THEORETICAL BASIS FOR FALLING-BALL RHEOMETRY IN SUSPENSIONS OF NEUTRALLY BUOYANT SPHERES

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Abstract—The effective viscosity μ of a dilute suspension of neutrally buoyant, polydisperse, rigid spheres of macroscopic size (characteristic radius $= c$) randomly distributed throughout a Newtonian liquid of viscosity μ_0 is derived theoretically. This result is obtained by two closely related, but nevertheless independent, schemes: (i) from the properties of the fundamental Stokeslet solution viewed at the suspension scale; and (ii) from the settling velocity U of a single, nonneutrally buoyant falling ball (radius $= b$) instantaneously (and quasistatically) settling through the unbounded suspension. To terms of leading order, both yield the classical Einstein result, $\mu = \mu_0(1 + \frac{5}{2}\phi)$, apparently independently of the ratio of c/b, as well as of the size distribution of the suspended spheres. Also studied are wall effects for the special case where the falling ball is instantaneously situated at the center of a hollow sphere (radius $= r_o$) that bounds the suspension externally. For circumstances where $b/r_o \ll 1$ and $c/r_o \ll 1$, it is demonstrated that classical falling-ball wall effects for a *homogeneous* Newtonian fluid of viscosity $\mu = \mu_0(1 + \frac{5}{2}\phi)$ apply equally well to the present suspension case. This example strongly suggests that the apparent viscosity of dilute suspensions can be experimentally measured via falling-ball rheometry using the well-known (circular cylindrical) wall-effect corrections developed for Newtonian liquids. This observation is in agreement with existing experimental data.

Key Words: suspension viscosity, Stokes law for suspensions, falling-ball suspension rheometry, viscosity of suspensions, rheology of suspensions

1. INTRODUCTION

Consider a viscous, incompressible Newtonian fluid in which neutrally buoyant spheres of macroscopic size are suspended randomly at infrequent intervals. Einstein (1906, 1911) derived his well-known formula for the apparent viscosity of such a suspension by subjecting the suspension to an elongational or irrotational flow field. Subsequently, Burgers (1939) obtained an identical result for simple shear flow between parallel plates. In this paper, we initially consider a suspension that is macroscopically at rest except for being perturbed by a steady point force or Stokeslet [after Hancock (1953)]. This singularity is asymptotically equivalent in its far-field behavior to a sphere animated by an externally imposed body force (e.g. a ball settling under the influence of gravity). Subsequently, we consider finite-size falling balls, polydisperse suspensions and bounded suspensions.

When the suspension is dilute, so that the suspended spheres do not interact hydrodynamically, only two-body interactions between the settling sphere and a single suspended sphere need be considered. Exact solutions for such unequal-size, two-sphere problems exist as both bispherical coordinate expansions (O'Neill & Majumdar 1970; Zinchencko 1980; Happel & Brenner 1983) and Taylor series expansions (Jeffrey & Onishi 1984). These solutions have been used to determine self-diffusion coefficients for monodisperse (Anderson & Reed 1976) and polydisperse (Batchelor 1976) suspensions of neutrally buoyant Brownian spheres, as well as to analyze hydrodynamic interactions in polydisperse sedimenting suspensions of spheres in which either Brownian or hydrodynamic forces are appreciable (Batchelor 1982; Batchelor & Wen 1982). As our analysis will require only asymptotic, rather than exact, knowledge of the detailed velocity and pressure fields generated by the moving ball, we shall not directly avail ourselves of these existing two sphere results but rather will use the "method of reflections" (Happel & Brenner 1983). Use of the method of reflections allows us to determine the perturbations to the velocity and pressure fields created by a Stokeslet arising from the presence of the suspended spheres and the bounding wall.

Section 2, which follows, formulates the governing equations and boundary conditions, in addition to providing an overview of the analytical methods employed in their solution. Section 3 furnishes the velocity and pressure fields generated by a Stokeslet in the presences of a *single* suspended sphere, and then effects the integration of these results over all allowable positions of the center(s) of the suspended sphere(s) to obtain the cumulative effect arising from the presence of *all* of the suspended spheres (in a dilute suspension). Section 3 also formally demonstrates that these results are independent of the degree of polydispersivity of the suspended spheres provided that the size distribution is uniform throughout the suspension. Next, section 4 extends the preceding analysis to the physically important case of a finite-size sphere moving in a (spherically) *bounded* suspension. Section 5 addresses the question of a relative slip velocity at the suspension scale. The final section summarizes our results.

2. COMPUTATIONAL OVERVIEW

This study focuses on a dilute suspension of neutrally buoyant, randomly distributed spheres in a viscous Newtonian liquid under conditions such that only hydrodynamic forces are sensible. Our objectives are initially to determine the suspension-scale velocity and pressure fields produced by a point force or Stokeslet in this dilute suspension, and subsequently to use these data to determine the analog of the Stokes law drag force experienced by a finite-size sphere moving through a dilute suspension, both for unbounded and bounded suspensions. The inclusion of external boundaries permits quantitative assessment of wall effects upon falling-ball rheometry in the case of suspensions.

2.1. Stokeslet

As in figure 1, let the Stokeslet be characterized by the vector force **F**. All velocities will be referred to a stationary coordinate system whose origin O concides with the Stokeslet (figure 2). In the latter figure, $\mathbf{r} \equiv \overrightarrow{OP}$ denotes the position vector of an arbitrary point P relative to O. Additionally, $r = |\mathbf{r}|$ is the magnitude of **r**, whereas $\hat{\mathbf{r}} = \mathbf{r}/r$ constitutes a unit vector in the direction of **r**.

The homogeneous suspending fluid is taken to be incompressible and Newtonain (viscosity $= \mu_0$). Accordingly, the fluid satisfies the continuity,

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

and the quasistatic, linearized Navier–Stokes equations,

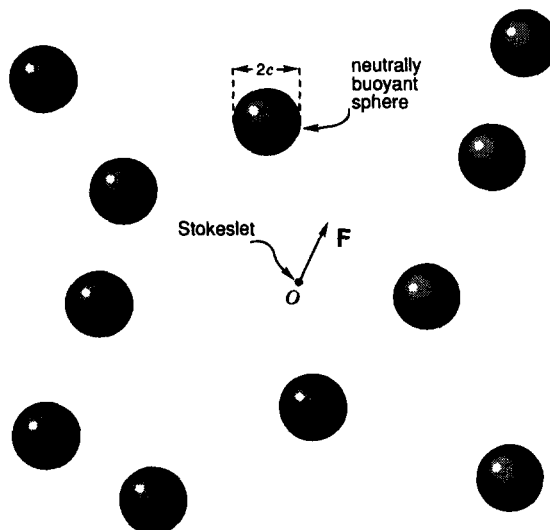$$\mathbf{V}p = \mu_0 \mathbf{V}^2 \mathbf{v} + \mathbf{J}, \tag{2}$$

Figure 1. A point force or Stokeslet (origin O) of strength **F** in a dilute suspension of neutrally buoyant spheres (radii c) that are randomly distributed throughout a homogeneous Newtonian fluid.

with $\mathbf{J}$ the external force per unit volume. The latter equation may be written alternatively as

$$\nabla \cdot \pi = -\mathbf{J}, \tag{3}$$

wherein

$$\pi = -p\mathbf{I} + \mu_0[\nabla\mathbf{v} + (\nabla\mathbf{v})\dagger], \tag{4}$$

with π the stress tensor, $\mathbf{I}$ the dyadic idemfactor and $(\nabla\mathbf{v})\dagger$ the transpose of $\nabla\mathbf{v}$.

Initially, the suspension is taken to be unbounded. Accordingly, we seek a solution of Stokes equations [1] and [2] for a Stokeslet,

$$\mathbf{J} = \mathbf{F}\delta(\mathbf{r}), \tag{5}$$

and satisfying the following boundary conditions in an otherwise quiescent suspension:

$$\mathbf{v} \to \mathbf{0}, \quad p \to p_\infty \quad \text{as} \quad r \to \infty, \tag{6a,b}$$

$$\int_{S_c^{(n)}} \mathbf{ds} \cdot \pi = \mathbf{0}, \quad \int_{S_c^{(n)}} \mathbf{r} \times (\mathbf{ds} \cdot \pi) = \mathbf{0} \quad \forall n, \tag{7a,b}$$

with $\delta(\mathbf{r})$ designating the Dirac delta function, p_∞ the uniform pressure at an infinite distance from the Stokeslet, ds a directed element of surface area and $S_c^{(n)}$ ($n = 1, 2, 3, \ldots$) the surface of the nth suspended sphere. Conditions [7a,b] embody the requirements that every suspended sphere be force and torque (or couple) free owing to its neutral buoyancy.

Linearity of the equations of motion and boundary conditions permits the boundary-value problem to be decomposed into a sequence of separate problems:

$$\mathbf{v} = \mathbf{v}' + \bar{\mathbf{v}}'', \quad p = p' + \bar{p}'', \tag{8a,b}$$

where each of the individual fields, defined below, satifies Stokes equations [1] and [2] (to which, we may imagine, primed and double-primed superscripts are added as needed). The primed solution corresponds to the homogeneous fluid Stokeslet, whereas the double-primed solution arises from the perturbation to the primed solution caused by the suspended spheres.

Using the method of reflections, $(\bar{\mathbf{v}}'', \bar{p}'')$ can be determined to terms of leading order as

$$\bar{\mathbf{v}}'' \approx \sum_n \mathbf{v}'', \quad \bar{p}'' \approx \sum_n p'', \tag{9a,b}$$

where $(\mathbf{v}'', p'')$ is the "reflection" of the Stokeslet field $\mathbf{v}'$ from a single sphere (of radius c), and the sum indicated by n is taken over all spheres in the suspension. The above supposes that hydrodynamic interactions among the suspended spheres are negligible. Subequently (cf. [33a,b]), these sums are replaced by comparable integrations.

A solution of Stokes equations satisfying the boundary conditions [6a,b] and [7a,b], correct to terms of first order in the sphere–Stokeslet and sphere–sphere separation distances, may therefore be obtained via the following algorithm: consider the field pair $(\mathbf{v}', p')$ generated by a Stokeslet in an unbounded fluid in which no suspended spheres are present. This corresponds to the choice $\mathbf{J}' = \mathbf{F}\delta(\mathbf{r})$ in [2] with conditions [7a,b] absent. This well-known Stokeslet solution is (Chwang & Wu 1975)

$$\mathbf{v}' = \frac{1}{8\pi\mu_0 r}(\mathbf{I} + \hat{\mathbf{r}}\hat{\mathbf{r}}) \cdot \mathbf{F}, \quad p' = \frac{\hat{\mathbf{r}} \cdot \mathbf{F}}{4\pi r^2} + p_\infty. \tag{10a,b}$$

However, if a (single) suspended sphere is present at an arbitrary point $\mathbf{R}$ (figure 2), this solution violates the condition that the sphere moves as a rigid body satisfying a no-slip condition on its surface. Another solution $(\mathbf{v}'', p'')$ is sought such that when added to $(\mathbf{v}', p')$ the resultant fields also satisfy conditions [7a,b] for a force- and couple-free sphere. Therefore, each of the double-primed fields in [8] must satisfy Stokes equations [1] and [2] together with the following conditions:

$$\mathbf{J}'' = \mathbf{0}, \tag{11}$$

$$\mathbf{v}'' \to \mathbf{0}, \quad p'' \to 0 \quad \text{as} \quad |\mathbf{r} - \mathbf{R}| \to \infty \tag{12a,b}$$

$$\mathbf{v}'' = -\dot{\gamma}_{\mathbf{R}} \cdot (\mathbf{r} - \mathbf{R}) \quad \text{on} \quad S_c, \tag{13}$$
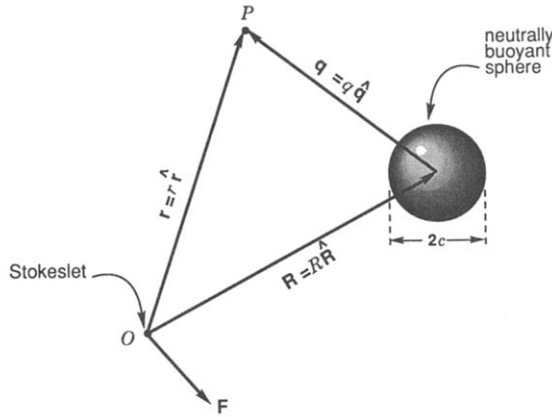
Figure 2. Stokeslet in an unbounded fluid with a single suspended sphere present. The center of the neutrally buoyant sphere is situated at the point whose position vector relative to the Stokeslet at O is $\mathbf{R}$. Point P is an arbitrary point in the fluid whose location is defined by the position vector $\mathbf{r}$ relative to the origin O, and by $\mathbf{q}$ relative to the center of the suspended sphere. Observe that $\mathbf{q} = \mathbf{r} - \mathbf{R}$, and hence that $q^2 = r^2 + R^2 - 2\mathbf{R} \cdot \mathbf{r}$.

with $\mathbf{R}$ the vector drawn from the Stokeslet to the center of the suspended sphere (figure 2) and $\dot{\gamma}_\mathbf{R} = \frac{1}{2}[\nabla \mathbf{v}' + (\nabla \mathbf{v}')\dagger]_\mathbf{R}$ the rate of strain tensor created by the Stokeslet and evaluated at the homogeneous fluid position $\mathbf{R}$ currently occupied by the sphere center (cf. [29]). The detailed derivation of the boundary condition [13] from Faxen's law will be given in section 3, with the field pair $(\mathbf{v}'', p'')$ corresponding to the "reflection" of the undisturbed Stokeslet field from the neutrally buoyant sphere; i.e. the resulting velocity field $\mathbf{v}' + \mathbf{v}''$ satisfies the condition that the suspended sphere be force free and couple free, and possess a rigid, impermeable, no-slip surface.

Next, we determine the cumulative effect of all the suspended spheres upon the velocity and pressure fields in the suspension. The sum of fields reflected from individual particles (shown schematically in figure 3) is determined by integrating the reflected velocity and pressure fields (weighted with the local number density of spheres) over all possible locations of the sphere centers.
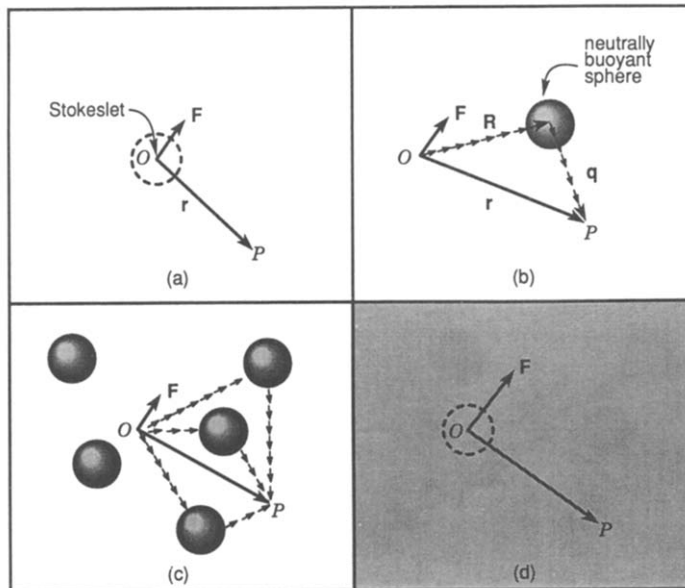

Figure 3. Symbolic representation of the computational algorithm. In (a) the far-field approximation to a moving sphere on which an external force $\mathbf{F}$ acts is represented by a Stokeslet. In (b) the solution is determined, correct to terms of first order, for the Stokeslet velocity field reflected off one of the suspended spheres. The cumulative effect at P produced by all the suspended spheres is determined in (c) by integrating the contribution of the suspended sphere in (b) (weighted with the local number density of suspended spheres) over all possible positions of the suspended sphere center. In (d) the collective suspension behavior is replaced by a hypothetical, homogeneous Newtonian liquid (shown shaded) possessing an apparent suspension viscosity, μ.

As will be shown in section 3, to terms of dominant order the velocity and pressure fields [8a,b] in the suspension (in the absence of any external boundaries) are given by the expressions

$$\mathbf{v} = \frac{1}{8\pi\mu r}(\mathbf{I} + \hat{\mathbf{r}}\hat{\mathbf{r}})\cdot\mathbf{F}, \quad p = \frac{\hat{\mathbf{r}}\cdot\mathbf{F}}{4\pi r^2} + p_\infty, \qquad [14a,b]$$

wherein

$$\mu = \mu_0(1 + \tfrac{5}{2}\phi). \qquad [15]$$

Comparison of [14a,b] with their homogeneous fluid counterparts [10a,b] reveals that a dilute suspension of spheres behaves macroscopically as a Newtonian fluid continuum with an apparent viscosity μ given by [15]. It will be demonstrated in section 3 that this result is independent of the size distribution of radii of the suspended spheres, and therefore holds for both monodisperse and polydisperse suspensions.

2.2. Finite-size sphere in a bounded suspension

The original Stokeslet [10a,b] may be regarded (Happel & Brenner 1983) as representing the far-field effect of the disturbance caused by a (nonneutrally buoyant) sphere settling under the influence of a net body force $\mathbf{F}$ through an unbounded suspending liquid. If instead the sphere now settles through a suspension, rather than a homogeneous fluid, it will suffer a reduction in its settling velocity owing to the presence of the suspended spheres (as well as any walls in a bounded suspension). In the preceding section we described how the sum of the Stokeslet velocity field $\mathbf{v}'$ and reflected velocity field $\mathbf{v}''$ give the correct leading-order velocity on the surface of a suspended sphere. However, this sum fails to produce a zero velocity at the container walls, as it must if the no-slip boundary condition on the solid wall is to be satisfied. Corrections for the presence of the wall are thus made by adding appropriate reflected fields such that the resulting velocity at the wall is zero. These corrections, $(\mathbf{v}^W, \mathbf{v}^{WP})$, are calculated in section 4. (Note that by satisfying the boundary condition on the wall, the boundary conditions imposed upon the suspended spheres and settling sphere are no longer *exactly* satisfied, although they are satisfied to the order of the approximation—as is always the case when using the method of reflections.)

Therefore, for a bounded suspension the method of reflections expresses the leading-order contribution to the velocity field $\mathbf{v}^T$ as

$$\mathbf{v}^T = \mathbf{v}' + \bar{\mathbf{v}}'' + \mathbf{v}^W + \bar{\mathbf{v}}^{WP}, \qquad [16]$$

where $\mathbf{v}^W$ and $\bar{\mathbf{v}}^{WP}$ are, respectively, the wall-effect velocity corrections due to the Stokeslet $(\mathbf{v}')$ interacting with the wall and the suspended spheres $(\bar{\mathbf{v}}'')$ interacting with the wall. These interactions are shown schematically in figure 4. While each of the Stokes velocity fields appearing in [16] will be calculated for the effect of the suspended spheres and bounding walls upon the (finite-size) settling sphere, it is rather the drag force exerted upon the latter that represents the quantity of ultimate physical interest.

An extension of Faxen's law (Faxen 1927; Brenner 1964b) may be used to determine the drag force $\mathbf{F}^*$ experienced by a finite-size sphere (radius $= b$) immersed in an arbitrary Stokes velocity field $\mathbf{v}^*$ that vanishes at infinity:

$$\mathbf{F}^* = -\tfrac{3}{2}\mu_0 b \int_{S_1} \mathbf{v}^*|_{r=b}\, d^2\hat{\mathbf{r}}. \qquad [17]$$

Here, $d^2\hat{\mathbf{r}}$ is a scalar element of surface area on a unit sphere, whereas S_1 denotes integration over a unit sphere. For a bounded suspension, Faxen's law can be expressed in the form (Happel & Brenner 1983)

$$\mathbf{F}^* = -\tfrac{3}{2}\mu b K \int_{S_1} \mathbf{v}^*|_{r=b}\, d^2\hat{\mathbf{r}} \qquad [18]$$

to account for both the increased suspension viscosity μ of the unbounded medium and the increased drag force K (relative to the unbounded fluid) on the settling sphere arising from the presence of the bounding walls. In both of the above equations, $\mathbf{v}^*|_{r=b}$ does not necessarily describe
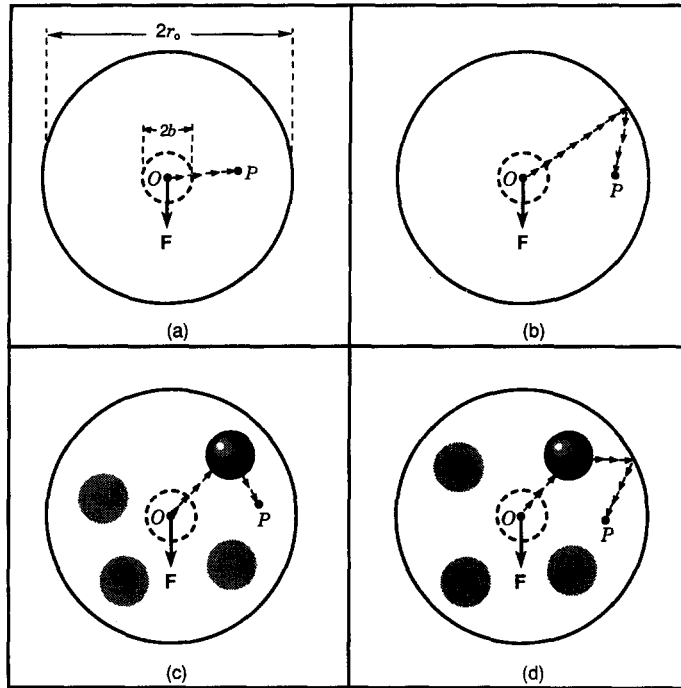
Figure 4. Symbolic representation of quasistatic wall effects for a falling ball of radius b instantaneously situated at the center of a hollow spherical shell of radius r_o. In (a) the fluid motion at an arbitrary point P in the fluid due to the presence of a Stokeslet is determined. The reflection of the Stokeslet velocity field from the containing wall is shown in (b). In (c) the fluid motion at P due to the perturbation caused by a suspended sphere is found; (d) represents the perturbation at the point P due to the indirect interaction of the Stokeslet with the wall by perturbing the suspended sphere. In (c) the cumulative result arising from all of the suspended spheres can be obtained by determining the effect on a single suspended sphere, and subsequently integrating over all possible sphere-center positions weighted with the local number density of the suspended spheres. The final result in (d) can be obtained by reflecting the integrated results of (c) from the containing wall.

a rigid-body motion; nevertheless, these formulas do indeed properly furnish the force exerted by the fluid upon the interior contents of the spherical domain of radius b.

Equation [18] may be employed to calculate the total drag force $\mathbf{F}^{\mathrm{T}}$ acting upon the settling sphere from knowledge of $\mathbf{v}^{\mathrm{T}}$. Alternatively, the linearity of [18] permits the total drag force upon the moving sphere to be expressed as the sum:

$$\mathbf{F}^{\mathrm{T}} = \mathbf{F}' + \overline{\mathbf{F}}'' + \mathbf{F}^{\mathrm{W}} + \overline{\mathbf{F}}^{\mathrm{WP}}, \qquad [19]$$

wherein each of the respective terms appearing therein may be derived from [18] by replacing $\mathbf{v}^*$ with the comparably superscripted velocity field appearing in [16].

The total drag force $\mathbf{F}^{\mathrm{T}}$, which represents the quasistatic force experienced by a sphere (radius b) moving through a dilute suspension at the instant that its center is positioned at the center of a spherical container (radius r_o), will be shown (cf. [18] and [65]) to possess the form

$$\mathbf{F}^{\mathrm{T}} = -\mathbf{F}\frac{\mu}{\mu_0}K\left(1 - \frac{5}{2}\phi\right)\left(1 - \frac{9}{4}\frac{b}{r_o}\right) + O\left(\phi^2\frac{b}{r_o}, \phi\frac{b^2}{r_o^2}\right). \qquad [20]$$

Since the drag force must be balanced by the body force $\mathbf{F}^{\mathrm{T}} = -\mathbf{F}$, the above equation gives

$$K = \left(1 - \frac{9}{4}\frac{b}{r_o}\right)^{-1}$$

and

$$\mu = \mu_0\left(1 + \frac{5}{2}\phi\right).$$

The additional drag coefficient K is the first-order, wall-correction factor (Happel & Brenner 1983) in a homogeneous fluid for the concentric-sphere geometry described above, and the suspension viscosity is that predicted from the unbounded suspension analysis.

To terms of first order, the presence of the walls therefore affects the ball's motion through the suspension exactly as if it were moving through a homogeneous Newtonian fluid of viscosity μ given by Einstein's law [15]. Going beyond the elementary concentric spherical-enclosure case, our analysis strongly suggests that the apparent viscosity of dilute suspensions can be measured via falling-ball rheometry using comparable wall corrections developed for *homogeneous* Newtonian liquids, e.g. for circular cylindrical walls (Happel & Brenner 1983). This tentative theoretical conclusion is strongly supported by existing experimental evidence (Mondy *et al.* 1986).

The following sections detail the calculations leading to the preceding results. We begin with the determination of the suspension-scale velocity and pressure fields produced by a Stokeslet in an externally *unbounded* dilute suspension.

3. STOKESLET IN A DILUTE, UNBOUNDED SUSPENSION

The velocity and pressure fields (v', p') associated with a Stokeslet in an unbounded homogeneous fluid of viscosity μ_0 are given by [10a,b]. To derive the perturbation (v'', p'') created by the presence of a single suspended sphere, we need to establish the boundary condition [13] governing the reflected velocity and pressure fields for a rigid, force- and couple-free sphere.

The neutrally buoyant sphere is animated solely by the point force. Let U and Ω, respectively, be the translational velocity of the center of this sphere and its angular velocity. These velocities are to be determined by the requirements [7a,b] that the suspended sphere be both force and couple free. If we consider this rigid sphere to be immersed in a fluid whose undisturbed motion is given by the Stokeslet velocity field v', Faxen's law together with the analogous torque equation (Happel & Brenner 1983) reduce to the respective forms

$$F = 6\pi\mu_0 c(v'_R - U) + \pi\mu_0 c^3 (\nabla^2 v')_R \tag{21}$$

and

$$T = 8\pi\mu_0 c^3 (\omega'_R - \Omega), \tag{22}$$

wherein $F = 0$ and $T = 0$, and in which

$$v'_R = \frac{1}{8\pi\mu_0 R} (I + \hat{R}\hat{R}) \cdot F \tag{23}$$

and

$$\omega'_R = \frac{1}{8\pi\mu_0 R^3} (F \times \hat{R}) \tag{24}$$

are, respectively, the Stokeslet velocity vector v' and angular velocity pseudovector $\omega' = \frac{1}{2}\nabla \times v'$, each evaluated at the fluid point $r = R$ currently occupied by the sphere center. From [10a] the $\nabla^2 v'_R$ term in [21] can be shown to be of $O(c/R)^2$ in comparison with the v'_R term. Therefore, [21] and [22] respectively yield for the translational and angular velocities of the suspended sphere,

$$U = v'_R \left[1 + O\left(\frac{c}{R}\right)^2 \right] \tag{25}$$

and

$$\Omega = \omega'_R. \tag{26}$$

Thus, the requirements [7a,b] that the suspended sphere be force and couple free will be satisfied to terms of lowest order in c/R if the velocity $v(r)$ on the surface of the suspended sphere whose center is located at R is chosen as

$$v(r) = v'_R + \omega'_R \times (r - R). \tag{27}$$

The latter constitutes a rigid-body motion for a sphere translating and rotating with the respective translational and angular velocities [23] and [24] of the homogeneous suspending fluid Stokeslet field [10a] evaluated at the sphere's center.

A Taylor series expansion of the homogeneous Stokeslet velocity field [10a] about the position $\mathbf{R} = R\hat{\mathbf{R}}$ (see figure 2) of the sphere center yields, for $|\mathbf{r} - \mathbf{R}|/|\mathbf{R}| \ll 1$,

$$\mathbf{v}'_r \approx \mathbf{v}'_R + \boldsymbol{\omega}'_R \times (\mathbf{r} - \mathbf{R}) + \dot{\gamma}_R \cdot (\mathbf{r} - \mathbf{R}),$$ [28]

in which $\mathbf{v}'_R$ and $\boldsymbol{\omega}'_R$ are given by [23] and [24], and

$$\dot{\gamma}_R = \frac{1}{8\pi\mu_0 R^2}(\mathbf{I} - 3\hat{\mathbf{R}}\hat{\mathbf{R}})(\hat{\mathbf{R}} \cdot \mathbf{F}).$$ [29]

The latter represents the rate-of-strain dyadic $\dot{\gamma} = \frac{1}{2}(\nabla\mathbf{v}' + \nabla\mathbf{v}'\dagger)$ evaluated at the point $\mathbf{r} = \mathbf{R}$.

The boundary conditions imposed upon $\mathbf{v}''$ are such that when the latter is added to $\mathbf{v}'$ the sum satisfies [27] on S_c, and, hence, satisfies the conditions of neutral buoyancy for a solid impenetrable sphere of radius c. Comparison of [27] with [28] reveals that satisfying [7a,b] requires that the boundary condition imposed at the surface of the suspended sphere be that cited in [13].

The solution $(\mathbf{v}'', p'')$ satisfying Stokes equations [1] and [2] and the boundary conditions [11]–[13] is readily obtained using the general scheme of Brenner (1964a) for solving boundary-value problems on spherical surfaces in the region exterior to a sphere. This yields

$$\mathbf{v}'' = \nabla\Phi_{-3} + \mathbf{q}\frac{p_{-3}}{2\mu_0}, \quad p'' = p_{-3},$$ [30a,b]

where $\mathbf{q} = \mathbf{r} - \mathbf{R}$ (figure 2) is the position vector originating at the center of the suspended sphere, and p_{-3} and Φ_{-3} are the respective solid spherical harmonics

$$p_{-3} = \frac{-5\mu_0 c^3\dot{\gamma}_R : \mathbf{q}\mathbf{q}}{q^5}, \quad \Phi_{-3} = \frac{-c^5\dot{\gamma}_R : \mathbf{q}\mathbf{q}}{2q^5}.$$ [31a,b]

The first and second terms of [30a] are respectively of orders $(1/q)(c/q)^5$ and $(1/q)(c/q)^3$. For $c/q \ll 1$, [30a] thus reduces to

$$\mathbf{v}'' \approx \mathbf{q}\frac{p_{-3}}{2\mu_0} \equiv \frac{-5c^3\mathbf{q}\mathbf{q}\mathbf{q} : \dot{\gamma}_R}{2q^5}.$$ [32]

Equations [30b], [31a] and [32] respectively represent the reflected pressure and velocity fields at the point P (defined by the position vector $\mathbf{r}$ as shown in figure 2) arising from the presence of a *single* suspended sphere whose center is situated at $\mathbf{R}$. The cumulative effect arising from *all* of the suspended spheres, obtained by effecting the sum in [8a,b], is given by

$$\bar{\mathbf{v}}''(\mathbf{r}) = \int \mathbf{v}''(\mathbf{r}, \mathbf{R})n(\mathbf{R}) \, \mathrm{d}^3\mathbf{R}, \quad \bar{p}''(\mathbf{r}) = \int p''(\mathbf{r}, \mathbf{R})n(\mathbf{R}) \, \mathrm{d}^3\mathbf{R},$$ [33a,b]

where the integration is over all positions $\mathbf{R}$ of the sphere centers, and in which $n = 3\phi(\mathbf{R})/4\pi c^3$, with $\phi(\mathbf{R})$ the local volume fraction of suspended spheres at $\mathbf{R}$. [The unnormalized probability that the center of a sphere lies in the volume element $\mathrm{d}^3\mathbf{R}$ centered at $\mathbf{R}$ is $n(\mathbf{R}) \, \mathrm{d}^3\mathbf{R}$.] For a random distribution of sphere centers [i.e. $\phi(\mathbf{R})$ constant] this yields upon integration,

$$\bar{\mathbf{v}}''(\mathbf{r}) = -\frac{15\phi}{8\pi} \int \frac{\mathbf{q}\mathbf{q}\mathbf{q} : \dot{\gamma}_R}{q^5} \mathrm{d}^3\mathbf{R}$$ [34]

and

$$\bar{p}''(\mathbf{r}) = -\frac{15\mu_0\phi}{4\pi} \int \frac{\mathbf{q}\mathbf{q} : \dot{\gamma}_R}{q^5} \mathrm{d}^3\mathbf{R}.$$ [35]

Details of the latter velocity integration are relegated to the appendix, the final result being

$$\bar{\mathbf{v}}'' = -\frac{5\phi}{16\pi\mu_0 r} \mathbf{F} \cdot \left\{ \mathbf{I}\left[1 - 2\frac{r}{r_o} + \frac{4}{5}\left(\frac{r}{r_o}\right)^3 \right] + \hat{\mathbf{r}}\hat{\mathbf{r}}\left[1 - \frac{2}{5}\left(\frac{r}{r_o}\right)^3 \right] \right\},$$ [36]

where r_o is an arbitrary upper bound on the integration. (Integration of the pressure field [35] is performed in the next subsection.) As $r_o \rightarrow \infty$, [36] reduces to

$$\bar{\mathbf{v}}'' = -\frac{5\phi}{16\pi\mu_0 r} \mathbf{F} \cdot (\mathbf{I} + \hat{\mathbf{r}}\hat{\mathbf{r}}).$$ [37]

Using the preceding results, [8a] yields

$$v = \frac{1}{8\pi\mu_0 r}(1 - \tfrac{5}{2}\phi)(\mathbf{I} + \hat{\mathbf{r}}\hat{\mathbf{r}})\cdot\mathbf{F} + O(\phi^2) \qquad [38]$$

for the suspension-scale velocity field at $\mathbf{r}$. Using Einstein's relation [15], the above may be written correctly to first order in ϕ in precisely the same form as that for the homogeneous fluid Stokeslet [10a], but now for a fluid of viscosity μ.

3.1. Pressure-field renormalization

Whereas the velocity [34] can be integrated straightforwardly, the pressure integral [35] constitutes a nonconvergent integral, requiring special analysis. In particular, a q^{-3} singularity appears, leading to a nonunique value for the latter integral. Hence, in [35] a spherical–polar integration scheme centered about the point P yields $\bar{p}'' = 0$, whereas a comparable integration centered about the Stokeslet O yields a nonzero value for $\bar{p}''$!

Nonconvergent integrals arise in a number of physical situations (Hinch 1977). Frequently, the integrals lead to infinite values as the limits of integration approach infinity. Renormalization methods reformulate the problem such that nonconvergent integrals become integrals with unique, finite values (Batchelor 1972; O'Brien 1979; Lu & Kim 1990). These methods may also be used to reformulate integrals like [35] which appears to give nonunique finite values that depend on the coordinate system used to evaluate the integral (Jeffrey 1977). While in practice all such methods necessary give the same results, Hinch's (1977) second renormalization scheme was chosen to evaluate [35].

In applying the scheme, [2] is first rewritten with the boundary condition [7a] represented as a distribution of singular force terms acting on the suspended spheres, such that here

$$\mathbf{J} = \mathbf{F}\delta(\mathbf{r}) - \int n(\mathbf{R})\left(\int_{q=c}\delta(\mathbf{r}'-\mathbf{r})\pi(\mathbf{r}')\cdot d\mathbf{s}'\right)d^3\mathbf{R}, \qquad [39]$$

with $d\mathbf{s}'$ a directed element of surface area on a suspended sphere centered at $\mathbf{R}$. Following Hinch (1977), a term equal to a continuous distribution of dipoles, modeling the far-field effects of the suspended spheres, is added to both sides of [3], thereby obtaining

$$-\nabla p + \mu_0\nabla^2 v - \int 5\mu_0\phi\dot{\gamma}_{\mathbf{R}}\cdot\nabla_{\mathbf{R}}\delta(\mathbf{r}-\mathbf{R})\,d^3\mathbf{R} = \int\left[n(\mathbf{R})\left(\int_{q=c}\delta(\mathbf{r}'-\mathbf{r})\pi(\mathbf{r}')\cdot d\mathbf{s}'\right)\right.$$
$$\left.-5\mu_0\phi\dot{\gamma}_{\mathbf{R}}\cdot\nabla_{\mathbf{R}}\delta(\mathbf{r}-\mathbf{R})\right]d^3\mathbf{R} - \mathbf{F}\delta(\mathbf{r}). \qquad [40]$$

Observe that $\nabla_{\mathbf{R}}\cdot\dot{\gamma}_{\mathbf{R}} = \tfrac{1}{2}\nabla_{\mathbf{R}}^2 v$ and

$$-\int 5\mu_0\phi\dot{\gamma}_{\mathbf{R}}\cdot\nabla_{\mathbf{R}}\delta(\mathbf{r}-\mathbf{R})\,d^3\mathbf{R} \equiv \int 5\mu_0\phi\delta(\mathbf{r}-\mathbf{R})\nabla_{\mathbf{R}}\cdot\dot{\gamma}_{\mathbf{R}}\,d^3\mathbf{R}$$

$$= \tfrac{5}{2}\phi\mu_0\nabla^2 v,$$

where $\nabla_{\mathbf{R}}$ is the gradient with respect to $\mathbf{R}$ ($\mathbf{r}$ being held constant). Hence, the extra term appearing on the l.h.s. of [40] acts as an effective increase in the homogeneous fluid viscosity for the velocity and pressure equations. This accords with intuition, since distant suspended spheres would appear to influence one another (i.e. hydrodynamically interact) not through the agency of the homogeneous fluid viscosity, but rather through that of the effective suspension viscosity. Incorporation of this effect allows [35] to be expressed in terms of convergent integrals.

In parallel with [8a,b], we now decompose the velocity field such that

$$v = v'_N + \bar{v}''_N, \quad p = p'_N + \bar{p}''_N, \qquad [41a,b]$$

where (v'_N, p'_N) arise from a Stokeslet in a fluid of viscosity $\mu_0(1 + \tfrac{5}{2}\phi)$, and $(\bar{v}''_N, \bar{p}''_N)$ constitute the reflected renormalized fields.

Use of the homogeneous Newtonian fluid Green's functions in the first term on the r.h.s. of [40] furnishes the solution of the additional reflected pressure field $\bar{p}_N''$ (Chwang & Wu 1975) explicitly as

$$\bar{p}_N'' = \int \left[p''n + 5\mu_0 \phi \dot{\gamma}_R : \mathbf{V_R} \left(\frac{2\mu_0 \mathbf{q}}{8\pi\mu_0 q^3} \right) \right] d^3 q. \qquad [42]$$

In consequence of the relation $\mathbf{q} = \mathbf{r} - \mathbf{R}$, observe that $\mathbf{V_R}(\mathbf{q}/q^3) = -(\mathbf{I} - 3\hat{\mathbf{q}}\hat{\mathbf{q}})/q^3$. This identity in conjunction with [30b] and [31a] yields

$$\bar{p}_N'' = \int \left[-\frac{15\mu_0 \phi}{4\pi} \frac{\mathbf{qq} : \dot{\gamma}_R}{q^5} - 5\mu_0 \phi \dot{\gamma}_R : (\mathbf{I} - 3\hat{\mathbf{q}}\hat{\mathbf{q}}) \frac{1}{4\pi q^3} \right] d^3 q \equiv 0. \qquad [43]$$

The latter equality follows from the identity $\mathbf{I} : \dot{\gamma}_R \equiv \mathbf{V_R} \cdot \mathbf{v}'' = 0$, which causes the above integrand to vanish. Therefore, the integral will also vanish irrespective of either the bounds delimiting the integration domain or the coordinate system used in effecting the integration. The vanishing of the above integral shows that the average additional or reflected pressure field created at any point $\mathbf{R}$ by the presence of the suspended spheres (beyond that modeled by the continuous distribution of dipoles) is zero to the order of the approximation.

Similarly, using the appropriate Green's function in the first term on the r.h.s. of [40], the additional velocity field may be obtained explicitly as

$$\bar{\mathbf{v}}_N'' = \int \left[\mathbf{v}''n + 5\mu_0 \phi \dot{\gamma}_R : \mathbf{V_R} \left(\frac{\mathbf{I} + \hat{\mathbf{q}}\hat{\mathbf{q}}}{8\pi\mu_0 q} \right) \right] d^3 q. \qquad [44]$$

Note that

$$\mathbf{V_R}[q^{-1}(\mathbf{I} + \hat{\mathbf{q}}\hat{\mathbf{q}})] = q^{-2}[\hat{\mathbf{q}} \bigcup - \bigcup\!\!\hat{\mathbf{q}} - \bigcup\hat{\mathbf{q}} + 3\hat{\mathbf{q}}\hat{\mathbf{q}}\hat{\mathbf{q}}], \qquad [45]$$

with $\bigcup \equiv \mathbf{I}$ the dyadic idemfactor [in the notation of Chapman & Cowling (1970)], used to relate nonconsecutive indices, such that if $\mathbf{C} = \bigcup\!\mathbf{a}$ then $C_{ijk} = \delta_{ik} a_j$. Since $\dot{\gamma}_R$ is symmetric and traceless, only the last term in [45] contributes to [44]. In conjunction with [32], [44] thus becomes

$$\bar{\mathbf{v}}_N'' = \int \left[-\frac{15\phi}{8\pi} \frac{\mathbf{qqq} : \dot{\gamma}_R}{q^5} + \frac{5\phi}{8\pi q^2} 3\hat{\mathbf{q}}\hat{\mathbf{q}}\hat{\mathbf{q}} : \dot{\gamma}_R \right] d^3 q \equiv 0, \qquad [46]$$

whence the average additional or reflected velocity field produced by the suspended spheres is identically zero at every point owing to the vanishing of the above integrand.

Equations [45] and [46] both derive from application of the Green's function to only the first term on the r.h.s. of [40]. Accordingly, the first term makes no net contribution to [40], whence the latter may be rewritten as

$$-\mathbf{V}p + \mu_0(1 + \tfrac{5}{2}\phi)\mathbf{V}^2\mathbf{v} = -\mathbf{F}\delta(\mathbf{r}). \qquad [47]$$

This is of precisely the same form as [2] except that μ_0 has been replaced by $\mu_0(1 + \tfrac{5}{2}\phi) \equiv \mu$. From [10b], the net change in the homogeneous fluid Stokeslet pressure field is zero since p' is independent of the viscosity. From [10a], to order ϕ the net change in the velocity field is that given by [37]. Consequently, the assumed field necessary to secure convergence of the pressure integral is identical to that derived by direct integration of [34].

In combination, [8a], [10a] and [37] for the velocity field, together with [8b], [10b] and [45] for the pressure field, yield

$$\mathbf{v} = \frac{1}{8\pi\mu r}(\mathbf{I} + \hat{\mathbf{r}}\hat{\mathbf{r}}) \cdot \mathbf{F}, \quad p = \frac{\hat{\mathbf{r}} \cdot \mathbf{F}}{4\pi r^2} + p_\infty, \qquad [48a,b]$$

with μ given by [15]. The latter equations reveal that in the presence of a point force, a dilute unbounded suspension behaves on average as a hypothetical, homogeneous, Newtonian liquid characterized by the Einstein scalar viscosity [15], at least in regard to the Stokeslet field engendered by the action of this point force. In the next subsection this result is shown to be independent of the distribution of radii of the suspended spheres in the case of polydisperse suspensions.

Equation [15] for the suspension viscosity, as *defined* by the suspension-scale Stokeslet field, is independent of the relative radii b/c of the suspended spheres to the falling sphere, at least to the

first order in ϕ. This conclusion is supported by (limited) experimental results (Graham et al. 1987) for $1 < b/c < 6$ and $\phi = 0.05$. [While data also exist for larger values of ϕ (Mondy et al. 1986; Milliken et al. 1989) these more concentrated suspensions fail to fulfill the criterion of diluteness, implicit in our analysis.]

3.2. Polydisperse suspensions

Consider a suspension in which the suspended spheres, though still uniformly distributed throughout the suspension, now possess at each point a distribution of radii rather than being monodisperse as was the case previously. This distribution is assumed to be independent of position R. Let the frequency distribution of radii of the suspended spheres be given by $f(c)$, whence by definition, the number dN of spheres with radii lying between c and $c + dc$ is $dN = f(c)\, dc$. We note that

$$\int_c f(c)\, dc = N,$$

where N is the total number of spheres of all sizes, and wherein the integration domain extends from $c = 0$ to ∞. With V_T the total suspension volume, the quantity $d\phi(c) = (4\pi c^3/3V_T)\, dN$ represents the volume fraction of spheres possessing radii between c and $c + dc$. Accordingly, the total volume fraction ϕ of spheres at each point of the suspension is

$$\phi = \int_c d\phi(c) \equiv \frac{4\pi}{3V_T} \int_c f(c) c^3\, dc. \qquad [49]$$

As the total number density of spheres in $n = N/V_T$, this makes

$$n = \frac{3}{4\pi} \int_c \frac{d\phi(c)}{c^3}. \qquad [50]$$

Referring to [33a,b], the total reflected velocity and pressure fields for spheres of all sizes are respectively given by the expressions

$$\bar{v}'' = -\frac{15}{8\pi} \iint_c \frac{\mathbf{qqq} : \dot{\gamma}_R}{q^5} d\phi\, d^3R \qquad [51]$$

and

$$\bar{p}'' = -\frac{15\mu_0}{4\pi} \iint_c \frac{\mathbf{qq} : \dot{\gamma}_R}{q^5} d\phi\, d^3R. \qquad [52]$$

As $d\phi(c)$ is independent of R, [51] and [52] become identical to [34] and [35], respectively, upon effecting the ϕ integration. Consequently, the suspension-scale Stokeslet fields [8a,b] are independent of the distribution of particle sizes, being dependent upon only the overall volume fraction ϕ of suspended spheres. This particle-size independence property arises as a consequence of having neglected hydrodynamic interactions among the suspended spheres. Therefore, in contrast with concentrated suspensions, whose viscosity depends upon the particle-size distribution (e.g. Chong et al. 1971), a dilute polydisperse suspension possesses the same viscosity [15] as does a monodisperse suspension with the same ϕ.

4. WALL EFFECTS

As is well-known (Happel & Brenner 1983), wall effects upon the motion of a sphere through a homogeneous Newtonian fluid can be appreciable. In this section we examine the comparable situation for the case of a dilute suspension rather than a homogeneous fluid. Though the most important case occurring in practice is that of a circular cylindrical boundary, the requisite algebra is daunting (Happel & Brenner 1983), especially so in the suspension case. Accordingly, in the interests of simplicity, attention will be confined to the illustrative case where the outer boundary is spherical rather than cylindrical. Moreover, the calculations are further simplified by restricting attention to the case where the falling ball is (instantaneously) at the center of the bounding spherical envelope. In this context it will be shown that the effect of the wall upon the falling ball

is the same for the suspension as it is for the usual homogeneous fluid case. This strongly suggests that the same conclusion would apply for the circular cylindrical boundary case, although a formal analysis of the latter would be required for an unequivocal demonstration.

In this section we calculate the quasistatic, low-Reynolds number wall effect experienced by a settling sphere of radius b instantaneously situated at the center of a hollow sphere of radius r_o filled with a dilute suspension of identical, randomly distributed spheres of radii c. (As in the preceding section, the results may be shown to apply equally well to a distribution of suspended sphere sizes provided that the same distribution of radii applies at each point of the suspension.) The analysis is confined to the case where $b/r_o \ll 1$ and $c/r_o \ll 1$.

The total drag force $\mathbf{F}^T$ on the falling ball can be calculated via [19]. Each of the terms appearing therein will be calculated for a fluid possessing an arbitrary suspension viscosity μ, and enclosed within a concentric spherical boundary, which imposes an additional drag factor of K. As the corrections to the velocity field have the net effect of reducing the unbounded suspending fluid settling velocity field $\mathbf{v}'$ to that of the bounded suspension settling velocity field $\mathbf{v}^T$ (cf. [16]), these velocity field corrections will reduce the drag force $\mathbf{F}'$ to the total drag force $\mathbf{F}^T$. At that point the *a priori* assumption of the existence of an effective suspension viscosity μ, modified by an additional wall drag factor K, can be confirmed *a posteriori* by determining their respective values.

As was done for the suspension velocity field $\mathbf{v}$ in the previous section, the drag force $\mathbf{F}'$ due to the sphere settling in the unbounded suspending fluid will be calculated first. Thus, substitution of [10a] into [18] yields, upon integration,

$$\mathbf{F}' = -\frac{\mu}{\mu_0} K\mathbf{F}. \tag{53}$$

The correction $\bar{\mathbf{F}}''$ to the drag force arising from the presence of the suspended spheres can be established by substituting the perturbed velocity field $\bar{\mathbf{v}}''$, given by [36], into [18]. Upon ignoring the higher-order, r_o^{-3}, terms in the perturbed velocity expression [36], the additional force becomes[†]

$$\bar{\mathbf{F}}'' = \frac{3}{2}\mu_0 b \frac{\mu}{\mu_0} K\left(\frac{5\phi}{16\pi\mu_0 b}\right)\mathbf{F} \cdot \int_{S_1}\left[\mathbf{I}\left(1-2\frac{b}{r_o}\right)+\hat{\mathbf{r}}\hat{\mathbf{r}}\right]d^2\hat{\mathbf{r}}$$

$$\equiv \frac{5}{2}\phi\left(1-\frac{3}{2}\frac{b}{r_o}\right)\frac{\mu}{\mu_0}K\mathbf{F}+O\left(\frac{b}{r_o}\right)^2. \tag{54}$$

Next we calculate the wall correction $\mathbf{F}^W$ due to the ball setting in the homogeneous fluid. In the absence of the wall, the dominant term in the far field is the Stokeslet velocity field $\mathbf{v}'$ [10a]. Therefore, we seek the reflection $(\mathbf{v}^W, p^W)$ of this field from the wall. This field pair is governed by Stokes equations [1] and [2] subject to the following boundary conditions:

$$\mathbf{J}^W = \mathbf{0}, \tag{55}$$

$$(\mathbf{v}^W, p^W)\text{finite} \quad \text{for} \quad 0 \leqslant r < r_o \tag{56}$$

$$\mathbf{v}^W = -\mathbf{v}'|_{r=r_o} \equiv -\frac{1}{8\pi\mu_0 r_o}\mathbf{F}\cdot(\mathbf{I}+\hat{\mathbf{r}}\hat{\mathbf{r}}) \quad \text{at} \quad r=r_o. \tag{57}$$

This boundary-value problem may be solved by techniques developed by Brenner (1981) and discussed in detail by Brenner *et al.* (1989). The technique evolves dividing the equations into two parts, the dependence on the driving force $\mathbf{F}$ and the bounding geometry. Since the equations are linear, $(\mathbf{v}^W, p^W)$ can be expressed as $(\mathbf{F}\cdot\mathbf{V}^W, \mathbf{F}\cdot\mathbf{\Pi}^W)$, reducing the problem to solving for the geometric dependence of $(\mathbf{V}^W, \mathbf{\Pi}^W)$. The result obtained by this scheme is

$$\mathbf{v}^W = -\frac{1}{8\pi\mu_0 r_o}\mathbf{F}\cdot\left\{\mathbf{I}\left[3-2\left(\frac{r}{r_o}\right)^2\right]+2\left(\frac{r}{r_o}\right)^2\hat{\mathbf{r}}\hat{\mathbf{r}}\right\}, \tag{58}$$

[†]That the error estimate in [54] is $O(b/r_o)^2$ rather than $O(b/r_o)^3$, which might otherwise be suggested from the magnitude of the terms neglected in [36], arises from the fact that Faxen's law [18] is only correct insofar as the leading-order contribution to the force is concerned.

together with the comparable pressure field p^W, explicit knowledge of which will not be required. Evaluation of [58] at $r = b$ followed by use of the generic formula [18] yields, upon integration over the unit sphere surface,

$$\mathbf{F}^W = \frac{9}{4}\frac{b}{r_o}\frac{\mu}{\mu_0}K\mathbf{F}\left[1 + O\left(\frac{b}{r_o}\right)\right].$$ [59]

The wall correction $\mathbf{\bar{F}}^{WP}$ due to all of the suspended spheres, randomly distributed within the spherical enclosure, may be similarly obtained. As in the preceding paragraph, in order to obtain the reflection of the field $\bar{v}''$ (cf. [36]) from the wall we seek a solution $(\bar{v}^{WP}, \bar{p}^{WP})$ of Stokes equations [1] and [2] satisfying the boundary conditions

$$\mathbf{J}^{WP} = \mathbf{0},$$ [60]

$$(\bar{v}^{WP}, \bar{p}^{WP}) \text{ finite} \quad \text{for} \quad 0 \leqslant r < r_o$$ [61]

and

$$\bar{v}^{WP} = -\bar{v}''|_{r=r_o} \equiv -\frac{\phi}{16\pi\mu_0 r_o}\mathbf{F}\cdot(\mathbf{I} - 3\hat{r}\hat{r}) \quad \text{at} \quad r = r_o.$$ [62]

Again following Brenner (1981), the solution of this boundary-value problem is

$$\bar{v}^{WP} = \frac{\phi}{16\pi u_0 r_o}\mathbf{F}\cdot\left\{\mathbf{I}\left[5 - 6\left(\frac{r}{r_o}\right)^2\right] + 3\left(\frac{r}{r_o}\right)^2\hat{r}\hat{r}\right\}.$$ [63]

Use of [18], with superscript WP written in place of the generic affix *, gives upon integration

$$\mathbf{\bar{F}}^{WP} = -\frac{15}{8}\frac{b}{r_o}\phi\frac{\mu}{\mu_0}K\mathbf{F}\left[1 + O\left(\frac{b}{r_o}\right)\right].$$ [64]

In the preceding calculation leading to [64] we first integrated the velocity disturbance v'' over all possible positions of the centers of the suspected spheres (to obtain $\bar{v}''$) and then corrected for the walls to obtain the correction $\mathbf{\bar{F}}^{WP}$. By direct calculation we have confirmed that [64] is one again obtained if, instead, we first consider a single suspended sphere intracting with the wall (to obtain $\mathbf{F}^{WP}$) and then integrate the result of that caculation over all possible positions of the center of that sphere within the bounded volume $0 \leqslant r \leqslant r_o$ (to obtain $\mathbf{\bar{F}}^{WP}$).

Upon substitution into [19], equations [53], [54], [59] and [64] combine to yield

$$\mathbf{F}^T = \frac{\mu}{\mu_0}K\left[-\mathbf{F} + \frac{5}{2}\phi\left(1 - \frac{6}{4}\frac{b}{r_o}\right)\mathbf{F} + \frac{9}{4}\frac{b}{r_o}\mathbf{F} - \frac{5}{2}\phi\frac{3}{4}\frac{b}{r_o}\mathbf{F}\right]$$

$$\equiv -\mathbf{F}\frac{\mu}{\mu_o}K\left(1 - \frac{5}{2}\phi\right)\left(1 - \frac{9}{4}\frac{b}{r_o}\right)$$ [65]

for the apparent force, correct to $O(b/r_o)$. Since the total drag force is balanced by the body force $\mathbf{F} = -\mathbf{F}^T$, this requires that

$$\frac{\mu}{\mu_0}K\left(1 - \frac{5}{2}\phi\right)\left(1 - \frac{9}{4}\frac{b}{r_o}\right) = 1.$$ [66]

Of the two parenthetical terms in the above equation, the former gives the effect arising from the suspended particles, whereas the latter gives the effect of the container geometry. Since the two terms are separable, the wall-effect coefficient for the suspension is thus identical to that found for a falling ball instantaneously situated at the center of a hollow sphere (radius r_o) filled with a homogeneous Newtonian liquid of viscosity μ (Happel & Brenner 1983) (as may also be seen by setting $\phi = 0$ in [15], whence $\mu/\mu_0 = 1$). Hence, from [66], the wall correction to Stokes law (using, of course, the appropriate viscosity in Stokes law, namely μ_0 for the homogeneous fluid and μ for the suspension) retains the same form for the suspension as it does for the homogeneous fluid.

5. STOKES-LAW RELATIVE SLIP VELOCITY AT THE SUSPENSION SCALE?

Despite the large body of existing literature giving alternative derivations of Einstein's viscosity law [15], we nevertheless believe that our analysis introduces several novel features not heretofore

addressed in rheological issues pertaining to the viscosity of suspensions. Among other things, our analysis appears to demonstrate (but see the discussion at the end of section 6) that Stokes law *without slip* applies on the suspension scale to the mean velocity of a ball settling through a (dilute) suspension; this conclusion is independent of the size of the settling ball in relation to either the sizes of the suspended spheres or the mean distance between them. This lack of suspension-scale slip at the surface of the falling ball is explictly demonstrated by the fact that Stokes law applies in the classical form

$$\mathbf{F} = 6\pi\mu b \mathbf{U},\qquad\qquad [67]$$

in which μ is given by Einstein's suspension viscosity formula [15]. Equation [67] corresponds to a no-slip condition in the sense that the 6π coefficient is valid only for the no-slip case. Were slip to occur (cf. Happel & Brenner 1983; O'Neill *et al.*, 1986) the coefficient of [67] would be diminished—becoming, for example, 4π in the case of perfect slip (Happel & Brenner 1983).

This no-slip conclusion seems very surprising. One would have thought, expecially in the case where the settling sphere is small compared with the size of the suspended spheres and the mean distance between them, that "Knudsen-type" effects would be manifest, leading to an effective slipping motion (cf. O'Neill *et al.*, 1986) at the surface of the settling sphere when viewing its motion on the suspension scale; i.e. given the mean-free-path limitation implicit in the continuum hypothesis for a suspension, it seems surprising to learn by direct calculation that Stokes law, *sans* slip, applies as in [67]. While this implicit, no-slip, Stokes-law-type behavior has been experimentally demonstrated heretofore (Mondy *et al.*, 1986) in nondilute suspensions and for relatively large falling-ball sizes (in relation to the suspended sphere sizes), these data do not seriously penetrate into the range where Knudsen-type issues would come to the fore.

6. SUMMARY AND DISCUSSION

An expression has been derived for the effective viscosity of a dilute, quiescent suspension of neutrally buoyant spheres by two independent, but closely related methods: (i) by calculating the suspension-scale Stokeslet velocity and pressure fields arising from a steady point force $\mathbf{F}$ acting as a fixed interstitial fluid point of the suspension, and subsequently comparing these fields with those arising in a hypothetical homogeneous Newtonain fluid continuum of viscosity μ given by [15]; and (ii) by calculating the retardation in the mean Stokes-law settling velocity of a ball falling through the suspension under the action of a net gravity force $\mathbf{F}$, as compared with the mean velocity achieved by the ball when settling through the particle-free homogeneous fluid of viscosity μ_0. Wall effects were included in the latter calculation.

Einstein's (1906, 1911) classic result, $\mu = \mu_0(1 + \tfrac{5}{2}\phi)$, for the suspension viscosity was reaffirmed in both cases. Morever, the suspension viscosity result was shown to be independent of the relative radii of the settling to suspended spheres (even when the sedimenting sphere is smaller than the suspended spheres), as well as of the degree of polydispersity of the suspended spheres.

Had the rheological calculation been approached instead as a mean field theory, the effective viscosity needed to make the pressure integral convergent would again be Einstein's result, as is assumed in Hinch's (1977) renormalization. Had Hinch's renormalization been used in section 4 (where renormalization was unnecessary since only velocity information was needed to calculate the wall corrections), our results would then have followed trivially from [47] upon replacing the fluid viscosity with the hypothetical homogeneous suspension viscosity.

The falling-ball wall correction to Stokes law obtained in section 4 for a dilute suspension bounded within a concentric hollow sphere was found to be identical to that for a homogeneous Newtonian liquid. These results strongly suggest that well-known (Happel & Brenner 1983) homogeneous Newtonain fluid wall corrections for other bounding geometries could equally well be applied to suspensions. In particular, one may apparently determine the viscosity of a suspension from falling-ball rheometry data (taken in circular cylinders) by *assuming* (*a priori*) that the classical Newtonian, cylindrical wall corrections to Stokes law also apply to suspensions.

Our analysis apparently demonstrates that Stokes law, *without slip*, applies on the suspension scale to the mean velocity of a sphere settling through a dilute suspension; this conclusion is independent of the size of the sedimenting sphere in relation to either the sizes of the suspended

spheres or the mean distance between them. However, this conclusion cannot strictly be claimed as being unequivocal in the absence of detailed calculations that include the higher-order terms neglected in our analysis. In particular, in [13] variations in velocity across the sphere beyond the linear gradient are neglected. Moreover, in applying Faxen's laws ([17] or [18]), third- and higher-order reflections have been neglected. Both of these approximations can obviously be justified asymptotically when the falling ball is large compared with the suspended spheres, namely $b/c \gg 1$. Consequently, the criterion constitutes a *sufficient* condition for the applicability of our analysis. Whether however, this inequality is also a *necessary* condition awaits a more detailed analysis. Existing experimental data (Mondy *et al.* 1986; Milliken *et al.* 1989), albeit obtained in much more concentrated systems than would be expected to lie within the purview of our dilute suspension analysis, appear to support the contention that the analysis applies at least in circumstances for which $b/c = O(1)$, if not $b/c \ll 1$. In particular, at $\phi = 0.25$ the Stokes law suspension viscosity data (Milliken *et al.*, 1989) are independent of the ratio b/c in the range $0.25 \leqslant b/c \leqslant 6.0$ of size ratios measured.

Irrespective of the resolution of the "slip" issue of the preceding paragraph, our analysis appears to be the first whereby Einstein's suspension-viscosity formula [15] has been derived from a *particulate* point of view by solving a nontrivial flow problem. Whereas Einstein's formula has always been derived heretofore by considering a macroscopically *homogeneous* linear shearing field as the underlying generator of the perturbed flow (the perturbation being caused by the presence of the suspended spheres), such is not the case here. Rather, in our analysis the Stokeslet field [10] is the generator, for which the basic flow field is intrinsically *inhomogeneous*.

Finally, we point out that our analysis depends neither on the concept of a (homogeneous) suspension-scale shear field nor stress tensor, concepts which permeate prior suspension-viscosity analyses. Indeed, foregoing completely the question of the range of applicability of our subsequent falling-ball analysis (and concomitant wall effects), our fundamental *Stokeslet* derivation (cf. [14] and [48a,b]) of the Einstein relation [15] eschews the use of these suspension-scale concepts.

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APPENDIX

Integration of [34]

The integral [34] for the cumulative addition to the velocity field arising from the suspended spheres possesses no singularities and decays like R^{-4} as $R \to \infty$. Hence the integration can be

effected directly without renormalization, either by integrating about the singularity with O as the origin or else about the point P as shown in figure 2. In either case, $\mathbf{r}$ is to be held constant during the integration. As our goal is to calculate the wall effect for a spherical shell concentric with the settling sphere, the integration will be performed about O. Upon writing $d^3\mathbf{R} = R^2\,dR\,d^2\hat{\mathbf{R}}$ for the volume element, [34] may be written as

$$\bar{\mathbf{v}}'' = -\frac{15\phi}{8\pi}\int q^{-3}\mathbf{q}\dot{\gamma}_{\mathbf{R}}:\hat{\mathbf{q}}\hat{\mathbf{q}}R^2\,dR\,d^2\hat{\mathbf{R}}, \qquad [A.1]$$

in which $d^2\hat{\mathbf{R}}$ denotes an element of surface area on a unit sphere centered at O in figure 2.

To perform the above integration it is convenient to transform the integrand into an expression containing only the variables $\mathbf{R}$ and $\mathbf{r}$, by eliminating the variable $\mathbf{q}$. An elegant way of effecting this transformation involves use of the identity

$$\frac{\hat{\mathbf{q}}\hat{\mathbf{q}}}{q^3} = \frac{1}{3}\left\{\mathbf{I}\left[\frac{1}{q^3}+\frac{4\pi}{3}\delta(\mathbf{q})\right]+\mathbf{\nabla}\mathbf{\nabla}\frac{1}{q}\right\}. \qquad [A.2]$$

In view of the further identity $\mathbf{I}:\dot{\gamma}_{\mathbf{R}} \equiv \mathbf{\nabla}\cdot\mathbf{v} = 0$, only the $\mathbf{\nabla}\mathbf{\nabla}q^{-1}$ term in [A.2] survives in the integrand of [A.1]. Upon substituting [29] into [A.1] and utilizing the identities $\mathbf{I}:\mathbf{\nabla}\mathbf{\nabla}=\nabla^2$ and $\hat{\mathbf{R}}\hat{\mathbf{R}}:\mathbf{\nabla}\mathbf{\nabla} = \partial^2/\partial R^2$, we obtain

$$\bar{\mathbf{v}}'' = -\frac{15\phi}{64\pi^2\mu_0}\mathbf{F}\cdot\int\left[\frac{\hat{\mathbf{R}}\mathbf{q}}{R^2}\left(\frac{1}{3}\nabla^2\frac{1}{q}-\frac{\partial^2}{\partial R^2}\frac{1}{q}\right)\right]R^2\,dR\,d^2\hat{\mathbf{R}}. \qquad [A.3]$$

However, $\nabla^2 q^{-1} = -4\pi(\mathbf{q})$. Given that $\mathbf{q} = \mathbf{r} - \mathbf{R}$, it readily follows that in [A.3]

$$\int\frac{\hat{\mathbf{R}}\mathbf{q}}{R^2}\delta(\mathbf{q})\,d^3\mathbf{R} \equiv \frac{\hat{\mathbf{r}}}{r^2}\int\mathbf{q}\delta(\mathbf{q})\,d^3\mathbf{q} = 0 \qquad [A.4]$$

since, given the basic properties of the delta function, the latter integral is merely the value of $\mathbf{q}$ evaluated at $\mathbf{q} = 0$. Hence, [A.3] reduces to

$$\bar{\mathbf{v}}'' = \frac{15\phi}{64\pi^2\mu_0}\mathbf{F}\cdot\int\left[\frac{\hat{\mathbf{R}}\mathbf{q}}{R^2}\frac{\partial^2}{\partial R^2}\left(\frac{1}{q}\right)\right]R^2\,dR\,d^2\hat{\mathbf{R}}. \qquad [A.5]$$

As is well-known in potential theory (MacRobert 1967), $1/q$ possesses the spherical harmonic expansion

$$\frac{1}{q} = \frac{1}{r}\sum_{n=0}^{\infty}H_n(s)P_n(\hat{\mathbf{r}}\cdot\hat{\mathbf{R}}), \qquad [A.6]$$

wherein, with

$$s = R/r, \qquad [A.7]$$

$$H_n(s) = \begin{cases} s^n & \text{for } |s| < 1, \\ s^{-(n+1)} & \text{for } |s| > 1. \end{cases} \qquad [A.8]$$

Additionally, $P_n(\hat{\mathbf{r}}\cdot\hat{\mathbf{R}})$ is the Legendre function of order n and argument $\hat{\mathbf{r}}\cdot\hat{\mathbf{R}} \equiv \cos(\hat{\mathbf{r}},\hat{\mathbf{R}})$. Introduction of [A.6] into [A.5] yields

$$\bar{\mathbf{v}}'' = \frac{15\phi}{64\pi^2\mu_0 r}\mathbf{F}\cdot(\mathbf{A}\hat{\mathbf{r}} - \mathbf{B}), \qquad [A.9]$$

with the respective vector and dyadic fields $\mathbf{A}(r_o/r,\hat{\mathbf{r}})$ and $\mathbf{B}(r_o/r,\hat{\mathbf{r}})$ defined as

$$\mathbf{A} = \sum_{n=0}^{\infty}\int_{s=0}^{r_o/r}G_n(s)\,ds\oint_{S_1}\hat{\mathbf{R}}P_n(\hat{\mathbf{r}}\cdot\hat{\mathbf{R}})\,d^2\hat{\mathbf{R}} \qquad [A.10]$$

and

$$\mathbf{B} = \sum_{n=0}^{\infty}\int_{s=0}^{r_o/r}G_n(s)s\,ds\oint_{S_1}\hat{\mathbf{R}}\hat{\mathbf{R}}P_n(\hat{\mathbf{r}}\cdot\hat{\mathbf{R}})\,d^2\hat{\mathbf{R}}, \qquad [A.11]$$

in which

$$G_n(s) = \frac{d^2H_n}{ds^2} \equiv r^2\frac{\partial^2H_n}{\partial R^2}. \qquad [A.12]$$

We have inserted an upper limit of r_o/r rather than ∞ in the s integration so as to permit us subsequently to employ the same scheme to treat the bounded suspension case for a spherical boundary of radius r_o.

In terms of the *tensor* surface spherical harmonics of polyadic rank k (Brenner 1964b), defined as

$$\mathbf{P}_k(\hat{\mathbf{R}}) = \frac{(-1)^k}{k!} r^{k+1} (\nabla)^k \frac{1}{R}, \quad (k = 0, 1, 2, \ldots),$$ [A.13]

we have the identities

$$\hat{\mathbf{R}} = \mathbf{P}_1(\hat{\mathbf{R}})$$ [A.14]

and

$$\hat{\mathbf{R}}\hat{\mathbf{R}} = \tfrac{1}{3}\mathbf{I}P_0(\hat{\mathbf{R}}) + \tfrac{2}{3}\mathbf{P}_2(\hat{\mathbf{R}}),$$ [A.15]

where $P_0 \equiv 1$. Use of these identities together with the orthogonality relationship (Brenner 1964b)

$$\oint_{S_1} \mathbf{P}_k(\hat{\mathbf{R}}) P_n(\hat{\mathbf{r}} \cdot \hat{\mathbf{R}}) \, d^2\hat{\mathbf{R}} = \delta_{kn} \frac{4\pi}{2n+1} \mathbf{P}_n(\hat{\mathbf{r}})$$ [A.16]

(with δ_{kn} the Kronecker delta) permits ready evaluation of the $\mathbf{A}$ and $\mathbf{B}$ integrals. In particular,

$$\mathbf{A} = \sum_{n=0}^{\infty} \oint_{S_1} \mathbf{P}_1 P_n(\hat{\mathbf{r}} \cdot \hat{\mathbf{R}}) \, d^2\hat{\mathbf{R}} \int_{s=0}^{r_o/r} G_n(s) \, ds$$ [A.17]

$$= \frac{4\pi}{3} \mathbf{P}_1 \int_{s=0}^{r_o/r} G_1(s) \, ds$$ [A.18]

$$= \frac{4\pi}{3} \hat{\mathbf{r}} \left[\int_0^1 \frac{d^2 s}{ds^2} \, ds - \int_1^{r_o/r} \frac{d^2}{ds^2} (s^{-2}) \, ds \right]$$ [A.19]

$$\equiv \frac{8\pi}{3} \hat{\mathbf{r}} \left[1 - \left(\frac{r}{r_o}\right)^3 \right].$$ [A.20]

Similarly,

$$\mathbf{B} = \sum_{n=0}^{\infty} \left[\frac{2}{3} \oint_{S_1} \mathbf{P}_2(\hat{\mathbf{R}}) P_n(\hat{\mathbf{r}} \cdot \hat{\mathbf{R}}) \, d^2\hat{\mathbf{R}} + \frac{1}{3} \mathbf{I} \oint_{S_1} P_0(\hat{\mathbf{R}}) P_n(\hat{\mathbf{r}} \cdot \hat{\mathbf{R}}) \, d^2\hat{\mathbf{R}} \right] \int_{s=0}^{r_o/r} G_n(s) s \, ds$$ [A.21]

$$= \frac{8\pi}{15} \mathbf{P}_2(\hat{\mathbf{r}}) \int_{s=0}^{r_o/r} G_2(s) s \, ds + \frac{4\pi}{3} \mathbf{I} P_0(\hat{\mathbf{r}}) \int_{s=0}^{r_o/r} G_0(s) s \, ds$$ [A.22]

$$= \frac{8\pi}{15} \mathbf{P}_2(\hat{\mathbf{r}}) \left[\int_0^1 \frac{d^2}{ds^2} (s^2) s \, ds + \int_1^{r_o/r} \frac{d^2}{ds^2} (s^{-3}) s \, ds \right]$$

$$+ \frac{4\pi}{3} \mathbf{I} \left[\int_0^1 \frac{d^2}{ds^2} (1) s \, ds + \int_1^{r_o/r} \frac{d^2}{ds^2} (s^{-1}) s \, ds \right]$$ [A.23]

$$= \frac{4\pi}{15} (3\hat{\mathbf{r}}\hat{\mathbf{r}} - \mathbf{I}) \left[1 + 4 - 4\left(\frac{r}{r_o}\right)^3 \right] + \frac{8\pi}{3} \mathbf{I} \left(1 - \frac{r}{r_o} \right)$$ [A.24]

$$\equiv 4\pi \hat{\mathbf{r}}\hat{\mathbf{r}} \left[1 - \frac{4}{5}\left(\frac{r}{r_o}\right)^3 \right] + \frac{4\pi}{3} \mathbf{I} \left[1 - 2\left(\frac{r}{r_o}\right) + \frac{4}{5}\left(\frac{r}{r_o}\right)^3 \right].$$ [A.25]

Substituting [A.20] and [A.25] into [A.9] for the cumulative contribution of the suspended spheres to the velocity field thereby gives the result cited in [36].