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Hydrodynamic interactions and properties of suspensions

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Abstract. A review is given of a recently developed scheme to evaluate the mobility tensors of an arbitrary number of spheres, also in the case that these spheres are enclosed in a spherical container. Self-diffusion and sedimentation are discussed as applications.

1. Introduction

Hydrodynamic interactions between spheres moving in a viscous fluid play an important role in many physical and physico-chemical problems. They were studied rather extensively by the so-called methods of reflections, inaugurated by Smoluchowski [1] for those situations in which the fluid can be described by the quasi-static Stokes equation, i.e. by the linearized Navier–Stokes equation for incompressible steady flow.

In practice, because of the increasing complexity of the problem, essentially only the two-sphere case was analyzed. Smoluchowski [1], Faxén [2], Dahl [3] and Happel and Brenner [4] calculated for this case the friction tensors to higher and higher order in the inverse distance between the spheres, while e.g. Burgers [5], Batchelor [6] and Felderhof [7] evaluated the mobility tensors (which are elements of the inverse of the friction tensor matrix).

When considering the properties of dilute suspensions of spherical (uncharged) particles one may of course restrict oneself to take only pair interactions into account [6, 8]. It was quite generally hoped or presumed that pair-wise additivity of hydrodynamic interactions would hold in concentrated suspensions as well (see in this connexion the excellent review of Pusey and Tough [9]). But in view of the long range nature of these interactions, such an assumption seemed rather doubtful. It is for this reason that we recently developed a systematic expansion to treat the full many-sphere problem [10]. Kynch [11] had already by a method reflection derived expressions for three and four spheres. However his work seems to have remained practically unnoticed. (In connection with the many-sphere problem we mention also work by Muthukumar [12] and Yoshizaki and Yamakawa [13].)

In this paper a review is given of the approach to, and the solution of, the many-sphere problem given in Ref. 10, an approach that is based on a method of induced forces. The review includes a later extension of the approach to incorporate also wall effects, in particular hydrodynamic interactions with the wall of a spherical container [14]. Finally self-diffusion and sedimentation are briefly discussed as properties of suspensions for which the many-body hydrodynamic interactions play an essential role. A similar review, without the discussion of wall effects in a spherical vessel, and of sedimentation, has also been published elsewhere [15].

2. Fluid equation of motion; formal solution

One considers a system of N macroscopic spheres of radii a_i ($i = 1, 2, \dots, N$) moving with velocities $\vec{u}_i$ and angular velocities $\vec{\omega}_i$ through an otherwise unbounded incompressible viscous fluid. The centres of the spheres are located at positions $\vec{R}_i$.

The motion of the fluid is described by the quasi-static Stokes equation

$$\vec{\nabla} \cdot \mathbf{P}(\vec{r}) = 0 \tag{2.1}$$

$$\text{for all } |\vec{r} - R_i| > a_i, \qquad i = 1, 2, \dots, N$$

$$\vec{\nabla} \cdot \vec{v}(\vec{r}) = 0 \tag{2.2}$$

Here $\vec{v}(\vec{r})$ is the fluid velocity field and $\mathbf{P}$ the pressure tensor whose components $P_{\alpha\beta}$ (Greek indices denote Cartesian components) are given by

$$P_{\alpha\beta} = p\delta_{\alpha\beta} - \eta\left(\frac{\partial v_\alpha}{\partial r_\beta} + \frac{\partial v_\beta}{\partial r_\alpha}\right) \tag{2.3}$$

In the last equation p is the hydrostatic pressure and η the viscosity of the fluid.

The force $\vec{K}_i$ and torque $\vec{T}_i$ exerted by the fiuid on sphere i are given by the following surface integrals over the pressure tensor.

$$\vec{K}_i = -\int_{S_i} dS\mathbf{P}(\vec{r}) \cdot \hat{n}_i, \tag{2.4}$$

$$\vec{T}_i = -\int_{S_i} dS(\vec{r} - \vec{R}_i) \wedge \mathbf{P} \cdot \hat{n}_i, \tag{2.5}$$

with S_i the surface of sphere i and $\hat{n}_i$ a unit vector normal to this surface pointing in the outward direction.

In order to solve the hydrodynamic equations (2.1)–(2.3), and then determine from (2.4) and (2.5) the forces and the torques, we take the case of stick boundary conditions

$$\vec{v}(\vec{r}) = \vec{u}_i + \vec{\omega}_i \wedge (\vec{r} - \vec{R}_i) \quad \text{for} \quad |\vec{r} - \vec{R}_i| = a_i. \tag{2.6}$$

The problem posed by equations (2.1)–(2.6) may be reformulated by introducing force densities $\vec{F}_j(\vec{r})$ induced on the spheres and extending the fluid equations to hold within the spheres. In this formulation the fluid equations are written in the equivalent form

$$\vec{\nabla}\cdot\mathbf{P}=\sum_{j=1}^{N}\vec{F}_j(\vec{r}) \qquad \text{for all } \vec{r}, \tag{2.7}$$
$$\vec{\nabla}\cdot\vec{v}(\vec{r})=0$$

with $\vec{F}_j(\vec{r})=0$ for $|\vec{r}-\vec{R}_j|>a_j$. Inside the spheres the fluid velocity and pressure field have the extensions

$$\vec{v}(\vec{r})=\vec{u}_i+\vec{\omega}_i\wedge(\vec{r}_i-\vec{R}_i) \quad \text{for} \quad |\vec{r}-\vec{R}_i|\leq a_i, \tag{2.8}$$

$$p(\vec{r})=0 \quad \text{for} \quad |\vec{r}-\vec{R}_i|<a_i. \tag{2.9}$$

As a consequence of these extensions it follows from (2.7) with (2.3) that the induced force density is of the form

$$\vec{F}_i(\vec{r})=a_i^{-2}\vec{f}_i(\hat{n}_i)\,\delta(|\vec{r}-\vec{R}_i|-a_i) \tag{2.10}$$

The factor a_i^{-2} has been introduced for convenience.

It follows with Gauss' theorem from equations (2.4), (2.5), (2.7) and (2.10) that the force $\vec{K}_i$ and the torque $\vec{T}_i$ can be expressed in terms of the induced surface forces $\vec{f}_i$ as

$$\vec{K}_i=-\int d\hat{n}_i\vec{f}_i(\hat{n}_i) \tag{2.11}$$

$$\vec{T}_i=-\int d\hat{n}_i\hat{n}_i\wedge\vec{f}_i(\hat{n}_i) \tag{2.12}$$

We shall now rewrite the equations of motion (2.7) in wave vector representation. One then has, together with (2.3)

$$\eta k^2\vec{v}(\vec{k})=-i\vec{k}p(\vec{k})+\sum_{j=1}^{N}e^{-i\vec{k}\cdot\vec{R}_j}\vec{F}_j(\vec{k}) \tag{2.13}$$

$$\vec{k}\cdot\vec{v}(\vec{k})=0 \tag{2.14}$$

where $\vec{v}(\vec{k})$ is the Fourier transformed fluid velocity field

$$\vec{v}(\vec{k})=\int d\vec{r}e^{-i\vec{k}\cdot\vec{r}}\vec{v}(\vec{r}). \tag{2.15}$$

The induced force density $\vec{F}_i(\vec{k})$, defined in a reference frame in which sphere i is at rest, is given by

$$\vec{F}_i(\vec{k})=\int d\vec{r}e^{-i\vec{k}\cdot(\vec{r}-\vec{R}_i)}\vec{F}_i(\vec{r}). \tag{2.16}$$

By applying the operator $\mathbf{1}-\hat{k}\hat{k}$, where $\hat{k}=\vec{k}/k$ is the unit vector in the direction of $\vec{k}$ and $\mathbf{1}$ the unit tensor, to both sides of equation (2.13) the pressure may be eliminated from this equation. If one then also assumes that the fluid unperturbed by the motion of the spheres is at rest, one has for the fluid velocity field, which also obeys equation (2.14), the formal solution of the equation of motion

$$\vec{v}(\vec{k}) = \sum_j \eta^{-1} k^{-2} e^{-i\vec{k}\cdot\vec{R}_j} (\mathbf{1} - \hat{k}\hat{k}) \cdot \vec{F}_j(\vec{k}). \tag{2.17}$$

From this equation one may calculate the forces and torques exerted by the fluid on the spheres and thereby evaluate the hydrodynamic interactions which are set up between the spheres when they move.

3. Induced force multipoles and velocity surface moments

It is convenient for the purpose of analyzing hydrodynamic interactions to define, in terms of the surface forces $\vec{f}_i(\hat{n}_i)$, cf. (2.10), irreducible induced force multipoles:

$$\mathbf{F}_i^{(l+1)} \equiv (l!)^{-1} \int d\hat{n}_i\, \overline{\hat{n}_i^l} \vec{f}_i(\hat{n}_i), \qquad l = 0, 1, 2, \ldots \tag{3.1}$$

Here $\overline{\hat{n}_i^l}$ is the irreducible tensor of rank l, i.e. the tensor traceless and symmetric in any pair of its indices, constructed with the vector $\hat{n}_i$.[1]) According to equations (2.11), (2.12) and (3.1)

$$\vec{K}_i = -\mathbf{F}_i^{(1)} \tag{3.2}$$

and

$$\vec{T}_i = a_i \boldsymbol{\varepsilon} : \mathbf{F}_i^{(2a)}, \qquad \mathbf{F}_i^{(2a)} = -\frac{1}{2a_i} \boldsymbol{\varepsilon} \cdot \vec{T}_i, \tag{3.3}$$

where $\boldsymbol{\varepsilon}$ is the Levi–Civita tensor, for which one has the identity $\boldsymbol{\varepsilon} : \boldsymbol{\varepsilon} = -2\,\mathbf{1}$, and $\mathbf{F}_i^{(2a)}$ the antisymmetric part of $\mathbf{F}^{(2)}$.

One can show that the surface force $\vec{f}_i(\hat{n}_i)$ has the following expansion in terms of irreducible force multipoles:

$$\vec{f}_i(\hat{n}_i) = \frac{1}{4\pi} \sum_{l=0}^{\infty} (2l+1)!!\, \overline{\hat{n}_i^l} \odot \mathbf{F}_i^{(l+1)} \tag{3.4}$$

where $(2l+1)!! = 1 \cdot 3 \cdot 5 \cdots (2l-1) \cdot (2l+1)$; the dot $\odot$ denotes a full l fold contraction between the tensors $\overline{\hat{n}^l}$ and $\mathbf{F}^{(l+1)}$, with the convention that the last index of $\overline{\hat{n}_i^l}$ is contracted with the first index of $\mathbf{F}_i^{(l+1)}$, etc. The expansion (3.4) is written in a coordinate-free way; it is equivalent to an expansion in spherical harmonics to which it can be reduced after introduction of polar coordinates.

The above expansion follows in a straightforward way if use is made of the following properties [10, 16] of the irreducible tensors $\overline{\hat{n}_i^l}$:

$$\frac{1}{4\pi} \sum_{l=0}^{\infty} \frac{(2l+1)!!}{l!} \overline{\hat{n}_i^l} \odot \overline{\hat{n}_i'^l} = \delta(\hat{n}_i - \hat{n}_i') \tag{3.5}$$

$$\frac{1}{4\pi} \int d\hat{n}_i \overline{\hat{n}_i^l}\, \overline{\hat{n}_i^m} = \frac{l!}{(2l+1)!!} \delta_{lm} \mathbf{\Delta}^{(l,l)} \tag{3.6}$$

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[1]) For $l = 1, 2, 3$ one has, see e.g. ref. 16, $\vec{b} = \vec{b}$, $b_\alpha b_\beta = b_\alpha b_\beta - \frac{1}{3} b^2 \delta_{\alpha\beta}$, $b_\alpha b_\beta b_\gamma = b_\alpha b_\beta b_\gamma - \frac{1}{5}(b_\alpha \delta_{\beta\gamma} + b_\beta \delta_{\alpha\gamma} + b_\gamma \delta_{\alpha\beta}) b^2$.

In equation (3.6) $\mathbf{\Delta}^{(l,l)}$ represents an isotropic tensor of rank $2l$ which projects out the irreducible part of a tensor of rank l.

For the Fourier transformed induced force density $\vec{F}_i(\vec{k})$ (cf. equations (2.10) and (2.16)) the expansion (3.4) leads to

$$\vec{F}_i(\vec{k}) = \sum_{l=0}^{\infty} (2l+1)!!\, i^{-l} j_l(ka) \overline{\hat{k}^l} \odot \mathbf{F}_i^{(l+1)} \tag{3.7}$$

with $j_l(x)$ the spherical Bessel function of order l. In obtaining (3.7) from (3.6) use has been made of the identity [17]

$$\frac{\overline{\partial^l}}{\partial \vec{k}^l} \frac{\sin k}{k} = (-1)^l\, \overline{\hat{k}^l}\, j_l(k). \tag{3.8}$$

Next to the irreducible induced force multipoles defined above, we also introduce irreducible surface moments of the fluid velocity field. The irreducible surface moment of order n is defined as

$$\begin{aligned}(2n+1)!!\,\overline{\overline{\hat{n}_i^n}\vec{v}(\vec{r})}^{S_i} &= \frac{(2n+1)!!}{4\pi a_i^2} \int d\vec{r}\, \overline{\hat{n}_i^n} \vec{v}(\vec{r})\, \delta(|\vec{r}-\vec{R}_i| - a_i) \\ &= i^n \frac{(2n+1)!!}{(2\pi)^3} \int d\vec{k} j_n(ka) \overline{\hat{k}^n} \vec{v}(\vec{k}) e^{i\vec{k}\cdot\vec{R}_i}, \quad n = 0, 1, 2, \ldots\end{aligned} \tag{3.9}$$

The numerical factor $(2n+1)!!$ is introduced for convenience. The velocity surface moments are essentially the coefficients of an expansion of the fluid velocity field at the surfaces of the spheres in irreducible tensors $\overline{\hat{n}_i^n}$. As a consequence of the boundary condition (2.6) and the orthogonality condition (3.6) all these moments vanish for $n \geqslant 2$, while the first two moments are

$$\overline{\vec{v}(\vec{r})}^{S_i} = \vec{u}_i, \qquad \overline{3\hat{n}_i \vec{v}(\vec{r})}^{S_i} = a_i \boldsymbol{\varepsilon} \cdot \vec{\omega}_i \tag{3.10}$$

We are now in a position to relate the induced force multipoles (3.1) to the surface multipoles of the fluid velocity field (3.9). If one uses the boundary conditions (2.6) at the left hand side of equation (3.9) and substitutes the formal solution (2.17) in combination with the series expansion (3.7) at the right hand side, one obtains indeed the following system of linear coupled equations

$$6\pi\eta a_i(\vec{u}_i\, \delta_{n,1} + a_i \boldsymbol{\varepsilon} \cdot \vec{\omega}_i\, \delta_{n,2}) = \sum_{j=1}^{N} \sum_{m=1}^{\infty} \mathbf{A}_{ij}^{(n,m)} \odot \mathbf{F}_j^{(m)} \tag{3.11}$$

where the coefficients, the so-called connectors, which are tensors of rank $n+m$ are given by

$$\begin{aligned}\mathbf{A}_{ij}^{(n,m)} &= \frac{3a_i(2n-1)!!\,(2m-1)!!}{4\pi^2}\, i^{n-m} \\ &\quad \times \int d\vec{k} e^{i\vec{k}\cdot\vec{R}_{ij}} \overline{\hat{k}^{n-1}} (\mathbf{1} - \hat{k}\hat{k}) \overline{\hat{k}^{m-1}} k^{-2} j_{n-1}(ka_i) j_{m-1}(ka_j)\end{aligned} \tag{3.12}$$

with $\vec{R}_{ij} = \vec{R}_j - \vec{R}_i$. For $i \neq j$ the connectors are only defined when $R_{ij} = |\vec{R}_{ij}| > a_i + a_j$. In equation (3.11) the dot $\odot$ denotes a full contraction between the last m indices of $\mathbf{A}_{ij}^{(n,m)}$ and the m indices of $\mathbf{F}_j^{(m)}$.

Before deriving below from the set of equations (3.11) expressions for the mobility tensors which relate the forces and the torques exerted by the fluid on the spheres to their velocities and angular velocities, we shall discuss in the next section some properties of the connectors as well as their explicit form.

4. The connectors and their properties

One may verify by inspection of expression (3.12), that the connectors satisfy the symmetry relation

$$a_j \mathbf{A}_{ij}^{(n,m)} = a_i \tilde{\mathbf{A}}_{ji}^{(m,n)} \tag{4.1}$$

where $\tilde{\mathbf{C}}$ is a generalized transpose of a tensor $\mathbf{C}$ of arbitrary rank p,

$$(\tilde{\mathbf{C}})_{\alpha_1\alpha_2 \cdots \alpha_{p-1}\alpha_p} = (\mathbf{C})_{\alpha_p\alpha_{p-1} \cdots \alpha_2\alpha_1} \tag{4.2}$$

As we shall see the symmetry relation (4.1) leads within the present scheme to the Onsager reciprocal relations, which the mobility tensors satisfy.

By defining the following $\vec{k}$ dependent tensors of rank $n+1$

$$\mathbf{S}_i^{(n+1)}(\vec{k}) = \frac{1}{2\pi}(2n-1)!!\, i^n e^{i\vec{k}\cdot\vec{R}_i} \overline{\hat{k}^{n-1}}(\mathbf{1} - \hat{k}\hat{k}) k^{-1} j_{n-1}(ka) \tag{4.3}$$

one can rewrite the integral (3.12) in a more compact form

$$A_{ij}^{(n,m)} = 3a_i \int d\vec{k}\, \mathbf{S}_i^{(n+1)} \cdot \tilde{\mathbf{S}}_j^{(m+1)*} \tag{4.4}$$

The asterisk denotes complex conjugation. One then has

$$\sum_{i,j} \sum_{n,m} \tilde{\mathbf{T}}_i^{(n)*} \odot \mathbf{A}_{ij}^{(n,m)} \odot \mathbf{T}_j^{(m)} = \int d\vec{k} \sum_{i,n} |3a_i \mathbf{T}_i^{(n)*} \cdot S_i^{(n+1)}|^2 \geqslant 0, \tag{4.5}$$

where the $\mathbf{T}_i^{(n)}$ are arbitrary $\vec{k}$ independent complex tensors of rank n. It follows from this inequality, which states that the matrix of connectors is positive definite, that the energy dissipation caused by the motion of the N spheres is also positive, as it should be. Indeed choosing for the tensors $\mathbf{T}_i^{(n)}$ the force multipoles $\mathbf{F}_i^{(n)}$, and using equation (3.11) as well as equations (3.2) and (3.3), one has

$$\sum_{i,j} \tilde{\mathbf{F}}_i^{(n)} \odot A_{ij}^{(n,m)} \odot \mathbf{F}_j^{(m)} = -\sum_i 6\pi\eta a_i(\vec{K}_i \cdot \vec{u}_i + \vec{T}_i \cdot \vec{\omega}_i) \geqslant 0. \tag{4.6}$$

We shall now consider in more detail the self-connectors $\mathbf{A}_{ii}^{(n,m)}$, which are tensors of rank $n+m$ independent of the index i. If $n+m$ is odd, $\mathbf{A}_{ii}^{(n,m)} = 0$, since in that case the integrand in expression (3.12) is an odd function of $\vec{k}$. Since furthermore the spherical Bessel functions have the property

$$\int_0^\infty dx\, j_{2n+\nu}(x) j_{2m+\nu}(x) = 0 \quad \text{for} \quad n \neq m \quad \text{and} \quad \nu = 0, 1, \tag{4.7}$$

it follows that the self-connectors are also diagonal in their upper indices (different multipoles in the same sphere do not couple),

$$\mathbf{A}_{ii}^{(n,m)} = -\mathbf{B}^{(n,n)}\,\delta_{nm}. \tag{4.8}$$

The tensors $\mathbf{B}^{(n,n)}$ have been calculated explicitly [21] in terms of elementary tensors $\mathbf{\Delta}^{(l,l)}$. We list here the first two only

$$\mathbf{B}^{(1,1)} = -\mathbf{1}, \tag{4.9}$$

$$B_{\alpha\beta\gamma\delta}^{(2,2)} = -\tfrac{9}{10}\,\Delta_{\alpha\beta\gamma\delta}^{(2,2)} - \tfrac{3}{4}(\delta_{\alpha\delta}\,\delta_{\beta\gamma} - \delta_{\alpha\gamma}\,\delta_{\beta\delta}). \tag{4.10}$$

Next we discuss the behaviour of the connectors $\mathbf{A}_{ij}^{(n,m)}$, $i \neq j$, as a function of the interparticle distance R_{ij}. Expression (3.1) can also be written as follows for $i \neq j$

$$\mathbf{A}_{ij}^{(n,m)} = \frac{3a_i(2n-1)!!\,(2m-1)!!}{2\pi}(-1)^{n+1}$$
$$\times \int_{-\infty}^{\infty} dk\, \frac{\overline{\partial^{n-1}}}{\partial \vec{R}_{ij}^{n-1}}\left(\mathbf{1} + k^{-2}\frac{\partial^2}{\partial \vec{R}_{ij}^2}\right)\frac{\overline{\partial^{m-1}}}{\partial \vec{R}_{ij}^{m-1}}\frac{\sin kR_{ij}}{kR_{ij}}\,k^{-(n+m-2)} j_{n-1}(ka_i) j_{m-1}(ka_j). \tag{4.11}$$

Here the integration over angles has been carried out, after replacing the tensors formed with the vector $\vec{k}$, by differentiations with respect to $\vec{R}_{ij}$. Expanding the Bessel functions around $k = 0$, one has

$$(2n-1)!!\,(2m-1)!!\,k^{-(n+m-2)} j_{n-1}(ka_i) j_{m-1}(ka_j)$$
$$= a_i^{n-1} a_j^{m-1}\left[1 - \left(\frac{1}{4n+2} + \frac{1}{4m+2}\right) a_i a_j k^2\right] + k^3 \mathcal{R}(k), \tag{4.12}$$

where $\mathcal{R}(z)$ is analytic in the complex plane and bounded for large $|z|$ by $\exp(2a\,|z|)$. Upon substitution of equation (4.12) into equation (4.11), the contribution of $\mathcal{R}(z)$ vanishes in view of the fact that $R_{ij} > a_i + a_j$. Straightforward evaluation of the remaining integral then leads to the results

$$\mathbf{A}_{ij}^{(n,m)} = \mathbf{G}_{ij}^{(n,m)} R_{ij}^{-(n+m-1)} + \mathbf{H}_{ij}^{(n,m)} R_{ij}^{-(n+m+1)}, \tag{4.13}$$

where the tensors $\mathbf{G}_{ij}^{(n,m)}$ and $\mathbf{H}_{ij}^{(n,m)}$, which only depend on the unit vector $\hat{r}_{ij} \equiv \vec{R}_{ij}/R_{ij}$ and the radii a_i and a_j, are given by

$$\mathbf{G}_{ij}^{(n,m)} = (-1)^{n+1}\tfrac{3}{4} a_i^n a_j^{m-1} R_{ij}^{n+m-1} \frac{\overline{\partial^{n-1}}}{\partial \vec{R}_{ij}^{n-1}} \frac{(\mathbf{1} + \hat{r}_{ij}\hat{r}_{ij})}{R_{ij}} \frac{\overleftarrow{\overline{\partial^{m-1}}}}{\partial \vec{R}_{ij}^{m-1}}, \tag{4.14}$$

$$\mathbf{H}_{ij}^{(n,m)} = (-1)^n \tfrac{3}{4} a_i^n a_j^{m-1} R_{ij}^{n+m+1}\left(\frac{1}{2n+1} + \frac{1}{2m+1}\right)$$
$$\times \frac{\overline{\partial^{n-1}}}{\partial \vec{R}_{ij}^{n-1}} \frac{\partial^2}{\partial \vec{R}_{ij}^{n-1}} \frac{\overline{\partial^{m-1}}}{\partial \vec{R}_{ij}^{m-1}} \frac{1}{R_{ij}}. \tag{4.15}$$

The arrow $\leftarrow$ on $\partial/\partial\vec{R}$ in equation (4.14) indicates a differentiation to the left.

The expression for $\mathbf{H}^{(n,m)}$ can easily be further simplified by carrying out the differentiations, and becomes

$$\mathbf{H}_{ij}^{(n,m)} = (-1)^m \tfrac{3}{4} a_i^n a_j^{m-1} \left(\frac{a_i^2}{2n+1} + \frac{a_j^2}{2m+1} \right) (2n+2m-1)!!\, \overline{\hat{r}_{ij}^{n+m}}. \tag{4.16}$$

For the tensor $\mathbf{G}^{(n,m)}$ the differentiations can in principle be carried out in a similar formal way. We list here the explicit results for the first few of these tensors

$$\mathbf{G}_{ij}^{(1,1)} = \tfrac{3}{4} a_i (\mathbf{1} + \hat{r}_{ij} \hat{r}_{ij}), \tag{4.17}$$

$$\mathbf{G}_{ij}^{(1,2s)} = -\tfrac{9}{4} a_i a_j \hat{r}_{ij} \overline{\hat{r}_{ij} \hat{r}_{ij}}, \tag{4.18}$$

$$\mathbf{G}_{ij}^{(2s,2s)} = -\tfrac{9}{4} a_i^2 a_j [3 \overline{\hat{r}_{ij} \hat{r}_{ij}}\, \overline{\hat{r}_{ij} \hat{r}_{ij}} + \mathbf{D}_{ij}]. \tag{4.19}$$

In equation (4.18) $\mathbf{G}_{ij}^{(1,2s)}$ denotes the part of $\mathbf{G}_{ij}^{(1,2)}$ which is (traceless) symmetric in its last two indices. A similar notation is adopted for $\mathbf{G}_{ij}^{(2s,2s)}$ in equation (4.19); the tensor $\mathbf{D}$ is traceless and symmetric in its first and last two indices and defined by

$$(\mathbf{D})_{\alpha\beta\gamma\delta} = 2 r_\alpha r_\beta r_\gamma r_\delta - \tfrac{1}{2}(r_\alpha r_\gamma\, \delta_{\beta\delta} + r_\alpha r_\delta\, \delta_{\beta\gamma} + r_\beta r_\delta\, \delta_{\alpha\gamma} + r_\beta r_\gamma\, \delta_{\alpha\delta}). \tag{4.20}$$

Further explicit expressions for $\mathbf{G}_{ij}^{(n,m)}$, with $n, m \leqslant 3$ and $n + m \leqslant 5$ may be found in Ref. 10.

5. Mobility tensors

In the linear regime considered, the velocities and angular velocities of the spheres are related to the forces and torques exerted on them by the fluid in a way described by the following set of linear coupled equations

$$\vec{u}_i = -\sum_j \boldsymbol{\mu}_{ij}^{TT} \cdot \vec{K}_j - \sum_j \boldsymbol{\mu}_{ij}^{TR} \cdot \vec{T}_j \tag{5.1}$$

$$\vec{\omega}_i = -\sum_i \boldsymbol{\mu}_{ij}^{RT} \cdot \vec{K}_j - \sum_j \boldsymbol{\mu}_{ij}^{RR} \cdot \vec{T}_j \tag{5.2}$$

In the above equations, $\boldsymbol{\mu}_{ii}^{TT}$ and $\boldsymbol{\mu}_{ij}^{RR}$ are translational and rotational mobility tensors respectively. The tensors $\boldsymbol{\mu}_{ij}^{TR}$ and $\boldsymbol{\mu}_{ij}^{RT}$ couple translational and rotational motion. The mobility tensors account for the hydrodynamic interactions between the spheres through their dependence on their relative positions.

The analysis given in the previous sections will enable us to express the mobilities in terms of connectors and thereby calculate these quantities as series in powers of inverse distances between the spheres.

To carry out this program we first rewrite equation (3.11) in a more compact form. For notational convenience we restrict ourselves henceforth to the case of equal-sized spheres, i.e. $a_i = a_j = a$. We define a formal vector $\mathscr{F}$ of which the components are the irreducible force multipoles of the N spheres

$$\{\mathscr{F}\}_j^1 = -\vec{K}_j,$$
$$\{\mathscr{F}\}_j^2 = \frac{-1}{2a}\boldsymbol{\varepsilon}\cdot\vec{T}_j, \tag{5.3}$$
$$\{\mathscr{F}\}_j^n = \mathbf{F}_j^{(n)}, \qquad n \geqslant 3.$$

We also define a second vector $\mathscr{U}$ with components

$$\{\mathscr{U}\}_i^1 = \vec{u}_i$$
$$\{\mathscr{U}\}_i^2 = a\boldsymbol{\varepsilon}\cdot\vec{\omega}_i, \tag{5.4}$$
$$\{\mathscr{U}\}_i^n = 0, \qquad n \geqslant 3.$$

Furthermore we introduce matrices $\mathring{\mathscr{A}}$ and $\mathscr{B}$ with elements (cf. equation (4.8))

$$\{\mathscr{B}\}_{ij}^{nm} = \mathbf{B}^{(n,n)}\delta_{nm}\,\delta_{ij}, \tag{5.5}$$
$$\{\mathring{\mathscr{A}}\}_{ij}^{nm} = \mathbf{A}_{ij}^{(n,m)} + \mathbf{B}^{(n,n)}\,\delta_{nm}\,\delta_{ij}. \tag{5.6}$$

With these notations the set of equations (3.11) becomes

$$6\pi\eta\alpha\mathscr{U} = -\mathscr{B}\mathscr{F} + \mathring{\mathscr{A}}\mathscr{F}. \tag{5.7}$$

Next we define projection operators $\mathscr{P}_1$, $\mathscr{P}_2$, and $\mathscr{Q} = 1 - \mathscr{P}_1 - \mathscr{P}_2$

$$\{\mathscr{P}_1\}_{ij}^{nm} = \delta_{n1}\,\delta_{m1}\,\delta_{ij}, \qquad \{\mathscr{P}_2\}_{ij}^{nm} = \delta_{n2}\,\delta_{m2}\,\delta_{ij}\mathbf{S},$$
$$\{\mathscr{Q}\}_{ij}^{nm} = (\delta_{nm}\mathbf{1}^{(n,n)} - \delta_{n1}\,\delta_{m1} - \delta_{n2}\,\delta_{m2}\mathbf{S})\,\delta_{ij}. \tag{5.8}$$

The tensor $\mathbf{S}$ of rank 4, which is an antisymmetrization operator, is defined by

$$S_{\alpha\beta\gamma\delta} = \tfrac{1}{2}\,\delta_{\alpha\delta}\,\delta_{\beta\gamma} - \tfrac{1}{2}\,\delta_{\alpha\gamma}\,\delta_{\beta\delta}. \tag{5.9}$$

$\mathbf{1}^{(n,n)}$ is the appropriate unit tensor of rank $2n$ which, when fully contracted with a tensor of rank n, projects out the part irreducible in the first $n-1$ indices. One verifies that

$$\mathscr{P}_1\mathscr{P}_2 = \mathscr{P}_2\mathscr{P}_1 = 0. \tag{5.10}$$

Note also that the matrix $\mathscr{B}$ commutes with the projection operators

$$\mathscr{P}_\nu\mathscr{B} = \mathscr{B}\mathscr{P}_\nu, \qquad \nu = 1, 2. \tag{5.11}$$

Now decompose $\mathscr{F}$ according to

$$\mathscr{F} = \mathscr{P}_1\mathscr{F} + \mathscr{P}_2\mathscr{F} + \mathscr{Q}\mathscr{F}, \tag{5.12}$$

and multiply equation (5.7) from the left by $\mathscr{P}_1$, $\mathscr{P}_2$ and $\mathscr{Q}$ respectively. This results, using also the properties (5.10) and (5.11), in the following set of equations

$$6\pi\eta a\mathscr{P}_i\mathscr{U} = \mathscr{P}_1(-\mathscr{B} + \mathring{\mathscr{A}})\mathscr{P}_1\mathscr{F} + \mathscr{P}_1\mathring{\mathscr{A}}\mathscr{P}_2\mathscr{F} + \mathscr{P}_1\mathring{\mathscr{A}}\mathscr{Q}\mathscr{F}, \tag{5.13}$$
$$6\pi\eta a\mathscr{P}_2\mathscr{U} = \mathscr{P}_2\mathring{\mathscr{A}}\mathscr{P}_1\mathscr{F} + \mathscr{P}_2(-\mathscr{B} + \mathring{\mathscr{A}})\mathscr{P}_2\mathscr{F} + \mathscr{P}_2\mathring{\mathscr{A}}\mathscr{Q}\mathscr{F}, \tag{5.14}$$
$$\mathscr{B}\mathscr{Q}\mathscr{F} = \mathscr{Q}\mathring{\mathscr{A}}\mathscr{P}_1\mathscr{F} + \mathscr{Q}\mathring{\mathscr{A}}\mathscr{P}_2\mathscr{F} + \mathscr{Q}\mathscr{A}\mathscr{Q}\mathscr{F}. \tag{5.15}$$

One may then solve equation (5.15) for $\mathscr{Q}\mathscr{F}$

$$\mathscr{Q}\mathscr{F} = (1 - \mathscr{B}^{-1}\mathscr{Q}\mathring{\mathscr{A}})^{-1}\mathscr{B}^{-1}(\mathscr{Q}\mathring{\mathscr{A}}\mathscr{P}_i\mathscr{F} + \mathscr{Q}\mathring{\mathscr{A}}\mathscr{P}_2\mathscr{F}). \tag{5.16}$$

The matrix $\mathscr{B}^{-1}$ has elements

$$\{\mathscr{B}^{-1}\}_{ij}^{nm} = \mathbf{B}^{(n,n)^{-1}}\,\delta_{nm}\,\delta_{ij} \quad \text{for} \quad n \neq 2, \tag{5.17}$$

$$\{\mathscr{B}^{-1}\mathscr{Q}\}_{ij}^{n2} = \mathbf{B}^{(2s,2s)^{-1}}\,\delta_{n2}\,\delta_{ij}. \tag{5.18}$$

The tensor $\mathbf{B}^{(n,n)^{-1}}$, $n \neq 2$, is the generalized inverse of $\mathbf{B}^{(n,n)}$ when acting on tensors of rank n that are irreducible in their first $n-1$ indices. For its construction see Ref. 21. The tensor $\mathbf{B}^{(2s,2s)^{-1}}$ is given by (cf. equation (4.10))

$$\mathbf{B}^{(2s,2s)^{-1}} = -\tfrac{10}{9}\mathbf{\Delta}^{(2,2)}. \tag{5.19}$$

Upon substitution of equation (5.16) into equations (5.13) and (5.14) one obtains, using also equations (4.9), (4.10), (5.3), (5.4) and (5.8), equations of the form (5.1)–(5.2) with the mobilities expressed in terms of connectors according to

$$\boldsymbol{\mu}_{ij}^{TT} = (6\pi\eta a)^{-1}[\delta_{ij} + \{\mathring{\mathscr{A}}(1 - \mathscr{B}^{-1}\mathscr{Q}\mathring{\mathscr{A}})^{-1}\}_{ij}^{11}] \tag{5.20}$$

$$\boldsymbol{\mu}_{ij}^{TR} = (12\pi\eta a^2)^{-1}\,\{\mathring{\mathscr{A}}(1 - \mathscr{B}^{-1}\mathscr{Q}\mathring{\mathscr{A}})^{-1}\}_{ij}^{12} : \boldsymbol{\varepsilon} = \tilde{\boldsymbol{\mu}}_{ji}^{RT} \tag{5.21}$$

$$\boldsymbol{\mu}_{ij}^{RR} = (24\pi\eta a^3)^{-1}[3\delta_{ij} - \boldsymbol{\varepsilon} : \{\mathring{\mathscr{A}}(1 - \mathscr{B}^{-1}\mathscr{Q}\mathring{\mathscr{A}})^{-1}\}_{ij}^{22} : \boldsymbol{\varepsilon}]. \tag{5.22}$$

The Onsager relation which is contained in equation (5.21) is a direct consequence of the symmetry property (4.1) of the connectors.

By expanding the inverse matrices in equations (5.20)–(5.22) in powers of the connector matrix $\mathring{\mathscr{A}}$, the mobilities are obtained in the form of a power series expansion in R^{-1}, where R is a typical distance between spheres.

For $\boldsymbol{\mu}_{ij}^{TT}$ one obtains in this way the series

$$\boldsymbol{\mu}_{ij}^{TT} = (6\pi\eta a)^{-1}\Big[\delta_{ij} + \mathbf{A}_{ij}^{(1,1)}(1 - \delta_{ij}) + \sum_{k \neq i,j}\Big\{\mathbf{A}_{ik}^{(1,2s)} \odot \mathbf{B}^{(2s,2s)^{-1}} \odot \mathbf{A}_{kj}^{(2s,1)} + \sum_{m=3}^{\infty} \mathbf{A}_{ik}^{(1,m)} \odot \mathbf{B}^{(m,m)^{-1}} \odot \mathbf{A}_{kj}^{(m,1)}\Big\} + \cdots\Big]. \tag{5.23}$$

Each term in this series has, as a function of a typical interparticle distance R, a given behaviour which is determined by the upper indices of the connectors and their number. Thus according to equation (4.13) a term in equation (5.23) with s connectors, $s = 1, 2, 3, \ldots$, gives contributions proportional to R^{-p} with p equal to

$$p = \begin{cases} 1, 3 & \text{for} \quad s = 1 \\ 3s - 2 + 2q & \text{for} \quad s \geq 2, \qquad q = 0, 1, 2, \ldots \end{cases} \tag{5.24}$$

This implies that $\boldsymbol{\mu}_{ij}^{TT}$ cannot contain terms proportional to R^{-2} and R^{-5}. We also note that each term in the expression (5.23) containing a sequence of s connectors involves the hydrodynamic interaction between at most $s+1$ spheres. Therefore the dominant n-sphere contributions, $n \geq 2$, are of order R^{-3n+5}, where equation (5.24) has been applied with $s = n - 1$ and $q = 0$.

Similar considerations lead to the conclusion that in the series for $\boldsymbol{\mu}_{ij}^{TR}$ contributions proportional to R^{-1}, R^{-3} and R^{-6} are excluded, and that the dominant n-sphere term is of order R^{-3n+4}. For $\boldsymbol{\mu}_{ij}^{RR}$ contributions proportional to R^{-1}, R^{-2}. R^{-4}, R^{-5} and R^{-7} are excluded; the dominant n-sphere contribution is of order R^{-3n+3} in this case.

Explicit expressions for the various terms in the expansions of formulae (5.20)–(5.22) can in principle be found, using formulae (4.13)–(4.15) and forming the necessary tensor products. Thus the three-sphere contributions of order R^{-7} to $\boldsymbol{\mu}_{ij}^{TT}$ is given by the product

$$\tfrac{100}{81} R_{ik}^{-2} R_{kl}^{-3} R_{li}^{-2} \mathbf{G}_{ik}^{(1,2s)} : \mathbf{G}_{kl}^{(2s,2s)} : \mathbf{G}_{li}^{(2s,1)}.$$

Into this product one then has to insert expressions (4.18) and (4.19) for the corresponding $\mathbf{G}$-tensors. In Ref. 10 all contributions to the tensors $\boldsymbol{\mu}_{ij}^{TT}$, $\boldsymbol{\mu}_{ij}^{TR}$ and $\boldsymbol{\mu}_{ij}^{RR}$ up to order R^{-7} are listed explicitly.

In the next two sections we shall consider two transport properties of suspensions viz. self-diffusion and sedimentation velocity, and briefly discuss the influence of the hydrodynamic interactions on their behaviour.

6. Self-diffusion; non-additivity of hydrodynamic interactions

In the preceding sections a consistent scheme has been developed to evaluate the static mobility tensors for an arbitrary number of spheres in an unbounded fluid. The mobility tensors characterize, as stated, the hydrodynamic interaction set up in a system of many-spheres through their motions. It is well-known that these interactions play an important role in determining the concentration dependence of transport properties of a suspension. A typical quantity that can be studied conveniently, both theoretically and experimentally [19], is the so-called short-time [20] self-diffusion coefficient D_s of uncharged spherical particles of identical radius a in suspension. This quantity, which characterizes self-diffusion on a time scale on which the relative configuration of the particles does not change appreciably, is related to the mobility tensor μ_{ii}^{TT} by the relation

$$D_s = \frac{k_B T}{3N} \operatorname{Tr}\left\langle \sum_i \boldsymbol{\mu}_{ii}^{TT} \right\rangle \tag{6.1}$$

where $\langle \cdots\cdot \rangle$ denotes an average over all configurations of N spheres inside a volume V; k_B is Boltzmann's constant and T the temperature, Tr denotes the trace of a tensor. By inserting into the above relation the series (5.23), one can in principle evaluate D_s as a power series in the density $n_0 = N/V$ (a so-called virial expansion). This has been done by C. W. J. Beenakker and the author [21] up to and including terms of second order in the density. Up to this order only two- and three-body hydrodynamic interactions need be considered, since the probability that a given sphere has s neighbours is of order n_0^s. Furthermore one needs to this order only knowledge of the hard sphere pair-distribution function to first order in n_0, and of the three-sphere distribution function to lowest order. It was found that

$$D_s = \frac{k_B T}{6\pi\eta a}(1 - 1.73\phi + 0.88\phi^2 + \mathcal{O}(\phi^3)), \tag{6.2}$$

where $\phi = \frac{4}{3}\pi a^3 n_0$ is the volume fraction of the spheres. Only two-body hydrodynamic interactions contribute to the well-known [6, 8] term of order ϕ are are therefore the only ones to contribute at sufficiently low densities. However at higher densities the many-sphere hydrodynamic interactions may not be neglected: two-sphere contributions alone would have lead to a value of $-0.93\phi^2$ for the term of order ϕ^2, instead of the value of $+0.88\phi^2$ in equation (6.2). This illustrates dramatically the non-additivity of hydrodynamic interactions.

In a concentrated suspension it is therefore essential to fully take into account the many-body hydrodynamic interactions between an arbitrary number of spheres. A virial expansion, however, is not appropriate at high densities. It turned out to be possible, starting from formula (5.20), to resum algebraically contributions due to hydrodynamic interactions between an arbitrary number of spheres [21] and moreover of a special class of self-correlations [22]. This algebraic resummation procedure leads to an expression equivalent to formula (5.20) but with, so to say, renormalized connectors, which account loosely speaking for the fact that density fluctuations interact hydrodynamically via the suspension rather than through the pure fluid. The equivalent expression could then be expanded in density-fluctuation correlation functions. Already to lowest order (and even somewhat better to quadratic order) the thus obtained numerical values for D_s agreed well with available experimental data [19], which exist only for the density interval $0.2 < \phi < 0.45$.

7. Sedimentation; the influence of container walls

To lowest order in the expansion in connectors the translational mobility tensor μ_{ij} (we omit in this section the upper indices TT) is, for $i \neq j$ explicitly given by

$$\boldsymbol{\mu}_{ij} = (6\pi\eta a)^{-1}\left\{\frac{3a}{4R_{ij}}(\mathbf{1} + \hat{r}_{ij}\hat{r}_{ij}) - \frac{3a^3}{2R_{ij}^3}(\hat{r}_{ij}\hat{r}_{ij} - \tfrac{1}{3}\mathbf{1}) + \mathcal{O}\left(\frac{1}{R_{ij}^4}\right)\right\}, \tag{7.1}$$

as follows from equations (5.20), (5.16) and (5.17). Thus the hydrodynamic interaction falls off very slowly. In an unbounded medium the long range natue of these interaction may give rise to difficulties. For self-diffusion, where only μ_{ii} need be considered, the above long range terms of orders R^{-1} and R^{-3} did not contribute.[2]) However the velocity of sedimentation in an unbounded medium

[2]) Even for collective diffusion these terms do not give rise to any complication, as a consequence of the "longitudinal" character (in wave-vector representation) of the diffusion phenomenon.

diverges, a fact sometimes referred to as the Smoluchowski paradox. It has been customary to circumvent the difficulty caused by the $1/R$ divergence by considering, following Pyun and Fixman [23], sedimentation with respect to the mean volume velocity, thus indirectly taking into account the "backflow" caused by container walls. Even then the $1/R^3$ term still gives rise to a conditionally convergent integral and poses the problem of a possible dependence of the sedimentation velocity on the shape of the vessel containing the suspension. Batchelor [24] was able to assign a definite value to the integral in question using an argument based on general considerations of a physical nature – valid for the unbounded system. Ultimately the difficulties mentioned should be resolved by a direct and explicit evaluation of the influence of containes walls on the mobilities of sedimenting particles. Such an evaluation is in fact possible along the lines of the analysis given in sections 2 to 5, and has been carried out for two geometries, first for the case of a plane wall [18, 25], and then also for the case of spherical particles inside a spherical container [14, 26]. It is the latter case we shall briefly review here.

The solution to the problem of N spheres moving in a viscous fluid inside a spherical container may be obtained from the solution to the problem of $N+1$ spheres in an unbounded medium studied above, by observing that the analysis given remains valid if one of the spherical boundaries, the container specified by the index $i=0$, encloses the other N spheres ($i=1, 2, \ldots, N$) and the viscous fluid, provided the induced force $\vec{F}_0$ on the container is chosen in such a way that

$$\vec{F}_0(\vec{r})=0 \quad \text{for} \quad |(\vec{r}-\vec{R}_0)|<a_0 \tag{7.2}$$

where $\vec{R}_0$ is the center of the container and a_0 its radius, and that the velocity field has, in addition to the extensions (2.8), the extension

$$\vec{v}(\vec{r})=0 \quad \text{for} \quad |\vec{r}-\vec{R}_0)| \geqslant a_0. \tag{7.3}$$

This condition implies that the translational velocity $\vec{u}_0$ and the rotational velocity of the container both vanish. The analysis of section 3 then leads to a set of equations of the form (3.11), with connectors $A_{ij}^{(n,m)}$ ($i, j=0, 1, 2, \ldots, N$) defined again by the integrals (3.12) with the additional conditions

$$R_{ij}>a_i+a_j \quad \text{for} \quad i, j=1, 2, \ldots, N; \qquad i \neq j \tag{7.4}$$

$$R_{0j}<a_0-a_j \quad \text{for} \quad j=1, 2, \ldots, N \tag{7.5}$$

Consequently the particle–particle connectors remain unchanged. The particle–container connectors are of a different type but can be evaluated as well using properties of integrals over Bessel functions.

Since the velocity $\vec{u}_0$ and angular velocity of the container vanish, one can reduce the hierarchy of equations of the type (3.11) for the $N+1$ spheres (particles and container) to a reduced set of the same form for the particles alone but now in terms of new connectors $\mathbf{A}_{ij;s.c.}^{(n,m)}$, which incorporate the effects of the spherical container, i.e. the hydrodynamic interaction with the container,

$$\mathbf{A}_{ij;s.c.}^{(n,m)}=\mathbf{A}_{ij}^{(n,m)}+\sum_{p=1}^{\infty}\mathbf{A}_{i0}^{(1,p)}\odot\mathbf{B}^{(p,p)^{-1}}\odot\mathbf{A}_{0j}^{(p,1)}, \qquad i, j=1, 2, \ldots, N \tag{7.6}$$

It is in terms of these new connectors that the translational mobility tensors $\boldsymbol{\mu}_{ij}$ which are again of the form (5.20) must now be evaluated.

To discuss sedimentation we shall also have to consider the flow of fluid caused by the motion of the particles, and described by the velocity field $\vec{v}(\vec{r})$ given by

$$\vec{v}(\vec{r}) = -\sum_{j=1}^{N} \mathbf{S}_j(\vec{r}) \cdot \vec{K}_j \tag{7.7}$$

The tensor field $\mathbf{S}_j(\vec{r})$ is closely related to the mobilities of the particles and may in fact be derived from these by studying a "test particle" of infinitesimal radius at point $\vec{r}$.

One is now in a position to evaluate the mean particle velocity $\vec{v}_p$ and the mean fluid velocity $\vec{v}_f$ of a homogeneous distribution of identical spheres, $a_i = a_j = a$, sedimenting inside a spherical container [26]. These two quantities, calculated at the center of the container and in the limit that its radius a_0 tends to infinity, may be written as conditional averages

$$\vec{v}_p = \lim_{a_0 \to \infty} \left\langle \sum_j \boldsymbol{\mu}_{ij} \mid R_{i0} = 0 \right\rangle \cdot \vec{F} \tag{7.8}$$

$$\vec{v}_f = \lim_{a_0 \to \infty} \left\langle \sum_j \mathrm{S}_j(\vec{r} = \vec{R}_0) \mid R_{j0} > a \quad \text{for all} \quad j \right\rangle \cdot \vec{F} \tag{7.9}$$

Here $\langle \cdots \mid R_{i0} = 0 \rangle$ denotes an average over those configurations for which $R_{i0} = 0$, while $\langle \cdots \mid R_{j0} > a$ for all $j \rangle$ denotes an average over configurations for which no suspended sphere overlaps the center of the container; $\vec{F}$ is the gravitational force (corrected for bouyancy on each of the particles).

To linear order in the volume fraction ϕ of suspended spheres calculation of $\vec{v}_p$ and $\vec{v}_f$ on the basis of equations (7.8) and (7.9) yields

$$\vec{v}_p = \{1 - 3.55\phi + \mathcal{O}(\phi^2)\}(6\pi\eta a)^{-1}\vec{F} \tag{7.9}$$

$$\vec{v}_f = \{2\phi + \mathcal{O}(\phi^2)\}(6\pi\eta a)^{-1}\vec{F}. \tag{7.10}$$

We can now also determine the average volume velocity given by $\vec{v}_v = \phi\vec{v}_p + (1 - \phi)\vec{v}_f$. From equations (7.9) and (7.10) it follows that

$$\vec{v}_v = \{3\phi + \mathcal{O}(\phi^2)\}(6\pi\eta a)^{-1}\vec{F}. \tag{7.11}$$

Since, because of incompressibility, the volume flux through any closed surface must vanish, this result, namely that there is a non-vanishing volume velocity at the center, implies the existence of a vortex of convective flow in the spherical container.

Finally one may evaluate the mean particle velocity with respect to the mean volume velocity. The result is

$$\vec{v}_p - \vec{v}_v = \{1 - 6.55\phi + \mathcal{O}(\phi^2)\}\vec{F}. \tag{7.12}$$

This is the result found for this quantity by Batchelor for an unbounded system. It is also what is found for sedimentation perpendicular to and towards a plane wall, in which case $\vec{v}_v$ vanishes, in the limit of an infinitely distant wall.

We therefore come to the following conclusions:

> The average local velocity of a sedimenting particle in a homogeneous suspension depends on the shape of the container, however far away the container walls. But this shape dependence disappears for the sedimentation velocity with respect to the average volume velocity.

These results illustrate once more the essential role played by hydrodynamic many-body interactions. They show that, for sedimentation, the "three-body" hydrodynamic interaction of two-particles and the container can in fact never be omitted from consideration, not even for sufficiently dilute suspensions.

The discussion of wall effects and of the non-additivity of hydrodynamic couplings in the previous section thus underscores the relevance and usefulness of the scheme developed, and summarized in this paper, for the evaluation of many-sphere hydrodynamic interactions.

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