Global homogenization of a dilute suspension of spheres.

Suspension rheology

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Abstract

A new method for rheologically homogenizing a dilute suspension composed of freely-suspended spherical particles dispersed in a Newtonian fluid is presented: The ensemble-averaged velocity and stress fields obtained for the neutrally-buoyant sphere suspension are compared with the respective velocity and stress fields obtained for a hypothetical homogeneous Newtonian fluid continuum possessing a spatially non-uniform viscosity for the same specified boundaries and ambient flow. The method is global in nature; that is, wall effects and spatial dependence of both the ambient flow and the particle number density are encountered, thereby confirming known classical results up to $O(c^2)$ terms ($c =$ volume concentration of spheres) for the suspension viscosity which have previously been obtained by assuming a priori that the suspension is both unbounded and statistically homogeneous.

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1 Introduction

The rheology of a dilute suspension composed of freely-suspended rigid spherical particles in a Newtonian fluid has been extensively studied since Einstein’s \(^7\) \(O(c)\) classical analysis. His calculation related the increased viscosity of the suspension to the additional dissipation occurring within a ‘suspension cell’ owing to the perturbing presence of a freely suspended sphere in an otherwise uniform shear field. This classical rheological result is supported by the analysis of Keller et al. \(^{11}\), who used variational principles to bound the overall dissipation in a suspension dispersed in a homogeneous shear field under the assumption that most particles lie relatively far from one other.

Such scalar dissipation arguments are viable only in cases where the suspension behaves macroscopically as a homogeneous isotropic fluid. In particular, these methods are inapplicable in circumstances where the suspension-scale stress/rate-of-strain relationship is anisotropic. Batchelor \(^2\) and Brenner \(^3\), building on the pioneering work of Kirkwood \(^{14},\ ^{13}\) and Giesekus \(^8\), developed a general theory from which the stress/strain-rate relation may be obtained. Their methods are based on calculations of the average interstitial-scale stress and velocity gradient tensors, such averaging being performed over a ‘suspension cell.’ In the case of freely suspended spherical particles an isotropic rheological constitutive relation is obtained.

Higher-order terms in the relative-viscosity/suspended-particle concentration expansion have been obtained by Batchelor & Green \(^4,\ ^5\). Their method is based on an ‘ensemble-average’ approach; that is, they obtain the relation between the averages over all possible \(N\)-sphere configurations of the stress and rate-of-strain tensors. To
obtain $O(c^2)$ terms in this expansion, only two-sphere configurations need be considered (a point to which we will subsequently return). Cox & Brenner [6] developed an alternative scheme to obtain these $O(c^2)$ terms, although their generic methods have not yet been implemented in the context of a specific rheological problem.

Each of the previously cited methods is essentially local in nature; that is, effects of bounding walls as well as spatial inhomogeneities in the ambient velocity gradient are neglected. When the ensemble-average approach is applied, and the existence of walls ignored, nonconvergent integrals arise (presumably owing to the non-uniformly valid nature of the Stokes-flow approximation in infinite domains). To overcome this difficulty, *ad hoc* renormalization methods [4],[3], based largely on intuitive arguments, have been invoked.

Hinch [10] developed another renormalization technique (his so-called “second renormalization”) which enabled the calculation of the permeability of a random fixed bed of spherical particles. He also offered further physical insights into the nature of Batchelor & Green’s [3] renormalization scheme, but did not formally resolve the underlying issues. A different insight into renormalization methods was later offered by O’Brien [17], who effectively removed the convergence problem at infinity by correctly adding a “macroscopic boundary integral”. Both Hinch’s [10] and O’Brien’s [17] analyses are again local in nature in the sense that the domain investigated is assumed *a priori* to be unbounded as well as statistically homogeneous.

The subsequent analysis develops an ensemble-average technique, via which we obtain the suspension’s average velocity and stress fields. Wall effects and spatial dependence of the ambient flow are encountered, and renormalization is not needed. Results
expressed in the form of integral representations, in which the kernel is Green’s function and the density function is the surface traction, are derived and compared with the respective velocity and stress fields obtained for a hypothetical homogeneous fluid possessing a *non-uniform* viscosity, for the same specified boundaries and ambient flow. It is demonstrated that for a certain (unique) choice of spatially *non-uniform* viscosity field, the velocity and stress fields obtained for the hypothetical homogeneous medium are asymptotically equal to the respective averages obtained for the suspension throughout the whole domain, except for a thin boundary layer near the walls. Though the results for the suspension-average velocity and stress fields depend upon the boundary’s size and shape, the results for the viscosity field of the hypothetical homogeneous medium are domain independent. The comparison is made up to $O(c^2)$ terms, and the known results of Einstein [7] and Batchelor & Green [4,3] formally confirmed.

Consider $N$ identical rigid spherical particles freely suspended in a homogeneous Newtonian fluid of viscosity $\mu$. The sphere centers are respectively situated at the points $(x_1, \ldots, x_N)$. (For later reference we shall term such a set of locations a ‘configuration’.) Denote by $\Omega$ the domain, and by $(\bar{u}, \bar{p})$ the ambient flow which satisfies the boundary conditions on $\partial\Omega$. Two different types of boundary conditions will be considered. On one subset of the boundary, denoted by $\partial\Omega_u$, we prescribe the velocity (‘adherence-to-walls’), whereas on the complementary subset, denoted by $\partial\Omega_f$ ($\partial\Omega = \partial\Omega_u \cup \partial\Omega_f$), we prescribe the surface traction.

We shall confine the discussion to finite domains ($\text{diam}(\Omega) = R'$) or though the analysis can be extended, in principle, to domains which are infinite in one direction,
e.g., a cylinder. In the latter case, $R'$ is determined by the cross section of the cylinder. We assume that the ambient flow varies on a length scale of $O(R')$. The length scale characterizing the number density $n(x)$ is assumed to be of the same order. This number density is defined as the number of particles per unit volume in any domain of characteristic size $L$ which is much smaller than $R'$ but much larger than the average distance $l = (N/V)^{-1/3}$ between neighboring particles, where $N$ is the total number of spheres in $\Omega$, and $V$ is the domain’s volume. (In the case of a cylinder, $N/V$ should be interpreted as the number density of particles per unit length.)

It is convenient to non-dimensionalize the spatial coordinate by $l$. Denote the dimensionless radius of the suspended spheres by $\epsilon$ ($\epsilon \ll 1$), supposed small since the suspension is assumed dilute. The dimensionless length scale $R'/l$ of the ambient flow will be denoted by $R$ ($R \gg 1$).

When inertial effects are negligible the velocity and pressure fields may be derived from the respective integral representations [16]:

\[ u_i(y, x_1, \ldots, x_N) = \bar{u}_i(y) + \sum_{n=1}^{N} \int_{\partial s_n} T_{ij}(x, y) f_j(x) ds_x, \quad (1.1) \]

\[ p(y, x_1, \ldots, x_N) = \bar{p}(y) + \sum_{n=1}^{N} \int_{\partial s_n} P_j(x, y) f_j(x) ds_x, \quad (1.2) \]

wherein $u$ and $p$ are the configuration-dependent velocity and pressure fields, respectively. The vector $f$ is the surface traction,

\[ f = \sigma(u) \cdot \hat{n}, \quad (1.3) \]

where $\sigma(u)$ is the stress tensor deriving from $(u, p)$, and $\hat{n}$ is the inward unit normal; $\partial s_n$ denotes the surface ($|x - x_n| = \epsilon$) of the $n$'th sphere, and $T(x, y), P(x, y)$ are the
Green’s functions respectively defined by

\[ T_{ij} = t_{ij} + \tau_{ij}, \quad P_i = p_i + \pi_i. \quad (1.4) \]

In the latter, \((t, p)\) denotes the Stokeslet

\[
\begin{align*}
    t_{ij}(x, y) &= \frac{1}{8\pi \mu} \left( \frac{\delta_{ij}}{r} + \frac{r_i r_j}{r^3} \right), \\
    p_i(x, y) &= \frac{1}{4\pi} \frac{r_i}{r^3}
\end{align*}
\]

\[ r = x - y, \tag{1.5} \]

with \((\tau, \pi)\) the Stokeslet image, the latter being a regular solution of the Stokes problem satisfying the boundary condition \(\tau_{ij}\big|_{\partial \Omega_a} = -t_{ij}\big|_{\partial \Omega_a}, \quad \sigma_{ij}(\tau, k) \cdot \hat{n}_j\big|_{\partial \Omega_f} = -\sigma_{ij}(t, k) \cdot \hat{n}_j\big|_{\partial \Omega_f}\), where \(t, k\) denotes the vector \((t_1, t_2, t_3)\). The representation (1.1) and (1.2) [as well as (1.6)] are still valid for cylindrical domains, where \(N\) can be infinite. For instance, since all particles are neutrally buoyant it is easy to show (cf. also appendix A) that as \(|x_n - y| \to \infty\),

\[
\int_{\partial s_n} T_{ij}(x, y)f_j(x)ds_x \sim O(|x_n - y|^{-2}).
\]

If the particles are appropriately numbered, then \(|x_n - y| \geq C(\epsilon, \Omega)n\), whence the series appearing in (1.1) is absolutely convergent.

It is easy to show using (1.1) and (1.2) that the stress field may be expressed in the form

\[
\sigma_{ij}(y, x_1, \ldots, x_N) = \bar{\sigma}_{ij}(y) + \sum_{n=1}^{N} \int_{\partial s_n} (\sigma_y)_{ij}(T_k(x, y))f_k(x, x_1, \ldots, x_N)ds_x. \quad (1.6)
\]

Upon letting \(y\) approach the surface of one of the particles, dot-multiplying by the inward normal, and using the 'jump condition' \([15]\), it may be shown \([16]\) that the
surface traction $f$ satisfies the boundary integral equation

$$\frac{1}{2} f_i(y, x_1, \ldots, x_N) = \tilde{f}_i(y) + \sum_{n=1}^{N} \int_{\partial s_n} (\sigma_y)_{ij}(T_k(x, y)) f_k(x, x_1, \ldots, x_N) ds_x \hat{n}_j(y)$$

(1.7)

for $y \in \bigcup_{n=1}^{N} \partial s_n$, wherein $\tilde{f}$ is the surface traction due to $(\bar{u}, \bar{p})$, and $\sigma_y(T_k)$ is the stress tensor due to $(T_k, P_k)$.

In the next section we introduce an iterative scheme used to approximate the solution of (1.7), thereby obtaining the average velocity and stress fields. Section 3 demonstrates the equivalence of the suspension with the homogeneous medium. Section 4 addresses several key points, insufficiently emphasized within the analysis. In appendix A we prove, upon invoking some mild assumptions, that the scheme presented in Section 2 is asymptotically accurate. Appendix B shows that the effect on the surface traction of any walls bounding the flow, as well as any spatial dependence of the ambient flow, has a negligible effect upon the average velocity field, thereby justifying the analysis of Section 3.

## 2 The average velocity field

We begin by introducing the iterative scheme by which we approximate the solution of (1.7) (or the surface traction field). The first iteration is obtained by solving the one-sphere problem,

$$\frac{1}{2} f_{(n)}^i(y, x_n) = \tilde{f}_i(y) + \int_{\partial s_n} (\sigma_y)_{ij}(T_k(x, y)) f_k(1^n)(x, x_n) ds_x \hat{n}_j(y),$$

(2.1)

for $y \in \partial s_n$. The superscript $(n)$ denotes the fact that unlike $f$, the distribution $f_{(n)}$ is defined only over $\partial s_n$. The second iteration is the solution of yet another one-sphere
Herein, \( f^{(n_1, n_2)}_0 \) is a solution of the following two-sphere problem:

\[
\frac{1}{2} f^{(n_1, n_2)}_0(y, x_{n_1}, x_{n_2}) = \tilde{f}_i(y) + \int_{\partial s_{n_1}} (\sigma_y)_{ij}(T_k) f^{(n_1)}_1(x, x_{1}, \ldots, x_N) ds_x \hat{n}_j(y) + \sum_{n_2=1, n_2 \neq n_1}^N \int_{\partial s_{n_2}} (\sigma_y)_{ij}(T_k) f^{(n_1, n_2)}_0(x, x_{n_1}, x_{n_2}) ds_x \hat{n}_j(y),
\]

(2.2)

Equation (2.2) includes the effect of touching, two-sphere hydrodynamic interactions.

The ideas underlying (2.2) closely resemble those employed in the method of scattering by groups [18]. Note that \( f^{(n_1, n_2)}_0(x, x_{n_1}, x_{n_2}) \sim f^{(n_2)}_0(x, x_{n_2}) \) for \( x \in \partial s_{n_2} \) when \( |x_{n_1} - x_{n_2}| \gg \epsilon \). Were we to substitute \( f^{(n_2)}_0 \) instead of \( f^{(n_1, n_2)}_0 \) into (2.2) (thereby neglecting the near-field effect), the resulting approximation for the average velocity field would be no more accurate than the one based on \( f_0 \) (cf. Appendix A). The usual explanation [3] underlying the need for addressing two-sphere, near-field interactions, is that since the probability of finding a pair of closely proximate spheres is of \( O(c^2) \), errors of \( O(1) \) in the configuration-dependent velocity field arising from the neglect of such near-field interactions cannot be allowed when calculating \( O(c^2) \) terms in the average velocity field. Some support for that intuitive argument is outlined in Appendix A.

It is more convenient to present the traction field in the form of series rather than as a successive sequence of iterations. In this context, the following result is easily
obtained:

\[
f_1^{(n_1)}(x, x_1, \ldots, x_N) - f_0^{(n_1)}(x, x_{n_1}) = \sum_{n_2=1 \atop n_2 \neq n_1}^{N} f_0^{(n_1,n_2)}(x, x_{n_1}, x_{n_2}) - f_0^{(n_1)}(x, x_{n_1}). (2.4)
\]

\[(x \in \partial s_{n_1}).\]

If the configuration is kept fixed and \( \epsilon \) allowed to tend towards zero (and thus \( c \to 0 \)), the right-hand side of the above will tend to zero and will explicitly be of \( O(\epsilon^3) \) relative to \( f_0 \), i.e.

\[
\sum_{n_2=1 \atop n_2 \neq n_1}^{N} |f_0^{(n_1,n_2)} - f_0^{(n_1)}| \leq C \epsilon^3 |f_0^{(n_1)}| \quad \forall 0 < \epsilon < \epsilon_0,
\]

for some \( \epsilon_0 > 0 \) and \( C \) which may depend on the configuration, the boundaries etc.

The next step consists of obtaining the configuration-dependent velocity field. To this end, set

\[
u(y, x_1, \ldots, x_N) = \bar{u}(y) + u_1(y, x_1, \ldots, x_N) + u_2(y, x_1, \ldots, x_N). \quad (2.5)
\]

The field \( u_1 \) is obtained by substituting the solution of (2.1) into (1.1):

\[
u_{1i}(y, x_1, \ldots, x_N) = \sum_{n=1}^{N} \int_{\partial s_n} T_{ij}(x, y) f_0^{(n_1)}(x, x_{n_1}) ds_x. \quad (2.6)
\]

Similarly, \( u_2 \) is obtained by first solving (2.2), substituting the result into (2.3), and subsequently introducing the result of the latter operation into (1.1). This yields

\[
u_{2i}(y, x_1, \ldots, x_N) = \sum_{n_2=1 \atop n_2 \neq n_1}^{N} \int_{\partial s_{n_1}} T_{ij}(x, y) [f_0^{(n_1,n_2)}(x, x_{n_1}, x_{n_2}) - f_0^{(n_1)}(x, x_{n_1})] ds_x. \quad (2.7)
\]

We next seek to obtain the average velocity field. Denote the configurational probability density at \((x_1, \ldots, x_N)\) at a specified instant \( t \) by the multivariable function
The average velocity,

$$\langle u \rangle (y) = \int u(y, x_1, \ldots, x_N) f_N(x_1, \ldots, x_N, t) dx_1 \ldots dx_N,$$

(2.8)
denoted here by $\langle u \rangle$, is obtained by averaging the configuration-dependent velocity over all possible configurations. All other averaged quantities will subsequently be defined and designated in the same manner.

Our first goal is that of obtaining $\langle u_1 \rangle$. In this context we note that

$$u_{1i} = \sum_{n=1}^{N} u'_{1i}(x, x_n),$$

(2.9)

wherein

$$u'_{1i}(x, x_n) = \int_{|\xi|=\epsilon} T_{ij}(x_n + \xi, x) f_0^{(n)}(\xi) ds_\xi.$$  

(2.10)

Substitution into (2.8) yields

$$\langle u_{1i} \rangle = \sum_{n=1}^{N} \int_{\Omega} u'_{1i}(x, x_n) f_1^{(n)}(x_n, t) dx_n,$$

(2.11)

where $f_1^{(n)}(x_n, t)$ is a first-order marginal probability density. The quantity $f_1^{(n)}(x_n, t) dx_n$ represents the probability of finding the n’th particle center $x_n$ in the box $x_{ni}^0 \leq x_{ni} \leq x_{ni}^0 + dx_{ni}$. Due to particle indistinguishability, it is plausible that $f_N(x_1, \ldots, x_N, t)$ is symmetric with respect to all particles. (Otherwise, it may be symmetrized with no changes in any of the following averages.) Hence,

$$f_1^{(n)}(x_n, t) = f_1^{(k)}(x_k, t) \text{ whenever } x_n = x_k.$$

The number density $n(x_1)$, which is the probability density for finding any of the $N$ particles at $x_1$, may be obtained as

$$n(x_1) = N f_1(x_1).$$
Consequently,

$$\langle u_{1i} \rangle (x) = \int_\Omega \int_{|\xi| = \epsilon} T_{ij}(x_1 + \xi, x)f_{0j}^{(1)}(\xi)ds_\xi n(x_1)dx_1. \quad (2.12)$$

In a similar manner, again taking advantage of the symmetry of $f_N(x_1, \ldots, x_N, t)$, we obtain

$$\langle u_{2i} \rangle (x) = \int_\Omega n(x_1)dx_1 \int_\Omega \int_{|\xi| = \epsilon} T_{ij}(x_1 + \xi, x)[f_{0j}^{(1,2)}(x_1, x_2, \xi) - f_{0j}^{(1)}(x_1, \xi)]ds_\xi P(x_2/x_1)dx_2, \quad (2.13)$$

where $P(x_2/x_1)$ is the conditional probability density of finding any particle at $x_2$ given the presence of another particle at $x_1$.

In appendix A we show, under several mild assumptions, that

$$|\langle u \rangle - \bar{u} - \langle u_1 \rangle| \sim O(c^2 \log \frac{R}{\epsilon}) \quad (2.14)$$

and

$$|\langle u \rangle - \bar{u} - \langle u_1 \rangle - \langle u_2 \rangle| \sim O(c^3 \log^2 \frac{R}{\epsilon}) \quad (2.15)$$

relative to $|\bar{u}|$. The error estimates in (2.14) and (2.15) arise respectively from the neglect of two- and three-sphere hydrodynamic interactions. The presence of the term $\ln R/\epsilon$ can be rationalized by the crudeness of the estimates derived in Appendix A. The fact that $\langle u_2 \rangle$ is of $O(c^2)$ (as will be shown later) supports this suggestion.

### 3 Homogenization

The goal of this section is to show that the average velocity field obtained in the preceding section may be approximated by the velocity field obtained for a homogeneous
fluid possessing a non-uniform viscosity, given the same boundaries and ambient flow field for the two cases. (The same can be shown for the comparable stress fields, but the demonstration is omitted in the interests of brevity.)

Our first step is to approximate the surface traction over the surface of a suspended sphere which is located far from the wall \([i.e., \, d(x_n, \partial \Omega) \gg \epsilon, \text{ where } x_n \text{ denotes the location of the sphere’s center}]\). Our approximation represents the solution of the following problem:

\[
\frac{1}{2} f^{(n)}_{0i}(y, x_n) = \left[2\mu G_{ij}(x_n) - \bar{p}(x_n)\delta_{ij}\right]\hat{n}_j(y) + \frac{3}{4\pi} \int_{\partial s_n} \frac{r_i r_j r_k}{r^5} f^{(n)}_{0k}(x, x_n) ds_n \hat{n}_j(y),
\]

(3.1)

wherein

\[
G_{ij} = \frac{1}{2} \frac{\partial \bar{u}_i}{\partial x_j} + \frac{\partial \bar{u}_j}{\partial x_i}.
\]

Equation (3.1) is almost identical with (2.1) except for two modifications: (a) neglect of the variation of \(G\) and \(\bar{p}\) over \(\partial s_n\), i.e., \(G(y) \equiv G(x_n)\,;\,\bar{p}(y) \equiv \bar{p}(x_n)\); (b) neglect of the Stokeslet image portion of the integral’s kernel. Item (a) results in an \(O(\epsilon/R)\) error, whereas (b) produces an \((\epsilon^3/[d(x_n, \partial \Omega)]^3)\) error, which means it is of \(O(1)\) near the walls. Appendix B provides an estimate of the overall effect of these neglections on the average velocity field. Equation (3.1) can readily be solved \([12]\), to obtain

\[
f^{(n)}_{0i} = 5\mu G_{ij}(x_n)\hat{n}_j(y) - \bar{p}(x_n)\hat{n}_i(y).
\]

(3.2)

Note that the portion of \(f_0\) due to shear has been increased by a factor of \(5/2\) relative to \(f\). This coefficient arises in the above context from the eigenvalue of the integral operator corresponding to shear flow over a sphere, which is \(1/5\).
As is shown in appendix B, the error due to the items neglected above is negligible even if we use (3.2) everywhere (including the vicinity of the walls). Substitution into (2.12) yields

\[
\langle u_{1i} \rangle = -\frac{4}{3}\pi\epsilon^3 \int_{\Omega/B(x,\epsilon)} \left[ 1 + \frac{1}{10}\epsilon^2 \frac{\partial^2}{\partial x_{1p}^2} \right] \frac{\partial}{\partial x_{1k}} [T_{ij}(x_1, x)] 5\mu G_{jk}(x_1)n(x_1)dx_1 . \tag{3.3}
\]

The domain \( B(x, \epsilon) \) has been omitted since

\[
\int_{|\xi| = \epsilon} T_{ij}(x_1 + \xi, x)f_{0j}^{(1)}(\xi)ds_\xi \cong G_{ij}(x_1 - x_j) + O(\frac{\epsilon}{R}) ,
\]

the \( O(\epsilon/R) \) error being a consequence of our approximation (3.2) to \( f_0 \). In view of (2.12) the integral over \( B(x, \epsilon) \) vanishes. The \( O(\epsilon^2) \) term appearing in (3.3) also impacts negligibly upon \( \langle u_{1i} \rangle \) since by applying the divergence theorem to it we obtain

\[
\int_{\Omega/B(x,\epsilon)} \frac{1}{10}\epsilon^2 \frac{\partial^2}{\partial x_{1p}^2} \frac{\partial}{\partial x_{1k}} [T_{ij}(x_1, x)] 5\mu G_{jk}(x_1)n(x_1)dx_1 =
\]

\[
\int_{\partial\Omega\cup\partial B(x,\epsilon)} \epsilon^2 \frac{\partial^2}{\partial x_{1p}^2} [T_{ij}(x_1, x)] 5\mu G_{jk}(x_1)n(x_1)\hat{n}_kds_{x_1} , \tag{3.4}
\]

The integral over \( \partial B(x, \epsilon) \) is \( O(\epsilon^3/R^3) \) since \( T \cong t \) in that domain. For those points \( x \) which lie well within the interior of \( \Omega \), i.e., \( d(x, \partial\Omega) \sim O(R) \), we have that \( \nabla \nabla T \sim O(1/R^3) \). Consequently, the above integral is bounded by \( C\epsilon^2/R^2\|G\|_R \sup_{x \in \Omega} n \). As such, it is then of \( O(\epsilon^2/R^2) \) with respect to the \( O(1) \) term in (3.3). Hence,

\[
\langle u_{1i} \rangle \cong -\frac{4}{3}\pi\epsilon^3 \int_{\Omega/B(x,\epsilon)} \frac{\partial}{\partial x_{1k}} [T_{ij}(x_1, x)] 5\mu G_{jk}(x_1)n(x_1)dx_1 , \tag{3.5}
\]

which upon applying the divergence theorem in conjunction with the fact that \( T(x_1, x) = 0 \) for \( x \in \partial\Omega_u \) yields (recall that \( \hat{n} \) is the inward normal)

\[
\langle u_{1i} \rangle \cong \frac{4}{3}\pi\epsilon^3 \left\{ \int_{\Omega} T_{ij}(x_1, x)5\mu\frac{\partial}{\partial x_{1k}} [G_{jk}(x_1)n(x_1)] dx_1 + \right. \\
\left. + \int_{\partial\Omega_j} T_{ij}(x_1, x)5\mu G_{jk}(x_1)n(x_1)\hat{n}_kds_{x_1} \right\} + O(\frac{\epsilon}{R}) . \tag{3.6}
\]
We have ignored here the fact that \( n(x_1) = 0 \) for \( d(x_1, \partial \Omega) < \epsilon \), and instead assumed it to be smooth throughout the whole domain. It can easily be shown that the error produced by this assumption is of \( O(\epsilon/R) \).

Our goal is to now obtain \( \langle u_1 \rangle \) as a solution of a Stokes problem in \( \Omega \) for a homogeneous fluid possessing a non-uniform (but continuous) viscosity distribution, given the same ambient flow in both cases. This Stokes problem has the following form:

\[
\begin{align*}
\frac{\partial v}{\partial x_i} &= 0 \quad \text{for } x \in \Omega, \\
\frac{\partial}{\partial x_j} \left[ \mu_s \left( \frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right) \right] &= \frac{\partial p}{\partial x_i} \quad \text{for } x \in \Omega, \\
v &= \bar{u} \quad \text{for } x \in \partial \Omega_u, \\
f_s(v) &= f(\bar{u}) \quad \text{for } x \in \partial \Omega_f.
\end{align*}
\]

In the latter, \( f_s \) is determined by the stress/rate-of-strain relation for the hypothetical homogeneous medium, i.e.,

\[
f_s(v) = \left\{ \mu_s \left[ \nabla v + \nabla v^\dagger \right] - pI \right\} \cdot \hat{n},
\]

in contrast to \( f \), which is determined by the stress/rate-of-strain relation for the Newtonian fluid (with \( \mu_s \) replaced by \( \mu \) in the above). We seek a solution of (3.8) for circumstances in which \( \mu_s \) is nearly constant. To this end we set

\[
\begin{align*}
\mu_s(x) &= \mu \left\{ 1 + a_1 \frac{4}{3} \pi \epsilon^3 n(x) + a_2 \left[ \frac{4}{3} \pi \epsilon^3 n(x) \right]^2 + O(\epsilon^9) \right\}, \\
v &= \bar{u} + \frac{4}{3} \pi \epsilon^3 v_1 + \left[ \frac{4}{3} \pi \epsilon^3 \right]^2 v_2 + O(\epsilon^9), \\
p &= \bar{p} + \frac{4}{3} \pi \epsilon^3 p_1 + \left[ \frac{4}{3} \pi \epsilon^3 \right]^2 p_2 + O(\epsilon^9),
\end{align*}
\]

where \( n(x) \) may be any positive continuous function. The latter choice of notation derives from the fact that, as will subsequently be shown, in order to obtain equality
between the velocity field obtained from (3.8) and (3.9) and the respective average velocity field obtained for the suspension, \( n(x) \) must be identical with the number density. Here, \( \epsilon^3 \) may be interpreted as the order of the viscosity fluctuation, since we use the same dimensionless spatial coordinate as before, and thus \( n(x) \sim O(1) \). The unknown constants \( a_1 \) and \( a_2 \) will be determined subsequently. Upon substituting (3.9) into (3.8) the following boundary-value problem is obtained for \( v_1 \):

\[
\frac{\partial v_{1i}}{\partial x_i} = 0, \quad (3.9a)
\]

\[
\mu \frac{\partial^2 v_{1i}}{\partial x_j^2} - \frac{\partial p_1}{\partial x_i} = -2a_1 \mu \frac{\partial}{\partial x_j}[nG_{ij}], \quad (3.9b)
\]

\[
v_{1i}|_{x \in \partial \Omega_u} = 0, \quad (3.9c)
\]

\[
[f_1 + 2a_1 n(x) \mu \mathbf{G} \cdot \mathbf{n}]|_{x \in \partial \Omega_f} = 0. \quad (3.9d)
\]

The solution of this set of equations may be expressed in the form of the following integral representation [9]:

\[
v_{1i} = - \int_{\partial \Omega} t_{ij}(x_1, x) \left[ \mu \left( \frac{\partial v_{1j}}{\partial x_1} + \frac{\partial v_{1k}}{\partial x_{1j}} \right) - p_1 \delta_{jk} \right] \mathbf{n}_k ds_{x_1} + 2a_1 \mu \int_{\Omega} t_{ij}(x_1, x) \frac{\partial}{\partial x_k}[nG_{jk}] dx_1. \quad (3.10)
\]

Using Green’s theorem together with (3.10c) it is easy to show that if we replace the Stokeslet \( t \) appearing on the right-hand side of (3.10) by its image \( \tau \), the left-hand side will vanish. Hence,

\[
v_{1i} = 2a_1 \mu \int_{\Omega} T_{ij}(x_1, x) \frac{\partial}{\partial x_{1k}}[G_{jk}(x_1)n(x_1)] dx_1 + 2a_1 \mu \int_{\partial \Omega_f} T_{ij}(x_1, x) G_{jk}(x_1)n(x_1) \mathbf{n}_k ds_{x_1}. \quad (3.11)
\]

For the choice \( a_1 = 5/2 \), Eq. (3.6) yields

\[
\langle u_1 \rangle = \frac{4}{3} \pi \epsilon^3 v_1. \quad (3.12)
\]
We have thus demonstrated that, to terms of $O(c)$, equality exists between the average velocity field obtained for the suspension and the velocity field obtained for a hypothetical homogeneous medium characterized by the non-uniform viscosity field $\mu_s = \mu[1 + 5/2 c(x)]$.

We next seek to obtain a comparable equality, but now up to terms of $O(c^2)$, between the suspension-average and homogeneous fields. To this end it is necessary to separately discuss the respective contributions to the average velocity field of the relatively distant and relatively close sphere pairs. For the case of relatively distant pairs of spheres we may use the approximation

$$
\left( f^{(n_1,n_2)}_{0i} - f^{(n_1)}_{0i} \right) \bigg|_{x \in \partial s_{n_1}} = \frac{4}{3} \pi \epsilon^3 \left\{ \frac{5}{2} \mu^2 G_{jm}(x_{n_2}) \cdot \left[ \frac{\partial T_{ij}}{\partial x_{n_1}^j} + \frac{\partial T_{kj}}{\partial x_{n_1}^i} \right]_{(x_{n_2},x_{n_1})} \hat{n}_k(x) - \left[ 5\mu G_{jk}(x_{n_2}) - \bar{p}(x_{n_2}) \delta_{jk} \right] \frac{\partial P_j}{\partial x_{n_2}^k} \bigg|_{(x_{n_2},x_{n_1})} \hat{n}_i(x) \right\}, \tag{3.13}
$$

which is valid for $|x_{n_1} - x_{n_2}| \gg \epsilon$. When $|x_{n_1} - x_{n_2}| \sim O(\epsilon)$, Eq. (2.3) has to be modified in the same manner used to obtain (3.1), and the exact solution found. [We do not however, provide an error estimate, as previously done in appendix B in order to justify (3.1)]. Substituting (3.13) into (2.13) for $|x_{n_1} - x_{n_2}| \geq L \gg \epsilon$ gives

$$
\langle u_{2i} \rangle (x) \cong \left( \frac{4}{3} \pi \epsilon^3 \right)^2 \frac{5}{2} \mu^2 \int_{\Omega/B(x_1,\epsilon)} \frac{\partial}{\partial x_{1k}} [T_{ij}(x_1,x)] n(x_1) dx_1 \cdot \int_{\Omega/B(x_1,L)} G_{pm}(x_2) \left[ \frac{\partial T_{pj}}{\partial x_{2k}} + \frac{\partial T_{kj}}{\partial x_{2p}} \right]_{(x_2,x_1)} n(x_2) dx_2 + \int_{\Omega} n(x_1) dx_1 \int_{2\epsilon \leq |x_2 - x_1| \leq L} \int_{|\xi| = \epsilon} T_{ij}(x_1 + \xi, x) \left[ f^{(1,2)}_{0ij}(x_1, x_2, \xi) - f^{(1)}_{0ij}(x_1, \xi) \right] d\xi \cdot P(x_2/x_1) dx_2 + O\left( \frac{\epsilon^2}{L^2} \right), \tag{3.14}
$$

where we have used two different properties of $P(x_2/x_1)$, namely: $P(x_2/x_1) \sim n$ for
\[|x_1 - x_2| \gg \epsilon, \text{ and } P(x_2/x_1) = 0 \text{ for } |x_1 - x_2| < 2\epsilon.\]

Upon applying the divergence theorem to the first term on the right-hand side of (3.14) and using the approximation

\[T_{ij}(x_1, x_2)|_{x_1=x_2}=L \cong t_{ij}(x_1, x_2) + O\left(\frac{L}{R}\right), \tag{3.15}\]

we obtain

\[
\langle u^2 \rangle(x) \cong -\frac{4}{3}\pi \varepsilon^3 \mu \int_{\Omega} \partial T_{ij} \left[ \frac{4}{3} \pi \varepsilon^3 G_{jk}(x_1)n(x_1) + G_{1jk}(x_1) \right] n(x_1) dx_1 +
\]

\[+ \int_{\Omega} n^2(x_1) dx_1 \int_{2\varepsilon \leq |x_2-x_1| \leq L} \int_{|\xi|=\varepsilon} T_{ij}(x_1 + \xi, \xi) [f^{(1,2)}_{ij}(x_1, x_2, \xi) - f^{(1)}_{ij}(x_1, \xi)] ds \cdot q(x_2/x_1) dx_2 + O\left(\varepsilon^2 R^{1/3}\right), \tag{3.16}\]

wherein

\[G_{1jk} = \frac{1}{2} \left[ \frac{\partial \langle u_{1i} \rangle}{\partial x_j} + \frac{\partial \langle u_{1j} \rangle}{\partial x_i} \right], \tag{3.17}\]

\[q(x_2/x_1) = P(x_2/x_1)/n(x_1), \text{ and wherein } L \text{ was chosen to be } (\varepsilon^2 R)^{1/3} \text{ so as to minimize the error.}\]

To facilitate evaluation of the near-field term we discuss the domains \(|x_1 - x| \geq \epsilon\) and \(|x_1 - x| < \epsilon\) separately. For \(|x_1 - x| \geq \epsilon\) we expand \(T\) in power series of \(\xi\), i.e.,

\[T_{ij}(x_1 + \xi, \xi) = \sum_{n=0}^{\infty} \frac{1}{n!} \frac{\partial}{\partial(x_1)_{m_1}} \cdots \frac{\partial}{\partial(x_1)_{m_n}} T_{ij}(x_1, \xi_1 \cdots \xi_m). \tag{3.18}\]

The next step entails obtaining \(f^{(1,2)}\) as a linear combination of the \(G\) components:

\[f^{(1,2)}_{ij} = A_{ijkl} G_{km} n_l.\]

Observe that due to the special symmetry of the two-sphere geometry,

\[A_{ijkl}(x_2 - x_1, \xi) = A'_{ijkl} (R \cdot (x_2 - x_1), R \cdot \xi),\]
where $\mathbf{R}$ is any rotation tensor, and the $A'_{jklm}$ are the components of $\mathbf{A}$ in a coordinate system rotated by $\mathbf{R}$; thus,

$$A_{jklm}(\mathbf{x}_2 - \mathbf{x}_1, \xi) = A'_{jklm}(\mathbf{x}_1 - \mathbf{x}_2, -\xi) = A_{jklm}(\mathbf{x}_1 - \mathbf{x}_2, \xi) .$$

(3.19)

Consequently, the contributions of all the even terms in (3.18) vanish if we assume

$$q(\mathbf{x}_2/\mathbf{x}_1) = q(2\mathbf{x}_1-\mathbf{x}_2/\mathbf{x}_1),$$

which appears to be reasonable hypothesis for $d(\mathbf{x}_1, \partial \Omega) \gg \epsilon$; (recall that $|\mathbf{x}_2 - \mathbf{x}_1| \leq L$).

The contribution of the term corresponding to $n = 1$ in (3.18) is

$$-\int_{\Omega/B(\mathbf{x}, \epsilon)} n^2 \partial T_{ij}(\mathbf{x}_1) dx_1 \int_{2\epsilon \leq |\mathbf{x}_2 - \mathbf{x}_1| \leq L} \left[ s^{(1,2)}_{jk}(\mathbf{x}_1, \mathbf{x}_2) - s^{(1)}_{jk}(\mathbf{x}_1) \right] q(\mathbf{x}_2/\mathbf{x}_1) dx_2 ,$$

(3.20)

wherein $s^{(1,2)}$ and $s^{(1)}$ are the stresslets of the sphere at $\mathbf{x}_1$ for the respective two- and one-sphere problems.

The contributions of all other odd terms can be shown to be negligible: applying the divergence theorem to the $2n + 1$ term yields

$$\int_{\partial \Omega \cup \partial B(\mathbf{x}, \epsilon)} \frac{\partial}{\partial (x_1)_{m_2}} \cdots \frac{\partial}{\partial (x_1)_{m_{2n+1}}} T_{ij}(\mathbf{x}_1, \mathbf{x}) \cdot \int_{|\xi| = \epsilon} \xi_{m_1} \cdots \xi_{m_{2n+1}} \left[ f^{(1,2)}_{0j}(\mathbf{x}_1, \mathbf{x}_2, \xi) - f^{(1)}_{0j}(\mathbf{x}_1, \xi) \right] ds_\xi g(\mathbf{x}_2/\mathbf{x}_1) dx_2 n^2(\mathbf{x}_1) n_{m_1} ds_{x_1} .$$

(3.21)

Upon pursuing the same general arguments as those following (3) we find that the above term is of $O(\epsilon^{2n+1}/R^{2n+1})$.

Thus, upon combining Eqs. (3.16) to (3), and applying the divergence theorem once
again, we obtain

\[ \langle u_{2i} \rangle (x) \cong -\frac{4}{3} \pi \epsilon^3 \mu \int_{\Omega} T_{ij} \frac{\partial}{\partial x_{1k}} \left\{ \left[ \frac{4}{3} \pi \epsilon^3 G_{jk}(x_1) n(x_1) + G_{1jk}(x_1) \right] n(x_1) \right\} dx_1 + \]

\[ + \int_{\partial \Omega_f} T_{ij} \left( \frac{4}{3} \pi \epsilon^3 G_{jk}(x_1) n(x_1) + G_{1jk}(x_1) \right) n(x_1) \hat{n}_k ds_{x_1} + \]

\[ + \int_{\Omega_f} T_{ij} \frac{\partial}{\partial x_{1k}} \left\{ \int_{2 \epsilon \leq |x_2 - x_1| \leq L} [s^{(1,2)}_{jk}(x_1, x_2) - s^{(1)}_{jk}(x_1)] q(x_2/x_1) dx_2 n^2(x_1) \right\} dx_1 + \]

\[ + \int_{\partial \Omega_f} T_{ij} \int_{2 \epsilon \leq |x_2 - x_1| \leq L} [s^{(1,2)}_{jk}(x_1, x_2) - s^{(1)}_{jk}(x_1)] q(x_2/x_1) dx_2 n^2(x_1) \hat{n}_k ds_{x_1} + \]

\[ + n^2(x) \int_{|x_1 - x| < \epsilon} \int_{2 \epsilon \leq |x_2 - x_1| \leq L} \int_{|\xi| = \epsilon} T_{ij}(x_1 + \xi, x)[f^{(1,2)}_{0j}(x_1, x_2, \xi) - f^{(1)}_{0j}(x_1, \xi)] d\xi \cdot \]

\[ q(x_2/x_1) dx_2 dx_1 + O(\frac{\epsilon^2}{L}). \quad (3.22) \]

Utilizing (3.19) it is easy to show that the last term on the right-hand side of (3.22) vanishes by applying the following transformation:

\[ \xi \rightarrow -\xi \quad ; \quad x_1 \rightarrow 2x - x_1 \quad ; \quad x_2 \rightarrow 2x_1 - x_2. \quad \text{Q.E.D.} \]

The function \( q(x_2/x_1) \) obtained by Batchelor & Green [3] for elongational flows considers only the effect of convection; [for simple shear flow the effect of convection alone is insufficient to determine \( q(x_2/x_1) \)]. For the case of elongational flows, \( J \) is a hydrodynamic coefficient defined in [4] and [3]. The coefficient 4.45 is taken from the numerical calculations of Yoon & Kim [20]. Substituting in (3.22) we have

\[ \int_{2 \epsilon \leq |x_2 - x_1| \leq L} [s^{(1,2)}_{jk}(x_1, x_2) - s^{(1)}_{jk}(x_1)] q(x_2/x_1) dx_2 \cong \]

\[ \cong \left( \frac{4}{3} \pi \epsilon^3 \right)^2 2 \mu G_{jk}(x_1) \frac{15}{2} \int_2^\infty J(\zeta) q(\zeta) d\zeta + O(\frac{L}{R}) \cong \]

\[ \cong 4.45 \left( \frac{4}{3} \pi \epsilon^3 \right)^2 2 \mu G_{jk}(x_1), \quad (3.23) \]

where \( J \) is a hydrodynamic coefficient defined in [4] and [3]. The coefficient 4.45 is taken from the numerical calculations of Yoon & Kim [20]. Substituting in (3.22) we have.
obtain

\[
\langle u_{2i} \rangle (x) \cong \frac{4}{3} \pi \epsilon^3 \left\{ 5\mu \int_{\Omega} T_{ij} \frac{\partial}{\partial x_{1k}} [G_{1jk}(x_1)n(x_1)] dx_1 + \\
+ \int_{\partial\Omega_f} T_{ij} G_{1jk}(x_1)n(x_1)\hat{n}_k ds_{x_1} \right\} + \\
+ \left( \frac{5}{2} + 4.45 \right) 2\mu \left[ \int_{\Omega} T_{ij} \frac{\partial}{\partial x_{1k}} \left( \frac{4}{3} \pi \epsilon^3 G_{jk}(x_1)n^2(x_1) \right) dx_1 \\
+ \int_{\partial\Omega_f} T_{ij} \frac{4}{3} \pi \epsilon^3 G_{jk}(x_1)n^2(x_1)\hat{n}_k ds_{x_1} \right] \right\}. \tag{3.24}
\]

The \(5/2\) coefficient appearing in the second integral on the right-hand side arises from far-field, two-sphere interactions. Batchelor & Green \[3\] arrive at the same conclusion by considering the somewhat artificial situation \(q(x_2/x_1) = 0\) for \(|x_1 - x_2| \leq L\). Here, it is derived more naturally through the separation of far-field and near-field contributions.

To obtain \(O(\epsilon^2)\) equality between the suspension-average velocity field and the velocity field obtained for a homogeneous fluid possessing a non-uniform viscosity, we consider the next higher-order balance equations for the homogeneous continuum problem, namely

\[
\frac{\partial v_{2i}}{\partial x_i} = 0, \tag{3.25a}
\]

\[
\mu \frac{\partial^2 v_{2i}}{\partial x_j^2} - \frac{\partial p_2}{\partial x_i} = -5\mu \frac{\partial}{\partial x_j} [nG_{1ij}'] - 2a_2 \mu \frac{\partial}{\partial x_j} [n^2 G_{ij}], \tag{3.25b}
\]

\[
v_1|_{x \in \partial\Omega_u} = 0, \tag{3.25c}
\]

\[
\{ f_2 + [5n(x)G_1'^\prime + 2a_2 n(x)\mu G_1] \cdot \hat{n} \}_{x \in \partial\Omega_f} = 0, \tag{3.25d}
\]
wherein \((4/3) \pi \epsilon^3 G' = G_1\) Similarly to (3.11) we obtain

\[
v_{2i}(x) = 5 \mu \left\{ \int_{\Omega} T_{ij} \frac{\partial}{\partial x_k} \left[ G'_{1jk}(x_1) n(x_1) \right] dx_1 + \int_{\partial \Omega} T_{ij} G'_{1jk}(x_1) n(x_1) \hat{n}_k ds_{x_1} \right\} +
\]

\[
+ 2a_2 \mu \left\{ \int_{\Omega} T_{ij} \frac{\partial}{\partial x_k} \left[ G_{jk}(x_1) n^2(x_1) \right] dx_1 + \int_{\partial \Omega} T_{ij} G_{jk}(x_1) n^2(x_1) \hat{n}_k ds_{x_1} \right\}. \quad (3.26)
\]

Thus, the choice \(a_2 = 6.95\) yields

\[
\langle u_2 \rangle = \left( \frac{4}{3} \pi \epsilon^3 \right)^2 v_1,
\]

thereby establishing an \(O(\epsilon^2)\) equality between the respective velocity fields.

## 4 Concluding remarks

This section addresses several key points insufficiently emphasized in the prior analysis:

1. **The error:** Two types of errors are produced in the process of approximation. The first arises from neglecting higher-order hydrodynamic interactions (e.g., three-sphere interactions in the case when the expansion addresses only two-sphere interactions) and is of \(O(c^{n+1})\), where \(n\) refers to the \(n\)-sphere interactions explicitly considered. The second error stems from ignoring ‘global’ effects (arising from the presence of boundaries and spatial inhomogeneities) on the surface traction, and is of \(O(\epsilon/R, c^2(\epsilon/R)(2/3))\). Thus, there is no point in considering more than \(n\) terms if \(c^n \sim O((\epsilon/R), c(\epsilon/R)(2/3))\). We may conclude, therefore, that for a dilute suspension the required number of terms in our expansion is rather small, whereas for a more concentrated suspension it might be useful to obtain more terms (assuming, of course, that the expansion is convergent, an issue which is not yet clear).
2. Comparison with the local approach: The major difference between the *global* approach, presented in the present work, and the classical *local* one (cf. [4], [3], [10], [17]) resides in the order in which limits are being taken. Local analysis assumes *a priori* that global effects are unimportant, i.e., that the medium is unbounded in extent and that the number density and ambient rate-of-strain are spatially uniform. Mathematically, this means that the ratio of particle size to macroscopic length scale, \( \epsilon/R \), is set to be equal to zero *before* performing any further analysis. This limit process, which is certainly an idealization, was based in prior analyses on the hope that in the real physical systems encountered in practice, neither the presence of boundaries nor of spatial inhomogeneities would have any effect on the suspension viscosity, which was intuitively expected to be a local quantity. In contrast to the local approach, we first calculate the velocity field – including the global effects – and only then do we pass to the above limit. Thereby, we show explicitly that

\[
\left| \langle u \rangle - \bar{u} - \frac{4}{3} \epsilon^3 v_1 - \left( \frac{4}{3} \epsilon^3 \right)^2 v_2 \right| \leq C \sup_{x \in \Omega} |\bar{u}| \left[ \epsilon \epsilon^2 \left( \frac{\epsilon^2}{R} \right)^{2/3} + \epsilon^3 \right],
\]

thus demonstrating that the suspension is equipollent, on average, to a homogeneous medium, possessing a non-uniform viscosity field. It should be emphasized that *this result is independent of the domain's size and shape.*

In addition for providing an estimate for the error generated by the presence of boundaries and spatial inhomogeneities, global analysis possesses another significant advantage over the local approach: It avoids the classical Stokesian divergence problem, and hence the need for renormalization [4], [3], [10], [17]! It is well known that the Stokesian divergence issue poses serious problems when seeking physically meaningful
solutions in unbounded domains, for both suspensions and homogeneous fluids. In fact, had we worked in $\mathbb{R}^3$, many of the integrals in sections 2 and 3 would have been divergent. The best way to avoid the Stokesian divergence is to discuss large but finite domains, which is exactly what was done in Ref. [19] for the sedimentation case, and what has been done here in the present paper for the suspension rheology case.

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A Error estimates for one- and two-sphere modes

The goal of this appendix is to demonstrate the validity of (2.14) and (2.15). To this end, it is first necessary to prove uniform boundedness of $f$.

Lemma 1

\[ \exists \varepsilon_0 > 0 \text{ such that } \|f\|_{L^\infty(\bigcup_{n=1}^{N} \partial s_n)} \leq C(N, \Omega) \mu \|G\| \quad \forall 0 < \varepsilon \leq \varepsilon_0 \quad \text{and} \quad (x_1, \ldots, x_N) \in D^N \]

for every $D \subset \Omega$ satisfying $d(\partial D, \partial \Omega) \geq \varepsilon$ and for smooth $\partial \Omega$.

The proof of lemma 1 [1] utilizes the fact that $f$ tends to the solution of $N$-particle problem in $\mathbb{R}^3$. Yet, it is still necessary to show that $f$ remains uniformly bounded even if some of the particles are allowed to approach the boundary proximities. To this end we first assume that $f$ tends, as $\varepsilon \to 0$ and for $d(x_n, \partial \Omega) \leq C\varepsilon \quad \forall 1 \leq n \leq N$ to
the solution of $N$-particle problem near a flat wall (which looks intuitively correct, but seems to be technically difficult to prove). Then, similar arguments to those presented in the proof of lemma 1 (where \[1\] can be applied in order to demonstrate uniform boundedness in this case as well.

The above uniform boundedness property suffices, as the subsequent analysis demonstrates, to prove \[2.14\] and \[2.15\] for fixed $N$ as $\epsilon \to 0$. Yet, a much more interesting limiting case arises when both $N \to \infty$ and $\epsilon \to 0$, for fixed $N/R^3$. In terms of the original physical variables, this case arises if we either let the boundaries approach infinity, or else let the average distance between particles tend to zero. Uniform boundedness of $f$ in that limit seems, however, to be difficult to prove. It would perhaps be easier to prove that the average of $f$ with respect to $N - 3$ particle locations is bounded, i.e.,

$$\int f(x_1, \ldots, x_N) dx_1 \cdots dx_N \leq C(\Omega) \mu \|G\| f_3(x_1, x_2, x_3),$$

which, if correct, suffices in order to prove both \[2.14\] and \[2.15\]. We shall, thus assume the validity of \[A.1\] in the limit $N \to \infty$. Further research is, however, necessary in order to prove this assumption.

Before deriving \[2.14\] and \[2.15\] we prove the following auxiliary result:

**Lemma 2**

$$\sup_{y \in \partial \Omega_1} \left| \int_{\partial \Omega_2} \sigma_y(T)(x, y) \cdot f(x) ds_x \cdot \hat{n}(y) \right| \leq C \epsilon^3 \frac{\|f\|_{L^\infty(\partial \Omega_2)}}{|x_{n_1} - x_{n_2}|^3}. \quad (A.2)$$

**proof:** Use of the fact that all spheres are neutrally buoyant furnishes the inequality
\[
\sup_{y \in \partial s_2} \left| \int_{\partial s_2} \sigma_y(T)(x,y) \cdot f(x) ds_x \cdot \hat{n}(y) \right| \leq \left\| (x - x_n) \cdot \nabla_x \sigma_y(T)(x',y) \cdot \hat{n}(y) \right\|_{L^1(\partial s_2)} \left\| f \right\|_{L^\infty(\partial s_2)} , \quad (A.3a)
\]

wherein
\[
x' = x_n + \theta(x - x_n), \quad (0 \leq \theta \leq 1).
\]

(A.3b)

It can be shown \([15]\) that
\[
\left| \nabla_x \sigma_y(T)(x',y) \right| \leq \frac{C}{|x' - y|^3}, \quad (x', y) \in \Omega. \quad (A.4)
\]

Consequently, (A.2) holds for \(|x_{n_2} - x_{n_1}| \geq 3\varepsilon\). For \(|x_{n_2} - x_{n_1}| < 3\varepsilon\) it is easy to show that for \(x \neq y\),
\[
\left| (x - x_n) \cdot \nabla_x \sigma_y(T)(x',y) \cdot \hat{n}(y) \right| \leq C_1 \left| (x - x_n) \cdot \nabla \frac{rrr}{r^5} \right|_{(r=x-y)} + \frac{C_2}{r} .
\]

(A.5)

Hence, since
\[
\sup_{y \in \partial s_2} \left\| (x - x_n) \cdot \nabla_x \frac{rrr}{r^5} \right\|_{(r=x-y)} \hat{n}(y) \right\|_{L^1(\partial s_n)}
\]

is finite, (A.2) remains valid for \(2\varepsilon \leq |x_{n_2} - x_{n_1}| < 3\varepsilon\) as well.

□

In the reminder of this appendix we derive first the one-sphere approximation (2.14), followed subsequently by the two-sphere approximation (2.15).

One-sphere case: The equation for \(f - f_0^{(n)}\) can be obtained via (1.7) and (2.1) as
\[
\frac{1}{2}(f_i - f_0^{(n_1)})(y,x,x_1,\ldots,x_N) = \bar{f}_i(y) + \int_{\partial s_{n_1}} (\sigma_y)_{ij}(T_k)(f_k - f_0^{(n_1)})(x,x_1,\ldots,x_N)ds_x \hat{n}_j(y) +
\]
\[
+ \sum_{n_2=1}^{N} \int_{\partial s_{n_2}} (\sigma_y)_{ij}(T_k)f_k(x,x_{n_1},\ldots,x_N)ds_x \hat{n}_j(y). \quad (A.6)
\]
Inasmuch as (A.6) is a Fredholm equation, use of (A.2) gives

\[ \| f - f^n_0 \|_{L^\infty(\partial s_{n_1})} \leq C \sum_{\substack{n_2=1 \\ n_2 \neq n_1}}^N \frac{\| f \| \epsilon^3}{|x_{n_2} - x_{n_1}|^3}. \]  

(A.7)

The error in the configuration-dependent velocity is

\[ \Delta u_{1i} = \sum_{n=1}^N \int_{\partial s_n} T_{ij}(f_j - f^n_{0j})dx. \]  

(A.8)

Invoking the fact that [15]

\[ |\nabla_x T|(x,y) \leq C \frac{|x - y|}{|x|}, \]

we obtain

\[ |\Delta u_1| \leq \frac{C}{\mu} \sum_{n=1}^N \frac{\| f \| \epsilon^6}{|x_{n_2} - x_{n_1}|^3|x_{n_1} - x|^2}. \]  

(A.9)

Assuming that (A.1) is valid, the error in the average velocity is bounded by

\[ |< u - \bar{u} - u_1>| \leq \epsilon^6 C \| G \| \int_{|x_2 - x_1| \geq \epsilon} n(x_1)dx_1 \int_{\Omega} \frac{P(x_2/x_1)dx_2}{|x_2 - x_1|^2}, \]  

(A.10)

where \( C \) is independent of \( N \). (For fixed \( N \) the above estimate follows from lemma 1.)

Since \( P(x_2/x_1) \sim n(x_1) \) for \( |x_2 - x_1| \gg \epsilon \), it can obviously be asserted that

\[ P(x_2/x_1) \leq C_1 n(x_1) \quad \forall |x_2 - x_1| \geq C_2 \epsilon, \]

where \( C_1 \) and \( C_2 \) are constants of \( O(1) \), i.e., independent of \( \epsilon \). Obviously, since

\[ \int_{|x_2 - x_1| \leq C \epsilon} P(x_2/x_1)dx_2 \approx \int_{|x_2 - x_1| \leq C \epsilon} n(x_2)dx_2 \]

for \( C \gg 1 \), we may assert that

\[ \int_{|x_2 - x_1| \leq C \epsilon} \frac{P(x_2/x_1)dx_2}{|x_2 - x_1|^3} \leq \frac{1}{2\epsilon^3} \int_{|x_2 - x_1| \leq C \epsilon} P(x_2/x_1)dx_2 \leq C_3 n(x_1). \]
In the above we have implicitly assumed that $P(\alpha(x_2 - x_1)/x_1, \alpha \epsilon) \leq CP(x_2 - x_1/x_1, \epsilon)$ for all $0 < \alpha \leq 1$, where $C$ is independent of $\alpha$, $\epsilon$, and $N$. Such an assumption is clearly necessary in order to guarantee the boundedness of $P(x_2/x_1)$ in the limit $N \to \infty$, $\epsilon \to 0$. The approximations we derive will thus be valid only in cases where the pair probability behaves according to the above assumption. It is expected, however, that in most cases $P(x_2/x_1)$ will satisfy this assumption, since it obeys simple conservation laws which do not depend upon the $\epsilon$ and $N$ values. (cf. Ref. [3] for instance).

Consequently,

$$\int_{\Omega} P(x_2/x_1) dx_2 \leq \bar{C} \int_{|x_2-x_1| \geq 2\epsilon} n(x_2) dx_2. \tag{A.11}$$

Hence, since $n(x_2) \leq n_{\text{max}} \sim O(1)$, one obtains via (A.10) and (A.11) the inequality

$$|\langle u - \bar{u} - u_1 \rangle| \leq \epsilon^6 n_{\text{max}}^2 \|G\| R \ln \left(\frac{R}{\epsilon}\right), \tag{A.12}$$

a result which coincides with (2.14).

**Two-sphere case:** Similarly to (A.6), one may obtain the equation

$$\frac{1}{2}(f_i - f_{0i}^{(n_1,n_2)})(y, x, x_1, \ldots, x_N) = \bar{f}_i(y) +$$

$$+ \int_{\partial s_{n_1} \cup \partial s_{n_2}} (\sigma_g)_{ij}(T_k)(f_k - f_{0k}^{(n_1,n_2)})(x, x_1, \ldots, x_N) ds_x \hat{n}_j(y) +$$

$$+ \sum_{n_3=1}^{N} \sum_{n_3 \neq n_1, n_2} \int_{\partial s_{n_3}} (\sigma_g)_{ij}(T_k)f_k(x, x_{n_1}, \ldots, x_N) ds_x \hat{n}_j(y). \tag{A.13}$$

Application of the same procedure used to obtain (A.7) gives, with the aid of lemma
We seek an estimate for \( \|f - f_0^{(n_1,n_2)}\|_{L^\infty(\partial s_{n_2})} \). We therefore first interpret (A.13) as a Fredholm equation over \( \partial s_{n_2} \) to obtain

\[
\|f - f_0^{(n_1,n_2)}\|_{L^\infty(\partial s_{n_2})} \leq C \mu \|G\| \epsilon^3 \sum_{n_3=1 \atop n_3 \neq n_1,n_2}^N \frac{1}{|x_n - x_n^2|^3} + \frac{\epsilon^3}{|x_n - x_n^1|^3 |x_n^2 - x_n^1|^3} \left| \bar{f}_n(y) - f_0^{(n_1,n_2)}(x_{n_1}, x_{n_2}) \right| ds_{x_n^2} \hat{n}_j(y). \tag{A.15}
\]

In combination, Eqs. (A.14) and (A.15) give

\[
\|f - f_0^{(n_1,n_2)}\|_{L^\infty(\partial s_{n_2})} \leq C \mu \|G\| \epsilon^3 \sum_{n_3=1 \atop n_3 \neq n_1,n_2}^N \frac{1}{|x_n - x_n^2|^3} + \frac{\epsilon^3}{|x_n - x_n^1|^3 |x_n^2 - x_n^1|^3} \sum_{n_2=1 \atop n_2 \neq n_1}^N \int_{\partial s_{n_2}} (\sigma y)_{ij} (T_{,k}) (f_k - f_0^{(n_1,n_2)}(x_{n_1}, x_{n_2})) ds_{x_k} \hat{n}_j(y). \tag{A.16}
\]

The equation governing \( f - f_1^{(n)} \) may be obtained from (1.7) and (2.3) as

\[
\frac{1}{2} (f_i - f_1^{(n_1)})(y, x_1, \ldots, x_N) = \bar{f}_i(y) + \int_{\partial s_{n_1}} (\sigma y)_{ij} (T_{,k}) (f_k - f_1^{(n_1)})(x, x_1, \ldots, x_N) ds_{x_k} \hat{n}_j(y) + \sum_{n_2=1 \atop n_2 \neq n_1}^N \int_{\partial s_{n_2}} (\sigma y)_{ij} (T_{,k}) [f_k - f_0^{(n_1,n_2)}(x_{n_1}, x_{n_2})] ds_{x_k} \hat{n}_j(y). \tag{A.17}
\]

Consequently, upon utilizing (A.16) one obtains

\[
\|f - f_1^{(n_1)}\|_{L^\infty(\partial s_{n_1})} \leq C \mu \|G\| \epsilon^6.
\]

\[
\sum_{n_2=1 \atop n_2 \neq n_1}^N \sum_{n_3=1 \atop n_3 \neq n_1,n_2}^N \frac{1}{|x_n - x_n^2|^3 |x_n^2 - x_n^1|^3} + \frac{\epsilon^3}{|x_n - x_n^1|^3 |x_n^2 - x_n^1|^3} \sum_{n_2=1 \atop n_2 \neq n_1}^N \int_{\partial s_{n_2}} (\sigma y)_{ij} (T_{,k}) [f_k - f_0^{(n_1,n_2)}(x_{n_1}, x_{n_2})] ds_{x_k} \hat{n}_j(y). \tag{A.18}
\]
The error in the average velocity is therefore

\[
\left| \langle \mathbf{u} - \bar{\mathbf{u}} - \mathbf{u}_1 - \mathbf{u}_2 \rangle \right| \leq \epsilon C \| G \| \int_{\Omega} \frac{n(x_1)dx_1}{|x_1 - x|^2} \left[ \int_{\Omega} \frac{P(x_2/x_1)dx_2}{|x_2 - x_1|^3} \int_{\Omega} \frac{P(x_3/x_2, x_1)dx_3}{|x_3 - x_2|^3} + \int_{\Omega} \frac{e^3 P(x_2/x_1)dx_2}{|x_2 - x_1|^6} \int_{\Omega} \frac{P(x_3/x_2, x_1)dx_3}{|x_3 - x_2|^3} \right] + \int_{\Omega} \frac{e^3 P(x_2/x_1)dx_2}{|x_2 - x_1|^6} \int_{\Omega} \frac{P(x_3/x_2, x_1)dx_3}{|x_3 - x_2|^3} \],
\]

which is valid in the limit \( N \to \infty \) if (A.1) is correct.

**B The error in the average velocity due to wall effects and spatial inhomogeneities**

Write (2.1) in the form

\[
\frac{1}{2} f_0^{(n)}(y) = [2\mu G_{ij}(x_n) - \bar{p}(x_n)]\hat{n}_j(y) + \frac{3}{4\pi} \int_{\partial s_n} \frac{r_i r_j r_k}{r^5} f_0^{(n)}(x, x_n)ds_x \hat{n}_j(y) + \{2\mu [G_{ij}(y) - G_{ij}(x_n)] - [\bar{p}(y) - \bar{p}(x_n)]\}\hat{n}_j(y) + \int_{\partial s_n} (\sigma_{ij}(\tau, k)(x, y)) f_0^{(n)}(x, x_n)ds_x \hat{n}_j(y). \quad (B.1)
\]

Obviously,

\[
\| G(y) - G(x_n) \|_{L^\infty(\Omega)} \leq C \frac{\epsilon}{R} \| G \|_{L^\infty(\Omega)}, \quad (B.2a)
\]

\[
\| \bar{p}(y) - \bar{p}(x_n) \|_{L^\infty(\Omega)} \leq C \frac{\epsilon}{R} \| \bar{p} \|_{L^\infty(\Omega)} \quad (B.2b)
\]

and

\[
\left| \int_{\partial s_n} (\sigma_{ij}(\tau, k)(x, y)) f_0^{(n)}(x, y)ds_x \hat{n}_j(y) \right| \leq C \epsilon^3 \left| \nabla_x \sigma_{ij}(\tau)(x_n, y) \right| |\mu| \| G \|. \quad (B.3)
\]

Upon utilizing the fact that \( \tau(x, y) \) is uniformly bounded throughout the whole domain, except when both \( x \) and \( y \) lie near \( \partial \Omega \), we obtain the rather crude estimate

\[
\nabla \tau(x, y) \leq \frac{C}{d(x, \partial \Omega)^3},
\]

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Hence,
\[\int_{\partial s_n} (\sigma_y)_{ij}(\mathbf{r}, k)(x, y) f_{nk}^{(n)} ds_x \nabla_j (y) \leq C \frac{\epsilon^3}{d(x_n, \partial \Omega)^3} \mu \| \mathbf{G} \|. \quad (B.4)\]

Thus,
\[\| f_0^n - 5\mu \mathbf{G} \cdot \mathbf{n} + \mathbf{p} \mathbf{n} \|_{L^\infty(\partial s_n)} \leq C \mu \| \mathbf{G} \| \left[ \frac{\epsilon^3}{d(x_n, \partial \Omega)^3} + \frac{\epsilon}{R} \right]. \quad (B.5)\]

The error in the configuration-dependent velocity field may now be estimated as
\[|\Delta \mathbf{u}_1| \leq C \| \mathbf{G} \| \sum_{n=1}^N \frac{e^3}{|x - x^1_n|^2} \left[ \frac{e^3}{d(x_n, \partial \Omega)^3} + \frac{\epsilon}{R} \right]. \quad (B.6)\]

Consequently, the error in the average velocity is
\[| \langle \Delta \mathbf{u}_1 \rangle | \leq C \| \mathbf{G} \| \left[ \int_{\Omega} \frac{e^6 n(x_1) dx_1}{|x - x_1|^2 d(x_1, \partial \Omega)^3} + e C \right]. \quad (B.7)\]

Motivated by the fact that \(d(x, \partial \Omega) \gg \epsilon\), we estimate the first term in brackets as
\[\int_{\Omega} \frac{e^6 n(x_1) dx_1}{|x - x_1|^2 d(x_1, \partial \Omega)^3} \leq \frac{8e^3}{d(x, \partial \Omega)^3} \int_{B(x, \epsilon^2 d(x, \partial \Omega))} \frac{e^3 n(x_1) dx_1}{|x - x_1|^2} + \frac{4e^2}{d(x, \partial \Omega)^2} \int_{\Omega \setminus B(x, \epsilon^2 d(x, \partial \Omega))} \frac{e^3 n(x_1) dx_1}{d(x_1, \partial \Omega)^3}\]

or
\[\int_{\Omega} \frac{e^6 n(x_1) dx_1}{|x - x_1|^2 d(x_1, \partial \Omega)^3} \leq cR \left[ C_1 \frac{e^3}{d(x, \partial \Omega)^3} + C_2 \frac{Re}{d(x, \partial \Omega)^2} \right], \quad (B.8)\]

whence
\[| \langle \Delta \mathbf{u}_1 \rangle | \leq C \| \mathbf{G} \| Re \frac{Re}{d(x, \partial \Omega)^2}. \]

More refined estimates can probably be derived from (B.7); however, for \(d(x, \partial \Omega) \sim O(R)\) the error is of \(O(\epsilon R)\), which is sufficiently small to justify the degree of omission made in utilizing (3.2).
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[1] Details of the proof can be obtained directly from the authors.

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