

## MANY-SPHERE HYDRODYNAMIC INTERACTIONS

### IV. WALL-EFFECTS INSIDE A SPHERICAL CONTAINER

C W J BEENAKKER\* and P MAZUR

*Instituut-Lorentz, Rijksuniversiteit te Leiden, Nieuwsteeg 18, 2311 SB Leiden, The Netherlands*

Received 2 January 1985

A previously developed scheme – to evaluate mobility and friction tensors of an arbitrary number of spherical particles suspended in an unbounded fluid – is extended to include the influence of a spherical wall bounding the suspension. If restricted to one particle the results generalize well-known formulae for two concentric spheres to the case of non-vanishing eccentricity.

#### 1. Introduction

This paper continues an investigation of many-sphere hydrodynamic interactions in a suspension, initiated by one of the authors<sup>1)</sup>. The purpose of the present work is to extend the theory of Mazur and van Saarloos<sup>2)</sup> for an unbounded suspension, to the case of a suspension bounded by a spherical container wall. Such an extension is of particular interest for the following reason.

First of all, as a consequence of the long range of hydrodynamic interactions, container walls have an essential influence on certain bulk properties of a suspension – even in the case of a very large container. A celebrated example is the sedimentation velocity, which is known to become infinitely large in an unbounded suspension – a paradoxical situation first noticed by Smoluchowski<sup>3)</sup>. Smoluchowski himself indicated already that this difficulty is a result of the neglect of backflow generated by boundary walls<sup>4)</sup>. Recently, we were able to show by explicit calculation that the divergency of the sedimentation velocity encountered in an unbounded suspension does indeed not occur if the presence of a wall supporting the suspension is accounted for<sup>5)</sup>. In this calculation essential use was made of formulae derived previously<sup>6)</sup> for mobil-

\* Present address Department of Chemistry, Stanford University, Stanford, California 94305, USA

ities of spheres in a fluid bounded in one direction by a plane wall of infinite extent

In a similar way, the formulae to be derived in this paper can serve as the starting point for a study of sedimentation in a suspension bounded in *all* directions by container walls. A study of this type is of great interest, as it is to be expected that the backflow in such a geometry is fundamentally different from the backflow in the geometry of ref. 5, where the suspension was assumed to be bounded only in the direction of the sedimentation velocity.

Although the problem studied in this paper is interesting in itself – and has in fact been studied extensively for the case of one single sphere having a common center with the container<sup>7-12</sup>) – it was with the above application in mind that we undertook our investigation.

In section 2 we formulate the problem of the motion of  $N$  spherical particles suspended inside a spherical container, which may itself be in arbitrary motion. Formally, this problem is very similar to that of the motion of  $N + 1$  spheres (of arbitrary radius) in an unbounded fluid, studied in ref. 2 (hereafter referred to as I). In this paper, and in ref. 6 (hereafter referred to as III), general expressions were given for the translational and rotational mobility and friction tensors of the spheres, as well as for the fluid velocity field, in terms of tensor objects called *connectors*. The circumstance that one of the spherical boundaries encloses the others does not invalidate the analysis leading to these expressions, but only affects the explicit evaluation of the connectors, cf. section 3. The formal similarity between the present problem and that of paper I thus enables us to immediately obtain expressions for the various quantities mentioned above. These expressions are given in section 4, for the case of a motionless container (Generalizations to e.g. a rotating container are, however, straightforward.)

Using the results for the connectors given in section 3, one then finds expansions of e.g. the mobility tensors of the suspended particles in the following three parameters: the ratio of particle radius to container radius, the ratio of particle radius to interparticle separation and the ratio of the center to center separation of particle and container to the radius of the container. Explicit results to third order in these parameters are given in section 5, to this order the hydrodynamic interactions of one and two particles and the container contribute. If restricted to one particle, these results generalize formulae known in the literature<sup>7-12</sup>) for the translational and rotational mobility of a spherical particle concentric with a spherical container, to the case of non-vanishing eccentricity<sup>3</sup>.

\* A previous generalization in this respect – but restricted to a situation with axial symmetry – has been given by Jeffery<sup>13</sup>) – who considered the rotation of two non-concentric spheres about their common diameter.

**2. Formulation of the problem and formal solution**

We consider  $N$  spherical particles with radii  $a_i$  and position vectors  $\mathbf{R}_i$  ( $i = 1, 2, \dots, N$ ) immersed in an incompressible fluid with viscosity  $\eta$ . We are interested in the influence of an external spherical boundary on the motion of the particles. Let  $a_0$  denote the radius, and  $\mathbf{R}_0$  the position vector of the center of a spherical container which encloses the  $N$  particles. The translational and angular velocities of the particles are denoted by  $\mathbf{u}_i$  and  $\boldsymbol{\omega}_i$  ( $i = 1, 2, \dots, N$ ), respectively. Similarly,  $\mathbf{u}_0$  and  $\boldsymbol{\omega}_0$  denote the translational and angular velocity of the container.

The motion of the fluid at position  $\mathbf{r}$  is described by the quasistatic Stokes equation for so-called creeping flow<sup>12</sup>),

$$\left. \begin{aligned} \nabla p(\mathbf{r}) - \eta \Delta \mathbf{v}(\mathbf{r}) &= 0 \\ \nabla \cdot \mathbf{v}(\mathbf{r}) &= 0 \end{aligned} \right\} \begin{array}{l} \text{for } |\mathbf{r} - \mathbf{R}_i| > a_i \quad (i = 1, 2, \dots, N) \\ \text{and } |\mathbf{r} - \mathbf{R}_0| < a_0. \end{array} \quad (2.1)$$

Here  $\mathbf{v}(\mathbf{r})$  is the velocity field and  $p(\mathbf{r})$  the hydrostatic pressure. Eq. (2.1) is supplemented by stick boundary conditions on the surfaces of the particles and on the container wall,

$$\mathbf{v}(\mathbf{r}) = \mathbf{u}_i + \boldsymbol{\omega}_i \wedge (\mathbf{r} - \mathbf{R}_i) \quad \text{for } |\mathbf{r} - \mathbf{R}_i| = a_i \quad (i = 0, 1, 2, \dots, N). \quad (2.2)$$

The forces  $\mathbf{K}_i$  and torques  $\mathbf{T}_i$  exerted by the fluid on the particles and on the container wall are defined as surface integrals of the pressure tensor  $\mathbf{P}(\mathbf{r})$ :

$$\left. \begin{aligned} \mathbf{K}_i &= - \int_{S_i} dS \mathbf{P}(\mathbf{r}) \cdot \hat{\mathbf{n}}_i \\ \mathbf{T}_i &= - \int_{S_i} dS (\mathbf{r} - \mathbf{R}_i) \wedge \mathbf{P}(\mathbf{r}) \cdot \hat{\mathbf{n}}_i \end{aligned} \right\} i = 0, 1, 2, \dots, N. \quad (2.3)$$

Here  $\hat{\mathbf{n}}_i$  is a unit vector perpendicular to the surface  $S_i$ , defined by  $|\mathbf{r} - \mathbf{R}_i| = a_i$ , and pointing in the direction of the fluid. The pressure tensor  $\mathbf{P}$  has cartesian components

$$P_{\mu\nu} = p\delta_{\mu\nu} - \eta \left( \frac{\partial v_\nu}{\partial r_\mu} + \frac{\partial v_\mu}{\partial r_\nu} \right). \quad (2.4)$$

The boundary value problem given by eqs. (2.1) and (2.2) was studied by Mazur and van Saarloos in paper I – for the case of an unbounded fluid, that is to say in the absence of the external boundary  $S_0$ . The method of solution used

in that paper was the so-called method of induced forces<sup>14</sup>), by which it was possible to reduce the problem to that of the solution of an infinite hierarchy of linear *algebraic* equations. (A similar approach had been taken by Yoshizaki and Yamakawa<sup>15</sup>.) The solution to the problem considered here, of  $N$  spherical particles inside a spherical container, may be obtained from the solution to the problem of  $N + 1$  spheres in an unbounded fluid – studied in paper I – by observing that the analysis of that paper, leading to the hierarchy of equations referred to above, *remains valid if one of the boundaries encloses the others*. This observation then leads us immediately to the result (cf. eqs. (I-5.2)–(I-5.5))

$$6\pi\eta a_i \mathbf{u}_i = -\mathbf{K}_i - \sum_{\substack{j=0 \\ j \neq i}}^N \mathbf{A}_{ij}^{(1,1)} \cdot \mathbf{K}_j - \sum_{\substack{j=0 \\ j \neq i}}^N (2a_j)^{-1} \mathbf{A}_{ij}^{(1,2a)} : \boldsymbol{\epsilon} \cdot \mathbf{T}_j + \sum_{\substack{j=0 \\ j \neq i}}^N \sum_{m=2}^{\infty} \mathbf{A}_{ij}^{(1,m)} \odot \mathbf{F}_j^{(m)}, \quad (2.5)$$

$$12\pi\eta a_i^2 \boldsymbol{\omega}_i = \sum_{\substack{j=0 \\ j \neq i}}^N \boldsymbol{\epsilon} : \mathbf{A}_{ij}^{(2a,1)} \cdot \mathbf{K}_j - 3(2a_i)^{-1} \mathbf{T}_i + \sum_{\substack{j=0 \\ j \neq i}}^N (2a_j)^{-1} \boldsymbol{\epsilon} : \mathbf{A}_{ij}^{(2a,2a)} : \boldsymbol{\epsilon} \cdot \mathbf{T}_j - \sum_{\substack{j=0 \\ j \neq i}}^N \sum_{m=2}^{\infty} \boldsymbol{\epsilon} : \mathbf{A}_{ij}^{(2a,m)} \odot \mathbf{F}_j^{(m)}, \quad (2.6)$$

$$\mathbf{F}_i^{(n)} = - \sum_{\substack{j=0 \\ j \neq i}}^N \mathbf{B}^{(n,n)^{-1}} \odot \mathbf{A}_{ij}^{(n,1)} \cdot \mathbf{K}_j - \sum_{\substack{j=0 \\ j \neq i}}^N (2a_j)^{-1} \mathbf{B}^{(n,n)^{-1}} \odot \mathbf{A}_{ij}^{(n,2a)} : \boldsymbol{\epsilon} \cdot \mathbf{T}_j + \sum_{\substack{j=0 \\ j \neq i}}^N \sum_{m=2}^{\infty} \mathbf{B}^{(n,n)^{-1}} \odot \mathbf{A}_{ij}^{(n,m)} \odot \mathbf{F}_j^{(m)} \quad (n = 2s, 3, 4, \dots), \quad (2.7)$$

where the index  $i$  runs from 0 to  $N$ .

Eqs. (2.5) and (2.6) relate translational and angular velocities of the particles and of the container to forces and torques exerted on them by the fluid, as well as to higher order multipoles of the induced forces, denoted by  $\mathbf{F}_j^{(m)}$ . These latter quantities (which are tensors of rank  $m$ ) need not be further specified in this context, as they can be eliminated iteratively by means of eq. (2.7), in favour of the forces and torques. The objects  $\mathbf{A}_{ij}^{(n,m)}$  and  $\mathbf{B}^{(n,m)^{-1}}$  appearing in eqs. (2.5)–(2.7) are tensors of rank  $n + m$ , whose definitions will be given below.

We list the further notations used: a single or double tensor contraction is denoted by a dot or colon, respectively. The symbol  $\odot$  in e.g.  $\mathbf{A}^{(m,n)} \odot \mathbf{F}^{(n)}$  prescribes a full  $n$ -fold contraction of the last  $n$  indices of the first tensor with

the first  $n$  indices of the second tensor. In these contractions the nesting convention is adopted, by which the last index of the first tensor is contracted with the first index of the second tensor, etc. For example,

$$(\mathbf{A}^{(1,3)} \odot \mathbf{F}^{(3)})_\alpha = \sum_{\beta\gamma\delta} A_{\alpha,\beta\gamma\delta}^{(1,3)} F_{\delta\gamma\beta}^{(3)}, \tag{2.8}$$

where Greek indices denote cartesian components. The value 2s or 2a for one of the upper indices of the tensors denotes respectively the traceless symmetric or anti-symmetric part, e.g.

$$F_{\alpha\beta}^{(2s)} = \frac{1}{2}F_{\alpha\beta}^{(2)} + \frac{1}{2}F_{\beta\alpha}^{(2)} - \frac{1}{3}\delta_{\alpha\beta} \sum_{\gamma} F_{\gamma\gamma}^{(2)},$$

$$A_{\alpha,\beta\gamma}^{(1,2a)} = \frac{1}{2}A_{\alpha,\beta\gamma}^{(1,2)} - \frac{1}{2}A_{\alpha,\gamma\beta}^{(1,2)}. \tag{2.9}$$

The prime in the summation  $\sum'_{m=2}^\infty$  over upper indices  $m$  indicates that for  $m = 2$  only the traceless symmetric part of the corresponding tensor has to be taken,

$$\sum'_{m=2}^\infty \mathbf{A}^{(1,m)} \odot \mathbf{F}^{(m)} = \mathbf{A}^{(1,2s)} : \mathbf{F}^{(2s)} + \sum_{m=3}^\infty \mathbf{A}^{(1,m)} \odot \mathbf{F}^{(m)}. \tag{2.10}$$

Furthermore, the symbol  $\epsilon$  denotes the Levi-Civita tensor, the completely anti-symmetric tensor of rank 3, with  $\epsilon_{123} = 1$ .

Finally, we give the definitions of the tensors  $\mathbf{A}_{jl}^{(n,m)}$  and  $\mathbf{B}^{(n,n)}$  (of which the tensor  $\mathbf{B}^{(n,n)^{-1}}$  is the inverse), cf. eqs. (I-4.31), (I-4.32), (III-3.8) and (III-3.13),

$$\mathbf{A}_{jl}^{(n,m)} = \frac{3}{8\pi} \left(\frac{a_j}{a_l}\right)^{1/2} i^{n-m} (2n-1)!! (2m-1)!! \int d\mathbf{k} e^{-i\mathbf{k}\cdot(\mathbf{R}_l-\mathbf{R}_j)}$$

$$\times k^{-3} J_{n-1/2}(a_j k) J_{m-1/2}(a_l k) \overline{\hat{\mathbf{k}}^{n-1}} (\mathbf{1} - \hat{\mathbf{k}}\hat{\mathbf{k}}) \overline{\hat{\mathbf{k}}^{m-1}}, \tag{2.11}$$

$$\mathbf{B}^{(n,n)} = -\mathbf{A}_{jl}^{(n,n)}. \tag{2.12}$$

Here we have defined  $(2n-1)!! = 1 \cdot 3 \cdot 5 \cdots (2n-3) \cdot (2n-1)$ ; the vector  $\mathbf{k}$  has magnitude  $k$  and direction  $\hat{\mathbf{k}} = \mathbf{k}/k$ ; the function  $J_p$  is the Bessel function of order  $p$ ; the notation  $\overline{\hat{\mathbf{k}}^p}$  denotes an irreducible tensor of rank  $p$  (i.e. a tensor traceless and symmetric in any pair of its indices) constructed from a  $p$ -fold ordered product of the vector  $\hat{\mathbf{k}}$ . For  $p = 1, 2$  one has e.g.

$$\overline{\hat{\mathbf{k}}} = \hat{\mathbf{k}}, \quad \overline{\hat{\mathbf{k}}\hat{\mathbf{k}}} = \hat{\mathbf{k}}\hat{\mathbf{k}} - \frac{1}{3}\mathbf{1}, \tag{2.13}$$

where  $\mathbf{1}$  denotes the second rank unit tensor. (See ref. 16 for useful formulae on irreducible tensors.)

This completes the definition of the objects appearing in the hierarchy (2.5)–(2.7). In section 4 we shall show, following papers I and III, how one can obtain from these equations series expansions for the various quantities of interest—such as the mobility and friction tensors of the particles or the fluid velocity field. In the next section, we shall first evaluate explicitly the integral expressions (2.11) and (2.12) of the so-called *connectors*  $\mathbf{A}$  and  $\mathbf{B}$ . It is only in this explicit evaluation that, as will be noted, the analysis of paper I has to be modified to account for the fact that one of the boundaries encloses the others.

### 3. Evaluation of the connectors

Consider the integral expression (2.11) of the connector  $\mathbf{A}_{jl}^{(n,m)}$ , for the case that  $j \neq l$ . If both indices  $j$  and  $l$  are unequal to zero (that is to say, each of the two indices refers to a particle inside the container) one necessarily has  $|\mathbf{R}_l - \mathbf{R}_j| > a_j + a_l$ . Under the assumption that this inequality holds, the integral (2.11) has been evaluated in papers I and III. The result is (cf. eqs. (III-5.4), (III-5.5) and (III-5.11))

$$\mathbf{A}_{jl}^{(n,m)} = \mathbf{G}_{jl}^{(n,m)} + \mathbf{H}_{jl}^{(n,m)} \quad (R_{jl} > a_j + a_l), \quad (3.1)$$

with

$$\mathbf{G}_{jl}^{(n,m)} = (-1)^{n+1} \frac{3}{4} a_j^n a_l^{m-1} \frac{\overset{\leftarrow}{\partial}^{n-1}}{\partial \mathbf{R}_{jl}^{n-1}} \frac{\mathbf{1} + \hat{\mathbf{r}}_j \hat{\mathbf{r}}_{jl}}{R_{jl}} \frac{\overset{\leftarrow}{\partial}^{m-1}}{\partial \mathbf{R}_{jl}^{m-1}}, \quad (3.2)$$

$$\mathbf{H}_{jl}^{(n,m)} = (-1)^m \frac{3}{4} a_j^n a_l^{m-1} \left( \frac{a_j^2}{2n+1} + \frac{a_l^2}{2m+1} \right) (2n+2m-1)!! \mathbf{R}_{jl}^{-(n+m+1)} \overset{\leftarrow}{\hat{\mathbf{r}}}_{jl}^{n+m}. \quad (3.3)$$

The particle–particle connector  $\mathbf{A}_{jl}^{(n,m)}$  is the sum of two terms, one of which is of order  $\mathbf{R}_{jl}^{-(n+m-1)}$ , the other of order  $\mathbf{R}_{jl}^{-(n+m+1)}$  in the interparticle separation  $\mathbf{R}_{jl} \equiv |\mathbf{R}_l - \mathbf{R}_j|$ . Each of these terms is a tensor of rank  $n+m$  which is irreducible in its first  $n-1$  and last  $m-1$  indices. (The tensor  $\mathbf{H}$  is in fact irreducible in all of its indices.) In eqs. (3.2) and (3.3), the vector  $\hat{\mathbf{r}}_{jl} \equiv \mathbf{R}_{jl}/R_{jl}$  denotes the unit vector in the direction of  $\mathbf{R}_{jl} \equiv \mathbf{R}_l - \mathbf{R}_j$ , i.e. pointing from the center of sphere  $j$  to the center of sphere  $l$ . The arrow  $\leftarrow$  on  $\overset{\leftarrow}{\partial}/\partial \mathbf{R}_{jl}$  in eq. (3.2) indicates a differentiation to the left.

We next consider the case where one of the indices  $j$  and  $l$  is unequal to zero, that is to say, one of the indices refers to a particle, the other index to the

container. Assume first that  $l=0$ . One then has the inequality  $a_0 > |\mathbf{R}_0 - \mathbf{R}_j| + a_j$ . The integral (2.11) can for this case also be evaluated explicitly, cf. the appendix, to yield the result

$$\mathbf{A}_{j_0}^{(n,m)} = \begin{cases} 0 & \text{if } n \geq m + 1, \\ -\left(\frac{a_j}{a_0}\right)^n \mathbf{B}^{(n,n)} & \text{if } n = m, \\ {}^1\mathbf{A}_{j_0}^{(n,m)} & \text{if } n = m - 1, \\ {}^1\mathbf{A}_{j_0}^{(n,m)} + {}^2\mathbf{A}_{j_0}^{(n,m)} + {}^3\mathbf{A}_{j_0}^{(n,m)} & \text{if } n \leq m - 2, \end{cases} \quad (3.4)$$

with the definitions

$${}^1\mathbf{A}_{j_0}^{(n,m)} = (-1)^{n+m} (2m-3)!! a_j^n a_0^{-m} \left[ \frac{(m-1)!}{(m-n)!} \mathbf{R}_{j_0}^{m-n} \odot^{m-n} \Delta^{(m-1 \text{ id}, m-1)} - \frac{3}{4} (2m+1)^{-1} \frac{\overline{\partial^{n-1}}}{\partial \mathbf{R}_{j_0}^{n-1}} \frac{\overline{\partial^2}}{\partial \mathbf{R}_{j_0}^2} \overline{\mathbf{R}_{j_0}^{m-1}} \mathbf{R}_{j_0}^2 \right], \quad (3.5)$$

$${}^2\mathbf{A}_{j_0}^{(n,m)} = (-1)^{n+m} \frac{3}{4} \frac{(m-1)!}{(m-n-2)!} (2m-5)!! a_j^n a_0^{2-m} \mathbf{R}_{j_0}^{m-n-2} \odot^{m-n-2} \Delta^{(m-1, m-1)}, \quad (3.6)$$

$${}^3\mathbf{A}_{j_0}^{(n,m)} = -\frac{2m-3}{2n+1} \left(\frac{a_j}{a_0}\right)^2 {}^2\mathbf{A}_{j_0}^{(n,m)}. \quad (3.7)$$

Here the symbol  $\odot^p$  denotes a  $p$ -fold contraction, with the nesting convention shown in eq. (2.8).

In the above equations we have introduced a class of isotropic tensors  $\Delta^{(n \ n)}$  of rank  $2n$ , which project out the irreducible part of a tensor of rank  $n$ :

$$\Delta^{(n,n)} \odot \mathbf{b}^n = \mathbf{b}^n \odot \Delta^{(n,n)} = \overline{\mathbf{b}^n}. \quad (3.8)$$

For  $n = 1, 2$  one has e.g.

$$\Delta_{\mu, \nu}^{(1,1)} = \delta_{\mu\nu}, \quad \Delta_{\mu\nu, \kappa\lambda}^{(2,2)} = \frac{1}{2} \delta_{\mu\kappa} \delta_{\nu\lambda} + \frac{1}{2} \delta_{\mu\lambda} \delta_{\nu\kappa} - \frac{1}{3} \delta_{\mu\nu} \delta_{\kappa\lambda}. \quad (3.9)$$

We refer again to ref. 16 for useful properties of such isotropic tensors (of which a few are listed in appendix A of ref. 17). The tensor  $\Delta^{(n \text{ id } n)}$  of rank  $2n+2$  used in eq. (3.5) has components

$$\Delta_{\mu_1 \mu_n, \nu_n \kappa\lambda, \nu_1 \nu_n}^{(n, \text{id}, n)} = \delta_{\kappa\lambda} \Delta_{\mu_1 \mu_n \nu_1 \nu_n}^{(n,n)}. \quad (3.10)$$

Eqs. (3.4)–(3.10), together with the formulae for  $\mathbf{B}^{(n,n)}$  given below, complete the expressions for the particle–container connectors  $\mathbf{A}_{j_0}^{(n,m)}$ . The container–particle connectors  $\mathbf{A}_{0j}^{(m,n)}$  can be obtained from these results by means of the general symmetry relation for connectors<sup>2)</sup>

$$(\mathbf{A}_{lj}^{(n,m)})_{\alpha_1 \alpha_2 \dots \alpha_{n+m-1} \alpha_{n+m}} = \frac{a_l}{a_j} (\mathbf{A}_{jl}^{(m,n)})_{\alpha_{n+m} \alpha_{n+m-1} \dots \alpha_2 \alpha_1}, \quad (3.11)$$

which follows from their definition (2.11). Note that these connectors, as well as the particle–particle connectors considered previously, are tensors of rank  $n+m$  which are irreducible in their first  $n-1$  and last  $m-1$  indices. Their dependence upon the quantities  $a_j/a_0$  and  $R_{j0}/a_0$  is as follows:

$$\begin{aligned} \mathbf{A}_{j_0}^{(n,n)} &\propto \left(\frac{a_j}{a_0}\right)^n, & \mathbf{A}_{0j}^{(n,n)} &\propto \left(\frac{a_j}{a_0}\right)^{n-1}, \\ \mathbf{A}_{j_0}^{(n,n+1)} &\propto \left(\frac{a_j}{a_0}\right)^n \left(\frac{R_{j0}}{a_0}\right), & \mathbf{A}_{0j}^{(n+1,n)} &\propto \left(\frac{a_j}{a_0}\right)^{n-1} \left(\frac{R_{j0}}{a_0}\right), \end{aligned} \quad (3.12)$$

for upper indices which differ by zero or one, whereas if the upper indices differ by two or more the connector decomposes into terms of order

$$\begin{aligned} {}^1\mathbf{A}_{j_0}^{(n,m)} &\propto \left(\frac{a_j}{a_0}\right)^n \left(\frac{R_{j0}}{a_0}\right)^{m-n}, & {}^1\mathbf{A}_{0j}^{(m,n)} &\propto \left(\frac{a_j}{a_0}\right)^{n-1} \left(\frac{R_{j0}}{a_0}\right)^{m-n}, \\ {}^2\mathbf{A}_{j_0}^{(n,m)} &\propto \left(\frac{a_j}{a_0}\right)^n \left(\frac{R_{j0}}{a_0}\right)^{m-n-2}, & {}^2\mathbf{A}_{0j}^{(m,n)} &\propto \left(\frac{a_j}{a_0}\right)^{n-1} \left(\frac{R_{j0}}{a_0}\right)^{m-n-2}, \\ {}^3\mathbf{A}_{j_0}^{(n,m)} &\propto \left(\frac{a_j}{a_0}\right)^{n+2} \left(\frac{R_{j0}}{a_0}\right)^{m-n-2}, & {}^3\mathbf{A}_{0j}^{(m,n)} &\propto \left(\frac{a_j}{a_0}\right)^{n+1} \left(\frac{R_{j0}}{a_0}\right)^{m-n-2}, \quad n \leq m-2, \end{aligned} \quad (3.13)$$

cf. eqs. (3.4)–(3.7) and (3.11). Note that, if the center of the particle coincides with that of the container, the particle–container connectors simplify to

$$\mathbf{A}_{j_0}^{(n,m)} \Big|_{R_j=R_0} = \begin{cases} 0 & \text{if } n \neq m \text{ and } n \neq m-2, \\ -\left(\frac{a_j}{a_0}\right)^n \mathbf{B}^{(n,n)} & \text{if } n = m, \\ \frac{3}{4}(n+1)!(2n-1)!! \left(\frac{a_j}{a_0}\right)^n \left(1 - \frac{a_j^2}{a_0^2}\right) \mathbf{\Delta}^{(n+1,n+1)} & \text{if } n = m-2. \end{cases} \quad (3.14)$$

We conclude with results for the tensors  $\mathbf{B}^{(n,n)} \equiv -\mathbf{A}_{jj}^{(n,n)}$ , and their inverses,

derived in ref. 17 (cf. also eq. (I-4.16)):

$$\mathbf{B}^{(1,1)} = -\mathbf{1}, \quad (3.15)$$

$$B_{\mu\nu, \kappa\lambda}^{(2,2)} = -\frac{3}{10}(4\delta_{\mu\lambda}\delta_{\nu\kappa} - \delta_{\mu\kappa}\delta_{\nu\lambda} - \delta_{\mu\nu}\delta_{\kappa\lambda}), \quad (3.16)$$

$$\mathbf{B}^{(2s, 2s)^{-1}} = -\frac{10}{9}\mathbf{\Delta}^{(2,2)}, \quad (3.16a)$$

$$\begin{aligned} \mathbf{B}^{(n,n)} = & -\frac{3}{2}(n-1)!(2n-3)!! \left[ \mathbf{\Delta}^{(n-1, \text{id}, n-1)} - \frac{n}{2n+1}\mathbf{\Delta}^{(n,n)} \right. \\ & \left. - \frac{n-1}{2n-1}\mathbf{\Delta}^{(n-1, n-1)} \odot^{n-2}\mathbf{\Delta}^{(n-1, n-1)} \right] \quad (n \geq 3), \end{aligned} \quad (3.17)$$

$$\begin{aligned} \mathbf{B}^{(n, n)^{-1}} = & -\frac{2}{3}[(n-1)!(2n-3)!!]^{-1} \left[ \mathbf{\Delta}^{(n-1, \text{id}, n-1)} + \frac{n}{n+1}\mathbf{\Delta}^{(n,n)} \right. \\ & \left. + \left(\frac{n-1}{n-2}\right)\left(\frac{2n-3}{2n-1}\right)\mathbf{\Delta}^{(n-1, n-1)} \odot^{n-2}\mathbf{\Delta}^{(n-1, n-1)} \right] \quad (n \geq 3). \end{aligned} \quad (3.17a)$$

We also record here for future use the formula

$$\mathbf{\Delta}^{(n-1, n-1)} \odot^n \mathbf{B}^{(n, n)^{-1}} = -\frac{2}{3}(n-2)^{-1}[(n-1)!(2n-5)!!]^{-1}\mathbf{\Delta}^{(n-1, n-1)} \quad (n \geq 3), \quad (3.18)$$

which may be derived with the help of eqs. (A.3)–(A.5) of ref. 17.

#### 4. The motion of particles and fluid

In this section we give expressions for the mobility and friction tensors of the particles, as well as for the fluid velocity field, resulting from the hierarchy of eqs. (2.5)–(2.7). We shall restrict ourselves here to the case of a *motionless container*. As extension of the formulae to the case of a moving (e.g. rotating) container is straightforward, as the fundamental eqs. (2.5)–(2.7) are completely general in this respect.

Substituting therefore  $\mathbf{u}_0 = 0$ ,  $\boldsymbol{\omega}_0 = 0$  in eqs. (2.5) and (2.6) and eliminating the force  $\mathbf{K}_0$  and torque  $\mathbf{T}_0$  on the container, as well as the quantities  $\mathbf{F}_j^{(m)}$  (by means of eq. (2.7)), one obtains linear relations between the velocities and angular velocities of the particles on one hand, and the forces and torques

exerted on them by the fluid on the other hand. These relations may be written in the form

$$\left. \begin{aligned} \mathbf{u}_i &= - \sum_{j=1}^N \boldsymbol{\mu}_{ij}^{\text{TT}} \cdot \mathbf{K}_j - \sum_{j=1}^N \boldsymbol{\mu}_{ij}^{\text{TR}} \cdot \mathbf{T}_j \\ \boldsymbol{\omega}_i &= - \sum_{j=1}^N \boldsymbol{\mu}_{ij}^{\text{RT}} \cdot \mathbf{K}_j - \sum_{j=1}^N \boldsymbol{\mu}_{ij}^{\text{RR}} \cdot \mathbf{T}_j \end{aligned} \right\} i = 1, 2, \dots, N. \quad (4.1)$$

Here  $\boldsymbol{\mu}_{ij}^{\text{TT}}$  is the translational mobility tensor,  $\boldsymbol{\mu}_{ij}^{\text{RR}}$  the rotational mobility tensor, and the tensors  $\boldsymbol{\mu}_{ij}^{\text{TR}}$  and  $\boldsymbol{\mu}_{ij}^{\text{RT}}$  couple translational and rotational motion. The expressions for these mobility tensors, which follow from eqs. (2.5)–(2.7), have the same form as in an unbounded fluid (cf. eqs. (I-5.16)–(I-5.19)).

$$6\pi\eta a_i \boldsymbol{\mu}_{ij}^{\text{TT}} = \mathbf{1}\delta_{ij} + \mathbf{C}_{ij}^{(1,1)} + \sum_{s=1}^{\infty} \sum_{m_1=2}^{\infty} \cdots \sum_{m_s=2}^{\infty} \sum_{j_1=1}^N \cdots \sum_{j_s=1}^N \\ \times \mathbf{C}_{ij_1}^{(1, m_1)} \odot \mathbf{B}^{(m_1, m_1)^{-1}} \odot \mathbf{C}_{j_1 j_2}^{(m_1, m_2)} \odot \cdots \odot \mathbf{B}^{(m_s, m_s)^{-1}} \odot \mathbf{C}_{j_s j'}^{(m_s, 1)}, \quad (4.2)$$

$$8\pi\eta a_i^2 a_j \boldsymbol{\mu}_{ij}^{\text{RR}} = \mathbf{1}\delta_{ij} - \frac{1}{3} \boldsymbol{\epsilon} : \mathbf{C}_{ij}^{(2a, 2a)} : \boldsymbol{\epsilon} - \frac{1}{3} \sum_{s=1}^{\infty} \sum_{m_1=2}^{\infty} \cdots \sum_{m_s=2}^{\infty} \sum_{j_1=1}^N \cdots \sum_{j_s=1}^N \\ \times \boldsymbol{\epsilon} : \mathbf{C}_{ij_1}^{(2a, m_1)} \odot \mathbf{B}^{(m_1, m_1)^{-1}} \odot \mathbf{C}_{j_1 j_2}^{(m_1, m_2)} \odot \cdots \odot \mathbf{B}^{(m_s, m_s)^{-1}} \odot \mathbf{C}_{j_s j'}^{(m_s, 2a)} : \boldsymbol{\epsilon}, \quad (4.3)$$

$$12\pi\eta a_i^2 \boldsymbol{\mu}_{ij}^{\text{RT}} = - \boldsymbol{\epsilon} : \mathbf{C}_{ij}^{(2a, 1)} - \sum_{s=1}^{\infty} \sum_{m_1=2}^{\infty} \cdots \sum_{m_s=2}^{\infty} \sum_{j_1=1}^N \cdots \sum_{j_s=1}^N \\ \times \boldsymbol{\epsilon} : \mathbf{C}_{ij_1}^{(2a, m_1)} \odot \mathbf{B}^{(m_1, m_1)^{-1}} \odot \mathbf{C}_{j_1 j_2}^{(m_1, m_2)} \odot \cdots \odot \mathbf{B}^{(m_s, m_s)^{-1}} \odot \mathbf{C}_{j_s j'}^{(m_s, 1)}, \quad (4.4)$$

$$12\pi\eta a_i a_j \boldsymbol{\mu}_{ij}^{\text{TR}} = \mathbf{C}_{ij}^{(1, 2a)} : \boldsymbol{\epsilon} + \sum_{s=1}^{\infty} \sum_{m_1=2}^{\infty} \cdots \sum_{m_s=2}^{\infty} \sum_{j_1=1}^N \cdots \sum_{j_s=1}^N \\ \times \mathbf{C}_{ij_1}^{(1, m_1)} \odot \mathbf{B}^{(m_1, m_1)^{-1}} \odot \mathbf{C}_{j_1 j_2}^{(m_1, m_2)} \odot \cdots \odot \mathbf{B}^{(m_s, m_s)^{-1}} \odot \mathbf{C}_{j_s j'}^{(m_s, 2a)} : \boldsymbol{\epsilon}. \quad (4.5)$$

The tensor  $\mathbf{C}_{ij}^{(n, m)}$  used in these equations (with  $i, j = 1, 2, \dots, N$ ) is defined in terms of the connectors by

$$\mathbf{C}_{ij}^{(n, m)} = \mathbf{A}_{ij}^{(n, m)} (1 - \delta_{ij}) - \mathbf{A}_{i0}^{(n, 1)} \cdot \mathbf{A}_{0j}^{(1, m)} - \frac{2}{3} \mathbf{A}_{i0}^{(n, 2a)} : \mathbf{A}_{0j}^{(2a, m)} \\ - \frac{10}{9} \mathbf{A}_{i0}^{(n, 2s)} : \mathbf{A}_{0j}^{(2s, m)} + \sum_{p=3}^{\infty} \mathbf{A}_{i0}^{(n, p)} \odot \mathbf{B}^{(p, p)^{-1}} \odot \mathbf{A}_{0j}^{(p, m)}. \quad (4.6)$$

The first term on the r.h.s. of eq. (4.6) reflects a hydrodynamic interaction between particle  $i$  and particle  $j \neq i$ . This term is also present in the case of an unbounded fluid, cf. paper I. The remaining terms may be seen as expressing hydrodynamic interactions between particles  $i$  and  $j$  *via the container*. (Note that these latter terms may also refer to a single particle, viz. for  $i = j$ .)

Substitution of expressions (3.1)–(3.7) for the connectors into the above equations yields the mobilities as an expansion in the three parameters  $a_p/a_0$ ,  $a_p/R_{pp'}$ , and  $R_{p0}/a_0$ . Here  $R_{pp'}$  and  $R_{p0}$  denote symbolically the typical separations of two particles and of a particle to the center of the container, respectively. Also,  $a_p$  is the typical radius of a particle and  $a_0$  the radius of the container. The dependence of the connectors on these parameters is given in section 3 (see in particular eqs. (3.12) and (3.13)).

As an aside, we note that one may easily verify that the mobilities defined in eqs. (4.2)–(4.5) satisfy the symmetry relations<sup>12</sup>

$$\boldsymbol{\mu}_{ij}^{\text{TT}} = \tilde{\boldsymbol{\mu}}_{ji}^{\text{TT}}, \quad \boldsymbol{\mu}_{ji}^{\text{RR}} = \tilde{\boldsymbol{\mu}}_{ji}^{\text{RR}}, \quad \boldsymbol{\mu}_{ij}^{\text{RT}} = \tilde{\boldsymbol{\mu}}_{ji}^{\text{TR}}, \quad (4.7)$$

where  $\tilde{\boldsymbol{\mu}}$  is the transpose of  $\boldsymbol{\mu}$ . As in the case of unbounded fluid<sup>2</sup>), these relations are within the present scheme a direct consequence of the symmetry (3.11) of the connectors.

We proceed to consider the flow of fluid caused by the motion of the particles inside the container at rest. The fluid velocity field  $\boldsymbol{v}(\boldsymbol{r})$  can be expressed in terms of the forces and torques on the particles by

$$\boldsymbol{v}(\boldsymbol{r}) = - \sum_{j=1}^N \boldsymbol{S}_j^{\text{T}}(\boldsymbol{r}) \cdot \boldsymbol{K}_j - \sum_{j=1}^N \boldsymbol{S}_j^{\text{R}}(\boldsymbol{r}) \cdot \boldsymbol{T}_j. \quad (4.8)$$

As noted in paper III, the tensors  $\boldsymbol{S}_j^{\text{T}}(\boldsymbol{r})$  and  $\boldsymbol{S}_j^{\text{R}}(\boldsymbol{r})$  defined above follow immediately from the general expressions for the mobilities of  $N + 1$  particles, by putting  $\boldsymbol{R}_{N+1} = \boldsymbol{r}$  and taking the limit  $a_{N+1} \rightarrow 0$ ,

$$\left. \begin{aligned} \boldsymbol{S}_j^{\text{T}}(\boldsymbol{r}) &= \lim_{a_{N+1} \rightarrow 0} \boldsymbol{\mu}_{N+1,j}^{\text{TT}} \Big|_{\boldsymbol{R}_{N+1}=\boldsymbol{r}} \\ \boldsymbol{S}_j^{\text{R}}(\boldsymbol{r}) &= \lim_{a_{N+1} \rightarrow 0} \boldsymbol{\mu}_{N+1,j}^{\text{TR}} \Big|_{\boldsymbol{R}_{N+1}=\boldsymbol{r}} \end{aligned} \right\} j = 1, 2, \dots, N. \quad (4.9)$$

So far, we have considered the forces and torques on the particles as given and have expressed the velocities in terms of these quantities. Alternatively, one may consider the velocities as given and ask for the forces and torques,

$$\left. \begin{aligned} \mathbf{K}_i &= - \sum_{j=1}^N \zeta_{ij}^{\text{TT}} \cdot \mathbf{u}_j - \sum_{j=1}^N \zeta_{ij}^{\text{TR}} \cdot \boldsymbol{\omega}_j \\ \mathbf{T}_i &= - \sum_{j=1}^N \zeta_{ij}^{\text{RT}} \cdot \mathbf{u}_j - \sum_{j=1}^N \zeta_{ij}^{\text{RR}} \cdot \boldsymbol{\omega}_j \end{aligned} \right\} \quad i = 1, 2, \dots, N. \quad (4.10)$$

The friction tensors  $\zeta$  may be obtained by inversion of the mobility tensor matrix, or more directly from the hierarchy (2.5)–(2.7). The resulting expressions for  $\zeta_{ij}^{\text{TT}}$ ,  $\zeta_{ij}^{\text{RR}}$ ,  $\zeta_{ij}^{\text{RT}}$  and  $\zeta_{ij}^{\text{TR}}$  will not be recorded here, as they are identical to those given in paper III for the case of a fluid bounded in one direction by a plane wall (eqs. (III-B.2)–(III-B.5))– with the proviso that the definition of the tensor  $\mathbf{C}_{ij}^{(n\ m)}$  given in that paper is replaced by eq. (4.6) in this paper.

## 5. Explicit results

Substitution of the expressions for the connectors derived in section 3 into the general formulae of section 4 enables one to calculate the (translational and rotational) mobility and friction tensors, as well as the fluid velocity field, to any desired order in the three expansion parameters which are\*: the ratio of particle radius to container radius ( $a_p/a_0$ ), the ratio of particle radius to interparticle separation ( $a_p/R_{pp'}$ ), and the ratio of the center to center separation of particle and container to the radius of the container ( $R_{p0}/a_0$ ).

We give below explicit expressions for the mobilities of spherical particles inside a *motionless* container, including terms of order  $(a_p/a_0)^n (a_p/R_{pp'})^m (R_{p0}/a_0)^l$  with  $n + m + l \leq 3$ . To this order specific hydrodynamic interactions of one and two particles and the container contribute. One finds

$$\begin{aligned} 6\pi\eta a_i \boldsymbol{\mu}_{ij}^{\text{TT}} &= \mathbf{1}\delta_{ij} + \mathbf{A}_{ij}^{(1,1)}(1 - \delta_{ij}) - \mathbf{A}_{i0}^{(1,1)} \cdot \mathbf{A}_{0j}^{(1,1)} - \frac{2}{3} \mathbf{A}_{i0}^{(1,2a)} : \mathbf{A}_{0j}^{(2a,1)} \\ &\quad - \frac{10}{9} \mathbf{A}_{i0}^{(1\ 2s)} : \mathbf{A}_{0j}^{(2s,1)} + ({}^1\mathbf{A}_{i0}^{(1,3)} + {}^2\mathbf{A}_{i0}^{(1,3)} + {}^3\mathbf{A}_{i0}^{(1,3)}) \odot \mathbf{B}^{(3,3)^{-1}} \odot {}^2\mathbf{A}_{0j}^{(3,1)} \\ &\quad + {}^2\mathbf{A}_{i0}^{(1,3)} \odot \mathbf{B}^{(3,3)^{-1}} \odot ({}^1\mathbf{A}_{0j}^{(3,1)} + {}^3\mathbf{A}_{0j}^{(3,1)}) + {}^2\mathbf{A}_{i0}^{(1,4)} \odot \mathbf{B}^{(4,4)^{-1}} \odot {}^2\mathbf{A}_{0j}^{(4,1)} \\ &= \mathbf{1}\delta_{ij} + \left[ \frac{3}{4} a_i \mathbf{R}_{ij}^{-1} (\mathbf{1} + \hat{\mathbf{r}}_{ij} \hat{\mathbf{r}}_{ij}) - \frac{3}{4} a_i (a_i^2 + a_j^2) \mathbf{R}_{ij}^{-3} (\hat{\mathbf{r}}_{ij} \hat{\mathbf{r}}_{ij} - \frac{1}{3} \mathbf{1}) \right] (1 - \delta_{ij}) \\ &\quad + \mathbf{1} \left[ -\frac{9}{4} a_i a_0^{-1} + \frac{5}{4} a_i (a_i^2 + a_j^2) a_0^{-3} \right] + \frac{3}{2} a_i a_0^{-3} \mathbf{1} [R_{i0}^2 + R_{j0}^2 - \frac{19}{8} \mathbf{R}_{i0} \cdot \mathbf{R}_{j0}] \\ &\quad + \frac{3}{4} a_i a_0^{-3} \left[ \frac{5}{2} \mathbf{R}_{i0} \mathbf{R}_{j0} - \frac{11}{4} \mathbf{R}_{j0} \mathbf{R}_{i0} - \mathbf{R}_{i0} \mathbf{R}_{i0} - \mathbf{R}_{j0} \mathbf{R}_{j0} \right], \quad (5.1) \end{aligned}$$

\* It should be explicitly remarked, that one cannot recover the results for the influence of a plane wall (obtained in paper III) by simply taking the limit  $a_0 \rightarrow \infty$ ,  $R_{p0} \rightarrow \infty$  (with  $a_0 - R_{p0}$  finite) in our expressions, as our third expansion parameter would tend to unity in this limit, thereby rendering the expansion unjustified.

$$\begin{aligned}
 8\pi\eta a_i^2 a_j \boldsymbol{\mu}_{ij}^{\text{RR}} &= \mathbf{1} \delta_{ij} - \frac{1}{3} \boldsymbol{\epsilon} : \mathbf{G}_{ij}^{(2a, 2a)} : \boldsymbol{\epsilon} (1 - \delta_{ij}) + \frac{2}{9} \boldsymbol{\epsilon} : \mathbf{A}_{i0}^{(2a, 2a)} : \mathbf{A}_{0j}^{(2a, 2a)} : \boldsymbol{\epsilon} \\
 &\quad - \frac{1}{3} \boldsymbol{\epsilon} : {}^2 \mathbf{A}_{i0}^{(2a, 4)} \odot \mathbf{B}^{(4, 4)^{-1}} \odot {}^2 \mathbf{A}_{0j}^{(4, 2a)} : \boldsymbol{\epsilon} \\
 &= \mathbf{1} \delta_{ij} + \frac{3}{2} a_i^2 a_j \mathbf{R}_{ij}^{-3} (\hat{\mathbf{r}}_i \hat{\mathbf{r}}_j - \frac{1}{3} \mathbf{1}) (1 - \delta_{ij}) - a_i^2 a_j a_0^{-3} \mathbf{1}, \quad (5.2)
 \end{aligned}$$

$$\begin{aligned}
 12\pi\eta a_i^2 \boldsymbol{\mu}_{ij}^{\text{RT}} &= 12\pi\eta a_i^2 \tilde{\boldsymbol{\mu}}_{ij}^{\text{TR}} = -\boldsymbol{\epsilon} : \mathbf{G}_{ij}^{(2a, 1)} (1 - \delta_{ij}) + \frac{2}{3} \boldsymbol{\epsilon} : \mathbf{A}_{i0}^{(2a, 2a)} : \mathbf{A}_{0j}^{(2a, 1)} \\
 &\quad - \boldsymbol{\epsilon} : \mathbf{A}_{i0}^{(2a, 3)} \odot \mathbf{B}^{(3, 3)^{-1}} \odot {}^2 \mathbf{A}_{0j}^{(3, 1)} - \boldsymbol{\epsilon} : {}^2 \mathbf{A}_{i0}^{(2a, 4)} \odot \mathbf{B}^{(4, 4)^{-1}} \odot {}^2 \mathbf{A}_{0j}^{(4, 1)} \\
 &= -\frac{3}{2} a_i^2 \mathbf{R}_{ij}^{-2} \boldsymbol{\epsilon} \cdot \hat{\mathbf{r}}_j (1 - \delta_{ij}) + \frac{3}{2} a_i^2 a_0^{-3} (\frac{5}{2} \boldsymbol{\epsilon} \cdot \mathbf{R}_{i0} - \boldsymbol{\epsilon} \cdot \mathbf{R}_{j0}). \quad (5.3)
 \end{aligned}$$

(Note the usefulness of formula (3.18) for evaluating tensor contractions in the above equations.)

In eqs. (5.1)–(5.3) the indices  $i, j = 1, 2, \dots, N$  label particles, whereas the index 0 is reserved for the container. We recall the notations used:  $\mathbf{R}_{i0}$  is a vector, with magnitude  $R_{i0}$ , pointing from the center of particle  $i$  to the center of the container;  $\mathbf{R}_{j0}$  and  $R_{j0} \equiv |\mathbf{R}_{j0}|$  are defined similarly for particle  $j$ ;  $\mathbf{R}_{ij}$  is a vector pointing from the center of particle  $i$  to the center of particle  $j$ ; this vector has magnitude  $R_{ij}$  and direction  $\hat{\mathbf{r}}_{ij} \equiv \mathbf{R}_{ij}/R_{ij}$ ;  $a_i$  and  $a_j$  are the radii of particles  $i$  and  $j$ ;  $a_0$  is the radius of the container;  $\mathbf{1}$  denotes the second-rank unit tensor and  $\boldsymbol{\epsilon}$  the third-rank Levi-Civita tensor, with the property  $\boldsymbol{\epsilon} : \mathbf{ab} = -\mathbf{a} \wedge \mathbf{b}$ ; finally,  $\tilde{\boldsymbol{\mu}}$  denotes the transpose of  $\boldsymbol{\mu}$ .

For a single particle inside the container,  $i = j$  and the expressions given above reduce to

$$6\pi\eta a_i \boldsymbol{\mu}_i^{\text{TT}} = \mathbf{1} \left[ 1 - \frac{9}{4} \frac{a_i}{a_0} + \frac{5}{2} \left( \frac{a_i}{a_0} \right)^3 \right] - \frac{9}{16} a_i a_0^{-3} (\mathbf{R}_{i0}^2 \mathbf{1} + 3 \mathbf{R}_{i0} \mathbf{R}_{i0}), \quad (5.4)$$

$$8\pi\eta a_i^3 \boldsymbol{\mu}_i^{\text{RR}} = \mathbf{1} \left[ 1 - \left( \frac{a_i}{a_0} \right)^3 \right], \quad (5.5)$$

$$12\pi\eta a_i^2 \boldsymbol{\mu}_i^{\text{RT}} = 12\pi\eta a_i^2 \tilde{\boldsymbol{\mu}}_i^{\text{TR}} = \frac{9}{4} a_i^2 a_0^{-3} \boldsymbol{\epsilon} \cdot \mathbf{R}_{i0}. \quad (5.6)$$

Note that eq. (5.6) implies that a single particle  $i$  moving under the influence of a hydrodynamic force  $\mathbf{K}$  will acquire an angular velocity  $\boldsymbol{\omega}$  equal to  $(3/16\pi\eta a_0^3) \mathbf{R}_{i0} \wedge \mathbf{K}$ , to the order considered. We have verified that the above equations with  $\mathbf{R}_{i0} = 0$  are consistent with the well-known<sup>7–12</sup>) *exact* results for the motion of one sphere *concentric* with a spherical container\*.

\* From Lamb's<sup>10</sup>) solution for a spherical particle rotating inside a concentric spherical container, one finds in fact that eq. (5.5) holds *exactly* for this particular case. Within the present theory this is a consequence of the fact that, if  $\mathbf{R}_{i0} = 0$ ,  $\mathbf{A}_{i0}^{(2a, n)} = 0$  unless  $n$  equals  $2a$  (cf. eq. (3.14)), so that eq. (5.5) contains indeed to all orders the contributions to the rotational mobility.

The flow of fluid in the container due to motion of the particles follows directly from eqs. (5.1) and (5.3), by virtue of relation (4.9). We have again verified for the case of a single particle having a common center with the container, that our results for the fluid velocity field agree with those in the literature<sup>11</sup>).

We remark, finally, that extensions of the formulae given above valid up to higher than third order in the expansion parameters, can be obtained in a straightforward way from the general results of sections 3 and 4 – by evaluating the appropriate contractions of the connector tensors.

**Acknowledgement**

This work was performed as part of the research programme of the “Stichting voor Fundamenteel Onderzoek der Materie” (F.O.M.), with financial support from the “Nederlandse Organisatie voor Zuiver-Wetenschappelijk Onderzoek” (Z.W.O.).

**Appendix**

*Evaluation of particle–container connectors*

The evaluation of the integral expression (2.11) of the particle–container connectors  $\mathbf{A}_{j_0}^{(n,m)}$  (with  $a_0 > R_{j_0} + a_j$ ) is simplest for the case  $n \geq m$ . We shall examine this case first, before proceeding to the more involved case of  $n < m$ .

We make use of the formula<sup>18</sup>)

$$\int_0^\infty dk k^{\mu-1} J_\mu(bk) f(k) = 2^{\mu-1} b^{-\mu} \Gamma(\mu) f(0), \quad \text{Re } \mu > 0, \tag{A.1}$$

valid for all functions  $f(z)$  which are: (i) analytic in the right complex half-plane,  $\text{Re } z \geq 0$ ; (ii) even along the imaginary axis; (iii) bounded for large  $|z|$  by  $\exp(b'|\text{Im } z|)$ , with  $b > b' \geq 0$ . If one makes the substitutions  $\mu = m - \frac{1}{2}$ ,  $b = a_j$ , and

$$f(k) = \frac{3}{8\pi} \left(\frac{a_j}{a_l}\right)^{1/2} i^{n-m} (2n-1)!! (2m-1)!! \times \int d\hat{k} \hat{k}^{n-1} (\mathbf{1} - \hat{k}\hat{k}) \hat{k}^{m-1} e^{-ik\hat{k}\cdot\mathbf{R}_j} k^{1/2-m} J_{n-1/2}(a_j k) \tag{A.2}$$

in the l.h.s. of eq. (A.1), one obtains precisely the integral (2.11) under consideration.

We now restrict ourselves to the case  $l = 0, n \geq m$ . One readily verifies that the function (A.2) then satisfies conditions (i)–(iii) above. Formula (A.1) may thus be applied to evaluate the integral (2.11), and one finds

$$\mathbf{A}_{j_0}^{(n, m)} = 0 \quad \text{if } n > m, \tag{A.3}$$

$$\begin{aligned} \mathbf{A}_{j_0}^{(n, n)} &= \left(\frac{a_l}{a_0}\right)^n \frac{3}{8\pi} [(2n - 1)!!]^2 (2n - 1)^{-1} \int d\hat{\mathbf{k}} \widehat{\mathbf{k}}^{n-1} (\mathbf{1} - \hat{\mathbf{k}}\hat{\mathbf{k}}) \widehat{\mathbf{k}}^{n-1} \\ &= -\left(\frac{a_l}{a_0}\right)^n \mathbf{B}^{(n, n)}. \end{aligned} \tag{A.4}$$

The last equality in (A.4) follows from definition (2.12) of the tensor  $\mathbf{B}^{(n, n)}$ ,

$$\mathbf{B}^{(n, n)} = -\frac{3}{8\pi} [(2n - 1)!!]^2 \int_0^\infty dk k^{-1} J_{n-1/2}^2(k) \int d\hat{\mathbf{k}} \widehat{\mathbf{k}}^{n-1} (\mathbf{1} - \hat{\mathbf{k}}\hat{\mathbf{k}}) \widehat{\mathbf{k}}^{n-1}, \tag{A.5}$$

upon carrying out the scalar integration<sup>19</sup>).

It will be noted that the restriction  $n \geq m$  in the above evaluation of particle–wall connectors is essential, as the function (A.2) has a pole in the origin for  $n < m$ , thereby violating the first condition necessary for eq. (A.1) to hold. To obtain the connectors for  $n < m$  as well, we proceed as follows.

We first write the integral expression (2.11) (with  $l = 0$ ) in the form

$$\begin{aligned} \mathbf{A}_{j_0}^{(n, m)} &= \frac{3}{8\pi} \left(\frac{a_l}{a_0}\right)^{1/2} (-1)^{n+1} (2n - 1)!! (2m - 1)!! \int_0^\infty dk \int d\hat{\mathbf{k}} k^{1-n-m} \\ &\quad \times J_{n-1/2}(a_l k) J_{m-1/2}(a_0 k) \frac{\widehat{\partial}^{n-1}}{\partial \mathbf{R}_{j_0}^{n-1}} (\mathbf{1} - \hat{\mathbf{k}}\hat{\mathbf{k}}) \frac{\widehat{\partial}^{m-1}}{\partial \mathbf{R}_{j_0}^{m-1}} e^{-i\mathbf{k} \cdot \mathbf{R}_{j_0}}. \end{aligned} \tag{A.6}$$

We next expand the exponential into irreducible tensors\*

$$e^{-i\mathbf{k} \cdot \mathbf{r}} = \sum_{p=0}^\infty (-i)^p \frac{(2p + 1)!!}{p!} (\pi/2kr)^{1/2} J_{p+1/2}(kr) \widehat{\mathbf{k}}^p \odot \widehat{\mathbf{r}}^p \tag{A.7}$$

\* Formula (A.7) is obtained by combining the well-known expansion<sup>20</sup>  $e^{-i\mathbf{k} \cdot \mathbf{r}} = \sum_{l=0}^\infty (-i)^l (2l + 1) (\pi/2kr)^{1/2} J_{l+1/2}(kr) P_l(\hat{\mathbf{k}} \cdot \hat{\mathbf{r}})$  with the expression for the Legendre polynomial given in ref. 16 (eq. (4.21)):  $P_l(\hat{\mathbf{k}} \cdot \hat{\mathbf{r}}) = [(2l - 1)!!/l!] \widehat{\mathbf{k}}^l \odot \widehat{\mathbf{r}}^l$ .

and substitute this expansion into eq. (A.6). The angular integration is then performed using the result<sup>16)</sup>

$$\frac{1}{4\pi} \int d\hat{k} (\mathbf{1} - \hat{k}\hat{k}) \sqrt{\hat{k}}^p = \frac{2}{3} \mathbf{1} \delta_{p0} - \frac{2}{15} \mathbf{A}^{(2,2)} \delta_{p2}; \tag{A.8}$$

the scalar integration is evaluated by means of formula (6.578.1) of ref. 19, valid for  $a_0 > R_{j0} + a_j$ . We thus obtain the expression

$$\begin{aligned} \mathbf{A}_{j_0}^{(n,m)} &= (-1)^{n+1} (2n-1)!! (2m-1)!! a_j^n a_0^{m-2} \sum_{p=0,2}^{\infty} \sum_{\substack{q,s=0 \\ q+s \leq m-p/2-1}}^{\infty} 2^{p/2-m+1} \\ &\times (-1)^{q+s} \frac{(p+2q+2s-1)!!}{q! s! (m-\frac{1}{2}p-q-s-1)! (2p+2q+1)!! (2n+2s-1)!!} \left(\frac{a_j}{a_0}\right)^{2s} \\ &\times \frac{\overline{\partial^{n-1}}}{\partial \mathbf{R}_{j_0}^{n-1}} \left( \mathbf{1} \delta_{p0} + \frac{3}{2} \overline{\hat{r}_{j_0} \hat{r}_{j_0}} \delta_{p2} \right) \left(\frac{R_{j0}}{a_0}\right)^{2q+p} \frac{\overline{\partial^{m-1}}}{\partial \mathbf{R}_{j_0}^{m-1}}. \end{aligned} \tag{A.9}$$

(The convention  $(-1)!! = 1$  has been adopted here.)

It remains to carry out the differentiations in eq. (A.9). Let us first examine the terms with  $p = 0$  in the r.h.s. of this equation and concentrate on the expression

$$\mathbf{E}_1 \equiv \frac{\overline{\partial^{n-1}}}{\partial \mathbf{R}^{n-1}} \mathbf{1} \frac{\overline{\partial^{m-1}}}{\partial \mathbf{R}^{m-1}} R^{2q} \quad (0 \leq q \leq m-1), \tag{A.10}$$

which they contain. (To simplify the notation, the indices of the vector  $\mathbf{R}_{j_0}$  have been omitted here.) By means of the formula

$$\frac{\overline{\partial^{m-1}}}{\partial \mathbf{R}^{m-1}} R^{2q} = \overline{\mathbf{R}^{m-1}} \left( R^{-1} \frac{\partial}{\partial R} \right)^{m-1} R^{2q} \tag{A.11}$$

(which is a special case of eq. (A.1) in ref. 21) one finds

$$\mathbf{E}_1 = \begin{cases} 2^{m-1} (m-1)! \frac{\overline{\partial^{n-1}}}{\partial \mathbf{R}^{n-1}} \overline{\mathbf{R}^{m-1}} & \text{if } q = m-1, \\ 0 & \text{if } q \leq m-2, \end{cases} \tag{A.12}$$

which is easily reduced to

$$E_1 = \begin{cases} 2^{m-1}[(m-1)!]^2[(m-n)!]^{-1} R^{m-n} \odot^{m-n} \Delta^{(m-1, id, m-1)} & \text{if } q = m-1 \\ & \text{and } n \leq m, \\ 0 & \text{if } q \leq m-2 \text{ or } n \geq m+1. \end{cases} \quad (A.13)$$

Next, consider the remaining terms in eq. (A.9) with  $p = 2$ . The expression to be evaluated is now

$$E_2 = \frac{\overline{\partial^{n-1}}}{\partial R^{n-1}} \overline{RRR} R^{2q} \frac{\overline{\partial^{m-1}}}{\partial R^{m-1}} = \frac{1}{4} [(q+1)(q+2)]^{-1} \frac{\overline{\partial^{n-1}}}{\partial R^{n-1}} \frac{\overline{\partial^2}}{\partial R^2} \frac{\overline{\partial^{m-1}}}{\partial R^{m-1}} R^{2q+4} \quad (0 \leq q \leq m-2). \quad (A.14)$$

Using again eq. (A.11) one finds

$$E_2 = 2^{m-3}(m-2)! \frac{\overline{\partial^{n-1}}}{\partial R^{n-1}} \frac{\overline{\partial^2}}{\partial R^2} \overline{R^{m-1}} R^2 \quad \text{if } q = m-2 \text{ and } n \leq m, \quad (A.15)$$

$$E_2 = 2^{m-3}(m-3)! \frac{\overline{\partial^{n-1}}}{\partial R^{n-1}} \frac{\overline{\partial^2}}{\partial R^2} \overline{R^{m-1}} \\ = 2^{m-3} \frac{(m-1)!(m-3)!}{(m-n-2)!} R^{m-n-2} \odot^{m-n-2} \Delta^{(m-1, m-1)} \quad \text{if } q = m-3 \text{ and } n \leq m-2, \quad (A.15a)$$

$$E_2 = 0 \quad \text{if } q \leq m-4 \text{ or } (q = m-3, n \geq m-1) \text{ or } (q = m-2, n \geq m+1). \quad (A.15b)$$

(We remark that it is possible to derive a general formula in which the remaining differentiations in eq. (A.15) have been carried out. This formula is, however, rather lengthy and will not be recorded here.)

Substitution of results (A.13) and (A.15) into eq. (A.9) gives, together with eqs. (A.3) and (A.4), the required expressions (3.4)–(3.7) for the particle–container connectors.

### References

- 1) P. Mazur, *Physica* **110A** (1982) 128.
- 2) P. Mazur and W. van Saarloos, *Physica* **115A** (1982) 21. (Paper I.)
- 3) M. Smoluchowski, *Bull. Acad. Sci. Cracow* **1a** (1911) 28.
- 4) M. Smoluchowski, *Proc. Vth Intern. Congr. of Mathematics* (Cambridge, 1912) Vol. II, p. 192.
- 5) C.W.J. Beenakker and P. Mazur, *Phys. Fluids* **28** (1985) 767.

- 6) C W J Beenakker, W van Saarloos and P Mazur, *Physica* **127A** (1984) 451 (Paper III)
- 7) E Cunningham, *Proc Royal Soc London* **83A** (1910) 357
- 8) W E Williams, *Phil Mag 6th series* **29** (1915) 526
- 9) H M Lee, M S Thesis, Univ of Iowa (Iowa City, 1947)
- 10) H Lamb, *Hydrodynamics* (Cambridge Univ Press, Cambridge, 1932)
- 11) L D Landau and E M Lifshitz, *Fluid Mechanics* (Pergamon, Oxford, 1959)
- 12) J Happel and H Brenner, *Low Reynolds Number Hydrodynamics* (Noordhoff, Leiden, 1973)
- 13) G B Jeffery, *Proc London Math Soc* **14** (1915) 327
- 14) P Mazur and D Bedeaux, *Physica* **76** (1974) 235
- 15) T Yoshizaki and H Yamakawa, *J Chem Phys* **73** (1980) 578
- 16) S Hess and W Kohler, *Formeln zur Tensor-Rechnung* (Palm und Enke, Erlangen 1980, ISBN 3-7896-0046-6)
- 17) C W J Beenakker and P Mazur, *Physica* **120A** (1983) 388
- 18) Ch Schwartz, *J Math Phys* **23** (1982) 2266
- 19) I S Gradshteyn and I M Ryzhik, *Table of Integrals, Series and Products* (Academic Press, New York, 1980)
- 20) L D Landau and E M Lifshitz, *Quantum Mechanics* (Pergamon, Oxford 1977) §34
- 21) A J Weisenborn and P Mazur, *Physica* **123A** (1984) 191