arbitrary, and no restrictions have been imposed on the interfacial-tension gradient distribution except that $\nabla_{\mathrm{s}}\sigma$ exist and be continuous on S_{d}.

3. The Stokes force on a spherical droplet

Brenner (1964*b*) used (4) which he had developed earlier, in (1963), to extend Faxén's laws to a rigid body of arbitrary shape placed in an arbitrary unbounded Stokes flow. In doing so, he obtained several useful intermediate results. One of them, when specialized suitably, gives the hydrodynamic force exerted on an object of spherical shape at whose surface an arbitrary velocity field $\boldsymbol{v}$ exists, provided the surrounding fluid (of viscosity μ) is quiescent at infinity:

$$\boldsymbol{F} = -\frac{3\mu}{2a}\int_{S_a} \boldsymbol{v}\, \mathrm{d}S. \tag{9}$$

Here, a is the radius of the sphere, and S_a its boundary. Again, the result is equally valid whether the object within the spherical boundary S_a is rigid or fluid.

It is interesting to note that (9) can be obtained by the application of the reciprocal theorem to two Stokes flows, over a spherical boundary S_a, one of which encloses a *rigid solid* translating at a constant velocity, and the other, a *fluid* droplet with an arbitrary velocity field on S_a.

A digression concerning the shape of the droplet is in order. In practice, as a consequence of the non-uniform normal stress imbalance at the interface, a fluid droplet will assume a non-spherical shape in the problem under consideration. However, in creeping motion induced by capillary effects, the variation in curvature will be $O(\Delta\sigma/\sigma)$ where $\Delta\sigma$ is the variation of the surface tension over the boundary of the droplet. Thus, if $\Delta\sigma/\sigma$, which serves the role of a capillary number in this problem, is small compared to unity, a spherical shape may be assumed for the droplet without introducing gross errors. This assumption is made from here onward in order to make further progress. It might be added that Brenner (1964*b*) provides a result for the force on arbitrary-shaped objects as well, and it might be possible, with a creative application of that result, to calculate the force on a droplet of arbitrary, but *given*, shape.

4. Expression of the force in terms of an arbitrary $\nabla_{\mathrm{s}}\sigma$ on the droplet surface

The next logical step is to use (8) and (9) together to obtain the hydrodynamic force exerted on a stationary spherical fluid droplet when $\nabla_{\mathrm{s}}\sigma$ is known on its surface. For this, let

$$\nabla_{\mathrm{s}}\sigma_1 = \boldsymbol{A} - (\boldsymbol{n}\cdot\boldsymbol{A})\,\boldsymbol{n}, \tag{10}$$

where $\boldsymbol{A}$ is an arbitrary, but *constant* spatial vector defined on S_d. Since $\boldsymbol{v}_2$ is tangent to S_d, (8) may be rewritten, taking (9) into account, as follows:

$$-\frac{2a}{3\mu}\boldsymbol{F}_2\cdot\boldsymbol{A} = \int_{S_a} \nabla_s\sigma_2\cdot\boldsymbol{v}_1\,\mathrm{d}S. \tag{11}$$

Note that, from here on, we shall assume $S_d \equiv S_a$ = surface of a spherical fluid droplet of radius a.

Now, inspired by Brenner (1964*a*), a second-rank tensor 'velocity' field $\boldsymbol{V}_1$, an associated vector 'pressure' field $\boldsymbol{P}_1$, and a third-rank (triadic) 'stress' tensor field $\boldsymbol{\Pi}_1$ are defined as follows.

$$\boldsymbol{v}_1 = \boldsymbol{V}_1\cdot\boldsymbol{A},\quad p_1 = \boldsymbol{P}_1\cdot\boldsymbol{A},\quad \boldsymbol{\pi}_1 = \boldsymbol{\Pi}_1\cdot\boldsymbol{A}. \tag{12a,b,c}$$

Similar fields for the droplet phase are defined in a completely analogous fashion.

Use of (12*a*) in (11) leads to

$$\left\{\int_{S_a} \nabla_s\sigma_2\cdot\boldsymbol{V}_1\,\mathrm{d}S\right\}\cdot\boldsymbol{A} = -\frac{2a}{3\mu}\boldsymbol{F}_2\cdot\boldsymbol{A}. \tag{13}$$

Since $\boldsymbol{A}$ is an arbitrary vector, and it will be seen shortly that $\boldsymbol{V}_1$ is independent of $\boldsymbol{A}$, this yields a result for the force $\boldsymbol{F}_2$,

$$\boldsymbol{F}_2 = -\frac{3\mu}{2a}\int_{S_a} \nabla_s\sigma_2\cdot\boldsymbol{V}_1\,\mathrm{d}S. \tag{14}$$

The properties of the tensor field $\boldsymbol{V}_1$, to be established shortly, will permit the rewriting of (14) in a form which only involves the surface gradient of σ_2 on S_a.

5. Calculation of the tensor fields $\boldsymbol{V}_1$ and $\hat{\boldsymbol{V}}_1$

The equations satisfied by the velocity fields $\boldsymbol{v}_1$ and $\hat{\boldsymbol{v}}_1$ are stated below:

$$\left.\begin{aligned} \nabla\cdot\boldsymbol{v}_1 = 0,\quad & \nabla\cdot\hat{\boldsymbol{v}}_1 = 0,\\ \mu\nabla^2\boldsymbol{v}_1 = \nabla p_1,\quad & \hat{\mu}\nabla^2\hat{\boldsymbol{v}}_1 = \nabla\hat{p}_1.\end{aligned}\right\} \tag{15}$$

The boundary conditions are

$$\boldsymbol{v}_1 \rightarrow \boldsymbol{0}\quad \text{as } r\rightarrow\infty, \tag{16a}$$

$$\hat{\boldsymbol{v}}_1 \text{ is bounded}\quad \text{at } r = 0 \tag{16b}$$

$$\left.\begin{aligned} \boldsymbol{v}_1 = \hat{\boldsymbol{v}}_1;\quad \boldsymbol{n}\cdot\boldsymbol{v}_1 = \boldsymbol{n}\cdot\hat{\boldsymbol{v}}_1 = 0,\\ \boldsymbol{n}\cdot\boldsymbol{\pi}_1\cdot\boldsymbol{t} = -\boldsymbol{A}\cdot\boldsymbol{t}+\boldsymbol{n}\cdot\hat{\boldsymbol{\pi}}_1\cdot\boldsymbol{t}\end{aligned}\right\}\ \text{on } S_a. \tag{16c}$$

Here, r is the distance from an origin located at the centre of the droplet, and $\boldsymbol{t}$ is an arbitrary unit vector lying in the tangent plane at any point on S_a. Since the capillary number is assumed negligible, the balance of normal stress at the interface will yield a spherical shape for the droplet, and will not be considered further. Equations (16*c*) represent the continuity of the velocity vector, the kinematic condition, and the tangential-stress balance at the interface S_a.

When the definitions in (12*a–c*) and analogous ones for the droplet phase are inserted in the above, use of the fact that $\boldsymbol{A}$ is an arbitrary vector yields the following equations for the tensor fields $\boldsymbol{V}_1$ and $\hat{\boldsymbol{V}}_1$. From here on, for convenience, the subscript 1 on these fields is dropped:

$$\left.\begin{aligned} \nabla\cdot\boldsymbol{V}=0, \quad & \nabla\cdot\hat{\boldsymbol{V}}=0, \\ \mu\nabla^2\boldsymbol{V}=\nabla\boldsymbol{P}, \quad & \hat{\mu}\nabla^2\hat{\boldsymbol{V}}=\nabla\hat{\boldsymbol{P}}, \end{aligned}\right\} \tag{17}$$

$$\boldsymbol{V}\rightarrow\boldsymbol{0} \quad \text{as } r\rightarrow\infty, \tag{18a}$$

$$\hat{\boldsymbol{V}}\text{ is bounded} \quad \text{at } r=0, \tag{18b}$$

$$\left.\begin{aligned} \boldsymbol{V}=\hat{\boldsymbol{V}};\quad \boldsymbol{n}\cdot\boldsymbol{V}=\boldsymbol{n}\cdot\hat{\boldsymbol{V}}=0, \\ \boldsymbol{n}\cdot\boldsymbol{\Pi}^{\dagger}\cdot\boldsymbol{t}=-\boldsymbol{t}+\boldsymbol{n}\cdot\hat{\boldsymbol{\Pi}}^{\dagger}\cdot\boldsymbol{t} \end{aligned}\right\} \text{ on } S_a. \tag{18c}$$

The fields $\boldsymbol{V}$ and $\hat{\boldsymbol{V}}$ are independent of $\boldsymbol{A}$. Unlike in Brenner's work, wherein it is possible to eliminate dependence on μ as well by suitable definition of $\boldsymbol{P}$ and $\boldsymbol{\Pi}$, this is not possible here. Adopting Brenner's definitions leads to the appearance of μ and $\hat{\mu}$ in the tangential-stress balance. Thus, we shall use the above definitions which yield tensor 'velocity' fields with units of velocity divided by interfacial-tension gradient.

The third-rank 'stress' field $\boldsymbol{\Pi}$ is given by

$$\boldsymbol{\Pi}=-\boldsymbol{I}\boldsymbol{P}+\mu[\nabla\boldsymbol{V}+{}^{\dagger}(\nabla\boldsymbol{V})]. \tag{19}$$

In (18c) and (19), standard conventions apply in defining the pre and post transpose of a triadic (Brenner 1964a) and $\boldsymbol{I}$ is the idemfactor. A similar result may be written for $\hat{\boldsymbol{\Pi}}$.

Recasting the tangential-stress balance in (18c) in a slightly different form reveals the intrinsic nature of the problem for the tensor fields $\boldsymbol{V}$ and $\hat{\boldsymbol{V}}$:

$$(\boldsymbol{n}\cdot\boldsymbol{\Pi}^{\dagger}-\boldsymbol{n}\cdot\hat{\boldsymbol{\Pi}}^{\dagger})\cdot\boldsymbol{t}=-\boldsymbol{I}\cdot\boldsymbol{t}. \tag{20}$$

The 'motion' arising from the action of (20) is driven by a tangential 'stress' imbalance given by an 'interfacial-tension gradient' which is the idemfactor. The analogy to Brenner's (1964a) formulation of problems of Stokes flow past rigid bodies of arbitrary shape is clear. There, Brenner defines second-rank tensor 'velocity' fields arising from a 'velocity' tensor equal to the idemfactor at the surface of the object.

It now remains to solve (17) and (18) for the fields $\boldsymbol{V}$ and $\hat{\boldsymbol{V}}$. This may be done most conveniently by decomposition of these fields into Cartesian components. We may write

$$\boldsymbol{V}=\boldsymbol{v}^{(1)}\boldsymbol{i}+\boldsymbol{v}^{(2)}\boldsymbol{j}+\boldsymbol{v}^{(3)}\boldsymbol{k}, \tag{21}$$

where $(\boldsymbol{i},\boldsymbol{j},\boldsymbol{k})$ are the usual Cartesian base vectors. Similar results may be written for the fields $\boldsymbol{P}$ and $\boldsymbol{\Pi}$, and for the appropriate counterparts within the droplet. Substitution in (17)–(19) leads to 'unit' problems for each of the vector fields $\boldsymbol{v}^{(j)}$. As an example, the problem for $\boldsymbol{v}^{(1)}$ is given below:

$$\left.\begin{aligned} \nabla\cdot\boldsymbol{v}^{(1)}=0, \quad & \nabla\cdot\hat{\boldsymbol{v}}^{(1)}=0, \\ \mu\nabla^2\boldsymbol{v}^{(1)}=\nabla p_1, \quad & \hat{\mu}\nabla^2\hat{\boldsymbol{v}}^{(1)}=\nabla\hat{p}_1, \end{aligned}\right\} \tag{22}$$

$$\boldsymbol{v}^{(1)}\rightarrow 0 \quad \text{as } r\rightarrow\infty, \tag{23a}$$

$$\hat{\boldsymbol{v}}^{(1)}\text{ is bounded} \quad \text{at } r=0, \tag{23b}$$

$$\left.\begin{aligned} \boldsymbol{v}^{(1)}=\hat{\boldsymbol{v}}^{(1)},\quad \boldsymbol{n}\cdot\boldsymbol{v}^{(1)}=\boldsymbol{n}\cdot\hat{\boldsymbol{v}}^{(1)}=0, \\ \boldsymbol{n}\cdot\boldsymbol{\pi}^{(1)}\cdot\boldsymbol{t}=-(\boldsymbol{t}\cdot\boldsymbol{i})+\boldsymbol{n}\cdot\hat{\boldsymbol{\pi}}^{(1)}\cdot\boldsymbol{t} \end{aligned}\right\} \text{ on } S_a. \tag{23c}$$

Here

$$\boldsymbol{\pi}^{(1)}=-\boldsymbol{I}p^{(1)}+\mu(\nabla\boldsymbol{v}^{(1)}+\nabla\boldsymbol{v}^{(1)\dagger}), \tag{24}$$

and a similar result may be written for $\hat{\boldsymbol{\pi}}^{(1)}$. The above equations describe the physical problem of a droplet held fixed in an unbounded fluid with a special distribution of the interfacial-tension gradient. This quantity is prescribed here as the tangential component of a unit vector which remains constant in direction everywhere on the droplet surface. In this case, this unit vector is parallel to the x-direction. In fact, a moment's reflection will convince one that the vector components of $\boldsymbol{V}$ and $\hat{\boldsymbol{V}}$ in any given spatial direction will satisfy a similar problem wherein the interfacial-tension gradient is the tangential component of a constant unit vector pointing in that direction.

Defining spherical polar coordinates (r, θ, ϕ) centred in the droplet, if the polar angle θ is measured from the (positive) x-direction, it is clear that the above $\nabla_s \sigma$ field on S_a will result in axisymmetric motion. The problem may be solved in a straightforward manner using general solutions of the Stokes equations given in Happel & Brenner (1965). Of particular interest here are the velocity components which are given below; for convenience, the superscript (1) has been omitted, and the radial coordinate is scaled by the sphere radius:

$$v_r = v_0\left(\frac{1}{r}-\frac{1}{r^3}\right)\cos\theta, \tag{25a}$$

$$v_\theta = -\frac{v_0}{2}\left(\frac{1}{r}+\frac{1}{r^3}\right)\sin\theta, \tag{25b}$$

$$\hat{v}_r = v_0(r^2-1)\cos\theta, \tag{25c}$$

$$\hat{v}_\theta = v_0(1-2r^2)\sin\theta. \tag{25d}$$

v_0 is given by

$$v_0 = \frac{a}{3\mu(1+\alpha)}, \tag{25e}$$

and the viscosity ratio α has been defined earlier. At the surface of the droplet, $r = 1$, $v_r = \hat{v}_r = 0$, and

$$\boldsymbol{v}^{(1)}|_{r=1} = -(v_0 \sin\theta)\,\boldsymbol{i}_\theta, \tag{26}$$

where the superscript (1) has been reintroduced.† Similar results for $\boldsymbol{v}^{(2)}$ and $\boldsymbol{v}^{(3)}$ may be written in suitable spherical polar coordinates. Thus, one finally arrives at the following representation of the tensor field $\boldsymbol{V}$ on S_a:

$$\frac{1}{v_0}\boldsymbol{V}\Big|_{\text{on } S_a} = \boldsymbol{W} = -\begin{pmatrix} -1+x^2 & xy & xz \\ xy & -1+y^2 & yz \\ xz & yz & -1+z^2 \end{pmatrix}. \tag{27}$$

Here, (x, y, z) are Cartesian coordinates of points on S_a scaled using the droplet radius, and, for convenience, a matrix representation is used on the right-hand side.

6. Properties of the tensor field $\boldsymbol{W}$ on S_a

$\boldsymbol{W}$, defined on S_a, is a symmetric tensor field. Thus, at every point on S_a, $\boldsymbol{W}$ possesses three orthogonal principal directions, and will be of diagonal form when represented in this privileged coordinate system at that point. It is quite straightforward to establish that one of these directions is normal to the spherical surface S_a, and hence is radial. The corresponding eigenvalue $\lambda_1 = 0$. This means that $\boldsymbol{W}$ operating on spatial-vector fields defined on S_a will annihilate their radial component.

The other two eigenvalues of $\boldsymbol{W}$ are both equal to unity, $\lambda_2 = \lambda_3 = 1$. Thus, $\boldsymbol{W}$

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† In (26), $\boldsymbol{i}_\theta$ is the unit vector in the θ direction.

maps a spatial vector lying in the tangent plane anywhere on S_a into itself. Due to the equality of λ_2 and λ_3, there is degeneracy of the other two principal directions, both lying on the tangent plane. Thus, any two orthogonal directions on that plane may be chosen as the principal directions. It is convenient to choose these as the θ and ϕ directions in a spherical polar coordinate system (r, θ, ϕ) centred in the droplet with the polar angle θ measured from any fixed, but arbitrary, spatial direction. In such a system, $\boldsymbol{W}$ has an extremely simple representation:

$$\boldsymbol{W} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \tag{28}$$

It is clear that $\boldsymbol{W}$ is merely a projection tensor mapping spatial vector fields defined on S_a into their projections in the tangent plane. It is quite likely that the high degree of symmetry exhibited by $\boldsymbol{W}$ is due to the symmetry in the problem for $\boldsymbol{V}$ induced by (20).

7. The force on the droplet

Now, one may insert the result for the tensor field $\boldsymbol{V}_1$ in terms of $\boldsymbol{W}$ from (27) into (14) for the hydrodynamic force on the droplet. Since $\boldsymbol{W}$ is a projection tensor, and $\nabla_s \sigma$ already lies in the tangent plane, the inner product with $\boldsymbol{W}$ leaves this vector unaffected. Using the definition of v_0 from (25*e*), and finally dropping the unnecessary subscript 2, we obtain the following principal result from this work:

$$\boldsymbol{F} = -\frac{1}{2(1+\alpha)} \int_{S_a} \nabla_s \sigma \, \mathrm{d}S. \tag{29}$$

Equation (29) is worthy of some comment. First, under the stated assumptions, the hydrodynamic force on a droplet is independent of the viscosity of either phase, depending instead only on the ratio of viscosities. Second, the force is directly proportional to the discontinuity in tangential stress across the interface integrated over the drop surface. Third, the force may be calculated without solving the detailed hydrodynamic problem, knowing only the distribution of the $\nabla_s \sigma$ field on S_a.

8. Some extensions and concluding remarks

So far, no assumptions have been necessary regarding the cause of the variation of interfacial tension on the droplet surface. This variation may result from temperature changes around the droplet induced by a non-uniform temperature distribution in the continuous phase. Or, in multicomponent systems, variations in individual species concentrations around the periphery of the droplet can give rise to $\nabla_s \sigma \neq \mathbf{0}$.

Typically, a droplet is inserted into a continuous-phase fluid possessing some arbitrary, but known, non-uniform temperature or concentration field. However, the distribution of temperature or concentration on the interface which determines the function $\nabla_s \sigma$ may not be known. Thus, it is worthwhile to explore the possibility of writing results similar in spirit to Faxén's force law in these situations. For simplicity, the case of temperature fields is discussed in detail below, but final results are given for the cases of both temperature and concentration fields.

It is assumed that the continuous-phase fluid in the absence of the droplet is quiescent and possesses a known temperature field $T_\infty(\boldsymbol{r})$. Introducing the droplet will disturb this distribution and result in temperature fields $T(\boldsymbol{r})$ and $\hat{T}(\boldsymbol{r})$ outside and within the droplet respectively. If it is assumed that $\mathrm{d}\sigma/\mathrm{d}T = \sigma'$ is constant over the range of temperatures encountered on the surface of the droplet, (29) may be rewritten as follows:

$$\boldsymbol{F} = -\frac{\sigma'}{2(1+\alpha)}\int_{S_a} \boldsymbol{\nabla}_s T\,\mathrm{d}S. \tag{30}$$

The motion generated by the interface can, in general, influence the temperature distributions within and outside the droplet. However, if we make the further assumption that the Péclet numbers for heat transfer within and outside the droplet are negligible, the convective heat-transport terms in the energy equations may be neglected in comparison to the conduction terms. In the absence of sources or sinks, then T and $\hat{T}$ will satisfy Laplace's equation, and the usual conditions of continuity of the temperature and normal energy flux are satisfied on S_a. To avoid diversion from the theme of this section, the following useful result is stated here (the derivation of this result via spherical harmonics solutions for T and $\hat{T}$ is relegated to the Appendix):

$$\int_{S_a} \boldsymbol{\nabla}_s T\,\mathrm{d}S = \frac{8\pi a^2}{2+\beta}(\boldsymbol{\nabla} T_\infty)_0. \tag{31}$$

Here, the subscript 0 stands for evaluation at the location of the droplet centre and $\beta = \hat{k}/k$ is the ratio of the thermal conductivity of the droplet phase to that of the continuous phase as defined in the introduction.

Thus, the following result may be written for the Stokes force on a fluid droplet inserted and held fixed in a continuous-phase fluid which, in the undisturbed state, is quiescent, and possesses an arbitrary harmonic temperature field $T_\infty(\boldsymbol{r})$:

$$\boldsymbol{F} = -\frac{4\pi a^2\sigma'}{(1+\alpha)(2+\beta)}(\boldsymbol{\nabla} T_\infty)_0. \tag{32}$$

The result is further restricted by the requirement that the temperature fields within and outside the droplet continue to be harmonic after insertion of the droplet. The result may be considered a useful first approximation for the case of small values of the Péclet numbers within and outside the droplet.

When the capillary force arises due to a gradient in concentration of a species which is transferred across the interface, the result for the force is modified slightly:

$$\boldsymbol{F} = -\frac{4\pi a^2\sigma'}{(1+\alpha)(2+K\beta)}(\boldsymbol{\nabla} C_\infty)_0. \tag{33}$$

Here, $C_\infty(\boldsymbol{r})$ is the harmonic concentration field in the undisturbed continuous phase fluid, and $\sigma' = \mathrm{d}\sigma/\mathrm{d}C$. K is an equilibrium constant, being the ratio of the concentration in the droplet phase to that in the continuous phase at the interface S_a. In this case, $\beta = \hat{D}/D$ is the ratio of diffusivities of the solute in the droplet and continuous phases.

When the undisturbed continuous phase is in Stokes flow with an arbitrary $\boldsymbol{u}_\infty(\boldsymbol{r})$ and the droplet translates with a velocity $\boldsymbol{U}$, a result may be given for the hydrodynamic force on the droplet by superposition. Since the temperature fields still have to be harmonic, it has to be assumed that convective transport of energy still is negligible compared to conduction. In this case, one can add the results for the force from (32) and (1) to arrive at the following final result:

$$\boldsymbol{F} = \frac{2\pi a}{1+\alpha}\left[-\frac{2a\sigma'}{2+\beta}(\nabla T_{\infty})_0 + \mu(2+3\alpha)\{(\boldsymbol{u}_{\infty})_0 - \boldsymbol{U}\} + \hat{\mu}\frac{a^2}{2}(\nabla^2 \boldsymbol{u}_{\infty})_0\right]. \tag{34}$$

The result for $C_{\infty}(\boldsymbol{r})$ is analogous and, therefore, omitted.

The analogy of (32) to (2) is evident. In fact, (2) is a special case of the more general result when $\nabla T_{\infty}(\boldsymbol{r})$ is simply a constant vector everywhere. It is interesting to note that, guided purely by intuition, (34) and variants thereof have been used without proof by this author and his collaborators in constructing elementary approximations in capillary problems. Examples may be found in Meyyappan, Wilcox & Subramanian (1983) and Meyyappan & Subramanian (1984).

One might note that in this section attention has been restricted to *linear* problems for the temperature (and concentration) fields. In such cases, as pointed out by Acrivos (personal communication, 1984), the isotropy of the sphere can be invoked to show that the force on the sphere can only depend linearly on $(\nabla T_{\infty})_0$ when T and $\hat{T}$ satisfy Laplace's equation. This will lead immediately to (32) or the analogous (33) since the constant of proportionality between $\boldsymbol{F}$ and $(\nabla T_{\infty})_0$ can be obtained from the known solution of Young *et al.* for the case of a constant ∇T_{∞} field. Of course, the more general (29) or (30) are not restricted by the requirement that the T and $\hat{T}$ fields satisfy linear equations in general, or Laplace's equation in particular.

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Appendix

Here, (31) is derived for an arbitrary harmonic field $T_{\infty}(\boldsymbol{r})$, given that T and $\hat{T}$ are harmonic functions. The field $T_{\infty}(\boldsymbol{r})$ possesses no singularities in the region of space occupied by the droplet. Therefore, in a spherical polar coordinate system (r, θ, ϕ) centred in the droplet, with θ being the polar angle measured from some fixed but arbitrary direction, one can write, quite generally,

$$T_{\infty}(\boldsymbol{r}) = \sum_{n=0}^{\infty}\left(\frac{r}{a}\right)^n F_n^m(\theta, \phi). \tag{A 1}$$

Here, the surface spherical harmonic $F_n^m(\theta, \phi)$ is defined as follows:

$$F_n^m(\theta, \phi) = \sum_{m=0}^{n}(C_n^m \cos m\phi + D_n^m \sin m\phi)\, T_n^m(s). \tag{A 2a}$$

Here,
$$s = \cos\theta. \tag{A 2b}$$

The Ferrer functions $T_n^m(s)$ are defined in MacRobert (1967), and are related to the Associated Legendre functions by a complex multiplicative constant. Any general solution of Laplace's equation in a domain *including the origin* may be written in the form of (A 1) and the constants C_n^m and D_n^m may be determined by the use of standard orthogonality relationships.

The usual problem for harmonic T and $\hat{T}$ fields is as follows. Here, to avoid repetition, the slightly more general problem for species concentration is posed. At the spherical boundary S_a of the droplet, equilibrium is assumed between phases, and it is required that the normal flux of species across the inte continuous (normal flux of energy if T is temperature):

$$\nabla^2 T = 0, \quad \nabla^2 \hat{T} = 0, \tag{A 3}$$

$$T \rightarrow T_\infty(\boldsymbol{r}) \quad \text{as } r \rightarrow \infty, \tag{A 4a}$$

$$\hat{T} \text{ is bounded} \quad \text{at } r = 0, \tag{A 4b}$$

$$\left.\begin{aligned} KT &= \hat{T}, \\ \frac{\partial T}{\partial r} &= \beta \frac{\partial \hat{T}}{\partial r} \end{aligned}\right\} \quad \text{on } r = 1. \tag{A 4c}$$

In (A 4c), if T is the temperature field, $K \equiv 1$, and $\beta = \hat{k}/k$ would be the ratio of thermal conductivities. If T is the species concentration, K would be an equilibrium constant and $\beta = \hat{D}/D$ would be the ratio of diffusivities.

The solutions of (A 3)–(A 4) may be represented in a straightforward manner in spherical harmonics. When account is taken of all the boundary conditions, recognizing that $T_\infty(\boldsymbol{r})$ satisfies Laplace's equation, one obtains:

$$T(\boldsymbol{r}) = T_\infty(\boldsymbol{r}) + \sum_{n=0}^{\infty} \left(\frac{a}{r}\right)^{n+1} G_n^m(\theta, \phi), \tag{A 5}$$

$$\hat{T}(\boldsymbol{r}) = K\left\{T_\infty(\boldsymbol{r}) + \sum_{n=0}^{\infty} \left(\frac{r}{a}\right)^{n} G_n^m(\theta, \phi)\right\}, \tag{A 6}$$

where
$$G_n^m = \sum_{m=0}^{n} (A_n^m \cos m\phi + B_n^m \sin m\phi)\, T_n^m(s) \tag{A 7}$$

also are spherical harmonics.

The constants A_n^m and B_n^m are related in a simple manner to C_n^m and D_n^m:

$$\begin{Bmatrix} A_n^m \\ B_n^m \end{Bmatrix} = \frac{n(1-K\beta)}{n(1+K\beta)+1} \begin{Bmatrix} C_n^m \\ D_n^m \end{Bmatrix}. \tag{A 8}$$

It is a straightforward matter now to establish the following result by direct integration:

$$\begin{aligned} \int_{S_a} \boldsymbol{\nabla}_{\mathrm{s}} T \,\mathrm{d}S &= -\tfrac{8}{3}\pi a[(A_1^1 + C_1^1)\,\boldsymbol{i} + (B_1^1 + D_1^1)\,\boldsymbol{j} - (A_1^0 + C_1^0)\,\boldsymbol{k}] \\ &= -\frac{8\pi a}{(2+K\beta)}\,[C_1^1\,\boldsymbol{i} + D_1^1\,\boldsymbol{j} - C_1^0\,\boldsymbol{k}]. \end{aligned} \tag{A 9}$$

Here, $(\boldsymbol{i}, \boldsymbol{j}, \boldsymbol{k})$ are the usual base vectors in a rectangular Cartesian coordinate system which shares the same origin as the spherical polar coordinate system chosen earlier. The vector $\boldsymbol{k}$ is the unit vector along the z-axis which is the axis from which the polar angle θ is measured.

It also is straightforward to establish that

$$a(\boldsymbol{\nabla} T_\infty)_0 = -[C_1^1\,\boldsymbol{i} + D_1^1\,\boldsymbol{j} - C_1^0\,\boldsymbol{k}]. \tag{A 10}$$

Equation (31) follows directly from equations (A 9) and (A 10), upon setting $K = 1$.

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