

## HYDRODYNAMIC INTERACTION OF A SMALL FLUID PARTICLE AND A SPHERICAL DROP IN LOW-REYNOLDS NUMBER FLOW

J. A. STOOS,† S.-M. YANG‡ and L. G. LEAL

Department of Chemical and Nuclear Engineering, University of California, Santa Barbara,  
CA 93106, U.S.A.

(Received 15 November 1991; in revised form 15 August 1992)

**Abstract**—The present study is concerned with estimating the hydrodynamic interactions between a small droplet and a much larger fluid drop when both drops are translating through an otherwise quiescent fluid. The method of solution is a matched asymptotic expansion involving resolution of the local undisturbed flow produced by the motion of the large drop into component flows that provide the far-field boundary conditions for calculating the disturbance flows produced by the small droplet. In the limit of very small size ratio, the surface of the large drop appears as locally planar. The theory yields a complete trajectory equation including a proper description of the effect of hydrodynamic interactions between the two neighboring drops. The trajectory of the small droplet on approaching the large drop does not deviate significantly from the streamlines of the undisturbed flow until it comes within range of the hydrodynamic repulsion from the surface of the large drop. The magnitude of hydrodynamic repulsion becomes weaker as the viscosity of the droplet is reduced, and this effect is a strong function of the separation distance from the surface of the large drop.

*Key Words:* flotation, particle interactions, creeping flow

### 1. INTRODUCTION

In this paper we consider the dynamics of a *small fluid* particle which moves freely in the slow streaming motion past a nearby much *larger* drop, for the general case in which all three fluids are different, see figure 1. Interest in this problem stems mainly from its role as a simple model problem relevant to the collection of very small (solid or fluid) particles at the surface of larger bubbles or drops in flotation processes (cf. Goldman *et al.* 1967a, b; Goren & O'Neill 1971; Spielman 1977; Dukhin & Rulev 1977; Stoos 1987; Stoos & Leal 1989; and references therein). It is also relevant to some aspects of drop coalescence (cf. Burrill & Woods 1973; Jones & Wilson 1978; Chen *et al.* 1984).

Nearly all previous attempts to incorporate hydrodynamic interactions into particle trajectory calculations for the flotation problem have relied on solutions for two rigid, no-slip spheres. The majority have followed the formulation of Goren & O'Neill (1971), in which it is assumed that the ratio of radii is  $a/A \ll 1$ , and thus rely on solutions for the interactions between a *rigid* sphere and a rigid plane wall (e.g. Spielman & Fitzpatrick 1973; Prieve & Ruckenstein 1974; Derjaguin *et al.* 1976). A more accurate formulation for two *solid* spheres of arbitrary size ratio has recently become possible, due to Jeffery & Onishi (1984), who developed comprehensive results for the full resistance and mobility tensors. Nevertheless, and in spite of the fact that very small bubbles tend to rise with the same terminal velocity as a solid sphere of the same size and density, there are many indications that the collector bubble does not behave as a solid sphere in the flotation process, especially in the final stages of the particle capture process. Among these are the observed *mobility* of particles that have been captured at the surface of such a bubble, and the expected deformation of the interface when the *separation* distance is small. One may also cite the measurements by Yuu & Fukui (1981) of the drag coefficient for a sphere approaching a fluid interface which showed significant deviation from the case of a sphere moving toward a solid wall. Finally, in the case of flotation or liquid–solid extraction processes (Puddington & Sparks 1975), where a liquid drop is

†,‡Present addresses: †Mobil Research and Development Corporation, Paulsboro, NH 08066, U.S.A. and ‡Department of Chemical Engineering, KAIST, Taejon 305-701 Korea.

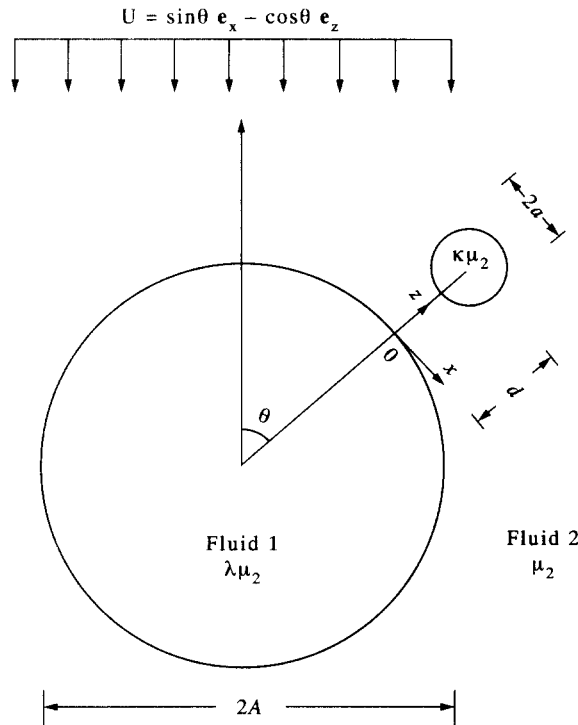


Figure 1. Schematic of the problem and the coordinate system.

used as the collector instead of a bubble, the drop is generally larger than in flotation, and thus may not behave as a solid sphere even in isolated motions.

Although there have been a number of prior studies of the relative motions of two drops in a viscous fluid, none is well-suited for use in modeling the hydrodynamic aspects of particle capture in flotation (or other) processes where there is a large difference in the size of the particle and the collector. The first comprehensive study of the relative motions of two spherical drops was due to Hetsroni & Haber (1978), who used the method of reflections in conjunction with eigenfunction expansions in bispherical coordinates to consider two arbitrarily sized but widely separated drops. Although this analysis is undoubtedly valuable for many applications, it is *not* well-suited to the case when one of the drops is very much smaller. This is primarily because Hetsroni & Haber (1978) considered approximations in which both of the parameters  $\alpha = a/(A + d)$  and  $\beta = A/(A + d)$  are assumed to be asymptotically small (here,  $d$  is the separation distance between drops, while  $a$  and  $A$  are the two radii). This analysis is best suited to  $A = O(a)$ , and obviously *loses accuracy* when, as in the present problem,  $A \gg a$ . This, and the limitation to large separations, are the primary factors which diminish the usefulness of Hetsroni & Haber's (1978) theory for application to the analysis of particle capture in the flotation process. However, several other restrictions are worth noting. First, the relation between the torque on the particle and its translational velocity does not reduce to the correct form in the limit as either particle becomes a solid (i.e. the particle translates without rotation), and this will affect the predicted trajectory for the small particle. Second, the relations describing the various resistance functions in Hetsroni & Haber (1978) are truncated at different orders,  $O(\alpha^m \beta^n)$ ,  $m + n = 3, 4, 5$ . Further, Stoops' (1987) comparison with the results of Jeffery & Onishi (1984)—described below—found that the truncated terms of  $O(\beta^m)$  are not small and significantly affect the particle trajectory. Finally, Hetsroni & Haber's results are algebraically complex, partly because they did not specifically consider the asymptotic limit  $a/A \ll 1$  in the problem *formulation*.

Recently, Fuentes *et al.* (1988, 1989) reconsidered the same asymptotic expansion scheme employed by Hetsroni & Haber (1978), but used fundamental solutions for a point force near a spherical drop, e.g. Stokeslets and their higher order moments, to extend the domain of applicability from large separations, to include all configurations except nearly touching spheres. Although the expansion scheme is best suited to cases where  $A = O(a)$ , as mentioned earlier,

Fuentes *et al.* (1988, 1989) were able to use their general solution in the limit  $a/d \ll 1$  to evaluate the *mobility* function (*only*) for translation of a spherical drop through a *quiescent* fluid near a plane interface. Their results for this special case are identical to those of Yang & Leal (1990), who used the same solution procedure as is followed here. In the present paper, we follow the direct and simpler approach [pioneered for two solid spheres by Goren & O'Neill (1971)] of introducing the limit  $a \ll A$  directly into the formulation, rather than approximating the complete solution of the full problem. Apart from the work reported here, there has been no prior analysis that is specifically directed at accurate results for hydrodynamic interactions of a very small particle or drop and a much larger bubble or drop.

In the present paper, we calculate trajectories for a very small *spherical* (solid or fluid) particle of radius,  $a$ , immersed freely in a slow streaming flow that is created by the motion of a much larger *spherical* fluid collector of radius,  $A$ . Following the precedent of Goren & O'Neill (1971), we evaluate the purely hydrodynamic interaction between the two neighboring spherical drops by constructing a rigorous asymptotic solution for the disturbance flow created by the small fluid particle in proximity to the larger drop for the limit  $\delta (\equiv a/A) \ll 1$ . Since the asymptotic solution in this limit is singular, we use the method of *matched* asymptotic expansions. The *outer* problem corresponds to resolution on the scale,  $A$ , in which the small particle to be collected appears only as a weak *point* disturbance very near (or at) the surface of the large drop. The *inner* problem, on the other hand, corresponds to resolution on the scale,  $a$ . On this scale, the undeformed surface of the large drop appears planar, and the outer flow provides the far-field boundary conditions through matching to describe the flow in the vicinity of the small particle. In particular, the outer *undisturbed* flow past the large drop, expanded about the point on the drop surface that is closest to the particle, contributes uniform, linear, quadratic and higher order flows as the far-field boundary conditions for the inner problem. In the present theory, we neglect deformation of the large drop both on the outer and inner scale of resolution. This approximation constitutes a very significant simplification because one can then use *superposition* of the results for the various undisturbed outer flow components to construct an analytic expression for the trajectories of relative motion.

We adopt the approach outlined above because it reduces the two-sphere problem to a series of problems involving the simpler geometry of a sphere near an infinite plane interface, and thus leads to relatively simple and accurate formulae compared to those that can be achieved via approximations of the "exact" solutions of the two spherical drop problem. However, in contrast to the solid collector case studied by Goren & O'Neill (1971), we must solve a *much* larger number of component problems for the present case of a *fluid* collector. Of course, an *exact* analytic solution is possible for each component problem using general eigenfunction expansions in bipolar spherical coordinates. However, this would defeat the purpose of Goren & O'Neill's asymptotic formulation procedure because the resulting solution forms would be too complex and unwieldy for use in trajectory calculations (note that each coefficient corresponding to an eigenfunction would have to be determined numerically at all times as the relative position and orientation of the two drops change). An attractive alternative which we pursue here is solution of the various component flow problems via a particular version of the *method of reflections* technique that was developed by Lee *et al.* (1979) and Yang & Leal (1990) using the fundamental solutions for point and higher order singularities near a plane fluid interface.

Part of the motivation for the present study is an evaluation of the difference in hydrodynamic interactions when the collector is treated as a bubble or drop, rather than as a solid sphere. Another motivation is the development of accurate and easily applied approximation techniques for evaluation of particle trajectories in the vicinity of a bubble/drop collector. Finally, it is important to develop approximation methods that are easily generalizable. For example, Anfruns & Kitchener (1977) demonstrated that collection efficiencies may vary greatly for different shape particles, i.e. rod-like particles vs spherical particles. Therefore, a solution technique that could easily be applied to nonspherical particles is needed, and that is true of the method developed here.

## 2. PROBLEM FORMULATION

We begin by considering the governing equations and boundary conditions for two spherical drops (which experience hydrodynamic interactions) for buoyancy-driven motions through an

otherwise quiescent immiscible fluid in an unbounded domain, as shown in figure 1. The three fluids are all assumed to be incompressible and Newtonian. The whole motion is further assumed to be dominated by viscous and pressure effects, so that the inertial terms in the equations of motion can be neglected entirely. In addition, we choose a coordinate reference system with an origin that is fixed, for convenience, at point O on the surface of the larger drop, thus translating relative to a fixed reference frame with a terminal velocity  $\mathbf{U} = -U_1(\sin \theta \mathbf{e}_\theta - \cos \theta \mathbf{e}_r)$  in the direction opposite to the gravity. We specialize our coordinate system by taking the centers of the two drops on the  $x$ - $z$  plane, i.e. the plane of the paper (see figure 1). The coordinate system is fixed on the larger-drop surface such that the  $z$ -axis points along the line of centers and the  $y$ -axis points into the plane of the paper, i.e. the base vectors  $(\mathbf{e}_z, \mathbf{e}_x, \mathbf{e}_y)$  in this local Cartesian coordinate system are identical to the base vectors  $(\mathbf{e}_r, \mathbf{e}_\theta, \mathbf{e}_\phi)$  in spherical polar coordinates, respectively. The orientation of the small drop relative to the larger drop can be specified in terms of the angle  $\theta$  measured between the line of centers (i.e. the  $z$ -axis) and the vertical. In order to write the governing differential equations and boundary conditions in nondimensional form, we use a characteristic length  $l_c$  and a characteristic velocity  $u_c = U_1$ , where  $l_c$  will be defined shortly. The stress tensors for the three fluids are nondimensionalized using  $p_c = \mu_2 U_1 / l_c$ . The position vector at a material point, measured relative to the origin, will be denoted as  $\mathbf{x}$ . With these conventions and assumptions, the equations of motion may be written in the familiar form

$$\nabla \cdot \mathbf{u}_i = 0, \quad \nabla \cdot \boldsymbol{\sigma}_i = 0; \quad [1]$$

with the stress  $\boldsymbol{\sigma}_i$  and pressure  $p_i$  given by

$$\boldsymbol{\sigma}_i = -p_i \mathbf{I} + \frac{\mu_i}{\mu_2} (\nabla \mathbf{u}_i + \nabla \mathbf{u}_i^T) \quad (i = 1, 2 \text{ and } 3), \quad [2]$$

in which  $\mu_i$  is the viscosity of fluid  $i$  and  $\mathbf{u}_i$  denotes the velocity field in fluid  $i$ . The boundary conditions in the moving frame of reference are

$$\mathbf{u}_2 \rightarrow \sin \theta \mathbf{e}_x - \cos \theta \mathbf{e}_z \quad \text{as } |\mathbf{x}| \rightarrow \infty \quad [3]$$

plus the interface conditions at the surfaces of the two neighboring drops, i.e. continuity of tangential velocity and stress, and zero normal velocity. Although the analysis will be carried out for two drops, it is convenient to refer to the smaller drop as a "particle" and the larger drop as the "drop", and we will follow this convention in the subsequent portions of the paper.

The operating characteristics of effluent flotation warrant several approximations that greatly simplify the analysis. Because of the very small particles generally encountered in effluent flotation, we will consider the asymptotic case in which the particle radius is very much smaller than the radius of the collector drop; i.e.  $\delta = a/A \ll 1$ . Further, the drop-particle separation,  $d$ , is assumed to be intermediate between the length scales  $A$  and  $a$ , so that  $A \gg d > a$ . Although the method of solution that we adopt for the component problems is strictly valid only for  $d \gg a$ , we will apply these solutions to problems where  $d \approx O(a)$ , and compare with exact solutions where they are available as a check on the accuracy of this *ad hoc* procedure. The particle's disturbance to the uniform flow should be small compared to the drop's disturbance, except locally in the vicinity of the particle, for the case  $(a/A) \ll 1$ . In the region close to the particle, however, the particle's effect on the flow obviously cannot be neglected. Since the boundary conditions must be satisfied on the particle's surface, the streamlines are different than they would be if the particle were not present. This local disturbance flow is significant over a distance from the particle of  $O(a)$ , but when the drop-particle separation is also of  $O(a)$ , interaction of this disturbance flow with the drop surface can significantly alter the particle trajectory. Clearly, the magnitude of this interaction will depend on the viscosity of each phase, both through the dependence of the magnitude of the "reflection" of the disturbance from the interface, and through the dependence of the form of the streaming flow around the drop.

Therefore, at least two characteristic length scales are involved in this process. The first,  $a$ , is a measure of the distance over which  $O(1)$  variations in the disturbance flow occur because of the presence of the particle. We shall call the region where this length scale applies the "inner" region,  $l_c = O(a)$ . The second,  $A$ , is a measure of the distance over which  $O(1)$  variations in the flow occur in the "outer" region where  $l_c = A$ , i.e. everything outside the immediate  $O(a)$  vicinity of the

particle. Hydrodynamic interactions, electroviscous interactions and interface deformation may be especially important in the “inner” region. In the present work, we will focus on the hydrodynamic interactions.

The existence of two characteristic length scales and the small parameter  $\delta = a/A \ll 1$  suggests the utility of applying the method of matched asymptotic expansions to solve the problem. Thus, if we begin with the “outer” region, characterized by the scale  $A$ , we may generally expect expansions in the following forms for the velocity and stress tensor, both inside and outside the drop:

$$\tilde{\mathbf{u}}_j = \tilde{\mathbf{u}}_j^{(0)} + \delta \tilde{\mathbf{u}}_j^{(1)} + \delta^2 \tilde{\mathbf{u}}_j^{(2)} + \dots \quad [4]$$

and

$$\tilde{\boldsymbol{\sigma}}_j = \tilde{\boldsymbol{\sigma}}_j^{(0)} + \delta \tilde{\boldsymbol{\sigma}}_j^{(1)} + \delta^2 \tilde{\boldsymbol{\sigma}}_j^{(2)} + \dots, \quad [5]$$

where  $\sim$  denotes the variable in the outer region, and the subscript  $j$  is taken to be 1 inside the drop and 2 outside the drop. The first terms in these expansions represent the “undisturbed” Hadamard–Rybczynski solution for streaming flow past the large drop, in the complete absence of the small particle, which appears from this outer view simply as a point force located on the surface of the drop. Subsequent terms represent the disturbance of the outer flow due to the particle.

In order to determine which of the terms in these expansions are expected to be nonzero in the limit  $\delta \rightarrow 0$ , we can apply a simple force balance to show that the disturbance flow due to the small particle is actually negligible at the first several orders of approximation in the *outer* region, relative to the undisturbed (Hadamard–Rybczynski) flow produced by the larger drop. The balance between the force acting on the fluid because of the buoyancy of the particle and the hydrodynamic force exerted by the fluid on the particle is given by

$$\frac{4}{3}\pi a^3(\rho_3 - \rho_2)\mathbf{g} + \mu U_1 A \int_{S_p} (\tilde{\boldsymbol{\sigma}}_2 \cdot \mathbf{n}_p) dS = 0. \quad [6]$$

Here  $\rho_j$  is the density of fluid  $j$ ,  $S_p$  indicates that the integration is to be performed over the surface of the particle,  $\mathbf{n}_p$  is the unit normal to the particle surface and  $dS$  is a nondimensionalized differential surface element. Similarly, for the large drop,

$$\frac{4}{3}\pi A^3(\rho_1 - \rho_2)\mathbf{g} + \mu U_1 A \int_{S_d} \tilde{\boldsymbol{\sigma}}_2 \cdot \mathbf{n}_d dS = 0, \quad [7]$$

where  $S_d$  indicates integration over the surface of the drop and  $\mathbf{n}_d$  is the unit normal to the drop surface. The ratio of the force exerted on the fluid by the small particle relative to that by the larger drop is therefore

$$\frac{F_p}{F_d} = \frac{a^3(\rho_3 - \rho_2)}{A^3(\rho_1 - \rho_2)} \approx O(\delta^3). \quad [8]$$

Thus, to  $O(\delta^3)$  the force exerted by the particle on the fluid is negligible in its effect on the flow in the “outer” region so that, at least to  $O(\delta^2)$ , the flow in the “outer” region should reduce to streaming flow around a drop with no particle present. Thus, the Hadamard–Rybczynski solution  $\mathbf{U}_j^{\text{HR}}$  ( $j = 1, 2$ ) provides the velocity fields inside ( $j = 1$ ) and outside ( $j = 2$ ) the drop for the “outer” region, through terms of  $O(\delta^2)$ .

Now the inner problem must be considered where the characteristic length scale is  $l_c = a$ . Since we are interested in the case for  $d \approx O(a)$ , where hydrodynamic interactions play a role, the “inner” region is assumed to be composed of sections both inside and outside the drop. Analogous to [4] and [5], the following “inner” domain velocity and stress tensor expansions are assumed:

$$\mathbf{u}_j = \mathbf{u}_j^{(0)} + \delta \mathbf{u}_j^{(1)} + \delta^2 \mathbf{u}_j^{(2)} + \dots \quad [9]$$

and

$$\boldsymbol{\sigma}_j = \boldsymbol{\sigma}_j^{(0)} + \delta \boldsymbol{\sigma}_j^{(1)} + \delta^2 \boldsymbol{\sigma}_j^{(2)} + \dots \quad [10]$$

Again, we denote the region inside the large drop as  $j = 1$ , and that outside as  $j = 2$ .

The boundary conditions must also be arranged into a suitable form so that the terms at  $O(1)$ ,  $O(\delta)$  and so forth can be isolated. To accomplish this, we note that the equation for the surface of the drop in terms of inner variables is

$$x^2 + y^2 + \left(z + \frac{A}{a}\right)^2 = \left(\frac{A}{a}\right)^2. \quad [11]$$

This yields the following asymptotic expression for the normal to the drop surface:

$$\mathbf{n}_d = \mathbf{e}_z + \delta(x\mathbf{e}_x + y\mathbf{e}_y) - \frac{1}{2}\delta^2(x^2 + y^2)\mathbf{e}_z + O(\delta^3). \quad [12]$$

Then the kinematic condition on the drop surface becomes

$$\begin{aligned} \mathbf{u}_j \cdot \mathbf{n}_d &= \mathbf{u}_j^{(0)} \cdot \mathbf{e}_z + \delta[x\mathbf{u}_j^{(0)} \cdot \mathbf{e}_x + y\mathbf{u}_j^{(0)} \cdot \mathbf{e}_y + \mathbf{u}_j^{(1)} \cdot \mathbf{e}_z] \\ &+ \delta^2[-\frac{1}{2}(x^2 + y^2)\mathbf{u}_j^{(0)} \cdot \mathbf{e}_z + x\mathbf{u}_j^{(1)} \cdot \mathbf{e}_x + y\mathbf{u}_j^{(1)} \cdot \mathbf{e}_y + \mathbf{u}_j^{(2)} \cdot \mathbf{e}_z] \\ &+ O(\delta^3) = 0 \quad \text{at } z = s, \end{aligned} \quad [13]$$

where  $z = s$  corresponds to the drop surface, which, according to [11], is

$$s = -\frac{1}{2}\delta(x^2 + y^2) + O(\delta^3). \quad [14]$$

The right-hand side of [13] contains the parameter  $\delta$  both explicitly and implicitly, since all the velocities are evaluated at  $z = s$ , which is itself a function of  $\delta$ . As in Van Dyke (1975), we perform a ‘‘domain perturbation’’ to remove the implicit dependence on the small parameter. This consists of utilizing a Taylor series expansion to obtain a boundary condition on the plane  $z = 0$ , which is asymptotically equivalent to the boundary condition given by [13] on the drop surface [14]. A Taylor series expansion for  $\mathbf{u}_j \cdot \mathbf{n}_d$  about  $z = 0$  yields

$$\begin{aligned} (\mathbf{u}_j \cdot \mathbf{n}_d)_{z=s} &= \mathbf{u}_j^{(0)} \cdot \mathbf{e}_z + \delta \left[ x\mathbf{u}_j^{(0)} \cdot \mathbf{e}_x + y\mathbf{u}_j^{(0)} \cdot \mathbf{e}_y + \mathbf{u}_j^{(1)} \cdot \mathbf{e}_z - \frac{1}{2}(x^2 + y^2) \frac{\partial}{\partial z} (\mathbf{u}_j^{(0)} \cdot \mathbf{e}_z) \right] \\ &+ \delta^2 \left[ \mathbf{u}_j^{(2)} \cdot \mathbf{e}_z + x\mathbf{u}_j^{(1)} \cdot \mathbf{e}_x + y\mathbf{u}_j^{(1)} \cdot \mathbf{e}_y - \frac{1}{2}(x^2 + y^2) \frac{\partial}{\partial z} (\mathbf{u}_j^{(1)} \cdot \mathbf{e}_z) \right] + O(\delta^3) \quad \text{at } z = 0. \end{aligned} \quad [15]$$

Note that the  $O(\delta)$  contribution to the normal velocity at  $z = s$  involves a  $z$ -derivative of the  $O(1)$  component  $\mathbf{u}_j^{(0)} \cdot \mathbf{e}_z$  evaluated at  $z = 0$ . Similar equations can be easily derived for the other boundary conditions, i.e. continuity of velocity and tangential stress on the drop surface. Since the drop is treated as spherical in the present analysis, the normal stress balance is not used. On the small particle surface, the boundary conditions to be satisfied are continuity of tangential velocity and stress and zero normal velocity if the particle is a fluid drop rather than the no-slip condition for a solid particle.

The requirement that the ‘‘inner’’ solution match with the ‘‘outer’’ solution in the overlap region provides the final boundary conditions, i.e.

$$\lim_{|\mathbf{x}| \rightarrow \infty} \mathbf{u}_j \leftrightarrow \lim_{|\tilde{\mathbf{x}}| \rightarrow 0} \tilde{\mathbf{u}}_j. \quad [16]$$

To utilize these matching conditions, the form of the leading order approximation to the solution in the ‘‘outer’’ region is determined for  $|\tilde{\mathbf{x}}| \rightarrow 0$ , via a Taylor series expansion of the Hadamard–Rybczynski solution,  $\mathbf{U}_j^{\text{HR}}$  ( $j = 1, 2$ ), around the point  $\tilde{\mathbf{x}} = 0$ . The result, outside the drop, in terms of inner variables is

$$\begin{aligned} \mathbf{U}_2^\infty(\mathbf{x}) &= \mathbf{U}_2^{\text{HR}}(\mathbf{0}) + \delta \mathbf{x} \cdot \nabla \mathbf{U}_2^{\text{HR}}(\mathbf{0}) + \frac{1}{2!} \delta^2 \mathbf{x} \mathbf{x} : \nabla \nabla \mathbf{U}_2^{\text{HR}}(\mathbf{0}) + O(\delta^3) \\ &= \frac{\sin \theta}{2(1 + \lambda)} \mathbf{e}_x + \delta \cdot \left\{ \frac{\cos \theta}{2(1 + \lambda)} (x\mathbf{e}_x + y\mathbf{e}_y - 2z\mathbf{e}_z) + \frac{\sin \theta}{2(1 + \lambda)} [(1 + 3\lambda)z\mathbf{e}_x - x\mathbf{e}_z] \right\} \\ &+ \delta^2 \cdot \left\{ -\frac{\cos \theta}{2} \frac{2 - 3\lambda}{1 + \lambda} (xz\mathbf{e}_x + yz\mathbf{e}_y - z^2\mathbf{e}_z) - \frac{\cos \theta}{1 + \lambda} (x^2 + y^2)\mathbf{e}_z + \frac{\sin \theta}{4(1 + \lambda)} [(1 + 3\lambda)y^2 \right. \\ &\left. - (2 + 9\lambda)z^2]\mathbf{e}_x - \frac{\sin \theta}{4(1 + \lambda)} [(1 - 3\lambda)x^2\mathbf{e}_x + 2xy\mathbf{e}_y - 2(2 - 3\lambda)xz\mathbf{e}_z] \right\} + O(\delta^3) \end{aligned} \quad [17]$$

in terms of the local Cartesian coordinates  $\mathbf{x} = (x, y, z)$  scaled with the *inner* scale,  $a$ . Here,  $\lambda \equiv \mu_1/\mu_2$  is the viscosity ratio of the large drop relative to the suspending fluid, and  $U_2^{\text{HR}}$  is the Hadamard–Rybczynski solution in fluid 2 for streaming flow past the larger drop. Similarly, for the “outer” region, the flow inside the large drop is expanded in a Taylor series around the origin, and expressed in terms of inner variables:

$$\begin{aligned}
 U_1^\infty(\mathbf{x}) = & \frac{\sin \theta}{2(1+\lambda)} \mathbf{e}_x + \delta \cdot \left[ \frac{\cos \theta}{2(1+\lambda)} (x\mathbf{e}_x + y\mathbf{e}_y - 2z\mathbf{e}_z) + \frac{\sin \theta}{2(1+\lambda)} (4z\mathbf{e}_x - x\mathbf{e}_z) \right] \\
 & + \delta^2 \cdot \left[ \frac{\cos \theta}{2(1+\lambda)} (xz\mathbf{e}_x + yz\mathbf{e}_y - z^2\mathbf{e}_z) - \frac{\cos \theta}{1+\lambda} (x^2 + y^2)\mathbf{e}_z \right. \\
 & \left. + \frac{\sin \theta}{1+\lambda} (y^2 + z^2)\mathbf{e}_x + \frac{\sin \theta}{2(1+\lambda)} (x^2\mathbf{e}_x - xy\mathbf{e}_y - xz\mathbf{e}_z) \right] + O(\delta^3). \tag{18}
 \end{aligned}$$

In [17] and [18] the superscript  $\infty$  denotes the velocity field at  $|\mathbf{x}| \rightarrow \infty$ . The terms of  $O(1)$ , in [17] and [18] represent a uniform streaming flow with the same velocity components as the local velocity of the outer flow solution at the origin,  $O$ , on the interface of the larger drop, i.e. the relative velocity of the Hadamard–Rybczynski solution. The remaining terms of  $O(\delta)$  and  $O(\delta^2)$  consist of several *linear* shear and extensional flows, and flows with various types of *quadratic* dependence on spatial position, all with a stagnation point at the origin, see figure 2. We have already explained that corrections in the outer flow solution due to the disturbance produced by the particle will not occur until  $O(\delta^3)$ , but we shall also see that this is true during the course of the analysis below.

It can be expected from [17] and [18] that the trajectories for a small (rigid or fluid) particle in the presence of a nearby larger drop should be qualitatively different from those obtained in earlier work by Goren & O’Neill (1971) on particle motion relative to a larger *no-slip* collector sphere. In the later case (i.e.  $\lambda \rightarrow \infty$ ), the leading term in the local Taylor-series approximation to the undisturbed flow around the *no-slip* collector sphere can be seen from [17] to be a simple shear flow of  $O(\delta)$  parallel to the collector surface with motion normal toward the surface appearing as

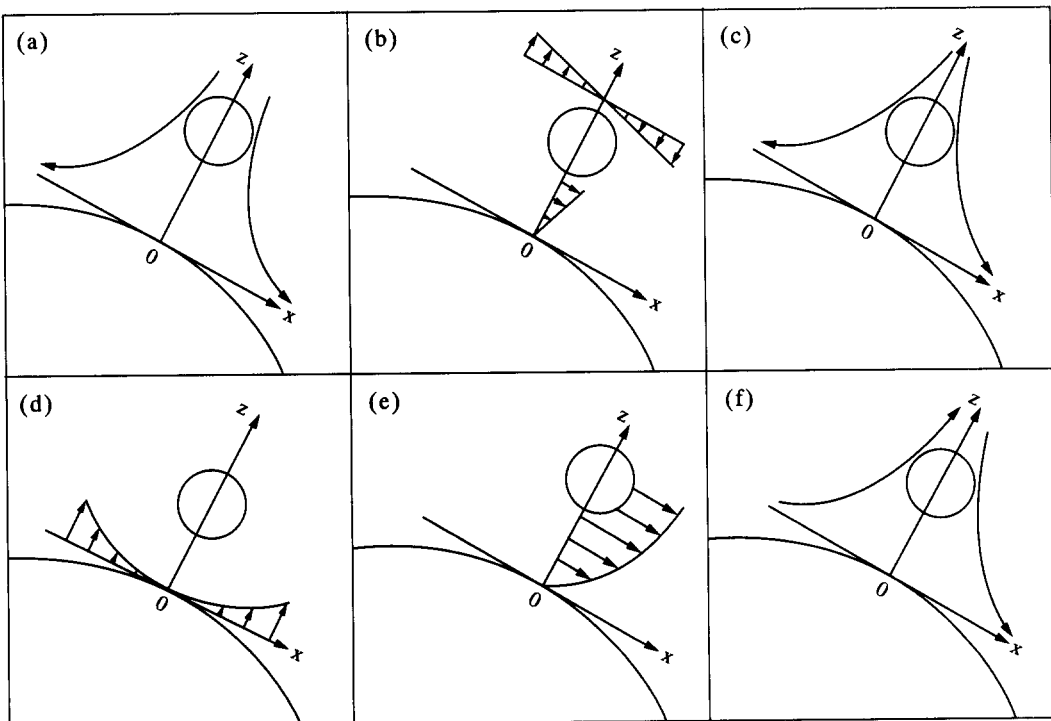


Figure 2. Components of local undisturbed flow: (a) uniaxial axisymmetric straining flow,  $U_2^\infty = \mathbf{E} \cdot \mathbf{x}$ ; (b) linear shear flows parallel and perpendicular to the  $x$ - $y$  plane,  $U_2^\infty = \mathbf{F} \cdot \mathbf{x}$ ; (c) quadratic stagnation flow,  $U_2^\infty = K_1(xz\mathbf{e}_x + yz\mathbf{e}_y - z^2\mathbf{e}_z)$ ; (d) axisymmetric paraboloidal flow,  $U_2^\infty = K_2(x^2 + y^2)\mathbf{e}_z$ ; (e) quadratic shear flow,  $U_2^\infty = (K_3y^2 + K_4z^2)\mathbf{e}_z$ ; (f) nonaxisymmetric quadratic stagnation flow.

a quadratic shear and stagnation flow at  $O(\delta^2)$  in agreement with Goren & O'Neill (1971). Thus, the particle is transported around a *solid* collector with a velocity of  $O(\delta)$ , and transported toward (or away from) the collector with a velocity of  $O(\delta^2)$ . When the collector is a *fluid* drop, the particle transport velocity is due to a uniform flow of  $O(1)$ , while the motion toward (or away from) the collector is due to an extensional flow at  $O(\delta)$ , as can be seen from [17]. A particle will thus remain near a fluid drop for a very much shorter period than it would near a large *no-slip* sphere, but the motion toward (or away from) the fluid drop is also much stronger. It may be expected that these differences between a solid *no-slip* and a *fluid* collector will have important consequence in the trajectories of relative motions.

The detailed trajectories of relative motion can be derived by solving the inner problem (i.e. the problem at the scale  $a$  of the small particle), in which we consider the motion of the small particle near the interface of the larger drop which appears as locally planar in its undeformed state (cf. [15]) with the combined streaming, linear and quadratic flows from the outer flow, [17] and [18], imposed via matching as the boundary condition at "infinity",

$$\left. \begin{aligned} \mathbf{u}_2 &\rightarrow \mathbf{U}_2^\infty(\mathbf{x}) \quad \text{as } |\mathbf{x}| \rightarrow \infty \quad (\text{outside the large drop}) \\ \mathbf{u}_1 &\rightarrow \mathbf{U}_1^\infty(\mathbf{x}) \quad \text{as } |\mathbf{x}| \rightarrow \infty \quad (\text{inside the large drop}). \end{aligned} \right\} \quad [19a]$$

If the interface is assumed to remain flat (i.e. the disturbance flow associated with the small drop produces negligible deformation on the inner scale of resolution), the problem is "linear" and can be solved by superposition of the small drop motions associated with each of the streaming, linear and quadratic flows in [17] taken separately. The main advantage of the flat, nondeforming interface approximation is that analytic equations can be derived for the relative trajectory. As indicated in the Introduction we shall approach this problem using the singularity method of Lee *et al.* (1979), who generalized the reciprocal theorem of Lorentz, to derive a general lemma for obtaining solutions of Stoke's equations that satisfy continuity of velocity and tangential stress on a flat interface, given only an arbitrary solution of Stoke's equations for an *unbounded* domain with no interface. We extend the singularity method of Lee *et al.* to consider the undisturbed flow past the small particle for the asymptotic limit  $\epsilon = (a/d) \ll 1$ . In this case, it is convenient to solve for the disturbance velocity field  $\mathbf{v}_j$  in the inner region due to the presence of the particle, rather than directly solving for  $\mathbf{u}_j$ . The boundary condition for the disturbance flow at infinity (i.e. the matching condition [19a]) is simply

$$\left. \begin{aligned} \mathbf{v}_2 (\equiv \mathbf{u}_2 - \mathbf{U}_2^\infty(\mathbf{x})) &\rightarrow 0 \\ \mathbf{v}_1 (\equiv \mathbf{u}_1 - \mathbf{U}_1^\infty(\mathbf{x})) &\rightarrow 0 \end{aligned} \right\} \quad \text{as } |\mathbf{x}| \rightarrow \infty. \quad [19b]$$

Further, the disturbance flow is required to "cancel" the far-field velocity  $-\mathbf{U}_2^\infty$  at the surface of the small fluid particle.

For the solution to the disturbance flow, the singularity method can be simplified to the superposition of fundamental solutions for a point force (i.e. Stokeslet), a potential dipole and higher order singularities (e.g. a stresslet, a rotlet, a potential quadrupole etc.) at the center of the small fluid particle. Thus, solutions for the problem are constructed in the following manner. First, we put singularities at the center of the particle which satisfy exactly the boundary conditions at the surface of the particle for an unbounded single-fluid domain. The resulting unbounded-domain solution does *not* satisfy the boundary conditions at the flat interface; instead, there is a mismatch of  $O(\epsilon)$  at the interface. To eliminate this "error", the simple transformation rule of Lee *et al.* (1979) is used to transform the unbounded-domain solution to a corresponding solution (in terms of the Green's functions for a bounded domain) that satisfies exactly the boundary conditions at the interface. In general, however, this new solution does not satisfy the boundary conditions any longer at the surface of the small particle, but induces an error of  $O(\epsilon)$ . Additional higher order singularities must then be included at the particle center to cancel this induced error of  $O(\epsilon)$ , and so on. The result of this procedure is an asymptotic approximation, in the form of a series in  $\epsilon$ , that is valid in the limit of  $\epsilon \rightarrow 0$ .

The complete solution for the inner problem is obtained by superposition of the solution for a uniform streaming flow  $\mathbf{U}^{\text{HR}}(\mathbf{0})$  and for the various linear and quadratic flows given by [17] and [18], with stagnation point at the origin on the approximately *planar interface* of the larger drop. However, a complete solution is already available for the uniform streaming flow problem,



corresponding to  $U^{HR}(\mathbf{0})$ , due to Yang & Leal (1990) who determined the relationship between the hydrodynamic drag force on the fluid particle and the streaming velocity. It thus remains only to solve the problems for the linear and quadratic flows. In the theoretical analysis that follows, we consider the hydrodynamic force acting on the small fluid particle in the presence of these linear and quadratic flows. The results are then used in section 5 to calculate the trajectories of a small drop in the vicinity of a larger drop.

### 3. SOLUTION AT $O(\delta)$ : LINEAR FLOWS

Let us begin with the contributions to the disturbance flow in the vicinity of the small particle associated with the outer flow field at  $O(\delta)$ , cf. [17] and [18]. The governing equations for the inner region at  $O(\delta)$  are just Stokes equations and the continuity equation. The boundary conditions for the disturbance velocity field obtained from expansions via domain perturbation are, at  $z = 0$  (the interface of the large drop):

$$\mathbf{v}_1^{(1)} = \mathbf{v}_2^{(1)}, \tag{20}$$

$$\mathbf{v}_1^{(1)} \cdot \mathbf{e}_z = \mathbf{v}_2^{(1)} \cdot \mathbf{e}_z = 0 \tag{21}$$

and

$$[[\sigma_{xz}^{(1)}]] = [[\sigma_{yz}^{(1)}]] = 0, \tag{22}$$

where the symbol  $[[(\cdot)]]$  represents the jump of quantity  $(\cdot)$  across the surface of the drop. At the surface of the small fluid particle, the disturbance flow must satisfy continuity of velocity and tangential stress and the kinematic condition:

$$\mathbf{v}_2^{(1)} \cdot \mathbf{n} = -U_2^\infty \cdot \mathbf{n} \quad \text{for } \mathbf{x} \in \text{small fluid particle surface.} \tag{23}$$

The far-field boundary conditions are

$$\mathbf{v}_1 \rightarrow 0, \quad \mathbf{v}_2 \rightarrow 0 \quad \text{as } |\mathbf{x}| \rightarrow \infty. \tag{24}$$

It can be seen by examining [17] that the flows imposed on the inner problem from the outer solution are, at  $O(\delta)$ , a uniaxial extension and a pair of simple shear flows.

The above problem for the inner region at  $O(\delta)$  can be simplified by using linearity to obtain a solution by superposition of the solutions for a stationary particle near a flat interface in the uniaxial straining flow and the linear shear flows.

#### 3.1. Uniaxial extensional flow

Let us first consider the creeping motion of a fluid in the vicinity of a stationary small particle that is located at point  $\mathbf{x}_p = (0, 0, d)^\dagger$  in fluid 2 when the undisturbed motion is an axisymmetric uniaxial straining flow with a stagnation point at the origin on the interface at  $z = 0$ :

$$U_2^\infty(\mathbf{x}) = \delta \cdot \frac{\cos \theta}{2(1 + \lambda)} \mathbf{E} \cdot \mathbf{x} \tag{25}$$

in which the dimensionless strain rate tensor  $\mathbf{E}$  has Cartesian components,  $E_{ij} = (\delta_{ij} - 3\delta_{i3}\delta_{j3})$ , see figure 2(a). The linearity of the problem enables us to decompose this undisturbed flow into a constant vector (i.e. uniform streaming flow),

$$U_2^\infty(\mathbf{x}) = \delta \cdot \frac{\cos \theta}{2(1 + \lambda)} \mathbf{E} \cdot \mathbf{x}_p = -\delta \cdot \frac{\cos \theta}{1 + \lambda} d \mathbf{e}_z, \tag{26}$$

and a uniaxial extensional flow with stagnation point at the particle center  $\mathbf{x}_p$ , i.e.

$$U_2^\infty(\mathbf{x}) = \delta \cdot \frac{\cos \theta}{2(1 + \lambda)} \mathbf{E} \cdot (\mathbf{x} - \mathbf{x}_p). \tag{27}$$

The uniform streaming flow problem was treated in Yang & Leal (1990). Here, we solve the problem with undisturbed flow  $U_2^\infty(\mathbf{x}) = \mathbf{E} \cdot (\mathbf{x} - \mathbf{x}_p)$ .

In an infinite fluid domain with no interface, the disturbance velocity field outside the small fluid particle, which satisfies the boundary conditions at the particle surface, including the kinematic

---

$^\dagger$ Note that  $d$  here, and throughout later sections, is assumed to be scaled with respect to the particle radius  $a$ .

condition [23] with undisturbed flow [27], can be represented by a superposition of the fundamental solutions for a potential quadrupole and a stresslet, both applied at the center of the particle. For a fluid particle with viscosity  $\mu_3 = \kappa\mu_2$ , the result satisfying the boundary conditions is of the form

$$\frac{5}{2}A(\kappa)\mathbf{u}_{\text{SS}}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_z) + \frac{1}{2}B(\kappa)\mathbf{u}_{\text{PQ}}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_z). \quad [28]$$

Here,  $\mathbf{u}_{\text{SS}}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_z)$  and  $\mathbf{u}_{\text{PQ}}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_z)$  denote the fundamental solutions for a stresslet ( $\mathbf{e}_z, \mathbf{e}_z$ ) and a potential quadrupole ( $\mathbf{e}_z, \mathbf{e}_z$ ) located at the center of the particle in an unbounded single fluid domain, cf. Chwang & Wu (1975) for the specific formulae of  $\mathbf{u}_{\text{SS}}$  and  $\mathbf{u}_{\text{PQ}}$ . The parameters  $A(\kappa)$  and  $B(\kappa)$  are defined as

$$A(\kappa) = \frac{\frac{2}{3} + \kappa}{1 + \kappa}, \quad B(\kappa) = \frac{\kappa}{1 + \kappa}. \quad [29a,b]$$

Since we consider only the limit  $\epsilon \equiv (1/d) \ll 1$ , the solution of the full problem, including the interface, is most conveniently obtained via the method of reflections, as explained in some detail by Lee *et al.* (1979). The zeroth-order approximation for the velocity field outside the particle in this procedure is the single-fluid unbounded-domain solution given by [28], which satisfies boundary conditions exactly at the particle surface but does not satisfy the interface boundary conditions at  $z = 0$ . However, Lee *et al.* have shown that a first correction which does satisfy these conditions can be obtained by simply utilizing the same form [28] as in the zeroth-order solution, but with the fundamental solutions  $\mathbf{u}_{2,\text{SS}}$  and  $\mathbf{u}_{2,\text{PQ}}$  for a stresslet and a potential quadrupole in the presence of a flat interface obtained by the generalized reciprocal theorem of Lee *et al.* (1979).

Although the solution given by [28] now satisfies the interface boundary conditions, it no longer satisfies the boundary conditions at the particle surface, and additional singularities are needed at the particle center in order to match the ‘‘first correction’’ velocity field at the particle surface: i.e. the interface reflection of the potential quadrupole and the stresslet, evaluated at the particle surface. The preceding two steps, leading to the first correction, could be carried out for arbitrary  $\epsilon$ . However, the expression for the interface reflection evaluated at the drop surface is highly complicated, and it is not possible for arbitrary  $\epsilon$  to determine singularities at the particle center which precisely satisfy the continuity of tangential velocity and stress and zero normal velocity boundary conditions at all points on the particle surface. Instead, we consider the asymptotic limit  $\epsilon \ll 1$ , and then choose singularities to match only the first few terms of the interface reflections at the particle surface in powers of  $\epsilon$ . The leading terms of the interface reflection near the particle surface, for small  $\epsilon$ , are

$$\frac{5}{8}A(\kappa) \left[ \epsilon^2 \cdot \frac{2 + 3\lambda}{1 + \lambda} \cdot \mathbf{e}_z + \epsilon^3 \cdot \frac{1 + 2\lambda}{1 + \lambda} \mathbf{E} \cdot (\mathbf{x} - \mathbf{x}_p) \right] + O(\epsilon^4), \quad [30]$$

in which  $\lambda$  is the viscosity ratio (i.e.  $\lambda = \mu_1/\mu_2$ ) of the two continuous fluid phases 1 and 2.

Examining [30], we see that the presence of the interface induces a steady streaming flow at  $O(\epsilon^2)$  normal to the interface. The term of  $O(\epsilon^3)$  in [30] is equivalent to an axisymmetric uniaxial straining flow with a stagnation point at the drop center, and the  $z$ -axis as the axis of symmetry. The singularities required to match the additional velocity field [30] at the particle surface can be readily evaluated. We have seen previously that an extensional flow of the type represented by the  $O(\epsilon^3)$  term in [30] is generated by a superposition of a stresslet and a potential quadrupole. It can be shown that a uniform streaming flow solution is generated in an unbounded single fluid by a Stokeslet and a potential dipole. To counter the term of  $O(\epsilon^2)$  in [30], we thus require the superposition of a Stokeslet and a potential dipole at the drop center. The resulting velocity field is

$$-\frac{15}{32} \cdot \frac{2 + 3\lambda}{1 + \lambda} A(\kappa) \epsilon^2 \left[ C(\kappa)\mathbf{u}_{\text{S}}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z) - \frac{1}{3}B(\kappa)\mathbf{u}_{\text{D}}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z) \right], \quad [31]$$

where the parameter  $C(\kappa)$  is defined as

$$C(\kappa) = \frac{\frac{2}{3} + \kappa}{1 + \kappa} \quad [32]$$

and  $\mathbf{u}_{\text{S}}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z)$  and  $\mathbf{u}_{\text{D}}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z)$  denote the fundamental solutions for a Stokeslet  $\mathbf{e}_z$  and a potential dipole  $\mathbf{e}_z$  at the drop center in an unbounded domain with no interface. It is important to note that the point force (i.e. Stokeslet) velocity of strength  $O(\epsilon^2)$ , corresponding to  $\mathbf{u}_{\text{S}}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z)$ ,

will itself generate a vertical velocity component of  $O(\epsilon^3)$  at the particle surface when it is “reflected” from the interface, cf. Lee *et al.* (1979). Thus, if we are to consider any correction terms of  $O(\epsilon^3)$  from [30] we must simultaneously include this additional  $O(\epsilon^3)$  correction to the velocity field near the drop. In order to match this  $O(\epsilon^2)$  term at the drop surface we require an additional point force and potential dipole at the drop center of the form:

$$\mathbf{u}_{2,ST}^{(3)} = -\frac{5}{4} \cdot \left(\frac{3}{8} \cdot \frac{2 \times 3\lambda}{1 + \lambda}\right)^2 A(\kappa)C(\kappa)\epsilon^3 [C(\kappa)\mathbf{u}_S(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z) - \frac{1}{3}B(\kappa)\mathbf{u}_D(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z)]. \quad [33]$$

Thus, the complete contribution to the velocity field that is required to match the first two terms of the interface reflection [30] at the drop surface is a superposition of:

*Stokeslet,*

$$-\frac{5}{4}A(\kappa)\mathbf{u}_S(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z) \left\{ \sum_{n=1}^2 \left[ \frac{9}{8}C(\lambda)C(\kappa)\epsilon \right]^n \cdot \epsilon + O(\epsilon^4) \right\}; \quad [34]$$

*potential dipole,*

$$\frac{5}{12}A(\kappa) \frac{B(\kappa)}{C(\kappa)} \mathbf{u}_D(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z) \left\{ \sum_{n=1}^2 \left[ \frac{9}{8}C(\lambda)C(\kappa)\epsilon \right]^n \cdot \epsilon + O(\epsilon^4) \right\}; \quad [35]$$

*stresslet,*

$$\frac{5}{2}A(\kappa)\mathbf{u}_{SS}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_z) \left[ 1 + \frac{5}{8}A(\kappa) \frac{1 + 2\lambda}{1 + \lambda} \epsilon^3 + O(\epsilon^4) \right] \quad [36]$$

and

*potential quadrupole,*

$$\frac{1}{2}B(\kappa)\mathbf{u}_{PQ}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_z) \left[ 1 + \frac{5}{8}A(\kappa) \frac{1 + 2\lambda}{1 + \lambda} \epsilon^3 + O(\epsilon^4) \right]. \quad [37]$$

The complete velocity field resulting from the superposition of [34]–[37] satisfies boundary conditions *exactly* at the interface  $z = 0$  and boundary conditions to  $O(\epsilon^2)$  at the particle surface. Higher order approximations could be obtained by straightforward continuation of the same procedure. However, the solution above is sufficient for present purposes.

The net force exerted on a fluid drop located at the stagnation point in the undisturbed flow field  $\hat{\mathbf{U}}_2^\infty = \mathbf{E} \cdot (\mathbf{x} - \mathbf{x}_p)$  can be evaluated simply from the Stokeslet strength:

$$\mathbf{F} = 10\pi A(\kappa)\epsilon \cdot \sum_{n=1}^2 \left[ \frac{9}{8}C(\lambda)C(\kappa)\epsilon \right]^n \mathbf{e}_z + O(\epsilon^4). \quad [38]$$

The force  $\mathbf{F}$  is always oriented *away* from the interface, and the magnitude is increased as the viscosity ratios,  $\lambda$  and  $\kappa$ , become larger. Thus, a negative external force  $-\mathbf{F}$  toward the interface would have to be applied to the particle to keep it from translating away from the stagnation point  $\mathbf{x}_p$  of the flow regardless of the particle position, or the viscosity ratio of the two fluids. It should be understood that, in this flow field  $\hat{\mathbf{U}}_2^\infty = \mathbf{E} \cdot (\mathbf{x} - \mathbf{x}_p)$ , the interface translates with velocity  $2d\mathbf{e}_z$  toward the stagnation point  $\mathbf{x}_p$  at which the drop center is held fixed. This “interface motion” can be viewed as the source of  $\mathbf{F}$ .

Now let us turn to the original problem of calculating the force acting on a stationary particle that is located at point  $\mathbf{x}_p$  in fluid 2 which is undergoing the axisymmetric uniaxial extension flow  $\hat{\mathbf{U}}_2^\infty = \mathbf{E} \cdot \mathbf{x}$  with origin at the interface [i.e. figure 2(a)]. Owing to the linearity of the problem, the hydrodynamic force exerted in this case can be determined by a superposition of the force for a uniform streaming flow with velocity  $\hat{\mathbf{U}}_2^\infty = \mathbf{E} \cdot \mathbf{x}_p$  and for a uniaxial straining flow  $\hat{\mathbf{U}}_2^\infty = \mathbf{E} \cdot (\mathbf{x} - \mathbf{x}_p)$  with stagnation point at the drop center. The result can be expressed in the following form:

$$\mathbf{F} = \mathbf{K}_T \cdot \mathbf{E} \cdot \mathbf{x}_p + 10\pi A(\kappa)\epsilon \cdot \sum_{n=1}^2 \left[ \frac{9}{8}C(\lambda)C(\kappa)\epsilon \right]^n \mathbf{e}_z + O(\epsilon^4). \quad [39]$$

The components of the translation resistance tensor  $\mathbf{K}_T$  were determined by Yang & Leal (1990) for motion of a fluid drop near a plane fluid–fluid interface.

In the study of the hydrodynamic interactions between a single bubble and a *no-slip* particle (i.e. for  $\lambda \rightarrow 0$  and  $\kappa \rightarrow \infty$ ), Dukhin & Rudev (1977) obtained an exact result for the drag force on

a *no-slip* sphere located at the axis of symmetry in an axisymmetric uniaxial extensional flow  $U_2^\infty = E \cdot x$ , near a gas-liquid interface (i.e.  $\lambda \rightarrow 0$ ), using the eigensolutions of Laplace's equation in bipolar coordinates. It is a simple matter to calculate the drag force  $F'$  on the fluid particle from the present asymptotic solution [39] with  $x_p = (0, 0, d)$ . The hydrodynamic drag scaled with respect to Stokes drag in an unbounded fluid is

$$-\frac{F'}{12\pi\mu_2 U_1 adE} = C(\kappa) \left\{ 1 + \sum_{n=1}^3 \left[ \frac{2}{8} C(\lambda) C(\kappa) \cdot \epsilon \right]^n - \frac{1}{8} B(\kappa) \cdot \frac{1+4\lambda}{1+\lambda} \cdot \epsilon^3 - \frac{15}{16} A(\kappa) C(\lambda) \epsilon^3 \right\} e_z + O(\epsilon^4). \tag{40}$$

In order to illustrate the effects of hydrodynamic interaction between the particle and the interface, the dimensionless drag ratio from [40] is plotted in figure 3 as a function of the dimensionless separation distance  $d$  for  $\lambda = 0$  and  $\infty$ . For each value of  $\lambda$ , we include two values of the viscosity ratio  $\kappa = 0$  (i.e. *inviscid* bubble) and  $\kappa = \infty$  (i.e. *no-slip* sphere). Also shown for comparison are the corresponding exact-solution results of Dukhin & Rlev for a *no-slip* sphere near a free surface (i.e.  $\lambda \rightarrow 0$ ). It can be seen from figure 3 that there is very good agreement between the two solutions, except in the region near  $d \approx 1$ . Indeed, the relative error associated with the asymptotic solution is  $< 1.77\%$  for  $d \geq 1.5$ . As expected, the difference between the two results becomes larger as the particle approaches the interface owing to the poor convergence of the asymptotic solution [40] in powers of  $\epsilon$ . Further, due to the presence of the interface, the magnitude of drag is increased for all values of  $\lambda$  and  $\kappa$  considered here, and this effect is a strong function of the drop position relative to the interface. Although the qualitative features of the dimensionless drag as a function of the dimensionless separation distance  $d$  are, in fact, quite similar for all viscosity ratios,  $\lambda$  and  $\kappa$ , the relative increase in the drag for an inviscid gas bubble ( $\kappa \rightarrow 0$ ) is much larger than that for a *no-slip* sphere ( $\kappa \rightarrow \infty$ ).

3.2. Linear shear flows

We now turn to the case of a fluid drop located at an arbitrary point  $x_p$  in a simple shear flow  $U_2^\infty = F \cdot x$ , either parallel or perpendicular to the interface as shown in figure 2(b). The shear rate tensor is given by

$$\Gamma = \{\Gamma_{ij}\} = \delta \cdot \frac{\sin \theta}{2(1+\lambda)} \cdot \begin{bmatrix} 0 & 0 & 1+3\lambda \\ 0 & 0 & 0 \\ -1 & 0 & 0 \end{bmatrix}. \tag{41}$$

Again the problem can be decomposed into a simple translation of the fluid system including the interface with uniform velocity  $U_2^\infty = F \cdot x_p$  past the stationary drop together with a linear shear

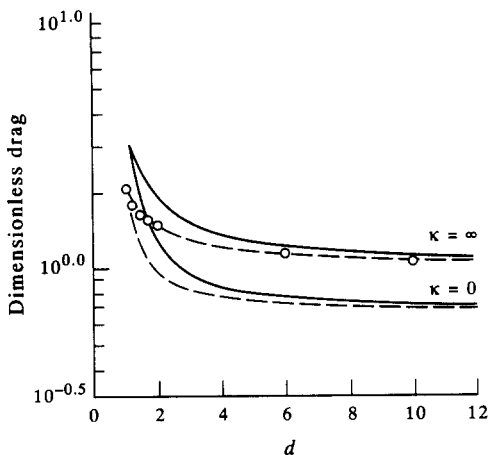


Figure 3. Drag ratio for axisymmetric extensional flow relative to Stoke's drag in an unbounded fluid as a function of the dimensionless distance,  $d$ , between the particle and the interface:  $U_2^\infty = E \cdot x$ ; — for  $\lambda = \infty$ , --- for  $\lambda = 0$  and  $\circ$  for the corresponding exact-solution results ( $\lambda = 0, \kappa = \infty$ ) of Dukhin & Rlev (1977).

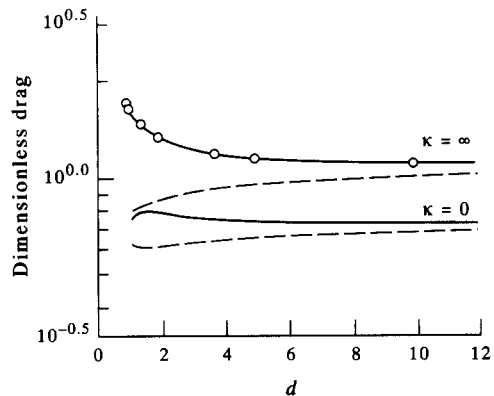


Figure 4. Drag ratio relative to the drag in an unbounded fluid as a function of the dimensionless distance,  $d$ , between the particle and the interface:  $U_2^\infty = \Gamma_{13} z e_x$ ; — for  $\lambda = \infty$ , --- for  $\lambda = 0$  and  $\circ$  for the corresponding exact-solution results ( $\lambda = \kappa = \infty$ ) of Goldman *et al.* (1976b) and Goren & O'Neill (1971).

flow  $\mathbf{U}_2^\infty = \mathbf{\Gamma} \cdot \mathbf{x} - \mathbf{\Gamma} \cdot \mathbf{x}_p$  with a stagnation point at the drop center. In view of the linearity of the problem we need only solve the case of  $\mathbf{U}_2^\infty = \Gamma_{13}(z - d)\mathbf{e}_x + \Gamma_{31}x\mathbf{e}_z$ . In order to analyze the velocity field we follow the method of reflections procedure of the preceding example. In this case, the solution for the disturbance flow in the unbounded fluid is simply a superposition of a stresslet, and a potential quadrupole, at the center of the drop, i.e.

*stresslet,*

$$-\frac{5}{6}A(\kappa)[\Gamma_{13}\mathbf{u}_{SS}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_x, \mathbf{e}_z) + \Gamma_{31}\mathbf{u}_{SS}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_x)]; \quad [42a]$$

and

*potential quadrupole,*

$$-\frac{1}{6}B(\kappa)[\Gamma_{13}\mathbf{u}_{PQ}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_x, \mathbf{e}_z) + \Gamma_{31}\mathbf{u}_{PQ}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_x)]. \quad [42b]$$

For a solid *no-slip* sphere (i.e.  $\kappa \rightarrow \infty$ ) we need an additional rotlet distribution that is associated with the hydrodynamic couple induced by the primary flow:

*rotlet,*

$$-\frac{1}{2}\Theta(\kappa)(\Gamma_{13} - \Gamma_{31})\mathbf{u}_R(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_y), \quad [42c]$$

in which the parameter  $\Theta(\kappa) = 1$  for a no-slip sphere, and otherwise  $\Theta(\kappa) = 0$ . The unbounded-domain solution represented by [42a–c] satisfies exactly the boundary conditions at the surface of the drop but generates a mismatch at  $O(\epsilon)$  at the flat interface.

As in the preceding example, the first correction for the presence of the interface in the reflections expansion can now be obtained easily from the unbounded-domain solution, [42a–c], by simply replacing the fundamental solutions  $\mathbf{u}_{SS}$ ,  $\mathbf{u}_{PQ}$  and  $\mathbf{u}_R$  (which pertain to an unbounded fluid) with the corresponding fundamental solutions  $\mathbf{u}_{2,SS}$ ,  $\mathbf{u}_{2,PQ}$  and  $\mathbf{u}_{2,R}$  that satisfy boundary conditions on the flat interface [and are generated using the lemma of Lee *et al.* (1979)]. The first correction for the presence of the interface does *not* satisfy the boundary conditions at the drop surface, because the interface reflection is nonzero at the drop surface. Following section 3.1, we examine the leading terms of the reflected velocity field at the particle surface as a power series in  $\epsilon$ :

$$-\frac{\epsilon^2}{16} \cdot \frac{5\lambda A(\kappa)(\Gamma_{13} + \Gamma_{31}) - 2\Theta(\kappa)(\Gamma_{13} - \Gamma_{31})}{1 + \lambda} \mathbf{e}_x + \epsilon^3 \cdot \mathbf{\Gamma}^* \cdot (\mathbf{x} - \mathbf{x}_p), \quad [43a]$$

where the nonzero components of the second-order shear rate tensor  $\mathbf{\Gamma}^*$  are given by

$$\Gamma_{13}^* = \frac{5\lambda A(\kappa)(\Gamma_{13} + \Gamma_{31}) - (2 - \lambda)\Theta(\kappa)(\Gamma_{13} - \Gamma_{31})}{16(1 + \lambda)} \quad [43b]$$

and

$$\Gamma_{31}^* = \frac{5(1 + 2\lambda)A(\kappa)(\Gamma_{13} + \Gamma_{31}) - (1 + 4\lambda)\Theta(\kappa)(\Gamma_{13} - \Gamma_{31})}{16(1 + \lambda)}. \quad [43c]$$

It can be seen from [43a–c] that the presence of the interface in this case is equivalent in its effect on the particle to a steady streaming flow at  $O(\epsilon^2)$  parallel to the interface, and a linear shear flow at  $O(\epsilon^3)$  either normal or parallel to the interface.

In order to satisfy the conditions of continuity of tangential velocity and stress and zero normal velocity at the particle surface, we need additional singularities at the particle center that match the reflected velocity field at the particle surface. For the term of  $O(\epsilon^2)$ , a point force and a potential dipole are required, which have the intensity and orientation:

$$\frac{3}{64} \cdot \frac{5\lambda A(\kappa)(\Gamma_{13} + \Gamma_{31}) - 2\Theta(\kappa)(\Gamma_{13} - \Gamma_{31})}{1 + \lambda} \epsilon^2 \cdot [C(\kappa)\mathbf{u}_S(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_x) - \frac{1}{3}B(\kappa)\mathbf{u}_D(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_x)]. \quad [44]$$

By induction, we also know that the interface reflection of the point force and potential dipole solutions corresponding to [44] will yield a nonzero contribution of  $O(\epsilon^3)$  to the  $x$ -component of velocity at the particle surface. In order to satisfy the boundary conditions on the particle surface

to  $O(\epsilon^3)$ , we thus require an additional point force and a potential dipole at the particle center with magnitude and orientation:

$$\frac{3}{16} \frac{3}{64} \frac{2 - 3\lambda}{1 + \lambda} \frac{5\lambda A(\kappa)(\Gamma_{13} + \Gamma_{31}) - 2\Theta(\kappa)(\Gamma_{13} - \Gamma_{31})}{1 + \lambda} \times C(\kappa)\epsilon^3 \cdot [C(\kappa)\mathbf{u}_S(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_x) - \frac{1}{3}B(\kappa)\mathbf{u}_D(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_x)]. \quad [45]$$

Further, the singularities required to match the  $O(\epsilon^2)$  contribution in [43a] have been previously seen to be a stresslet, a rotlet and a potential quadrupole at the drop center at  $O(\epsilon^3)$ .

Consequently, for the linear shear flow past a drop, the singularities required at the center of the drop through  $O(\epsilon^3)$  are:

*Stokeslet,*

$$\frac{3}{64} \frac{5\lambda A(\kappa)(\Gamma_{13} + \Gamma_{31}) - 2\Theta(\kappa)(\Gamma_{13} - \Gamma_{31})}{1 + \lambda} C(\kappa)\epsilon^2 \cdot [1 - \frac{9}{16}D(\lambda)C(\kappa)\epsilon]\mathbf{u}_S(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_x); \quad [46]$$

*potential dipole,*

$$\frac{1}{64} \frac{5\lambda A(\kappa)(\Gamma_{13} - \Gamma_{31}) - 2\Theta(\kappa)(\Gamma_{13} - \Gamma_{31})}{1 + \lambda} B(\kappa)\epsilon^2 \cdot [1 - \frac{9}{16}D(\lambda)C(\kappa)\epsilon]\mathbf{u}_D(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_x); \quad [47]$$

*stresslet,*

$$-\frac{5}{6}A(\kappa)\left\{(\Gamma_{13} + \Gamma_{31}) + \left[\frac{5(1 + 3\lambda)A(\kappa)(\Gamma_{13} + \Gamma_{31})}{16(1 + \lambda)} - \frac{3\Theta(\kappa)(\Gamma_{13} - \Gamma_{31})}{16}\right]\epsilon^3\right\}\mathbf{u}_{SS}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_x, \mathbf{e}_z); \quad [48]$$

*potential quadrupole,*

$$-\frac{1}{6}B(\kappa)\left\{(\Gamma_{13} + \Gamma_{31}) + \left[\frac{5(1 + 3\lambda)A(\kappa)(\Gamma_{13} + \Gamma_{31})}{16(1 + \lambda)} - \frac{3\Theta(\kappa)(\Gamma_{13} - \Gamma_{31})}{16}\right]\epsilon^3\right\}\mathbf{u}_{PQ}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_x, \mathbf{e}_z); \quad [49]$$

and

*rotlet,*

$$-\frac{1}{2}\Theta(\kappa)\left\{(\Gamma_{13} - \Gamma_{31}) + \left[\frac{(5\lambda - 1)\Theta(\kappa)(\Gamma_{13} - \Gamma_{31})}{16(1 + \lambda)} - \frac{5A(\kappa)(\Gamma_{13} + \Gamma_{31})}{16}\right]\epsilon^3\right\}\mathbf{u}_R(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_y); \quad [50]$$

in which the parameter  $D(\lambda)$  is defined as

$$D(\lambda) = \frac{\frac{2}{3} - \lambda}{1 + \lambda}. \quad [51]$$

The net force exerted on the drop located at the center of the simple shear flow can be evaluated simply from the Stokeslet distribution and expressed in the following form:

$$\mathbf{F} = \mathbf{K}_{SF} : \boldsymbol{\Gamma}. \quad [52a]$$

Here, the nonzero components of the third-order hydrodynamic tensor  $\mathbf{K}_{SF}$  are given by

$$K_{SF}^{113} = -\frac{3\pi}{8} \frac{5\lambda A(\kappa) - 2\Theta(\kappa)}{1 + \lambda} C(\kappa)\epsilon^2 [1 - \frac{9}{16}D(\lambda)C(\kappa)\epsilon] + O(\epsilon^4) \quad [52b]$$

$$K_{SF}^{131} = -\frac{3\pi}{8} \frac{5\lambda A(\kappa) - 2\Theta(\kappa)}{1 + \lambda} C(\kappa)\epsilon^2 [1 - \frac{9}{16}D(\lambda)C(\kappa)\epsilon] + O(\epsilon^4), \quad [52c]$$

which include the rotlet contribution for a *no-slip* sphere case, see [42c]. In an unbounded single fluid the net force on a drop at the center of a linear shear flow would be zero. For a fluid drop (with finite  $\kappa$ ) the force is always oriented towards the negative  $x$ -axis. Thus, the interface will induce a translation in the  $-\mathbf{e}_x$  direction parallel to the interface in the absence of an applied force  $-\mathbf{F}$ . It can be noted that a fluid drop ( $\kappa \neq \infty$ ) near a *free* surface (i.e.  $\lambda \rightarrow 0$ ) will not experience the induced translation. On the other hand, in the case of a *no-slip* sphere, the direction of the force component for shear component  $\Gamma_{13}$ , i.e.  $\mathbf{U}_2^\infty = \Gamma_{13}(z - d)\mathbf{e}_x$  depends on the viscosity ratio  $\lambda$ . Indeed when  $\lambda < 2/5$ , the induced translation is in the *positive*  $\mathbf{e}_x$  direction parallel to the interface.

A solid particle ( $\kappa = \infty$ ), in addition to the induced hydrodynamic force [52a], will experience a hydrodynamic torque in the linear shear flow, which is readily determined from the rotlet singularity, [50]:

$$\mathbf{T} = \mathbf{K}_{ST} : \boldsymbol{\Gamma}, \tag{53a}$$

where the nonzero components of the third-order hydrodynamic tensor  $\mathbf{K}_{ST}$  are given by

$$K_{ST}^{213} = 4\pi \left[ 1 - \frac{3}{8(1+\lambda)} \epsilon^3 \right] \tag{53b}$$

and

$$K_{ST}^{231} = 4\pi \left[ 1 + \frac{5\lambda + 2}{8(1+\lambda)} \epsilon^3 \right]. \tag{53c}$$

All of the preceding discussion is concerned with the force and torque on a particle in a shear flow with a stagnation at the drop center. In order to determine the force and torque when the particle is located at an arbitrary point  $\mathbf{x}_p$  in the undisturbed flow  $\mathbf{U}_2^\infty(\mathbf{x}) = \boldsymbol{\Gamma} \cdot \mathbf{x}$ , which is zero at the origin on the interface  $z = 0$  [cf. figure 2(b)], the present results, [52a] and [53a], must be combined with the corresponding result for the uniform streaming flow  $\mathbf{U}_2^\infty = \boldsymbol{\Gamma} \cdot \mathbf{x}_p$ :

$$\mathbf{F} = \mathbf{K}_T \cdot \boldsymbol{\Gamma} \cdot \mathbf{x}_p + \mathbf{K}_{SF} : \boldsymbol{\Gamma} \tag{54a}$$

and

$$\mathbf{T} = \mathbf{K}_C \cdot \boldsymbol{\Gamma} \cdot \mathbf{x}_p + \mathbf{K}_{ST} : \boldsymbol{\Gamma}, \tag{54b}$$

where  $\mathbf{K}_C$  is the coupling tensor and  $\mathbf{K}_C = \mathbf{0}$  for a fluid particle (i.e.  $\kappa \neq \infty$ ). The components of the coupling tensor  $\mathbf{K}_C$  have been determined elsewhere for a solid particle in the vicinity of a fluid interface (cf. Lee *et al.* 1979; Yang & Leal 1984).

In this case, one basis to check our solution is to compare it with the hydrodynamic torque on a *no-slip* sphere for a linear shear flow parallel to a *rigid* plane boundary  $\mathbf{U}_2^\infty(\mathbf{x}) = \Gamma_{13} z \mathbf{e}_x$ , calculated by Goldman *et al.* (1967b) and Goren & O'Neill (1971) using the eigenfunctions of Laplace's equation in bipolar coordinates. The approximate drag scaled with respect to Stokes drag in an infinite fluid is simply obtained from the present asymptotic solution [54a] and given as

$$\frac{\mathbf{F}'}{6\pi\mu_2 U_1 \Gamma_{13} da} = C(\kappa) \left\{ 1 + \sum_{n=1}^3 \left[ -\frac{9}{16} D(\lambda) C(\kappa) \epsilon \right]^n - \frac{1}{16} \cdot B(\kappa) \frac{1+2\lambda}{1+\lambda} \epsilon^3 - \frac{5\lambda A(\kappa) - 2\Theta(\kappa)}{16(1+\lambda)} \epsilon^3 \right\} \mathbf{e}_x + O(\epsilon^4) \tag{55}$$

In figure 4 the drag ratio [55] is plotted as a function of  $d$ , the dimensionless separation distance between the drop and the interface, for the same set of parameters as in figure 3. Also shown for comparison is the corresponding drag for the case of  $\lambda = \kappa \rightarrow \infty$  determined by Goldman *et al.* (1976b) and Goren & O'Neill (1979). In many respects, the results are similar to those for parallel translation of a drop obtained by Yang & Leal (1990). As mentioned previously, we presume  $\epsilon \ll 1$  in the derivation of [55]. Thus, for  $\epsilon \ll 1$  (i.e.  $d \gg 1$ ) the asymptotic solution [55] coincides almost exactly with the earlier works, which are the exact solutions for simple shear flow parallel to a *solid* wall. Even for  $d \approx 1.5$ , the approximation solution shows reasonably good agreement with the exact solutions. Indeed, for a *no-slip* sphere the relative error is within 1.38% for  $d \geq 1.5$ . It can be noted that, for all  $\kappa$ , the drag either increases or decreases relative to Stokes' drag for an unbounded fluid (depending upon the viscosity ratio  $\lambda$ ) owing to the presence of the interface. However, the drag for an inviscid gas bubble ( $\kappa \rightarrow 0$ ) does not increase (or decrease) as fast as the result for a *no-slip* sphere in the limit as  $d \rightarrow 1$ . On the other hand, as noted in figure 3, the drag for an inviscid gas bubble in a uniaxial straining flow increases *rapidly* as the bubble approaches the interface compared to the result for a *no-slip* sphere. Thus, the magnitude of the effect of the interface on drag for an inviscid gas bubble in a linear flow is *considerably* larger in motion normal to the interface than in motion parallel to the interface.

We now have a complete set of solutions for a *stationary* particle at arbitrary point  $\mathbf{x}_p$  in the linear part of the local component flows formulated in section 2. It should be noted, however, that

for a force and torque-free particle (as would be typical in trajectory calculations), the translational and rotational motion of the particle must generate Stokeslets, potential dipoles and rotlets that exactly cancel the Stokeslets [34] and [46], the potential dipoles [35] and [47] and the rotlets [50] produced at the center of a *stationary* particle in the linear flows. Thus, at  $O(\delta)$  the net disturbance velocity field due to the presence of a particle freely suspended in the linear flows corresponds to stresslet singularities, [36] and [48], and potential quadrupole singularities, [37] and [49] at the particle center  $\mathbf{x}_p$ . The stresslet velocity fields,  $\mathbf{u}_{ss}$  decay like  $1/r^2$  and the potential quadrupole velocity fields decay like  $1/r^4$  ( $r = |\mathbf{x} - \mathbf{x}_p|$ ). Thus, these singularities produce disturbance flows in the matching region with the outer solution, which are of  $O(\delta^3)$  and  $O(\delta^5)$ , respectively [since the strength of the singularities is  $O(\delta)$ ]. Thus, the first nonzero correction to the outer solution would appear to be  $O(\delta^3)$ , as was, in fact, suggested earlier via an overall force balance argument.

#### 4. SOLUTION AT $O(\delta^2)$

For a solid collector (i.e.  $\lambda = \infty$ ), the first component of the undisturbed flow, [17] and [18], which produces a contribution in the inner region that forces the particle toward the interface, occurs at  $O(\delta^2)$ . Thus, to obtain accurate trajectories over the whole range of possible viscosity ratios for the collector, we must consider the  $O(\delta^2)$  inner problem.

In the context of the solution procedure outlined above, contributions to the inner problem at  $O(\delta^2)$  come from two sources. One is the  $O(\delta^2)$  terms in the undisturbed outer flow, given by [17] and [18]. We may note, as indicated above, that the first two terms in the inner expansion at  $O(1)$  and  $O(\delta)$  produce no disturbance flow contribution in the matching region that is larger than  $O(\delta^3)$ , and thus the outer flow solution is just the undisturbed flow [17] and [18] through terms of  $O(\delta^2)$ . The second contribution to the inner disturbance flow at  $O(\delta^2)$  is generated by the nonhomogeneous terms that arise from the domain perturbation approximation of the boundary conditions at the large drop surface. The specific form of these nonhomogeneous boundary terms is obtained by substituting the solutions for the  $O(1)$  and  $O(\delta)$  velocity and stress fields in the inner region into the domain perturbation terms at  $O(\delta^2)$ ; e.g. the zero normal velocity condition [15], which becomes

$$\mathbf{u}_j^{(2)} \cdot \mathbf{e}_z + x \mathbf{u}_j^{(1)} \cdot \mathbf{e}_x + y \mathbf{u}_j^{(1)} \cdot \mathbf{e}_y - \frac{1}{2}(x^2 + y^2) \frac{\partial}{\partial z} (\mathbf{u}_j^{(1)} \cdot \mathbf{e}_z) = 0 \quad \text{at } z = 0; \quad [56a]$$

the continuity of velocity condition, which becomes

$$\left[ \mathbf{u}^{(2)} - \frac{1}{2}(x^2 + y^2) \frac{\partial \mathbf{u}^{(1)}}{\partial z} \right] = 0 \quad \text{at } z = 0; \quad [56b]$$

and the continuity of tangential stress condition, which becomes

$$\left[ \sigma_{zx}^{(2)} + x \sigma_{xx}^{(1)} + y \sigma_{yx}^{(1)} - x \sigma_{zz}^{(1)} - \frac{1}{2}(x^2 + y^2) \frac{\partial \sigma_{zx}^{(1)}}{\partial z} \right] = 0 \quad \text{at } z = 0 \quad [56c]$$

and

$$\left[ \sigma_{zy}^{(2)} + x \sigma_{xy}^{(1)} + y \sigma_{yy}^{(1)} - y \sigma_{zz}^{(1)} - \frac{1}{2}(x^2 + y^2) \frac{\partial \sigma_{zy}^{(1)}}{\partial z} \right] = 0 \quad \text{at } z = 0. \quad [56d]$$

Thus, in the boundary conditions at  $O(\delta^2)$ , inhomogeneous contributions occur due to the  $O(\delta)$  disturbance flows produced by the singularities in [36], [37], [48] and [49]. To reduce the complexity of the individual problems, we will consider each of the two types of  $O(\delta^2)$  contributions separately.

##### 4.1. Quadratic flows arising from the undisturbed outer flows at $O(\delta^2)$

First, we will consider the disturbance flows for a stationary particle in the  $O(\delta^2)$  flows that arise from matching with [17] and [18]. Since the undisturbed flows [17] and [18] satisfy the boundary conditions at  $z = 0$ , the disturbance flows are required to satisfy the boundary conditions on the surface of a stationary particle and on the interface  $z = 0$  (i.e. continuity of tangential velocity and stress and zero normal velocity on both the surfaces), in addition to the requirement that the disturbance velocity decay to zero as  $|\mathbf{x}| \rightarrow \infty$ , so that the net velocity matches the outer solution.



Thus, we follow the same method as used in the previous section to obtain solutions for the net disturbance singularities at the particle center.

*4.1.1. Axisymmetric paraboloidal and stagnation flows.* The first case that we consider is the flow with an axisymmetric paraboloidal and stagnation-like velocity profile with vanishing velocity at the interface:

$$\mathbf{U}_2^\infty(\mathbf{x}) = K_1(xz\mathbf{e}_x + yz\mathbf{e}_y - z^2\mathbf{e}_z) + K_2(x^2 + y^2)\mathbf{e}_z \quad [57]$$

in which the flow parameters are

$$K_1 = -\frac{\cos\theta(2-3\lambda)}{2} \frac{\delta^2}{1+\lambda} \quad \text{and} \quad K_2 = -\frac{\cos\theta}{1+\lambda} \delta^2,$$

see figures 2(c, d). This problem can be treated, as in the preceding cases, by decomposing the undisturbed flow into a simple uniform streaming flow  $\mathbf{U}_2^\infty(\mathbf{x}) = -K_1 d^2 \mathbf{e}_z$ , a linear extensional flow with vanishing velocity at the particle center  $\mathbf{U}_2^\infty(\mathbf{x}) = K_1 d \mathbf{E} \cdot (\mathbf{x} - \mathbf{x}_p)$  and the remaining quadratic flows with a stagnation point at the particle center. The first two problems were treated in our earlier publication, Yang & Leal (1990), and in the previous section 3.1, respectively.

The problem of a particle in the quadratic flows with a stagnation at the *particle* center can be solved for an *unbounded* domain (with no interface) utilizing the fundamental singularity solutions of Chwang & Wu (1975). In particular, we can show that a Stokeslet, a Stokes quadrupole, a potential dipole and a potential octupole, of the form

*Stokeslet,*

$$\frac{B(\kappa)}{4} (K_1 - 2K_2) \mathbf{u}_S(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z), \quad [58a]$$

*Stokes quadrupole,*

$$\frac{2K_1 + K_2}{24} \cdot \frac{2 + 7\kappa}{1 + \kappa} \frac{\partial^2}{\partial z^2} \mathbf{u}_S(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z), \quad [58b]$$

*potential dipole,*

$$\frac{1}{12} \left( K_1 \frac{2 + \kappa}{1 + \kappa} - K_2 \frac{2 - 5\kappa}{1 + \kappa} \right) \mathbf{u}_D(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z) \quad [58c]$$

and

*potential octupole,*

$$-\frac{B(\kappa)}{24} (2K_1 + K_2) \frac{\partial^2}{\partial z^2} \mathbf{u}_D(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z), \quad [58d]$$

are required at the particle center to satisfy the boundary conditions, i.e. continuity of tangential velocity and stress and zero normal velocity at the particle surface. Since the interface reflection in Lee *et al.*'s (1979) reflection technique generates a mismatch at the particle surface, we again require additional singularities at the particle center to match the reflected flow field. Following the preceding analysis, we examine the leading terms of the reflected field at the particle surface as a power series in  $\epsilon$ :

$$\frac{3}{8} (K_1 - 2K_2) C(\lambda) B(\kappa) [-\epsilon \mathbf{e}_z - \epsilon^2 \mathbf{E} \cdot (\mathbf{x} - \mathbf{x}_p) + O(\epsilon^3)]. \quad [59]$$

In order to satisfy the boundary condition at all points of the particle surface, additional singularities are required at the particle center. These can be determined from the corresponding solutions for a surface velocity distribution of the form [59] in an unbounded domain. In particular, the Stokeslet contribution is sufficient to evaluate the hydrodynamic force exerted on the particle. The result is

$$\mathbf{F} = -2\pi B(\kappa) (K_1 - 2K_2) \left\{ \sum_{n=0}^2 [\frac{3}{8} C(\lambda) C(\kappa) \epsilon]^n \mathbf{e}_z + O(\epsilon^3) \right\}. \quad [60]$$

Because of symmetry there is no torque exerted on the particle by these flows. The existence of the hydrodynamic force  $\mathbf{F}$ , [60], implies that a freely suspended particle located at the center of the quadratic stagnation and paraboloidal flows will move away from the stagnation point along the streamline that would pass through this point in the absence of the particle. However, it can

be noted that an inviscid gas bubble [i.e.  $\kappa \rightarrow 0$  and thus  $B(\kappa) = 0$ ] will not experience any hydrodynamic drag and thus remains at the stagnation point of the flows.

It is a simple matter to calculate the hydrodynamic drag force on the particle immersed in the axisymmetric paraboloidal and stagnation flows of [57] with a stagnation point at the *interface* by a superposition of the corresponding solutions for a uniform streaming flow, a uniaxial extensional flow and the solution [60] for the quadratic flows with vanishing velocity at the particle center. The resulting expression for the drag

$$\frac{\mathbf{F}'}{6\pi\mu_2 U_1 K_1 d^2 a} = C(\kappa) \left\{ \sum_{n=0}^3 \left[ \frac{9}{8} C(\lambda) C(\kappa) \epsilon \right]^n - \frac{1}{8} B(\kappa) \frac{1 + 4\lambda}{1 + \lambda} \epsilon^3 - \frac{15}{8} A(\kappa) C(\lambda) \epsilon^3 \right. \\ \left. + \frac{1}{3} \frac{K_1 - 2K_2}{K_1} \frac{B(\kappa)}{C(\kappa)} \epsilon^2 \left[ 1 + \frac{9}{8} C(\lambda) C(\kappa) \epsilon \right] \right\} \mathbf{e}_z + O(\epsilon^4). \quad [61]$$

This can be compared directly with the corresponding result from Goren & O'Neill's (1971) exact solution for a *no-slip* small particle (i.e.  $\kappa \rightarrow \infty$ ) in the presence of a nearby *rigid* "collector" (i.e.  $\lambda \rightarrow \infty$ ). As shown in figure 5, the approximate solution gives a remarkably accurate representation of the exact result, over almost the whole range of possible particle positions. Although the discrepancy between the two solutions become larger as  $d \rightarrow 1$ , it still remains relatively small (e.g. the relative error at  $d = 1.01$  is only 2.40% and the error is within 0.90% for  $d > 1.5$ ).

4.1.2. *Quadratic shear flows.* A second quadratic flow problem that appears from matching the undisturbed flow, [17] and [18], is the steady quadratic shear flows past a fluid particle near a plane fluid interface. In this problem, the fluid velocity at infinity is

$$\mathbf{U}_2^\infty(\mathbf{x}) = (K_3 y^2 + K_4 z^2) \mathbf{e}_x, \quad [62]$$

in which the flow parameters are

$$K_3 = \delta^2 \frac{\sin \theta}{4} \frac{1 + 3\lambda}{1 + \lambda} \quad \text{and} \quad K_4 = -\delta^2 \frac{\sin \theta}{4} \frac{2 + 9\lambda}{1 + \lambda},$$

see figure 2(e). A general solution for this problem can be obtained by superimposing the results for a uniform streaming flow with velocity  $\mathbf{U}_2^\infty(\mathbf{x}) = K_4 d^2 \mathbf{e}_x$ , a linear shear flow  $\mathbf{U}_2^\infty(\mathbf{x}) = 2K_4 d(z - d) \mathbf{e}_x$  and, finally, a quadratic shear flow with a stagnation point at the particle center.

First, we consider the case of a particle in the quadratic flow which vanishes at the particle center. It can be demonstrated that a Stokeslet, a potential dipole, a Stokes quadrupole and a potential octupole are necessary to produce such a flow in an unbounded fluid. However, as usual, the reflected velocity field from the interface does not satisfy boundary conditions at  $O(\epsilon)$  on the

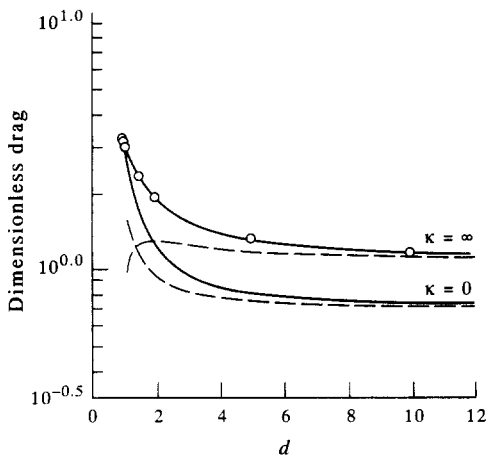


Figure 5. Dimensionless drag of [61] as a function of the dimensionless distance,  $d$ , between the particle and the interface:  $\mathbf{U}_2^\infty = K_1(xz \mathbf{e}_x + yz \mathbf{e}_y - z^2 \mathbf{e}_z) + K_2(x^2 + y^2) \mathbf{e}_z$ ; — for  $\lambda = \infty$ , --- for  $\lambda = 0$  and  $\circ$  for the corresponding exact-solution results ( $\lambda = \kappa = \infty$ ) of Goren & O'Neill (1971).

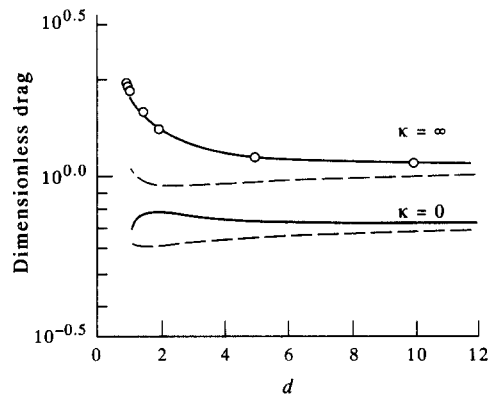


Figure 6. Dimensionless drag of [65] as a function of the dimensionless distance,  $d$ , between the particle and the interface:  $\mathbf{U}_2^\infty = K_4 z^2 \mathbf{e}_x$ ; — for  $\lambda = \infty$ , --- for  $\lambda = 0$  and  $\circ$  for the corresponding exact-solution results ( $\lambda = \kappa = \infty$ ) of Goren & O'Neill (1971).

particle surface, and thus additional singularities are required at the center of the particle. We may examine the leading terms of this reflected field, expressed as a power series in  $\epsilon$ :

$$-\frac{1}{4}(K_3 + K_4)B(\kappa)\left\{\frac{3}{4}\epsilon \cdot D(\lambda)\mathbf{e}_x - \frac{3}{8}\epsilon^2 \cdot [D(\lambda)(z-d)\mathbf{e}_x - C(\lambda)x\mathbf{e}_z] + O(\epsilon^3)\right\}. \quad [63]$$

The required additional singularities to match the corresponding reflected flows at the particle surface are determined using the approach outlined in the previous example. The hydrodynamic force and torque on the particle in the quadratic flow with vanishing velocity at the particle center can be evaluated from the resulting Stokeslet and rotlet contributions:

$$\mathbf{F} = 2\pi B(\kappa)(K_3 + K_4)\left\{\sum_{n=0}^2 \left[-\frac{9}{16}D(\lambda)C(\kappa)\epsilon^n + O(\epsilon^3)\right]\right\}\mathbf{e}_x \quad [64a]$$

$$\mathbf{T} = \frac{\pi}{2}B(\kappa)\Theta(\kappa)(K_3 + K_4)\left[\epsilon^2 \frac{1}{1+\lambda} + O(\epsilon^3)\right]\mathbf{e}_y. \quad [64b]$$

These are the negatives of the force and torque that are required to keep the particle from translating and rotating at the stagnation point. In can be seen that for an inviscid gas particle  $\mathbf{F} = \mathbf{T} = \mathbf{0}$ .

Goren & O'Neill (1971) determined the exact solution for a solid *no-slip* sphere (i.e.  $\kappa \rightarrow \infty$ ) in the flow field, which in the absence of the sphere is a combination of a uniform streaming, and linear and parabolic shear flows with spatial dependence on  $z$ , near a *rigid* plane wall (i.e.  $K_3 = 0$  in the problem of Goren & O'Neill). It is a simple matter to calculate the approximate drag force on the fluid particle immersed in the undisturbed flow, [62], from the present asymptotic solution [64], combined with the corresponding solutions for the uniform streaming and linear shear flows. The drag ratio is simply given as:

$$\begin{aligned} \frac{\mathbf{F}'}{6\pi\mu_2 U_i K_4 d^2 a} = C(\kappa)\left\{\sum_{n=0}^3 \left[-\frac{9}{16}D(\lambda)C(\kappa)\epsilon^n - \frac{1}{16}B(\kappa)\frac{1+2\lambda}{1+\lambda}\epsilon^3 - \frac{5\lambda A(\kappa) - 2\Theta(\kappa)}{8(1+\lambda)}\epsilon^3\right] \right. \\ \left. + \frac{K_3 + K_4}{3K_4} \cdot \frac{B(\kappa)}{C(\kappa)}\epsilon^2 \left[1 - \frac{9}{16}D(\lambda)C(\kappa)\epsilon\right]\right\}\mathbf{e}_x + O(\epsilon^4). \quad [65] \end{aligned}$$

In figure 6 the drag ratio [65] is plotted, for  $K_3 = 0$ , as a function of  $d$ , the dimensionless distance between the particle and the interface, for the same set of parameters as in figure 5. The "exact" results calculated numerically by Goren & O'Neill (1971) are also shown in the figure. There is very good agreement between the two solutions, except in the region near  $d \approx 1$ . As expected, the discrepancy between the two results becomes larger as the particle approaches the interface. However, a detailed comparison shows that, even for  $d \approx 2$ , there is very good agreement between the two solutions, and the relative error in the asymptotic solution, [65], compared to the exact solution of Goren & O'Neill, is  $< 1.74\%$  for  $d \geq 1.5$ . For particle separations from the interface exceeding 2 or 3 particle radii, the existence of a critical viscosity ratio  $\lambda$  separating cases of increasing or decreasing drag, evident in figure 6, is similar to the result of Yang & Leal (1990) for parallel translation of a drop near a flat interface. As suggested from the results for the linear flows (e.g. figures 3 and 4 in section 3), the variation in the hydrodynamic resistances acting on an *inviscid* gas bubble in the quadratic flow is much more sensitive to the separation distance  $d$  for normal motion than for parallel motion relative to the interface, cf. figures 5 and 6.

**4.1.3. Nonaxisymmetric quadratic stagnation flow.** Finally, we consider a nonaxisymmetric quadratic stagnation flow

$$\mathbf{U}_2^\infty(\mathbf{x}) = \frac{1}{2}(K_5 + K_6)x^2\mathbf{e}_x - K_5xy\mathbf{e}_y - K_6xz\mathbf{e}_z, \quad [66]$$

where

$$K_5 = \delta^2 \cdot \frac{\sin \theta}{2(1+\lambda)} \quad \text{and} \quad K_6 = -\delta^2 \cdot \frac{2-3\lambda}{2(1+\lambda)} \sin \theta,$$

see figure 2(f). This is the remaining contribution at  $O(\delta^2)$  from matching with the undisturbed flow, [17] and [18], in the outer region. This problem can also be treated conveniently by decomposing the undisturbed flow with vanishing velocity at the origin on the interface  $z = 0$  into a linear shear flow  $\mathbf{U}_2^\infty(\mathbf{x}) = -K_6 dx\mathbf{e}_z$  and a quadratic flow with stagnation point at the particle center. The former problem was solved in section 3. The *unbounded-domain* solution for the latter

problem can be shown to be represented by a Stokeslet, a potential dipole, a Stokes quadrupole and a potential octupole. The Stokeslet is required to produce a drag, and the potential dipole is associated with it to account for the body-thickness effect. We also require the Stokes quadrupole and the potential octupole to balance the power-law variations of the solution in  $r (= |\mathbf{x} - \mathbf{x}_p|)$ . In this case, the leading terms of the mismatch generated by the reflected velocity field at the particle surface are:

$$\mathbf{u}_2^{(1)}(\mathbf{x}) = -\frac{1}{8}(K_5 + K_6)B(\kappa)\left\{\frac{3}{4}\epsilon \cdot D(\lambda)\mathbf{e}_x - \frac{3}{8}\epsilon^2 \cdot [D(\lambda)(z - d)\mathbf{e}_x - C(\lambda)x\mathbf{e}_z] + O(\epsilon^3)\right\}. \quad [67]$$

The additional singularities needed to match those interface reflections are those for a uniform streaming flow at  $O(\epsilon)$  parallel to the interface, and a linear shear flow at  $O(\epsilon^2)$  either normal or parallel to the interface. The hydrodynamic force and torque acting on the particle at the center of the quadratic stagnation flow can be readily evaluated from the resulting Stokeslet and rotlet contributions:

$$\mathbf{F} = \pi(K_5 + K_6)B(\kappa)\left\{\sum_{n=0}^2 \left[-\frac{9}{16}D(\lambda)C(\kappa)\epsilon^n + O(\epsilon^3)\right]\right\} \quad [68a]$$

and

$$\mathbf{T} = \frac{\pi}{4}(K_5 + K_6)B(\kappa)\Theta(\kappa)\left[\epsilon^2 \frac{1}{1 + \lambda} + O(\epsilon^3)\right]\mathbf{e}_y. \quad [68b]$$

We now have a complete solution for a fluid particle in the quadratic stagnation flow with vanishing velocity at the particle center. Combining the results of the present solution [68a,b] with those for the simple shear flow  $\mathbf{U}^\infty(\mathbf{x}) = -K_6 dx\mathbf{e}_z$ , we can obtain directly the hydrodynamic force and torque on the particle in the undisturbed flow centered at the origin defined in [67].

This completes our calculations of the disturbance flows for a stationary particle in the  $O(\delta^2)$  flows from matching with [17] and [18]. As we noted earlier, however, there remain contributions from the inhomogeneous boundary conditions at  $O(\delta^2)$  due to the domain-perturbation approximation [56a–d] at the interface: these consist of terms from the stresslet and potential quadrupole singularities, [36], [37], [48] and [49] in the  $O(\delta)$  disturbance flows.

#### 4.2. Nonhomogeneous contributions from the domain perturbation

Thus, we now consider the additional  $\mathbf{u}_2^{(2)}$  flows at  $O(\delta^2)$  that arise from the domain perturbation conditions [56a–d] when the nonhomogeneous terms are evaluated using the  $O(\delta)$  disturbance flow solutions that are represented by the singularities in [36], [37], [48] and [49]. We thus require solutions that satisfy inhomogeneous boundary conditions at the interface  $z = 0$  from [56a–d] and the boundary conditions on the surface of a stationary particle, but vanish at “infinity”. This velocity field, generated by nonhomogeneous terms on the plane  $z = 0$ , is in some aspects, similar to the corrections associated with application of the original boundary conditions on the flat interface. The solution technique is similar to the method used to derive the general lemma in Lee *et al.* (1979). The problems, corresponding to each term in [36], [37], [48] and [49], are repetitive and exceedingly tedious; thus, only one example will be worked through in detail here. This is the velocity field  $\mathbf{u}_2^{(2)}$  corresponding to the stresslet normal to the interface  $\mathbf{u}_{SS}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_z)$  in [36]. For simplicity of presentation, we drop the multiplicative coefficient that appears in [36], since the problem is linear and we can multiply by any constant at the end. Upon substitution into [56a–d], the “normalized” stresslet velocity field  $\mathbf{u}_{SS}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_z)$  produces the following nonhomogeneous boundary conditions at  $O(\epsilon^3)$  on the interface  $z = 0$ :

$$\mathbf{u}_2^{(2)} \cdot \mathbf{e}_z = \mathbf{u}_2^{(2)} \cdot \mathbf{e}_z = -\frac{\delta(x^2 + y^2)}{1 + \lambda} \epsilon^3 + O(\epsilon^4), \quad [69a]$$

$$\llbracket \sigma_{xz}^{(2)} \rrbracket = \frac{24(1 - \lambda)x}{1 + \lambda} \epsilon^3 + O(\epsilon^4), \quad [69b]$$

$$\llbracket \sigma_{yz}^{(2)} \rrbracket = \frac{24(1 - \lambda)y}{1 + \lambda} \epsilon^3 + O(\epsilon^4) \quad [69c]$$

and

$$\llbracket \mathbf{u}^{(2)} \rrbracket = O(\epsilon)^4. \quad [69d]$$

As stated above, we require a solution that satisfies [69a–d], the boundary conditions on the surface of a stationary fluid particle (i.e continuity of tangential velocity and stress and zero normal velocity), and decays to zero as one moves far from the interface, i.e. into the outer region.

To solve the above problem, we assume a solution of the form

$$\mathbf{u}_1 = \frac{8}{(1 + \lambda)(z - d)^3} [xz \mathbf{e}_x + yz \mathbf{e}_y + (x^2 + y^2) \mathbf{e}_z] + zG_1(\mathbf{x}) \mathbf{e}_z + z^2 \mathbf{H}_1(\mathbf{x}) \tag{70a}$$

and

$$\mathbf{u}_2 = -\frac{8}{(1 + \lambda)(z + d)^3} [xz \mathbf{e}_x + yz \mathbf{e}_y + (x^2 + y^2) \mathbf{e}_z] + zG_2(\mathbf{x}) \mathbf{e}_z + z^2 \mathbf{H}_2(\mathbf{x}), \tag{70a}$$

which will still satisfy the normal velocity condition [69a] and the continuity of tangential stress on the interface  $z = 0$ . From the continuity equation and the symmetry of the problem, appropriate choices for the unknown functions  $G_j$  and  $\mathbf{H}_j$  are made. The base velocity field, which vanishes at infinity and satisfies boundary conditions [69a–d], is found to be

$$\begin{aligned} \mathbf{u}_1 = & \frac{8}{(1 + \lambda)(z - d)^3} \left[ xz \mathbf{e}_x + yz \mathbf{e}_y + \frac{(x^2 + y^2)(4z - d)}{z - d} \mathbf{e}_z \right] \\ & - \frac{12z^2}{(1 + \lambda)(z - d)^4} \left\{ \left[ x - \frac{5(x^2 + y^2)x}{(z - d)^2} \right] \mathbf{e}_x + \left[ y - \frac{5(x^2 + y^2)y}{(z - d)^2} \right] \mathbf{e}_y \right\} \\ & - \frac{8z^2}{(1 + \lambda)(z - d)^3} \left[ 1 - \frac{6(x^2 + y^2)}{(z - d)^2} \right] \mathbf{e}_z \end{aligned} \tag{71a}$$

$$\begin{aligned} \mathbf{u}_2 = & -\frac{8}{(1 + \lambda)(z + d)^3} \left[ xz \mathbf{e}_x + yz \mathbf{e}_y + \frac{(x^2 + y^2)(4z + d)}{z + d} \mathbf{e}_z \right] \\ & - \frac{12z^2}{(1 + \lambda)(z + d)^4} \left\{ \left[ x - \frac{5(x^2 + y^2)x}{(z + d)^2} \right] \mathbf{e}_x + \left[ y - \frac{5(x^2 + y^2)y}{(z + d)^2} \right] \mathbf{e}_y \right\} \\ & - \frac{8z^2}{(1 + \lambda)(z + d)^3} \left[ 1 - \frac{6(x^2 + y^2)}{(z + d)^2} \right] \mathbf{e}_z. \end{aligned} \tag{71b}$$

This flow field does *not* satisfy boundary conditions on the surface of the particle (i.e. continuity of tangential velocity and stress and zero normal velocity). Hence, the next step is to add a disturbance velocity field so that the sum satisfies the boundary conditions on the surface of the fluid particle. This is accomplished by expanding the above flow in a Taylor series expansion about the center of the particle for small  $\epsilon$ , and introducing singularities at the particle center to match the component flows from this expansion at the sphere surface. These singularities are then reflected from the interface, using the relations of Lee *et al.* (1979). The resulting force arising from the domain perturbation of the net stresslet normal to the interface at  $O(\delta)$  (i.e. including the coefficient from [36]) is

$$\mathbf{F} = -\pi\delta^2 \frac{15 \cos \theta}{2(1 + \lambda)^2} A(\kappa)C(\kappa) \left[ \epsilon + \frac{2}{3}C(\lambda)C(\kappa)\epsilon^2 + O(\epsilon^3) \right] \cdot \mathbf{e}_z. \tag{72}$$

It will be noted that the  $O(\epsilon^3)$  contribution from the stresslet  $\mathbf{u}_{SS}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_z)$  to the boundary conditions [69a–d] requires a flow field that produces an  $O(\epsilon)$  force on the particle. This appears surprising at first, since an  $\epsilon^2$  point force at the particle center will produce an  $\epsilon^3$  normal velocity on the interface. However, the  $\epsilon^3$  stress difference on the interface, [69b], requires an  $\epsilon^2$  tangential velocity on the interface. To satisfy the continuity equations, a component of the normal velocity, although zero on the interface, has a strength of  $O(\epsilon)$  at the particle center. Thus, the contribution to the force on the particle is  $O(\epsilon)$ . Following the same line of reasoning, it is apparent that the  $O(\epsilon^4)$  contribution from the stresslet through the domain perturbation conditions may contribute to the force at  $O(\epsilon^2)$ , while the  $O(\epsilon^5)$  term may contribute to the force at  $O(\epsilon^3)$ . Thus, to obtain the complete force expression to  $O(\epsilon^2)$ , as is required to be compatible with the force contributions calculated earlier at  $O(\delta^2)$  from the match with the undisturbed outer flow, it is necessary to include

the stresslet contributions to the domain perturbation conditions through  $O(\epsilon^4)$ . A stresslet normal to the interface,  $\mathbf{u}_{\text{SS}}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_z)$ , generates the following boundary conditions at  $O(\epsilon^4)$ :

$$\mathbf{u}_1^{(2)} \cdot \mathbf{e}_z = \mathbf{u}_2^{(2)} \cdot \mathbf{e}_z = 0 + O(\epsilon^5), \quad [73a]$$

$$[\sigma_{zx}^{(2)}] = O(\epsilon^5), \quad [73b]$$

$$[\sigma_{zy}^{(2)}] = O(\epsilon^5) \quad [73c]$$

and

$$[\mathbf{u}^{(2)}] = \frac{18(1-\lambda)}{1+\lambda} (x^2 + y^2)(x\mathbf{e}_x + y\mathbf{e}_y)\epsilon^4 + O(\epsilon^5). \quad [73d]$$

The  $O(\epsilon^4)$  terms can be shown to yield a net force of  $O(\epsilon^3)$  on a stationary fluid particle. It is obvious that the hydrodynamic torque is zero in this case due to the symmetry of the stresslet  $\mathbf{u}_{\text{SS}}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_z)$ .

The other contributions from the singularities [37], [48] and [49] can be treated in a similar manner. The net force and torque on a stationary fluid particle arising from the stresslet,  $\mathbf{u}_{\text{SS}}(\mathbf{x}, \mathbf{x}_p, \mathbf{e}_x, \mathbf{e}_z)$  of [48], are

$$\mathbf{F} = O(\epsilon^3\delta^2)\mathbf{e}_x \quad [74a]$$

and

$$\mathbf{T} = \pi\delta^2 \frac{15 \sin \theta \lambda (5\lambda + 6)}{8(1 + \lambda)^2} A(\kappa)\Theta(\kappa)[\epsilon^2 + O(\epsilon^3)]\mathbf{e}_y. \quad [74b]$$

The net forces on a stationary particle arising from the potential quadrupoles,  $\mathbf{u}_{\text{PQ}}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_z, \mathbf{e}_z)$  of [37] and  $\mathbf{u}_{\text{PQ}}(\mathbf{x}, \mathbf{x}_p; \mathbf{e}_x, \mathbf{e}_z)$  of [49], are both  $O(\epsilon^3\delta^2)$  and the hydrodynamic torques are zero and  $O(\epsilon^4\delta^2)$ , respectively.

We now have a complete set of solutions for a fluid particle located at an arbitrary position relative to the larger drop. These solutions provide the necessary relationships between the various flow parameters, and the hydrodynamic force and torque for calculation of particle trajectories in the vicinity of the collector, which we shall consider in the following section.

## 5. TRAJECTORY CALCULATIONS

Let us now turn to considering the trajectory of a small fluid particle in the slow streaming motion past a large spherical drop. It has been shown in section 2 that the appropriate trajectory equation can be derived by using an asymptotic model in which the small fluid particle is in a semi-infinite fluid that is bounded by a fluid interface that appears planar at the leading order of approximation, and which undergoes a flow at infinity that is determined from the first few terms of the classical Hadamard–Rybczynski solution in an asymptotic expansion about the nearest point on the interface to the small particle for  $\delta (\equiv a/A) \ll 1$ .

Whenever the creeping motion approximation is applicable, general relationships can be written between the force on a fluid particle and its translational velocity as follows:

$$\mathbf{U} = \mathbf{M} \cdot [\mathbf{F}_t - \mathbf{K}_c^t \cdot \mathbf{K}_R^{-1} \cdot \mathbf{T}_t], \quad [75a]$$

where  $\mathbf{F}_t$  and  $\mathbf{T}_t$  are the total net force and torque acting on the particle, respectively, and  $\mathbf{M}$  is the so-called mobility tensor defined by

$$\mathbf{M} = [\mathbf{K}_T - \mathbf{K}_c^t \cdot \mathbf{K}_R^{-1} \cdot \mathbf{K}_c] \quad [75b]$$

in terms of the translational resistance tensor  $\mathbf{K}_T$ , the rotation tensor  $\mathbf{K}_R$  and the coupling tensor  $\mathbf{K}_c$ . The components of these tensors were evaluated by Yang & Leal (1990) for a spherical fluid drop in the vicinity of a plane fluid interface.

In the preceding analysis, we have evaluated the hydrodynamic force  $\mathbf{F}$  and torque  $\mathbf{T}$  acting on a *stationary* fluid particle, as a function of its position relative to the interface, due to the local component flows associated with the undisturbed Hadamard–Rybczynski flow around the collector drop. If external forces are acting on the particle these must be included in the total force,  $\mathbf{F}_t$ . One

such force is the buoyancy of the particle. The nondimensional buoyant force acting on the particle is

$$\mathbf{F}_B = \frac{4}{3}\pi a^3(\rho_3 - \rho_2)\mathbf{g} / \mu_2 U_t a. \quad [76a]$$

After substituting for the terminal velocity of a large drop in an unbounded fluid, the buoyancy force in terms of inner variables is

$$\mathbf{F}_B = 6\pi \frac{(\rho_3 - \rho_2)}{(\rho_2 - \rho_1)} C(\lambda) \delta^2 [\sin \theta \mathbf{e}_\theta - \cos \theta \mathbf{e}_r], \quad [76b],$$

which is  $O(\delta^2)$ . Similarly, approximate forms for the unretarded London attractive force between a particle and a plane wall, as in Spielman & Cukor (1973), must also be included at  $O(\delta^2)$  in terms of the inner variables if they are to be treated in the analysis; i.e.

$$\mathbf{F}_L = -\frac{3QC(\lambda)}{a^4 g(\rho_2 - \rho_3)} \cdot \frac{\delta^2}{(d^2 - 1)^2} \cdot \mathbf{e}_r, \quad [77]$$

where  $Q$  is the Hamaker constant, characteristic of the attractive forces between the two bodies. Also, double-layer forces can be included using the approximate relations of Spielman & Cukor (1973):

$$\mathbf{F}_{DL} = \frac{9D_e \zeta_p \zeta_d \tau C(\lambda)}{2a^4 g(\rho_2 - \rho_1)} \cdot \frac{e^{-\tau(d-1)}}{[1 \pm e^{-\tau(d-1)}]} \cdot \delta^2 \mathbf{e}_r, \quad [78]$$

where  $D_e$ ,  $\zeta_p$ ,  $\zeta_d$  and  $\tau$  are the outer fluid dielectric constant, the  $\zeta$ -potentials for the particle and the large drop and the dimensionless reciprocal double-layer thickness.

In the present study, we consider a particle which is free of any external force or torque (i.e.  $\mathbf{F}_t = \mathbf{F}$  and  $\mathbf{T}_t = \mathbf{T}$ ). In this case, given the initial position of the particle, [75a, b] provide its complete trajectory, with the hydrodynamic force and torque ( $\mathbf{F}$  and  $\mathbf{T}$ ) obtained by superimposing the results for each component flow. It is a simple matter to show that the velocity of the particle  $\mathbf{U} = U_r \mathbf{e}_r + U_\theta \mathbf{e}_\theta$  is given by

$$U_r = -\cos \theta \left\{ \delta \cdot \frac{1}{1+\lambda} \left[ d - \frac{15}{16} C(\lambda) A(\kappa) \frac{1}{d^2} + O\left(\frac{1}{d^4}\right) \right] + \delta^2 \cdot \left[ -\frac{3}{2} D(\lambda) d^2 + \frac{1}{2} \frac{B(\kappa)}{C(\kappa)} C(\lambda) \right. \right. \\ \left. \left. + \frac{45}{16} D(\lambda) C(\lambda) A(\kappa) \frac{1}{d} - \frac{5A(\kappa)}{4(1+\lambda)^2} \frac{1}{d} + O\left(\frac{1}{d^3}\right) \right] \right\} + O(\delta^2) \quad [79a]$$

and

$$U_\theta = \sin \theta \left( \frac{1}{2(1+\lambda)} + \delta \cdot \left\{ \frac{1}{2} \cdot \frac{1+3\lambda}{1+\lambda} d - \frac{15}{32} [B(\lambda)]^2 A(\kappa) \frac{1}{d^2} + O\left(\frac{1}{d^4}\right) \right\} \right. \\ \left. + \delta^2 \cdot \left\{ -\frac{1}{4} \cdot \frac{2+9\lambda}{1+\lambda} d^2 - \frac{1}{4} \frac{B(\kappa)}{C(\kappa)} C(\lambda) + \frac{15}{8} [B(\lambda)]^2 A(\kappa) \frac{1}{d} + O\left(\frac{1}{d^3}\right) \right\} \right) + O(\delta^3), \quad [79b]$$

in which the velocity components,  $U_r$  and  $U_\theta$ , are normal and tangential to the surface of the larger drop, respectively.

First, we begin with the special case of a small fluid particle near a large *no-slip* collector sphere (i.e.  $\lambda \rightarrow \infty$ ). Clearly, in the absence of the hydrodynamic interaction between the particle and the solid collector,

$$U_r^\infty \approx -\frac{3}{2} \delta^2 d^2 \cos \theta \quad [80a]$$

and

$$U_\theta^\infty \approx \frac{3}{2} \delta d \sin \theta (1 - \frac{3}{2} \delta d), \quad [80b]$$

which are the leading order forms for the velocity components in the asymptotic limit,  $\delta \rightarrow 0$ , in which the particle reduces to a material point. However, owing to the presence of hydrodynamic interactions between the particle and the collector, both of the velocity components of the particle decrease more rapidly as it approaches the surface of the collector than they would with no interaction. This is illustrated in figures 7 and 8, where the velocity components  $U_r/U_r^\infty$  and  $U_\theta/U_\theta^\infty$  are given as a function of the dimensionless separation distance  $d$  between the particle and the

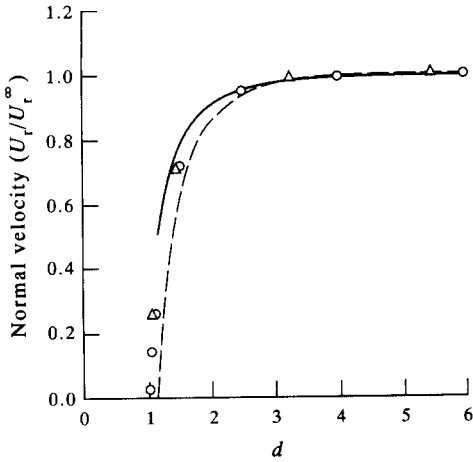


Figure 7. Dimensionless normal velocity,  $U_r/U_r^\infty$ , as a function of the dimensionless separation distance,  $d$ , when  $\delta = a/A = 0.1$ : — for  $\kappa = 0$  and --- for  $\kappa = \infty$ ,  $\circ$  and  $\triangle$  are the corresponding exact-solution results ( $\lambda = \kappa = \infty$ ) of Goren & O'Neill (1971) and Jeffery & Onishi (1984), respectively.

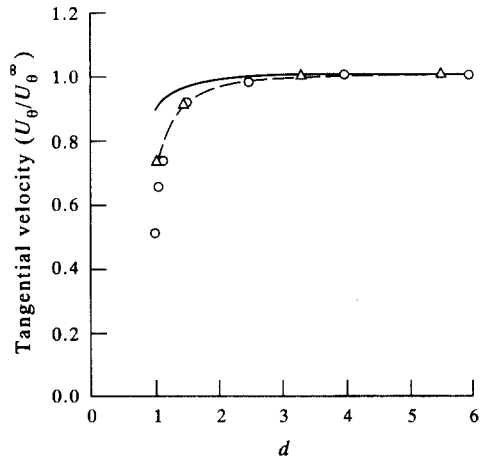


Figure 8. Dimensionless tangential velocity,  $U_\theta/U_\theta^\infty$ , as a function of the dimensionless separation distance,  $d$ , when  $\delta = a/A = 0.1$ : — for  $\kappa$  and --- for  $\kappa = \infty$ ;  $\circ$  and  $\triangle$  are the corresponding exact-solution results ( $\lambda = \kappa = \infty$ ) of Goren & O'Neill (1971) and Jeffery & Onishi (1984), respectively.

collector surface. Also included for comparison are the corresponding exact-solution results of Goren & O'Neill (1971) and Jeffery & Onishi (1984) for two *solid* spherical particles. It can be seen from the figures that the magnitude of the effect of the *no-slip* boundary on particle velocity is larger for the motion toward the collector surface than for the motion tangential to the collector surface. As indicated in figure 7, the discrepancy between the approximate and exact-solution results for the velocity component  $U_r$ , becomes relatively large as  $d \rightarrow 1$ , but it still remains relatively small for  $d > \sim 2$ . The present asymptotic result for the velocity component  $U_\theta$  is in remarkable agreement with the exact solution in the entire region  $d > 1$ . Indeed, the asymptotic and "exact" predictions for the velocity component  $U_\theta$  agree within 0.03% up to  $d = 1.5$ . Even when  $d = 1.1$ , the error associated with the asymptotic solution is only 4.94%. It can also be noted that the particle velocities around and toward the collector surface become larger for the less viscous particle, and this effect is a strong function of the particle position relative to the collector surface. The variation of the velocity with the separation distance  $d$  suggests that a particle approaching the collector surface does not immediately respond to the curving of the undisturbed fluid streamlines as they divide past the collector, but when the particle comes within range of the hydrodynamic repulsion from the collector, it then moves *permanently* to the outside of the fluid streamline it was initially following upstream.

We now consider a more general case for finite  $\lambda$ . In this case, the leading terms of  $U_r$  and  $U_\theta$  are simply given as

$$U_r^\infty \approx -\frac{1}{1 + \lambda} \cdot \delta d \cos \theta \tag{81a}$$

and

$$U_\theta^\infty \approx \frac{1}{2} \frac{1}{1 + \lambda} \sin \theta. \tag{81b}$$

Thus, the small particle is transported *around* the fluid-collector sphere with a velocity of  $O(1)$  and transported *toward* the collector surface with a velocity of  $O(\delta)$ . In contrast, we have just seen in the case of a *rigid* collector sphere, that the transport motion becomes considerably weaker and the velocity components,  $U_\theta$  and  $U_r$ , are only  $O(\delta)$  and  $O(\delta^2)$ , respectively. A small particle will thus remain near a large fluid-collector for a much shorter period than it would near a solid collector, but the motion toward the collector surface is also much stronger.

In order to investigate the consequence of these differences in the relative trajectory, it is convenient to derive a trajectory equation that relates the rate of change of the separation distance



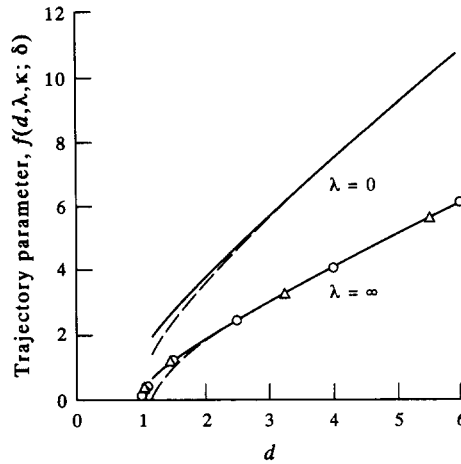


Figure 9. Trajectory function  $f(d, \lambda, \kappa; \delta)$  as a function of the dimensionless separation distance,  $d$ , when  $\delta = a/A = 0.1$ : — for  $\kappa = 0$  and --- for  $\kappa = \infty$ ; ○ and △ are the corresponding exact-solution results ( $\lambda = \kappa = \infty$ ) of Goren & O'Neill (1971) and Jeffrey & Onishi (1984), respectively.

$d$  to that of the orientation angle  $\theta$  of the small particle, rather than to examine the velocity components separately. The trajectory equation may be deduced readily from the asymptotic solution [79a, b]. The result is

$$-\frac{\partial d}{\partial \theta} = f(d, \lambda, \kappa; \delta) \frac{\cos \theta}{\sin \theta}, \tag{82}$$

in which the trajectory parameter  $f(d, \lambda, \kappa; d)$  is defined by

$$f(d, \lambda, \kappa; \delta) \equiv -\frac{(1 + \delta d) U_r \sin \theta}{\delta U_\theta \cos \theta} \tag{83}$$

It is worth pointing out that, owing to the symmetry of the “outer” solution for streaming flow past the collector, the trajectories exhibit a *fore* and *aft* symmetry with respect to  $\theta = 90^\circ$ , as the small particle is transported around the collector surface.

When the small particle is located at sufficiently large distances from the surface of the collector with respect to the characteristic scale of the inner problem,  $a$  (but still *close* to the surface relative to the characteristic lengthscale of the outer problem A), the influence of hydrodynamic interactions is vanishingly small. In the case of no interaction,

$$f(d, \lambda, \kappa; \delta) \approx d \tag{84a}$$

for  $\lambda \rightarrow \infty$ , and

$$f(d, \lambda, \kappa; \delta) \approx 2d \tag{84b}$$

for finite  $\lambda$ . Thus, a small particle at an initial position near a fluid collector will be transported closer to the surface of the *fluid*-collector than it would from the same initial position in the vicinity of a *solid*-collector. In figure 9, the trajectory function  $f(d, \lambda, \kappa; \delta)$  is plotted as a function of the separation distance from the surface of the larger collector. Also shown are the corresponding exact-solution results of Goren & O'Neill (1971) and Jeffrey & Onishi (1984). It can be seen that the hydrodynamic repulsion from the surface of the collector is increased as the viscosity of the particle is increased. It can also be observed that although the individual force components deviate from the corresponding unbounded domain solutions (e.g. see figures 3–6), the deviation of the trajectory function  $f(d, \lambda, \kappa; \delta)$  is much smaller and is actually insignificant until  $d < \sim 3$ . In fact, for  $d > 3$  the slope of  $f(d, \lambda, \kappa; \delta)$  vs  $d$  in the plot is about 2 (or 1 for  $\lambda \rightarrow \infty$ ), which corresponds exactly to the case of no interaction. Thus, in practice for sufficiently small  $\delta (= a/A)$ , a particle is not expected to deviate from a streamline of the flow until its center is less than 3 particle radii from the surface of the collector. Thereafter, its motion can be accurately approximated by the method developed in this paper.

## REFERENCES

- ANFRUNS, J. F. & KITCHENER, J. A. 1977 Rate of capture of small particles in flotation. *Inst. Min. Metall.* **86**, C9–C15.
- BURRILL, K. A. & WOODS, D. R. 1973 Film shapes for deformable drops at liquid–liquid interface. II. The mechanism of film drainage. *J. Colloid Interface Sci.* **42**, 15–34.
- CHEN, J. D., HAHN, P. S. & SLATTERY, J. C. 1984 Coalescence time for a small drop or bubble at a fluid–fluid interface. *AIChE JI* **30**, 622–630.
- CHWANG, A. T. & WU, T. Y.-T. 1975 Hydromechanics of low-Reynolds-number flow. Part 2. Singularity method for Stokes flow. *J. Fluid Mech.* **67**, 787–815.
- DERJAGUIN, B. V., DUKHIN, S. S. & RULEV, N. N. 1976 Importance of hydrodynamic interaction in the flotation of fine particles. *Coll. J. U.S.S.R.* **38**, 227–232.
- DUKHIN, S. S. & RULEV, N. N. 1977 Hydrodynamic interaction between a solid spherical particle and a bubble in the elementary act of flotation. *Coll. J. U.S.S.R.* **39**, 231–236.
- FUENTES, Y. O., KIM, S. & JEFFREY, D. J. 1988 Mobility functions for two unequal viscous drops in Stokes flow I. Axisymmetric motions. *Phys. Fluids* **31**, 2445–2455.
- FUENTES, Y. O., KIM, S. & JEFFREY, D. J. 1989 Mobility functions for two unequal viscous drops in Stokes flow II. Asymmetric motions. *Phys. Fluids* **A1**, 61–76.
- GOLDMAN, A. J., COX, R. G. & BRENNER, H. 1967a Slow viscous motion of a sphere parallel to a plane wall. I. Motion through a quiescent fluid. *Chem. Engng Sci.* **22**, 637–651.
- GOLDMAN, A. J., COX, R. G. & BRENNER, H. 1967b Slow viscous motion of a sphere parallel to a plane wall. II. Couette flow. *Chem. Engng Sci.* **22**, 653–660.
- GOREN, S. L. & O'NEILL, M. E. 1971 On the hydrodynamic resistance to a particle of a dilute suspension when in the neighborhood of a large obstacle. *Chem. Engng Sci.* **26**, 325–338.
- HETSRONI, G. & HABER, S. 1978 Low Reynolds number flow of two drops. *Int. J. Multiphase Flow* **4**, 1–17.
- JAMESON, G. J., NAM, S. & YOUNG, M. M. 1977 Physical factors affecting recovery rates in flotation. *Miner. Sci. Engng* **9**, 103–118.
- JEFFERY, D. J. & ONISHI, Y. 1984 Calculation of the resistance and mobility functions for two unequal rigid spheres in low-Reynolds-number flow. *J. Fluid Mech.* **139**, 261–290.
- JONES, A. F. & WILSON, S. D. R. 1978 The film drainage problem in drop coalescence. *J. Fluid Mech.* **87**, 263–288.
- LEE, S. H., CHADWICK, R. S. & LEAL, L. G. 1979 Motions of a sphere in the presence of a plane interface. Part I. An approximate solution by generalization of the method of Lorentz. *J. Fluid Mech.* **93**, 705–726.
- PRIEVE, D. C. & RUCKENSTEIN, E. 1974 Effect of London forces upon the rate of deposition of Brownian particles. *AIChE JI* **20**, 1178–1187.
- PUDDINGTON, I. E. & SPARKS, B. D. 1975 Spherical agglomeration processes. *Miner. Sci. Engng* **17**, 282–291.
- RULEV, N. N. 1977 Efficiency of particle capture by bubbles in noninertial flotation. *Coll. J. U.S.S.R.* **40**, 747–756.
- SPIELMAN, L. A. 1977 Particle capture from low-speed laminar flows. *A. Rev. Fluid Mech.* **9**, 297–319.
- SPIELMAN, L. A. & CUKOR, P. M. 1973 Deposition of non-Brownian particles under colloidal forces. *J. Colloid Interface Sci.* **43**, 51–65.
- SPIELMAN, L. A. & FITZPATRICK, J. A. 1973 Theory for particle collection under London and gravity forces. *J. Colloid Interface Sci.* **42**, 608–623.
- STOOS, J. A. 1987 Ph.D. dissertation, California Inst. of Technology, Pasadena, CA.
- STOOS, J. A. & LEAL, L. G. 1989 Particle motion in axisymmetric stagnation flow toward an interface. *AIChE JI* **35**, 196–212.
- VAN DYKE, M. 1975 *Perturbation Method in Fluid Mechanics*. Parabolic Press, Stanford, CA.
- YANG, S.-M. & LEAL, L. G. 1984 Particle motion in Stokes flow near a plane fluid–fluid interface. Part 2, Linear shear and axisymmetric straining flows. *J. Fluid Mech.* **149**, 272–304.
- YANG, S.-M. & LEAL, L. G. 1990 Motions of a fluid drop near a deformable interface. *Int. J. Multiphase Flow* **16**, 597–616.
- YUU, S. & FUKUI, Y. 1981 Measurement of fluid resistance correction factor for a sphere moving through a viscous fluid toward a plane surface. *AIChE JI* **27**, 168–170.