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MANY-SPHERE HYDRODYNAMIC INTERACTIONS AND MOBILITIES IN A SUSPENSION

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A general scheme is presented to evaluate the mobility tensors of an arbitrary number of spheres, immersed in a viscous fluid, in a power series expansion in R^{-1}, where R is a typical distance between spheres. Some general properties of these (translational and rotational) mobility tensors are discussed. Explicit expressions are derived up to order R^{-7}. To this order, hydrodynamic interactions between two, three and four spheres contribute.

1. Introduction

The hydrodynamic interactions between spheres moving in a viscous fluid play an important role in many physical and physico-chemical problems. They have been studied extensively for this reason, in particular in situations in which the fluid can be described by the linearized Navier–Stokes equations for incompressible steady flow[1]). In general, these interactions are studied by the so-called methods of reflection. These methods, in which one calculates the relevant quantities as a series expansion in the inverse particle distances, were inaugurated by Smoluchowski[2]). Because of their complexity, they have essentially only been applied to the case of two spheres, with one important exception, however, to which we will return.

For the two-sphere problem, Smoluchowski[2]) considers the velocities as given and calculates the hydrodynamic forces exerted on them. In doing so, he calculated the friction tensors up to order R_{12}^{-2}, where R_{12} is the distance between the spheres. Later, Faxén[3]) determined the friction for the case that the spheres move along their line of centers up to order R_{12}^{-5}. For the special case of spheres with equal radii, Dahl[4]) carried Faxén's calculation to order R_{12}^{-9}. On the other hand, Burgers[5]) considered the forces to be given and then calculated the velocities of the two spheres. He evaluated in this way the mobility tensors (the elements of the inverse of the friction tensor matrix) to order R_{12}^{-4}. Happel and Brenner[1]), in their monograph, present a calculation of the friction tensors to order R_{12}^{-5} by considering not only Faxén's special arrangement but also the case that the spheres move perpendicular to their

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line of centers. More recent calculations of the mobility tensors were presented by Batchelor[6]), to order R_{12}^{-5}, and by Felderhof[7])*, to order R_{12}^{-7}.

In all those above mentioned treatments in which one directly obtains the mobility tensors, it was assumed that the two spheres were freely rotating, i.e. that the hydrodynamic torques exerted on the spheres vanish. Results for the speed of rotation in the case of free rotation as well as for the torque in the case of hindered rotation, in which the angular velocities of the spheres vanish, were obtained by Happel and Brenner[1]), in their approach to the problem which directly yielded expressions for the friction tensors.

In his paper published in 1959, Kynch[8]) considers the motion of *two* or *more* spheres through a viscous fluid. His work on many-sphere hydrodynamic interactions does not seem to have received due attention. Thus, Happel and Brenner write in their monograph[1])* "*Kynch has presented general formulas indicating how analytical solutions can be obtained for the case of three or more particles, but the expressions are so complicated that generalizations are possible only for special arrangements of the particles.*" Nevertheless, Kynch[8]), who considers the case of an arbitrary configuration of freely rotating spheres, explicitly gives expressions for all contributions to the mobility tensors up to order R^{-7}, where R is a typical distance between the spheres, and concludes that to this order two-, three- and four-particle interactions contribute. As we shall show, these expressions are correct. We were however unable to follow Kynch's[8]) particular analysis and line of reasoning which are based on the use of a reflection method.

In this paper, we shall treat the same problem as Kynch[8]) did, along the lines of a recent paper by one of us[9]), to be referred to as paper I. A method of induced forces[10,11]) was used there to derive a generalization of Faxén's theorem[12,10]) to the case of an arbitrary number of spheres of different radii, immersed in a fluid in non-uniform steady flow. In addition, this method also yielded expressions for the mobility tensors up to order R^{-3}. Here, we shall consider a system consisting of N spheres moving in a fluid at rest if unperturbed by their motion. We will extend for this case the analysis of paper I and construct a scheme which allows us to calculate the mobility tensors to any desired order of approximation as an expansion in powers of R^{-1}. In paper I, just as in the work of Kynch[8]), only the translational mobility tensors, which relate the hydrodynamic forces to their velocities, were studied. In this paper, on the other hand, we shall derive general expressions for all elements of the mobility tensor matrix, i.e. also for the tensors which

*Felderhof uses mixed slip-stick boundary conditions at the surface of the spheres. The other authors exclusively use stick boundary conditions. For further references on the two-sphere problem and in particular concerning exact solutions, see also refs. 1 and 7.

*Loc. cit. p. 276.

relate the velocities of the spheres to the hydrodynamic torques exerted on them, as well as for the tensors which relate the angular velocities of the spheres to the forces and torques. All these expressions will be evaluated explicitly up to order R^{-7}.

Our method differs from the usual methods of reflection[1-8]) used to study hydrodynamic interactions, in that it does not require the knowledge and therefore the explicit construction of the fluid velocity field. The desired results follow directly from an appropriate use of the boundary conditions for this field at the surfaces of the spheres. This proves to be very efficient in studying more than two spheres.

In section 2, we review the equations of motion. We also give the formal solution in wave-vector representation of the quasi-static Stokes equation, describing the motion of the fluid, on which the analysis is based (eq. (2.19)). In section 3 we discuss the expansion of the force induced in each sphere in irreducible induced-force-multipoles. This expansion is used in section 4 to obtain a hierarchy of equations for these force multipoles. This hierarchy is summarized in eqs. (5.2)–(5.5) of section 5: the force multipoles induced in the spheres are connected to each other as well as to the velocities and angular velocities of the spheres, via tensor objects called connectors, which are defined as three dimensional integrals in section 4. The hierarchy of equations is then used in section 5 to obtain expressions for the mobilities by elimination of all irreducible force multipoles except the total force and the total torque exerted on each sphere. Various properties of the mobilities, which are obtained in the form of a power series expansion in R^{-1}, are discussed. It is shown that they satisfy the required Onsager symmetry relations and that certain powers of R^{-1} do not occur in the expansion. Moreover, it is demonstrated that for e.g. the translational mobilities, the dominant contributions from clusters of n spheres, $n \geq 2$, are of order $R^{-(3n-5)}$. Finally, in section 6, the various mobility tensors (translational and rotational) are explicitly evaluated up to order R^{-7}. These expressions can be found in table II and eqs. (6.19)–(6.38).

Our analysis indicates that specific hydrodynamic interactions of 3 spheres may not a priori be neglected when evaluating the diffusion coefficient of a suspension which is not dilute. Indeed, as we shall see, these interactions are of order R^{-4}, i.e. of the same order as the short range part of the translational mobility tensors arising from binary hydrodynamic interactions. These two-sphere short range interactions have been shown to contribute considerably to the diffusion coefficient[13]).

Finally, it should be stressed that our scheme leads to a straightforward algorithm for calculating contributions of even higher order to the mobilities (cf. in this connection the concluding remarks of section 7).

2. Equations of motion

As in paper I, we consider N macroscopic spheres of masses m_i and radii a_i ($i = 1, \ldots, N$) immersed in an otherwise unbounded incompressible fluid. The centers of the spheres have positions $\boldsymbol{R}_i(t)$ at time t. We shall summarize in this section the basic equations of motion of the fluid and the spheres on which our subsequent analysis of the hydrodynamic interactions is based.

The motion of the fluid obeys the quasistatic Stokes equation,

$$\left.\begin{aligned} \boldsymbol{\nabla} \cdot \boldsymbol{P}(\boldsymbol{r}, t) = 0, \\ \boldsymbol{\nabla} \cdot \boldsymbol{v}(\boldsymbol{r}, t) = 0, \end{aligned}\right\} \quad \text{for all } |\boldsymbol{r} - \boldsymbol{R}_i(t)| > a_i, \qquad \begin{aligned} (2.1) \\ (2.2) \end{aligned}$$

with

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

Here $\boldsymbol{v}$ is the velocity field, $\boldsymbol{P}$ the pressure tensor, p the hydrostatic pressure and η the viscosity of the fluid. Here and henceforth, the index i runs from 1 to N and labels the spheres (and so do the indices j, k and l, to be used later), and Greek indices run from 1 to 3 and denote the Cartesian components. The velocity $\boldsymbol{u}_i(t)$ and the angular velocity $\boldsymbol{\omega}_i(t)$ of the ith sphere obey the equations of motion

$$m_i \frac{\mathrm{d}\boldsymbol{u}_i(t)}{\mathrm{d}t} = - \int\limits_{S_i(t)} \mathrm{d}S\boldsymbol{P}(\boldsymbol{r}, t) \cdot \hat{\boldsymbol{n}}_i + \boldsymbol{K}_i^{\mathrm{ext}}(t) \equiv \boldsymbol{K}_i(t) + \boldsymbol{K}_i^{\mathrm{ext}}(t), \tag{2.4}$$

$$I_i \frac{\mathrm{d}\boldsymbol{\omega}_i(t)}{\mathrm{d}t} = - \int\limits_{S_i(t)} \mathrm{d}S[\boldsymbol{r} - \boldsymbol{R}_i(t)] \wedge \boldsymbol{P} \cdot \hat{\boldsymbol{n}}_i + \boldsymbol{T}_i^{\mathrm{ext}} \equiv \boldsymbol{T}_i(t) + \boldsymbol{T}_i^{\mathrm{ext}}(t). \tag{2.5}$$

Here $\boldsymbol{K}_i$, $\boldsymbol{T}_i$, $\boldsymbol{K}_i^{\mathrm{ext}}$ and $\boldsymbol{T}_i^{\mathrm{ext}}$ are, respectively, the force and torque exerted by the fluid on sphere i, and the external force and torque on this sphere. $S_i(t)$ is the surface of sphere i at time t*, $\hat{\boldsymbol{n}}_i$ a unit vector normal to this surface and pointing in the outward direction, and $I_i = 2m_i a_i^2/5$ its moment of inertia (where a homogeneous mass distribution has been assumed). The set of equations (2.1)–(2.5) must be supplemented by boundary conditions at the surfaces of the spheres. We assume stick conditions, i.e.

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

Within the context of the method of induced forces (see paper I and ref. 10)

*The surface S_i is considered as the surface of a sphere centred at $\boldsymbol{R}_i$ with radius $a_i + \epsilon$ in the limit $\epsilon \downarrow 0$.

the above set of equations is replaced by an equivalent one in which the fluid equations are extended inside the spheres and are written in the form

$$\left.\begin{aligned} \nabla \cdot \mathsf{P}(r,t) &= \sum_{j=1}^{N} F_j(r,t), \\ \nabla \cdot v(r,t) &= 0, \end{aligned}\right\} \quad \text{for all } r, \tag{2.7, 2.8}$$

with $F_j(r,t) \equiv 0$, for $|r - R_j(t)| > a_j$. The extension of the fluid velocity field is chosen in such a way that

$$v(r,t) = u_i(t) + \omega_i(t) \wedge [r - R_i(t)], \quad \text{for } |r - R_i(t)| \leq a_i. \tag{2.9}$$

For the hydrostatic pressure we impose the condition

$$p(r,t) = 0, \quad \text{for } |r - R_i(t)| < a_i. \tag{2.10}$$

Clearly the problem posed by the set of equations including induced forces is completely equivalent with the original boundary value problem. Moreover, since according to eqs. (2.9) and (2.10) the pressure tensor in the induced force method is constant (zero) within the spheres and has discontinuities at their surface, it follows from eq. (2.7) that the induced force density must be of the form

$$F_i(r,t) = a_i^{-2} f_i(\hat{n}_i, t)\delta(|r - R_i(t)| - a_i). \tag{2.11}$$

The factor a_i^{-2} is introduced here for convenience.

If we make use of eq. (2.7), we can express the force K_i and the torque T_i which the fluid exerts on the ith sphere in terms of the induced force density. One has indeed

$$K_i(t) = -\int_{S_i(t)} \mathrm{d}S\mathsf{P}(r,t) \cdot \hat{n}_i = -\int_{|r-R_i(t)|\leq a_i} \mathrm{d}r \nabla \cdot \mathsf{P}(r,t) = -\int \mathrm{d}r F_i(r,t), \tag{2.12}$$

$$T_i(t) = -\int_{S_i(t)} \mathrm{d}S[r - R_i(t)] \wedge \mathsf{P}(r,t) \cdot \hat{n}_i = -\int \mathrm{d}r[r - R_i(t)] \wedge F_i(t). \tag{2.13}$$

In order to solve formally the equation of motion of the fluid we introduce Fourier transforms of e.g. the velocity field,

$$v(k) = \int \mathrm{d}r\, \mathrm{e}^{-ik \cdot r} v(r). \tag{2.14}$$

We have omitted here, and will do so henceforth, for all quantities and fields, the time argument of v. We also define the Fourier transform of the induced force density $F_i(t)$ in a reference frame in which sphere i is at rest at the origin,

$$F_i(k) = \int dr\, e^{-ik\cdot(r-R_i)} F_i(r). \tag{2.15}$$

The equations of motion (2.7) and (2.8), together with (2.3) then become in wavevector representation

$$\eta k^2 v(k) = -ikp(k) + \sum_{j=1}^{N} e^{-ik\cdot R_j} F_j(k), \tag{2.16}$$

with

$$k \cdot v(k) = 0. \tag{2.17}$$

If one applies the operator $\mathbf{1} - kk/k^2$ to both sides of eq. (2.16), one obtains with equation (2.17)

$$\eta k^2 v(k) = \sum_j e^{-ik\cdot R_j} (\mathbf{1} - \Omega\Omega) \cdot F_j(k), \tag{2.18}$$

where $\Omega \equiv k/k$ is the unit vector in the direction of k and $\mathbf{1}$ the unit tensor. This equation has the formal solution

$$v(k) = v^0(k) + \sum_{j=1}^{N} \eta^{-1} k^{-2} e^{-ik\cdot R_j} (\mathbf{1} - \Omega\Omega) \cdot F_j(k), \tag{2.19}$$

where $v^0(k)$ is the solution of eq. (2.16) in the absence of induced forces and is therefore the velocity field unperturbed by the presence of the N spheres. It is the above equation which will be the starting point for the calculation of the forces and the torques exerted by the fluid on the spheres. The spheres will be allowed to move with arbitrary velocity through the fluid, which may itself, in principle, be in arbitrary stationary non-uniform unperturbed flow. In our subsequent analysis we shall however assume that the unperturbed fluid is at rest,

$$v^0(r) \equiv 0, \tag{2.20}$$

and study the hydrodynamic interactions which are set up between the spheres when they move (see, however, in this connection section 7).

3. Induced force multipoles

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

$$\boldsymbol{u}_i = -\sum_j \boldsymbol{\mu}_{ij}^{\mathrm{TT}} \cdot \boldsymbol{K}_j - \sum_j \boldsymbol{\mu}_{ij}^{\mathrm{TR}} \cdot \boldsymbol{T}_j, \tag{3.1}$$

$$\boldsymbol{\omega}_i = -\sum_j \boldsymbol{\mu}_{ij}^{\mathrm{RT}} \cdot \boldsymbol{K}_j - \sum_j \boldsymbol{\mu}_{ij}^{\mathrm{RR}} \cdot \boldsymbol{T}_j. \tag{3.2}$$

Here $\boldsymbol{\mu}_{ij}^{\mathrm{TT}}$ is the translational mobility tensor, $\boldsymbol{\mu}_{ij}^{\mathrm{RR}}$ the rotational mobility tensor, and the matrices $\boldsymbol{\mu}_{ij}^{\mathrm{TR}}$ and $\boldsymbol{\mu}_{ij}^{\mathrm{RT}}$ couple translational and rotational motion. The various mobility tensors must obey the following (Onsager) symmetry relations[1])

$$\boldsymbol{\mu}_{ij}^{\mathrm{TT}} = \tilde{\boldsymbol{\mu}}_{ji}^{\mathrm{TT}}, \quad \boldsymbol{\mu}_{ij}^{\mathrm{RT}} = \tilde{\boldsymbol{\mu}}_{ji}^{\mathrm{TR}}, \quad \boldsymbol{\mu}_{ij}^{\mathrm{RR}} = \tilde{\boldsymbol{\mu}}_{ji}^{\mathrm{RR}}, \tag{3.3}$$

where $\tilde{\boldsymbol{\mu}}_{ij}$ is the transposed of $\boldsymbol{\mu}_{ij}$.

It is the aim of the present analysis to calculate these mobility tensors, using eq. (2.19), up to a given order of approximation in a series expansion in powers of inverse distances between the spheres. In paper I this program was carried out for the translational mobility tensor alone up to an order of approximation in which only hydrodynamic interactions between two spheres contribute. Here we intend to carry the analysis for all mobility tensors to higher order, in which case also interactions between three and four spheres must be taken into account. In the next section we shall derive for this purpose a hierarchy of equations for the force multipoles induced in the spheres. These force multipoles are the coefficients in an expansion of the induced force density $\boldsymbol{F}_i(\boldsymbol{k})$ in powers of the wave-vector $\boldsymbol{k}$:

$$\boldsymbol{F}_i(\boldsymbol{k}) = \sum_{p=0}^{\infty} (-ia_i k)^p \boldsymbol{\Omega}^p \odot \boldsymbol{M}_i^{(p+1)}, \tag{3.4}$$

where

$$\boldsymbol{M}_i^{(p+1)} \equiv (i/a_i)^p (p\,!)^{-1} \left[\frac{\partial^p}{\partial \boldsymbol{k}^p} \boldsymbol{F}_i(\boldsymbol{k})\right]_{\boldsymbol{k}=0}$$
$$= a_i^{-p} (p\,!)^{-1} \int \mathrm{d}\boldsymbol{r} (\boldsymbol{r} - \boldsymbol{R}_i)^p \boldsymbol{F}_i(\boldsymbol{r}) = (p\,!)^{-1} \int \mathrm{d}\hat{\boldsymbol{n}}_i \hat{\boldsymbol{n}}_i^p \boldsymbol{f}_i(\hat{\boldsymbol{n}}_i). \tag{3.5}$$

In the last member, use has been made of the property (2.11) of the induced forces. The notation $\boldsymbol{b}^p$ denotes a p-fold ordered product of a vector $\boldsymbol{b}$ (e.g. $\boldsymbol{b}^3 = \boldsymbol{bbb}$), while the dot $\odot$ in eq. (3.4) denotes a full p-fold contraction between the tensor $\boldsymbol{\Omega}^p$ and $\boldsymbol{M}_i^{(p+1)}$, e.g.

$$(\boldsymbol{\Omega}^3 \odot \boldsymbol{M}_i^{(4)})_\alpha = \sum_{\beta\gamma\delta} \Omega_\beta \Omega_\gamma \Omega_\delta M_{i,\delta\gamma\beta\alpha}^{(4)}. \tag{3.6}$$

We note that the pth force multipole $\boldsymbol{M}_i^{(p+1)}$ is a tensor of rank $p+1$, which is symmetric in its first p indices and which has the dimension of a force. Since $\hat{n}_i \cdot \hat{n}_i = 1$, each multipole $\boldsymbol{M}_i^{(p+1)}$ reduces to a multipole $\boldsymbol{M}_i^{(p-1)}$ by taking its trace with respect to any pair of its first p indices. In this way multipoles of higher rank contain contributions of multipoles of lower rank. It is therefore convenient to introduce irreducible multipoles $\boldsymbol{F}^{(p+1)}$ which are traceless in any pair of their first p indices. These multipoles are defined as

$$\boldsymbol{F}_i^{(p+1)} \equiv (p\,!)^{-1} \int \mathrm{d}\hat{n}_i \overline{\hat{n}_i^p} f_i(\hat{n}_i) = (i/a_i)^p (p\,!)^{-1} \left[\frac{\overline{\partial^p}}{\partial \boldsymbol{k}^p} \boldsymbol{F}_i(\boldsymbol{k})\right]_{k=0} (p \geq 0). \tag{3.7}$$

Here $\overline{\boldsymbol{b}^p}$ is the irreducible tensor of rank p, i.e. the tensor traceless and symmetric in any pair of its indices, constructed with the vector $\boldsymbol{b}$. For $p = 1, 2, 3$, one has* (see e.g. ref. 14)

$$\overline{b_\alpha} = b_\alpha, \quad \overline{b_\alpha b_\beta} = b_\alpha b_\beta - \tfrac{1}{3}\delta_{\alpha\beta} b^2,$$

$$\overline{b_\alpha b_\beta b_\gamma} = b_\alpha b_\beta b_\gamma - \tfrac{1}{5}(\delta_{\alpha\beta} b_\gamma + \delta_{\alpha\gamma} b_\beta + \delta_{\beta\gamma} b_\alpha) b^2. \tag{3.8}$$

For the first three multipoles, one has the relations

$$\boldsymbol{F}_i^{(1)} = \boldsymbol{M}_i^{(1)}, \qquad \boldsymbol{F}_i^{(2)} = \boldsymbol{M}_i^{(2)},$$

$$F_{i,\alpha\beta\gamma}^{(3)} = M_{i,\alpha\beta\gamma}^{(3)} - \tfrac{1}{3}\delta_{\alpha\beta} \sum_\delta M_{i,\delta\delta\gamma}^{(3)} = M_{i,\alpha\beta\gamma}^{(3)} - \tfrac{1}{6}\delta_{\alpha\beta} F_{i,\gamma}^{(1)}. \tag{3.9}$$

Note that according to eqs. (2.11), (2.12) and (3.7), one has

$$\boldsymbol{K}_i = -\boldsymbol{F}_i^{(1)}. \tag{3.10}$$

Similarly, it follows from eqs. (2.11), (2.13) and (3.7) that the hydrodynamic torque $\boldsymbol{T}_i$ is related to the antisymmetric part $\boldsymbol{F}_i^{(2a)}$ of $\boldsymbol{F}_i^{(2)}$ according to

$$\boldsymbol{T}_i = a_i \boldsymbol{\epsilon} : \boldsymbol{F}_i^{(2a)}, \tag{3.11}$$

where $\boldsymbol{\epsilon}$ is the Levi-Civita tensor. The inverse of eq. (3.11) is

$$\boldsymbol{F}_i^{(2a)} = -(2a_i)^{-1} \boldsymbol{\epsilon} \cdot \boldsymbol{T}_i, \tag{3.12}$$

which may be checked using the identity

$$\boldsymbol{\epsilon} : \boldsymbol{\epsilon} = -2\boldsymbol{1}. \tag{3.13}$$

The induced force $\boldsymbol{F}_i(\boldsymbol{k})$ can alternatively be expanded in terms of the irreducible multipoles $\boldsymbol{F}_i^{(p+1)}$. Using properties of the Legendre polynomials,

*The normalisation of the irreducible part of the tensor of rank p has been chosen such that the coefficient of the term $b_{\alpha_1} b_{\alpha_2} \ldots b_{\alpha_p}$ in the expression for $\overline{b_{\alpha_1} b_{\alpha_2} \ldots b_{\alpha_p}}$ is equal to 1.

we show in appendix A that one has

$$F_i(k\Omega) = \sum_{p=0}^{\infty} (2p+1)!!(i/a_i)^p \left(\overline{\frac{\partial^p}{\partial k^p}} \frac{\sin ka_i}{ka_i}\right) \odot F_i^{(p+1)}, \tag{3.14}$$

where $(2p+1)!! = 1 \cdot 3 \cdot 5 \cdots (2p-1) \cdot (2p+1)$. In appendix A we also show that

$$(2p+1)!!a_i^{-p} \left(\overline{\frac{\partial^p}{\partial k^p}} \frac{\sin ka_i}{ka_i}\right) = (-1)^p \overline{\Omega^p} a_i^p k^p (1 - a_i^2 k^2/(4p+6) + \mathcal{O}(a_i k)^4). \tag{3.15}$$

From this equation it is clear that the expansion (3.14) is essentially still an expansion in powers of k; however, each irreducible multipole $F_i^{(p+1)}$ is now multiplied by k^p times a power series in k^2.

We finally list a number of identities for the multipoles. From eq. (2.11) and the fact that the function f_i in this equation is bounded, it follows that for complex values of k

$$|F_i(k\Omega)| \leq M\, e^{|k''|a_i}, \tag{3.16}$$

where k'' is the imaginary part of k. One can then show by complex integration that the following integral relation holds*

$$F_i^{(1)} = \frac{1}{\pi} \int_{-\infty}^{+\infty} dk \frac{\sin a_i k}{k} F_i(k\Omega). \tag{3.17}$$

Analogously, one has

$$\Omega \cdot F_i^{(2)} = \frac{i}{a_i \pi} \int_{-\infty}^{+\infty} dk \frac{\sin a_i k}{k} \frac{\partial F_i(k\Omega)}{\partial k}, \tag{3.18}$$

$$\overline{\Omega\Omega} : F_i^{(3)} = -\frac{1}{2a_i^2 \pi} \int_{-\infty}^{+\infty} dk \frac{\sin a_i k}{k} \left(\frac{\partial^2}{\partial k^2} + \frac{1}{3} a_i^2\right) F_i(k\Omega). \tag{3.19}$$

These relations, which can be generalized to higher order, and which can also be derived with the aid of the expansion (3.14), will be helpful in establishing the hierarchy of equations for the force multipoles.

4. Evaluation of velocity surface moments

In order to derive for the force multipoles induced in the spheres the desired hierarchy of equations we shall determine so-called surface moments

*To derive this relation one has to replace a_i in eq. (3.17) by $a_i + \epsilon$ and take the limit $\epsilon \downarrow 0$; cf. the footnote in section 2.

of the fluid velocity field. These moments are appropriate averages over the surfaces of the spheres. If one uses in the evaluation of these averages eq. (2.19) as well as the boundary condition (2.9) and the multipole expansion (3.11) one obtains for each force multipole $\boldsymbol{F}_i^{(n)}$ an equation which relates this multipole to *all* the multipoles induced in the *other* spheres. From the lowest (zeroth) order moment one obtains an equation which relates the total force acting on a sphere not only to the force multipoles induced in the other spheres but also to the velocity of that particular sphere. Similarly the next (first) surface moment yields an analogous equation which involves also the angular velocity of a sphere. Higher moments lead to equations which couple solely force multipole moments.

To illustrate the scheme we shall derive here the first three equations of the hierarchy. Using the identities

$$\overline{\overline{\hat{n}_i^p}\boldsymbol{v}(\boldsymbol{r})}^{S_i} \equiv \frac{1}{4\pi a_i^2}\, a_i^{-p} \int \mathrm{d}\boldsymbol{r}\,\overline{(\boldsymbol{r}-\boldsymbol{R}_i)^p}\,\boldsymbol{v}(\boldsymbol{r})\,\delta(|\boldsymbol{r}-\boldsymbol{R}_i|-a_i)$$
$$= \frac{(-\mathrm{i}/a_i)^p}{(2\pi)^3} \int \mathrm{d}\boldsymbol{k}\left(\overline{\frac{\partial^p}{\partial \boldsymbol{k}^p}}\,\frac{\sin ka_i}{ka_i}\right) \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_i}\boldsymbol{v}(\boldsymbol{k}) \tag{4.1}$$

and

$$\frac{\partial}{\partial \boldsymbol{k}} = \boldsymbol{\Omega}\,\frac{\partial}{\partial k} + \frac{1}{k}(\boldsymbol{1}-\boldsymbol{\Omega}\boldsymbol{\Omega})\cdot\frac{\partial}{\partial \boldsymbol{\Omega}}, \tag{4.2}$$

we find for the first three surface moments for the ith sphere of the fluid velocity field

$$\overline{\boldsymbol{v}(\boldsymbol{r})}^{S_i} = \frac{1}{(2\pi)^3}\int \mathrm{d}\boldsymbol{k}\,\boldsymbol{v}(\boldsymbol{k})\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_i}\,\frac{\sin ka_i}{ka_i}, \tag{4.3}$$

$$3!!\,\overline{\hat{\boldsymbol{n}}_i\boldsymbol{v}(\boldsymbol{r})}^{S_i} = \frac{3!!(-\mathrm{i}/a_i)}{(2\pi)^3}\int \mathrm{d}\boldsymbol{k}\,\boldsymbol{\Omega}\boldsymbol{v}(\boldsymbol{k})\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_i}\left(\frac{\partial}{\partial k}\,\frac{\sin ka_i}{ka_i}\right), \tag{4.4}$$

$$5!!\,\overline{\overline{\hat{\boldsymbol{n}}_i\hat{\boldsymbol{n}}_i}\boldsymbol{v}(\boldsymbol{r})}^{S_i} = \frac{5!!(-\mathrm{i}/a_i)^2}{(2\pi)^3}\int \mathrm{d}\boldsymbol{k}\left(\overline{\frac{\partial^2}{\partial \boldsymbol{k}^2}}\,\frac{\sin ka_i}{ka_i}\right)\boldsymbol{v}(\boldsymbol{k})\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_i}$$
$$= -\frac{3\cdot 5!!}{2a_i^2(2\pi)^3}\int \mathrm{d}\boldsymbol{k}\,\overline{\boldsymbol{\Omega}\boldsymbol{\Omega}}\boldsymbol{v}(\boldsymbol{k})\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_i}\left[\left(\frac{\partial^2}{\partial k^2}+\frac{1}{3}a_i^2\right)\frac{\sin ka_i}{ka_i}\right]. \tag{4.5}$$

The numerical factors 3!! and 5!! in eqs. (4.4) and (4.5) have been introduced for convenience. In the last number of eq. (4.5), use was made of the identity

$$\overline{\frac{\partial}{\partial \boldsymbol{k}}\frac{\partial}{\partial \boldsymbol{k}}}\,\frac{\sin ka_i}{ka_i} = \frac{3}{2}\,\overline{\boldsymbol{\Omega}\boldsymbol{\Omega}}\left(\frac{\partial^2}{\partial k^2}+\frac{1}{3}a_i^2\right)\frac{\sin ka_i}{ka_i}, \tag{4.6}$$

which follows from eq. (4.2) and the fact that

$$k^{-1}\left(\frac{\partial}{\partial k}\frac{\sin ka_i}{ka_i}\right) = -\frac{1}{2}\left(\frac{\partial^2}{\partial k^2} + a_i^2\right)\frac{\sin ka_i}{ka_i}. \tag{4.7}$$

We shall now consider these first three moments in succession.

(i) Substituting in eq. (4.3) the formal solution (2.19) and using the boundary condition (2.9) one obtains the following set of N equations

$$u_i = (6\pi\eta a_i)^{-1}\frac{3a_i}{8\pi^2}\int \mathrm{d}\boldsymbol{\Omega}\int_{-\infty}^{+\infty} \mathrm{d}k(\mathbf{1}-\boldsymbol{\Omega\Omega})\frac{\sin ka_i}{ka_i}\boldsymbol{F}_i(k\boldsymbol{\Omega})$$

$$+ (6\pi\eta a_i)^{-1}\sum_{j\neq i}\frac{3a_i}{8\pi^2}\int \mathrm{d}\boldsymbol{\Omega}\int_{-\infty}^{+\infty} \mathrm{d}k(\mathbf{1}-\boldsymbol{\Omega\Omega})\frac{\sin ka_i}{ka_i}\,\mathrm{e}^{-\mathrm{i}kR_{ij}\boldsymbol{\Omega}\cdot\hat{r}_{ij}}\boldsymbol{F}_j(k\boldsymbol{\Omega}). \tag{4.8}$$

Here $R_{ij} \equiv |\boldsymbol{R}_i - \boldsymbol{R}_j|$ is the distance between the centres of sphere i and j and $\hat{r}_{ij} \equiv (\boldsymbol{R}_j - \boldsymbol{R}_i)/R_{ij}$ is a unit vector pointing from sphere i to sphere j. Use has also been made of the fact that the integrands in eq. (4.8) are invariant under the transformation $k \to -k$, $\boldsymbol{\Omega} \to -\boldsymbol{\Omega}$*. If we now use the identity (3.17) in the first integral at the right hand side of eq. (4.8) as well as the multipole expansion (3.14) in the other integrals, we obtain a set of equations of the form

$$u_i = (6\pi\eta a_i)^{-1}\boldsymbol{F}_i^{(1)} + (6\pi\eta a_i)^{-1}\sum_{j\neq i}\sum_{m=1}^{\infty}\boldsymbol{A}_{ij}^{(1,m)} \odot \boldsymbol{F}_j^{(m)}. \tag{4.9}$$

The dimensionless quantities $\boldsymbol{A}_{ij}^{(1,m)}$, $j \neq i$, which will be called connectors since they connect force multipoles $\boldsymbol{F}_i^{(1)}$ to force multipoles $\boldsymbol{F}_j^{(m)}$, are tensors of rank $m+1$. The dot $\odot$ denotes here an m-fold contraction between the last m indices of $\boldsymbol{A}^{(1,m)}$ and the m indices of $\boldsymbol{F}^{(m)}$ with the convention that the *last* index of $\boldsymbol{A}^{(1,m)}$ is contracted with the *first* of $\boldsymbol{F}^{(m)}$, etc. Cf. in this connection also eq. (3.6). The general expression for the connector $\boldsymbol{A}^{(1,m)}$ is**

$$\boldsymbol{A}_{ij}^{(1,m)} = \frac{3a_i(2m-1)!!}{8\pi^2}\left(\frac{\mathrm{i}}{a_j}\right)^{m-1}\int \mathrm{d}\boldsymbol{\Omega}\int_{-\infty}^{+\infty} \mathrm{d}k(\mathbf{1}-\boldsymbol{\Omega\Omega})\left(\overline{\frac{\partial^{m-1}}{\partial k^{m-1}}}\frac{\sin ka_j}{ka_j}\right)$$

$$\times\frac{\sin ka_i}{ka_i}\,\mathrm{e}^{-\mathrm{i}kR_{ij}\boldsymbol{\Omega}\cdot\hat{r}_{ij}}. \tag{4.10}$$

*It should be mentioned that in paper I, an erroneous argument was used regarding the properties of eq. (4.8) under inversion. This erroneous argument does, however, not invalidate the analysis up to the order considered in that paper, but would have affected an analysis to higher order.

** By convention all connectors are zero for $j = i$. All expressions given for connectors are only valid for $j \neq i$.

Properties of these connectors will be discussed in more detail in section 5.

It should be noted that if there is only one sphere, the second term in the right hand side of eq. (4.9) is absent; in view of eq. (3.10), the resulting equation then reduces to Stokes' law for a single sphere.

(ii) Consider now eq. (4.4). We again use the boundary condition (2.9) at the left hand side; the resulting surface average is easily evaluated. After substitution of the formal solution (2.19) in the right-hand side, we obtain the set of equations

$$a_i \boldsymbol{\epsilon} \cdot \boldsymbol{\omega}_i = -(6\pi\eta a_i)^{-1} \frac{3i3!!}{8\pi^2} \int \mathrm{d}\boldsymbol{\Omega} \int_{-\infty}^{+\infty} \mathrm{d}k \boldsymbol{\Omega}(\mathbf{1} - \boldsymbol{\Omega}\boldsymbol{\Omega}) \cdot \boldsymbol{F}_i(k\boldsymbol{\Omega}) \left(\frac{\partial}{\partial k} \frac{\sin ka_i}{ka_i}\right)$$

$$- (6\pi\eta a_i)^{-1} \sum_{j\neq i} \frac{3i3!!}{8\pi^2} \int \mathrm{d}\boldsymbol{\Omega} \int_{-\infty}^{+\infty} \mathrm{d}k \boldsymbol{\Omega}(\mathbf{1} - \boldsymbol{\Omega}\boldsymbol{\Omega}) \cdot \boldsymbol{F}_j(k\boldsymbol{\Omega})$$

$$\times \mathrm{e}^{-\mathrm{i}kR_{ij}\boldsymbol{\Omega}\cdot\hat{\boldsymbol{r}}_{ij}} \left(\frac{\partial}{\partial k} \frac{\sin ka_i}{ka_i}\right). \tag{4.11}$$

In this case, the identity (3.18) applies to the first term on the right hand side of eq. (4.11). Substituting again the force multipole expansion (3.14) into the other integrals, one now obtains equations of the form

$$a_i \boldsymbol{\epsilon} \cdot \boldsymbol{\omega}_i = -(6\pi\eta a_i)^{-1} \boldsymbol{B}^{(2,2)} : \boldsymbol{F}_i^{(2)} + (6\pi\eta a_i)^{-1} \sum_{j\neq i} \sum_{m=1}^{\infty} \boldsymbol{A}_{ij}^{(2,m)} \odot \boldsymbol{F}_j^{(m)}. \tag{4.12}$$

The explicit expressions for $\boldsymbol{B}^{(2,2)}$ and the connectors $\boldsymbol{A}^{(2,m)}$ are

$$\boldsymbol{B}^{(2,2)} = -\frac{3\cdot 3!!}{8\pi} \int \mathrm{d}\boldsymbol{\Omega}\boldsymbol{\Omega}(\mathbf{1} - \boldsymbol{\Omega}\boldsymbol{\Omega})\boldsymbol{\Omega}, \tag{4.13}$$

$$\boldsymbol{A}_{ij}^{(2,m)} = \frac{3a_i 3!!(2m-1)!!}{8\pi^2} \left(\frac{-\mathrm{i}}{a_i}\right)\left(\frac{\mathrm{i}}{a_j}\right)^{m-1} \int \mathrm{d}\boldsymbol{\Omega} \int_{-\infty}^{+\infty} \mathrm{d}k \boldsymbol{\Omega}(\mathbf{1} - \boldsymbol{\Omega}\boldsymbol{\Omega})$$

$$\times \left(\overline{\frac{\partial^{m-1}}{\partial k^{m-1}}} \frac{\sin ka_j}{ka_j}\right)\left(\frac{\partial}{\partial k} \frac{\sin ka_i}{ka_i}\right) \mathrm{e}^{-\mathrm{i}kR_{ij}\boldsymbol{\Omega}\cdot\hat{\boldsymbol{r}}_{ij}}. \tag{4.14}$$

Note that $\boldsymbol{B}^{(2,2)}$ as well as all connectors $\boldsymbol{A}^{(2,m)}$ are traceless in their first two indices. This property is related to the fact that in view of eq. (2.8)

$$\overline{\hat{\boldsymbol{n}}_i \cdot \boldsymbol{v}(\boldsymbol{r})}^{S_i} = \frac{1}{4\pi a_i^2} \int_{|\boldsymbol{r}-\boldsymbol{R}_i|\leq a_i} \mathrm{d}\boldsymbol{r} \boldsymbol{\nabla} \cdot \boldsymbol{v}(\boldsymbol{r}) = 0, \tag{4.15}$$

irrespective of the boundary conditions.

From eq. (4.13), we obtain for $\boldsymbol{B}^{(2,2)}$ the explicit expression (see appendix B for useful formulae in connection with the evaluation of this and similar integrals)

$$B^{(2,2)}_{\alpha\beta\gamma\delta} = -\tfrac{3}{10}(4\delta_{\alpha\delta}\delta_{\beta\gamma} - \delta_{\alpha\gamma}\delta_{\beta\delta} - \delta_{\alpha\beta}\delta_{\gamma\delta}). \tag{4.16}$$

The evaluation of the expressions for the first three connectors $\boldsymbol{A}^{(2,m)}_{ij}$ will be deferred to sections 5 and 6.

It is convenient to write

$$\boldsymbol{B}^{(2,2)} = \boldsymbol{B}^{(2s,2s)} + \boldsymbol{B}^{(2a,2a)}, \tag{4.17}$$

with

$$B^{(2s,2s)}_{\alpha\beta\gamma\delta} = -\tfrac{9}{10}(\tfrac{1}{2}\delta_{\alpha\delta}\delta_{\beta\gamma} + \tfrac{1}{2}\delta_{\alpha\gamma}\delta_{\beta\delta} - \tfrac{1}{3}\delta_{\alpha\beta}\delta_{\gamma\delta}) \equiv -\tfrac{9}{10}\boldsymbol{\Delta}_{\alpha\beta\gamma\delta} \tag{4.18}$$

$$B^{(2a,2a)}_{\alpha\beta\gamma\delta} = -\tfrac{3}{2}(\tfrac{1}{2}\delta_{\alpha\delta}\delta_{\beta\gamma} - \tfrac{1}{2}\delta_{\alpha\gamma}\delta_{\beta\delta}). \tag{4.19}$$

$\boldsymbol{B}^{(2s,2s)}$ is (traceless) symmetric and $\boldsymbol{B}^{(2a,2a)}$ is antisymmetric in both the first and the last pair of indices, respectively. The tensor $\boldsymbol{\Delta}$ is the tensor which projects out the irreducible part of a tensor of rank 2. We shall also split the connectors $\boldsymbol{A}^{(2,m)}$ into terms which are traceless symmetric and antisymmetric in the first two indices, $\boldsymbol{A}^{(2,m)} = \boldsymbol{A}^{(2s,m)} + \boldsymbol{A}^{(2a,m)}$. We also split the multiples $\boldsymbol{F}^{(2)}$ according to* $\boldsymbol{F}^{(2)} = \boldsymbol{F}^{(2s)} + \boldsymbol{F}^{(2a)} + \tfrac{1}{3}\boldsymbol{1}\,\mathrm{Tr}\,\boldsymbol{F}^{(2)}$, where $\boldsymbol{F}^{(2s)}$ and $\boldsymbol{F}^{(2a)}$ are the traceless symmetric and antisymmetric part of $\boldsymbol{F}^{(2)}$ respectively.

Eq. (4.12) is essentially an equation for a tensor of rank 2. Writing the equations for the symmetric and antisymmetric part separately, we obtain for the symmetric part

$$\boldsymbol{B}^{(2s,2s)}:\boldsymbol{F}^{(2)}_i = -\tfrac{9}{10}\boldsymbol{F}^{(2s)}_i = \sum_{j\neq i}\sum_{m=1}^{\infty}\boldsymbol{A}^{(2s,m)}_{ij}\odot\boldsymbol{F}^{(m)}_j. \tag{4.20}$$

If on the other hand we contract eq. (4.12) with the Levi-Civita tensor $\boldsymbol{\epsilon}$ and use the fact that

$$\boldsymbol{B}^{(2a,2a)}:\boldsymbol{F}^{(2)}_i = -\tfrac{3}{2}\boldsymbol{F}^{(2a)}_i, \tag{4.21}$$

we obtain

$$-2a_i\boldsymbol{\omega}_i = (4\pi\eta a_i)^{-1}\boldsymbol{\epsilon}:\boldsymbol{F}^{(2a)}_i + (6\pi\eta a_i)^{-1}\sum_{j\neq i}\sum_{m=1}^{\infty}\boldsymbol{\epsilon}:\boldsymbol{A}^{(2a,m)}_{ij}\odot\boldsymbol{F}^{(m)}_j. \tag{4.22}$$

Using eq. (3.11), this relation becomes

$$\boldsymbol{\omega}_i = -(8\pi\eta a_i^3)^{-1}\boldsymbol{T}_i - (12\pi\eta a_i^2)^{-1}\sum_{j\neq i}\sum_{m=1}\boldsymbol{\epsilon}:\boldsymbol{A}^{(2a,m)}_{ij}\odot\boldsymbol{F}^{(m)}_j. \tag{4.23}$$

*It should be noted that $\boldsymbol{F}^{(2)}$ itself is not traceless. However, its trace does not play a role in the analysis, since it is related only to the pressure and does not contribute to the mobility.

For a single sphere, this equation reduces to the well-known result for the rotational friction.

(iii) Finally one obtains in a similar way from eq. (4.5) and (2.19)

$$0 = (6\pi\eta a_i)^{-1} \frac{9 \cdot 5!!(-\mathrm{i}/a_i)^2}{16\pi^2} \int \mathrm{d}\boldsymbol{\Omega} \int_{-\infty}^{+\infty} \mathrm{d}k \overline{\boldsymbol{\Omega}\boldsymbol{\Omega}}(\mathbf{1} - \boldsymbol{\Omega}\boldsymbol{\Omega}) \cdot \boldsymbol{F}_i(k\boldsymbol{\Omega})$$
$$\times \left(\frac{\partial^2}{\partial k^2} + \frac{1}{3} a_i^2\right) \frac{\sin k a_i}{k a_i}$$
$$+ \sum_{j \neq i} (6\pi\eta a_i)^{-1} \frac{9 \cdot 5!!(-\mathrm{i}/a_i)^2}{16\pi^2} \int \mathrm{d}\boldsymbol{\Omega} \int_{-\infty}^{+\infty} \mathrm{d}k \overline{\boldsymbol{\Omega}\boldsymbol{\Omega}}(\mathbf{1} - \boldsymbol{\Omega}\boldsymbol{\Omega}) \cdot \boldsymbol{F}_j(k\boldsymbol{\Omega})$$
$$\times \mathrm{e}^{-\mathrm{i}kR_{ij}\boldsymbol{\Omega} \cdot \hat{r}_{ij}} \left(\frac{\partial^2}{\partial k^2} + \frac{1}{3} a_i^2\right) \frac{\sin k a_i}{k a_i}. \tag{4.24}$$

With the aid of the identity (3.17) and the multipole expansion (3.14), we then get the set of equations

$$\boldsymbol{B}^{(3,3)} \odot \boldsymbol{F}_i^{(3)} = \sum_{j \neq i} \sum_{m=1}^{\infty} \boldsymbol{A}_{ij}^{(3,m)} \odot \boldsymbol{F}_j^{(m)}, \tag{4.25}$$

where $\boldsymbol{B}^{(3,3)}$ and the connectors $\boldsymbol{A}^{(3,m)}$ are given by

$$\boldsymbol{B}^{(3,3)} = -\frac{9 \cdot 5!!}{8\pi} \int \mathrm{d}\boldsymbol{\Omega} \overline{\boldsymbol{\Omega}\boldsymbol{\Omega}}(\mathbf{1} - \boldsymbol{\Omega}\boldsymbol{\Omega}) \overline{\boldsymbol{\Omega}\boldsymbol{\Omega}}, \tag{4.26}$$

$$\boldsymbol{A}_{ij}^{(3,m)} = \frac{9 a_i 5!!(2m-1)!!}{16\pi^2} \left(\frac{-\mathrm{i}}{a_i}\right)^2 \left(\frac{\mathrm{i}}{a_j}\right)^{m-1} \int \mathrm{d}\boldsymbol{\Omega} \int_{-\infty}^{+\infty} \mathrm{d}k \overline{\boldsymbol{\Omega}\boldsymbol{\Omega}}(\mathbf{1} - \boldsymbol{\Omega}\boldsymbol{\Omega})$$
$$\times \left(\frac{\overline{\partial^{m-1}}}{\partial \boldsymbol{k}^{m-1}} \frac{\sin k a_j}{k a_j}\right) \left[\left(\frac{\partial^2}{\partial k^2} + \frac{1}{3} a_i^2\right) \frac{\sin k a_i}{k a_i}\right] \mathrm{e}^{-\mathrm{i}kR_{ij}\boldsymbol{\Omega} \cdot \hat{r}_{ij}}. \tag{4.27}$$

The above scheme may be continued by considering irreducible surface moments of higher order, which are all zero,

$$(2n-1)!!\overline{\overline{\hat{n}_i^n}\boldsymbol{v}(\boldsymbol{r})}^{S_i} = (2n-1)!!\overline{\overline{\hat{n}_i^n}(\boldsymbol{u}_i + a_i \boldsymbol{\omega}_i \wedge \hat{n}_i)}^{S_i} = 0, \quad n \geq 3. \tag{4.28}$$

The fact that all these surface moments vanish follows from the general property (see eq. (A.8))

$$\overline{\overline{\hat{n}_i^n}\,\overline{\hat{n}_i^m}}^{S_i} = 0, \quad \text{for } n \neq m. \tag{4.29}$$

If, on the other hand, the surface moments are evaluated with the aid of the formal solution (2.19), one obtains along similar lines as above, equations of the form

$$\boldsymbol{B}^{(n,n)} \odot \boldsymbol{F}_i^{(n)} = \sum_{m=1}^{\infty} \sum_{j \neq i} \boldsymbol{A}_{ij}^{(n,m)} \odot \boldsymbol{F}_j^{(m)}, \quad n \geq 4. \tag{4.30}$$

The fact that on the left hand side of this equation only the nth multipole appears (just as in the previous case, eq. (4.25)) is shown in appendix C. The general expressions for $\boldsymbol{B}^{(n,n)}$ and the connectors $\boldsymbol{A}_{ij}^{(n,m)}$ are

$$\boldsymbol{B}^{(n,n)} = -\left(\frac{(2n-1)!!}{a_i^{n-1}}\right)^2 \frac{3a_i}{8\pi^2} \int \mathrm{d}\boldsymbol{\Omega} \int_{-\infty}^{+\infty} \mathrm{d}k \times \left(\overline{\frac{\partial^{n-1}}{\partial \boldsymbol{k}^{n-1}}} \frac{\sin ka_i}{ka_i}\right)(\boldsymbol{1} - \boldsymbol{\Omega\Omega})\left(\overline{\frac{\partial^{n-1}}{\partial \boldsymbol{k}^{n-1}}} \frac{\sin ka_i}{ka_i}\right), \tag{4.31}$$

$$\boldsymbol{A}_{ij}^{(n,m)} = \frac{3a_i(2n-1)!!(2m-1)!!}{8\pi^2} \left(\frac{-\mathrm{i}}{a_i}\right)^{n-1} \left(\frac{\mathrm{i}}{a_j}\right)^{m-1} \int \mathrm{d}\boldsymbol{\Omega} \int_{-\infty}^{+\infty} \mathrm{d}k \, \mathrm{e}^{-\mathrm{i}kR_{ij}\boldsymbol{\Omega}\cdot\hat{\boldsymbol{r}}_{ij}} \times \left(\overline{\frac{\partial^{n-1}}{\partial \boldsymbol{k}^{n-1}}} \frac{\sin ka_i}{ka_i}\right)(\boldsymbol{1} - \boldsymbol{\Omega\Omega})\left(\overline{\frac{\partial^{m-1}}{\partial \boldsymbol{k}^{m-1}}} \frac{\sin ka_j}{ka_j}\right). \tag{4.32}$$

These equations are in fact valid for all n and m. One can verify using also eq. (4.6) that eq. (4.31) reduces to eqs. (4.13) and (4.26) for $n = 2$ and 3, respectively, and that $\boldsymbol{B}^{(1,1)} = -\boldsymbol{1}$. Furthermore, all previously derived connectors are contained in formula (4.32). Note that the $\boldsymbol{B}^{(n,n)}$ are tensors of rank $2n$, and the connectors $\boldsymbol{A}^{(n,m)}$ tensors of rank $n + m$.

It is clear from expression (4.31) that the tensor $\boldsymbol{B}^{(n,n)}$ satisfies the symmetry relation

$$\widetilde{\boldsymbol{B}^{(n,n)}} = \boldsymbol{B}^{(n,n)}, \tag{4.33}$$

where $\tilde{\boldsymbol{C}}$ is a generalized transposed of a tensor $\boldsymbol{C}$ of arbitrary rank p defined by

$$(\tilde{\boldsymbol{C}})_{\alpha_1\alpha_2\ldots\alpha_{p-1}\alpha_p} = (\boldsymbol{C})_{\alpha_p\alpha_{p-1}\ldots\alpha_2\alpha_1}. \tag{4.34}$$

On the other hand, using the fact that $\hat{r}_{ij} = -\hat{r}_{ji}$, one verifies that the connectors satisfy the symmetry relation

$$a_j\widetilde{\boldsymbol{A}_{ij}^{(n,m)}} = a_i\boldsymbol{A}_{ji}^{(m,n)}. \tag{4.35}$$

The numerical factor $(2n-1)!!$ was introduced in eq. (4.28) so as to ensure that the connectors satisfy symmetry relations of the form (4.35) (see also eqs. (4.3)–(4.5)). It will turn out that the symmetry (3.3) of the mobilities is within the present scheme a direct consequence of the symmetries (4.33) and (4.35).

5. General properties of connectors and mobilities

In order to summarize the set of equations derived in the previous section for the induced force multipoles, we decompose connectors $\mathbf{A}^{(n,2)}$ into terms which are traceless symmetric and antisymmetric in the last two indices, $\mathbf{A}^{(n,2s)}$ and $\mathbf{A}^{(n,2a)}$ respectively. In addition we had defined in the previous section a similar decomposition for $\mathbf{A}^{(2,m)}$. We therefore have

$$\mathbf{A}_{ij}^{(2,m)} = \mathbf{A}_{ij}^{(2s,m)} + \mathbf{A}_{ij}^{(2a,m)}, \quad \mathbf{A}_{ij}^{(n,2)} = \mathbf{A}_{ij}^{(n,2s)} + \mathbf{A}_{ij}^{(n,2a)}. \tag{5.1}$$

With the help of eqs. (3.10), (3.12) and (5.1), eqs. (4.9), (4.20), (4.23), (4.25) and (4.30) can then be written in the form

$$6\pi\eta a_i \boldsymbol{u}_i = -\boldsymbol{K}_i - \sum_{j\neq i} \mathbf{A}_{ij}^{(1,1)} \cdot \boldsymbol{K}_j - \sum_{j\neq i} (2a_j)^{-1}(\mathbf{A}_{ij}^{(1,2a)} : \boldsymbol{\epsilon}) \cdot \boldsymbol{T}_j + \sum_{j\neq i} \sum_{m=2}^{\infty}{}' \mathbf{A}_{ij}^{(1,m)} \odot \boldsymbol{F}_j^{(m)}, \tag{5.2}$$

$$8\pi\eta a_i^3 \boldsymbol{\omega}_i = \tfrac{2}{3} a_i \sum_{j\neq i} \boldsymbol{\epsilon} : \mathbf{A}_{ij}^{(2a,1)} \cdot \boldsymbol{K}_j - \boldsymbol{T}_i + \frac{a_i}{3} \sum_{j\neq i} (a_j^{-1} \boldsymbol{\epsilon} : \mathbf{A}_{ij}^{(2a,2a)} : \boldsymbol{\epsilon}) \cdot \boldsymbol{T}_j - \tfrac{2}{3} a_i \sum_{j\neq i} \sum_{m=2}^{\infty}{}' \boldsymbol{\epsilon} : \mathbf{A}_{ij}^{(2a,m)} \odot \boldsymbol{F}_j^{(m)}, \tag{5.3}$$

$$\mathbf{B}^{(2s,2s)} : \boldsymbol{F}_i^{(2s)} = -\sum_{j\neq i} \mathbf{A}_{ij}^{(2s,1)} \cdot \boldsymbol{K}_j - \sum_{j\neq i} ((2a_j)^{-1} \mathbf{A}_{ij}^{(2s,2a)} : \boldsymbol{\epsilon})) \cdot \boldsymbol{T}_j + \sum_{j\neq i} \sum_{m=2}^{\infty}{}' \mathbf{A}_{ij}^{(2s,m)} \odot \boldsymbol{F}_j^{(m)}, \tag{5.4}$$

$$\mathbf{B}^{(n,n)} \odot \boldsymbol{F}_i^{(n)} = -\sum_{j\neq i} \mathbf{A}_{ij}^{(n,1)} \cdot \boldsymbol{K}_j - \sum_{j\neq i} ((2a_j)^{-1} \mathbf{A}_{ij}^{(n,2a)} : \boldsymbol{\epsilon}) \cdot \boldsymbol{T}_j + \sum_{j\neq i} \sum_{m=2}^{\infty}{}' \mathbf{A}_{ij}^{(n,m)} \odot \boldsymbol{F}_j^{(m)}, \quad n \geq 3. \tag{5.5}$$

In these equations, $\Sigma_{m=2}^{\prime\infty}$ denotes a summation over all integer values $m \geq 2$, with the proviso that for $m = 2$ only the connectors $\mathbf{A}^{(n,2s)}$ and multipoles $\boldsymbol{F}^{(2s)}$ are included in the summation, e.g.

$$\sum_{m=2}^{\infty}{}' \mathbf{A}_{ij}^{(1,m)} \odot \boldsymbol{F}_j^{(m)} \equiv \mathbf{A}_{ij}^{(1,2s)} : \boldsymbol{F}_j^{(2s)} + \sum_{m=3}^{\infty} \mathbf{A}_{ij}^{(1,m)} \odot \boldsymbol{F}_j^{(m)}. \tag{5.6}$$

The above equations summarize the hierarchy of equations we set out to derive. This hierarchy of equations will enable us to obtain expressions for the mobilities in terms of the interparticle distance.

We shall first study the behaviour of the connectors $\mathbf{A}_{ij}^{(n,m)}$ as a function of the interparticle distance R_{ij} in greater detail. Substituting the expansion (3.15) into eq. (4.32) we have

$$\boldsymbol{A}_{ij}^{(n,m)} = \frac{3a_i}{4\pi}(\mathrm{i}a_i)^{n-1}(-\mathrm{i}a_j)^{m-1}\int \mathrm{d}\boldsymbol{\Omega}\overline{\boldsymbol{\Omega}^{n-1}}(\boldsymbol{1}-\boldsymbol{\Omega\Omega})\overline{\boldsymbol{\Omega}^{m-1}}$$

$$\times\frac{1}{2\pi}\int_{-\infty}^{+\infty}\mathrm{d}k k^{n+m-2}\left[1-\left(\frac{a_i^2}{4n+2}+\frac{a_j^2}{4m+2}\right)k^2+\mathcal{O}(k^4)\right]\mathrm{e}^{-\mathrm{i}k\xi_{ij}R_{ij}}, \qquad (5.7)$$

where

$$\xi_{ij} = \hat{r}_{ij}\cdot\boldsymbol{\Omega} \qquad (5.8)$$

is the cosine of the angle between the unit vectors $\hat{r}_{ij}$ and $\boldsymbol{\Omega}$. In a reference frame in which the z-axis is parallel to the unit vector $\hat{r}_{ij}$, we may write $\mathrm{d}\boldsymbol{\Omega} = -\mathrm{d}\xi_{ij}\,\mathrm{d}\phi_{ij}$, where ϕ_{ij} is the other polar angle. We now make use of the identity

$$\frac{1}{2\pi}\int_{-1}^{+1}\mathrm{d}x x^p\int_{-\infty}^{+\infty}\mathrm{d}y y^q\,\mathrm{e}^{-\mathrm{i}xy} = \mathrm{i}^q\int_{-1}^{+1}\mathrm{d}x x^p\frac{\mathrm{d}^q}{\mathrm{d}x^q}\delta(x) = \delta_{pq}p\,!(-\mathrm{i})^p. \qquad (5.9)$$

It then follows that the terms of order k^4 and higher between square brackets in the integrand at the right-hand side of eq. (5.7) give vanishing contributions upon integration, since any element of the tensor $\overline{\boldsymbol{\Omega}^{n-1}}(\boldsymbol{1}-\boldsymbol{\Omega\Omega})\overline{\boldsymbol{\Omega}^{m-1}}$ will after integration over ϕ_{ij} be a polynomial in ξ_{ij} of which the term of highest order is proportional to ξ_{ij}^{n+m}. We therefore have the *exact* result

$$\boldsymbol{A}_{ij}^{(n,m)} = \boldsymbol{G}_{ij}^{(n,m)}R_{ij}^{-(n+m-1)} + \boldsymbol{H}_{ij}^{(n,m)}R_{ij}^{-(n+m+1)}, \qquad (5.10)$$

where

$$\boldsymbol{G}_{ij}^{(n,m)} = (-1)^{n-1}\frac{3}{4\pi}a_i^n a_j^{m-1}\int \mathrm{d}\boldsymbol{\Omega}\overline{\boldsymbol{\Omega}^{n-1}}(\boldsymbol{1}-\boldsymbol{\Omega\Omega})\overline{\boldsymbol{\Omega}^{m-1}}\frac{\partial^{n+m-2}}{\partial\xi_{ij}^{n+m-2}}\delta(\xi_{ij}), \qquad (5.11)$$

$$\boldsymbol{H}_{ij}^{(n,m)} = (-1)^{n-1}\frac{3}{4\pi}a_i^n a_j^{m-1}\left(\frac{a_i^2}{4n+2}+\frac{a_j^2}{4m+2}\right)$$

$$\times\int \mathrm{d}\boldsymbol{\Omega}\overline{\boldsymbol{\Omega}^{n-1}}(\boldsymbol{1}-\boldsymbol{\Omega\Omega})\overline{\boldsymbol{\Omega}^{m-1}}\frac{\partial^{n+m}}{\partial\xi_{ij}^{n+m}}\delta(\xi_{ij}). \qquad (5.12)$$

As discussed in appendix D, the tensor $\boldsymbol{H}^{(n,m)}$ can be evaluated in a simple manner. One finds

$$\boldsymbol{H}_{ij}^{(n,m)} = (-1)^m\tfrac{3}{2}(2n+2m-1)!!a_i^n a_j^{m-1}\left(\frac{a_i^2}{4n+2}+\frac{a_j^2}{4m+2}\right)\overline{\hat{r}_{ij}^{n+m}}, \qquad (5.13)$$

so that $\boldsymbol{H}^{(n,m)}$ is irreducible. It is clear from this result that the tensors $\boldsymbol{H}^{(2a,m)}$ and $\boldsymbol{H}^{(n,2a)}$ vanish. We have not been able to find as general a formula for the tensor $\boldsymbol{G}^{(n,m)}$. In section 6 we will give the explicit expressions of these tensors for $n, m \leqslant 3$, $n+m \leqslant 5$.

In view of eq. (5.10) and the fact that $\boldsymbol{B}^{(n,n)}$ is independent of the inter-particle distance R_{ij}, we note that eqs. (5.4) and (5.5) are essentially expansions for $\boldsymbol{F}_i^{(2s)}$ and $\boldsymbol{F}_i^{(n)}$ ($n \geqslant 3$) in terms of R_{ij}^{-1}, of which the dominant terms are of order R_{ij}^{-2} and R_{ij}^{-n}, respectively. We shall rewrite these equations in the form

$$\boldsymbol{F}_i^{(2s)} = -\sum_{j\neq i} \boldsymbol{B}^{(2s,2s)^{-1}} : \boldsymbol{A}_{ij}^{(2s,1)} \cdot \boldsymbol{K}_j - \sum_{j\neq i} ((2a_j)^{-1} \boldsymbol{B}^{(2s,2s)^{-1}} : \boldsymbol{A}_{ij}^{(2s,2a)} : \boldsymbol{\epsilon}) \cdot \boldsymbol{T}_j + \sum_{j\neq i} \sum_{m=2}^{\infty}{}' \boldsymbol{B}^{(2s,2s)^{-1}} : \boldsymbol{A}_{ij}^{(2s,m)} \odot \boldsymbol{F}_j^{(m)}, \tag{5.14}$$

$$\boldsymbol{F}_i^{(n)} = -\sum_{j\neq i} \boldsymbol{B}^{(n,n)^{-1}} \odot \boldsymbol{A}_{ij}^{(n,1)} \cdot \boldsymbol{K}_j - \sum_{j\neq i} ((2a_j)^{-1} \boldsymbol{B}^{(n,n)^{-1}} \odot \boldsymbol{A}_{ij}^{(n,2a)} : \boldsymbol{\epsilon}) \cdot \boldsymbol{T}_j + \sum_{j\neq i} \sum_{m=2}^{\infty}{}' \boldsymbol{B}^{(n,n)^{-1}} \odot \boldsymbol{A}_{ij}^{(n,m)} \odot \boldsymbol{F}_j^{(m)}, \quad n \geqslant 3, \tag{5.15}$$

where $\boldsymbol{B}^{(n,n)^{-1}}$ is the generalized inverse of $\boldsymbol{B}^{(n,n)}$ when acting on tensors of rank n which are irreducible in their first $n-1$ indices*. By iteration of these equations, we may now eliminate the multipoles $\boldsymbol{F}_i^{(2s)}$ and $\boldsymbol{F}_i^{(n)}$ in the right-hand sides of these equations in favour of $\boldsymbol{K}_i$ and $\boldsymbol{T}_i$. The sums of products of connectors, appearing in these equations, constitute series expansions in the inverse particle distance. When the resulting equations are the substituted into eqs. (5.2) and (5.3), one gets equations of the form (3.1), (3.2) with the mobilities expressed in terms of connectors according to

$$\boldsymbol{\mu}_{ij}^{\mathrm{TT}} = (6\pi\eta a_i)^{-1} \Bigg[\boldsymbol{1}\delta_{ij} + \boldsymbol{A}_{ij}^{(1,1)} + \sum_{s=1}^{\infty} \Bigg(\sum_{m_1=2}^{\infty}{}' \sum_{m_2=2}^{\infty}{}' \cdots \sum_{m_s=2}^{\infty}{}' \sum_{j_1\neq i} \sum_{j_2\neq j_1} \cdots \sum_{\substack{j_s\neq j_{s-1}\\ j_s\neq j}} \boldsymbol{A}_{ij_1}^{(1,m)} \odot \boldsymbol{B}^{(m_1,m_1)^{-1}} \odot \boldsymbol{A}_{j_1j_2}^{(m_1,m_2)} \odot \cdots \odot \boldsymbol{A}_{j_{s-1}j_s}^{(m_{s-1},m_s)} \odot \boldsymbol{B}^{(m_s,m_s)^{-1}} \odot \boldsymbol{A}_{j_sj}^{(m_s,1)} \Bigg) \Bigg], \tag{5.16}$$

$$\boldsymbol{\mu}_{ij}^{\mathrm{RR}} = (8\pi\eta a_i^3)^{-1} \Bigg[\boldsymbol{1}\delta_{ij} - \frac{a_i}{3a_j} \boldsymbol{\epsilon} : \boldsymbol{A}_{ij}^{(2a,2a)} : \boldsymbol{\epsilon} - \frac{a_i}{3a_j} \sum_{s=1}^{\infty} \Bigg(\sum_{m_1=2}^{\infty}{}' \sum_{m_2=2}^{\infty}{}' \cdots \sum_{m_s=2}^{\infty}{}' \sum_{j_1\neq i} \sum_{j_2\neq j_1} \cdots \sum_{\substack{j_s\neq j_{s-1}\\ j_s\neq j}} \boldsymbol{\epsilon} : \boldsymbol{A}_{ij_1}^{(2a,m_1)} \odot \boldsymbol{B}^{(m_1,m_1)^{-1}} \odot \boldsymbol{A}_{j_1j_2}^{(m_1,m_2)} \odot \cdots \boldsymbol{A}_{j_{s-1}j}^{(m_{s-1},m_s)} \odot \boldsymbol{B}^{(m_s,m_s)^{-1}} \odot \boldsymbol{A}_{j_sj}^{(m_s,2a)} : \boldsymbol{\epsilon} \Bigg) \Bigg], \tag{5.17}$$

*According to eq. (4.18) $\boldsymbol{B}^{(2s,2s)}$ is proportional to the projection operator $\boldsymbol{\Delta}$ (defined in eq. (4.18)) and $\boldsymbol{B}^{(2s,2s)^{-1}} = -(10/9)\boldsymbol{\Delta}$. The existence of $\boldsymbol{B}^{(3,3)^{-1}}$ is explicitly demonstrated in section 6.

$$\boldsymbol{\mu}_{ij}^{RT} = -(12\pi\eta a_i^2 a_j)^{-1}\Bigg[a_j\boldsymbol{\epsilon}:\boldsymbol{A}_{ij}^{(2a,1)} + a_j \sum_{s=1}^{\infty}\Bigg(\sum_{m_1=2}^{\infty}{}' \cdots \sum_{m_s=2}^{\infty}{}' \sum_{j_1\neq i} \cdots \sum_{\substack{j_s\neq j_{s-1}\\ j_s\neq j}}$$
$$\times\, \boldsymbol{\epsilon}:\boldsymbol{A}_{ij_1}^{(2a,m_1)} \odot \boldsymbol{B}^{(m_1,m_1)^{-1}} \odot \boldsymbol{A}_{j_1j_2}^{(m_1,m_2)} \odot \cdots \odot \boldsymbol{A}_{j_{s-1}j_s}^{(m_{s-1},m_s)}$$
$$\odot\, \boldsymbol{B}^{(m_s,m_s)^{-1}} \odot \boldsymbol{A}_{j_sj}^{(m_s,1)}\Bigg)\Bigg], \tag{5.18}$$

$$\boldsymbol{\mu}_{ij}^{TR} = (12\pi\eta a_i^2 a_j)^{-1}\Bigg[a_i\boldsymbol{A}_{ij}^{(1,2a)}:\boldsymbol{\epsilon} + a_i \sum_{s=1}^{\infty}\Bigg(\sum_{m_1=2}^{\infty}{}' \cdots \sum_{m_s=2}^{\infty}{}' \sum_{j_1\neq i} \cdots \sum_{\substack{j_s\neq j_{s-1}\\ j_s\neq j}}$$
$$\times\, \boldsymbol{A}_{ij_1}^{(1,m_1)} \odot \boldsymbol{B}^{(m_1,m_1)^{-1}} \odot \boldsymbol{A}_{j_1j_2}^{(m_1,m_2)} \odot \cdots \odot \boldsymbol{A}_{j_{s-1}j_2}^{(m_{s-1},m_s)} \odot \boldsymbol{B}^{(m_s,m_s)^{-1}} \odot \boldsymbol{A}_{j_sj}^{(m_s,1)}\Bigg)\Bigg]. \tag{5.19}$$

Here, in an expression of the form $\boldsymbol{A}^{(n,m)} \odot \boldsymbol{B}^{(m,m)^{-1}}$ the dot denotes, as follows from our previous convention, a full contraction over the last m indices of the first tensor and the first m indices of the second.

We shall now list a number of general properties of these expressions for the mobilities.

(i) As may easily be verified, they satisfy the Onsager symmetry relation (3.3) as a consequence of the symmetries (4.33) (which also holds for $\boldsymbol{B}^{-1}$) and (4.35) obeyed by the connectors. Use must be made here of the fact that

$$\widetilde{\boldsymbol{A}_{ij}^{(n,m)} \odot \boldsymbol{B}^{(m,m)^{-1}}} = \widetilde{\boldsymbol{B}^{(m,m)^{-1}}} \odot \widetilde{\boldsymbol{A}_{ij}^{(n,m)}}, \tag{5.20}$$

which follows from definition (4.34), and also of the fact that $\tilde{\boldsymbol{\epsilon}} = -\boldsymbol{\epsilon}$.

(ii) Each term in the expressions (5.16)–(5.19) which is essentially a product of $s+1$ connectors 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 eq. (5.10) a term in eq. (5.16) with $s+1$ connectors $\boldsymbol{A}$ with upper indices $m_0 \equiv 1$ and $m_i \geq 2$ $(i = 1, \dots, s)$, yields contributions proportional to $R^{-M_{TT}}$ where M_{TT} can assume the values

$$M_{TT} = \begin{cases} 1, 3, & \text{for } s = 0, \\ \sum_{i=1}^{s} 2m_i + 2 + j, & s \geq 1,\, m_i \geq 2,\, j = -s-1, -s+1, \dots, s+1. \end{cases} \tag{5.21}$$

This implies that the translational mobility can not contain terms proportional to R^{-2} and R^{-5}, since no sequences of connectors occur which give such contributions. This result is already implicit in the work of Kynch[8]). The absence of the terms proportional to R^{-5} in addition to those proportional to R^{-2} was also noticed for the case of two spheres by Batchelor[6]) and Felderhof[7]). All other inverse powers of R exist according to eq. (5.21). In table I we have listed all sequences contributing to $\boldsymbol{\mu}^{TT}$ up to order R^{-7}.

TABLE I

Products of connectors contributing in each order to the mobility tensors up to order R^{-7}, taking into account the form (5.10) of each connector. Use has been made of the fact that $\boldsymbol{B}^{(2s,2s)^{-1}} = -10/9\boldsymbol{\Delta}$

$6\pi\eta a_i \boldsymbol{\mu}_{ij}^{TT}$	$12\pi\eta a_i a_j \boldsymbol{\mu}_{ij}^{TR}$
$R^0: \delta_{ij}\boldsymbol{1}$	$R^0: -$
$R^{-1}: \boldsymbol{A}_{ij}^{(1,1)}$	$R^{-1}: -$
$R^{-2}: -$	$R^{-2}: \boldsymbol{A}_{ij}^{(1,2a)}: \boldsymbol{\epsilon}$
$R^{-3}: \boldsymbol{A}_{ij}^{(1,1)}$	$R^{-3}: -$
$R^{-4}: \frac{-10}{9} \boldsymbol{A}_{ik}^{(1,2s)}: \boldsymbol{A}_{kj}^{(2s,1)}$	$R^{-4}: -$
$R^{-5}: -$	$R^{-5}: \frac{-10}{9} \boldsymbol{A}_{ik}^{(1,2s)}: \boldsymbol{A}_{kj}^{(2s,2a)}: \boldsymbol{\epsilon}$
$R^{-6}: \frac{-10}{9} \boldsymbol{A}_{ik}^{(1,2s)}: \boldsymbol{A}_{kj}^{(2s,1)};\ \boldsymbol{A}_{ik}^{(1,3)} \vdots \boldsymbol{B}^{(3,3)^{-1}} \vdots \boldsymbol{A}_{kj}^{(3,1)}$	$R^{-6}: -$
$R^{-7}: \left(\frac{-10}{9}\right)^2 \boldsymbol{A}_{ik}^{(1,2s)}: \boldsymbol{A}_{kl}^{(2s,2s)}: \boldsymbol{A}_{lj}^{(2s,1)}$	$R^{-7}: \frac{-10}{9} \boldsymbol{A}_{ik}^{(1,2s)}: \boldsymbol{A}_{kj}^{(2s,2a)}: \boldsymbol{\epsilon};\ \boldsymbol{A}_{ik}^{(1,3)} \vdots \boldsymbol{B}^{(3,3)^{-1}} \vdots \boldsymbol{A}_{kj}^{(3,2a)}: \boldsymbol{\epsilon}$

$12\pi\eta a_i^2 \boldsymbol{\mu}_{ij}^{RT}$	$8\pi\eta a_i^3 \boldsymbol{\mu}_{ij}^{RR}$
$R^0: -$	$R^0: \delta_{ij}\boldsymbol{1}$
$R^{-1}: -$	$R^{-1}: -$
$R^{-2}: -\boldsymbol{\epsilon}: \boldsymbol{A}_{ij}^{(2a,1)}$	$R^{-2}: -$
$R^{-3}: -$	$R^{-3}: \frac{-a_i}{3a_j} \boldsymbol{\epsilon}: \boldsymbol{A}_{ij}^{(2a,2a)}: \boldsymbol{\epsilon}$
$R^{-4}: -$	$R^{-4}: -$
$R^{-5}: \frac{10}{9} \boldsymbol{\epsilon}: \boldsymbol{A}_{ik}^{(2a,2s)}: \boldsymbol{A}_{kj}^{(2s,1)}$	$R^{-5}: -$
$R^{-6}: -$	$R^{-6}: \frac{10}{9} \frac{a_i}{3a_j} \boldsymbol{\epsilon}: \boldsymbol{A}_{ik}^{(2a,2s)}: \boldsymbol{A}_{kj}^{(2s,2a)}: \boldsymbol{\epsilon}$
$R^{-7}: \frac{10}{9} \boldsymbol{\epsilon}: \boldsymbol{A}_{ik}^{(2a,2s)}: \boldsymbol{A}_{kj}^{(2s,1)};\ -\boldsymbol{\epsilon}: \boldsymbol{A}_{ik}^{(2a,3)} \vdots \boldsymbol{B}^{(3,3)^{-1}} \vdots \boldsymbol{A}_{kj}^{(3,1)}$	$R^{-7}: -$

Similarly one finds for $\boldsymbol{\mu}^{RR}$, eq. (5.17), contributions proportional to $R^{-M_{RR}}$ where M_{RR} can now assume the values

$$M_{RR} = \begin{cases} 3, & \text{for } s = 0, \\ \sum_{i=1}^{s} 2m_i + 4 + j, & s \geq 1, m_i \geq 2, j = -s-1, -s+1, \ldots, s-3. \end{cases} \tag{5.22}$$

The fact that the values of j now range from $-s-1$ to $s-3$ is a consequence of the fact that the connector $\boldsymbol{A}^{(2a,m)}$ is of order $R^{-(m+1)}$ only, as follows from the symmetry of the tensor $\boldsymbol{H}$ in eq. (5.10) (cf. eq. (5.13)). Eq. (5.22) implies that $\boldsymbol{\mu}^{RR}$ does not contain contributions proportional to R^{-1}, R^{-2}, R^{-4}, R^{-5} and

R^{-7}. Again we have listed in table I all connector sequences giving contributions to $\boldsymbol{\mu}^{RR}$ up to and including order R^{-7}.

Finally, for $\boldsymbol{\mu}^{RT}$ and $\boldsymbol{\mu}^{TR}$ one has contributions proportional to $R^{-M_{TR}}$, where

$$M_{TR} = \begin{cases} 2, & \text{for } s = 0, \\ \sum_{i=1}^{s} 2m_i + 3 + j, & s \geq 1, m_i \geq 2, j = -s-1, -s+1, \ldots, s-1. \end{cases} \tag{5.23}$$

Consequently, contributions proportional to R^{-1}, R^{-3}, R^{-4} and R^{-6} are excluded. The existing terms up to order R^{-7} are given in table I.

(iii) Each term in the expressions for the mobilities containing a sequence of s connectors involves the hydrodynamic interaction between at most $s+1$ spheres. This implies in view of the results discussed above that for $\boldsymbol{\mu}^{TT}$ the dominant n-sphere contributions are of order R^{-3n+5} and are due to sequences $\boldsymbol{A}^{(1,2s)} : \boldsymbol{A}^{(2s,2s)} : \ldots : \boldsymbol{A}^{(2s,1)}$, i.e. to dipole–dipole interactions. Thus, e.g. the lowest order 5-sphere contribution to $\boldsymbol{\mu}^{TT}$ is of order R^{-10}.

It is clear from the above discussion that if one wishes to evaluate the mobilities up to and including terms of order R^{-7}, only those connectors appearing in table I have to be evaluated. This corresponds to truncating the hierarchy eq. (5.2)–(5.5) after the quadrupole $\boldsymbol{F}^{(3)}$ of the induced force density. In the next section we shall explicitly calculate the connectors needed to this order of approximation.

6. Evaluation of the mobilities

In view of table I and eq. (5.10), the explicit expressions for the mobilities up to order R^{-7} are those given below (table II). Since the tensors $\boldsymbol{H}^{(n,m)}$ are known for all n and m (cf. eq. (5.13)), it is sufficient to evaluate the tensors $\boldsymbol{G}^{(n,m)}$ occurring in this table. We shall first calculate $\boldsymbol{G}^{(1,1)}$,

$$\boldsymbol{G}_{ij}^{(1,1)} = \frac{3}{4\pi} a_i \int_0^{2\pi} d\phi_{ij} \int_{-1}^{+1} d\xi_{ij} (\boldsymbol{1} - \boldsymbol{\Omega}\boldsymbol{\Omega}) \delta(\xi_{ij}). \tag{6.1}$$

This tensor must be of the form

$$\boldsymbol{G}_{ij}^{(1,1)} = g_1 \boldsymbol{1} + g_2 \hat{\boldsymbol{r}}_{ij} \hat{\boldsymbol{r}}_{ij}. \tag{6.2}$$

From these two equations we obtain

$$\hat{\boldsymbol{r}}_{ij} \hat{\boldsymbol{r}}_{ij} : \boldsymbol{G}_{ij}^{(1,1)} = g_1 + g_2 = \tfrac{3}{2} a_i, \tag{6.3}$$

$$\mathrm{Tr}\, \boldsymbol{G}_{ij}^{(1,1)} = 3g_1 + g_2 = 3a_i, \tag{6.4}$$

so that

$$\boldsymbol{G}_{ij}^{(1,1)} = \tfrac{3}{4}a_i(\boldsymbol{1} + \hat{r}_{ij}\hat{r}_{ij}). \tag{6.5}$$

The tensor $\boldsymbol{G}_{ij}^{(1,2s)}$ which is irreducible (symmetric and traceless) in its last two indices must be of the form

$$\boldsymbol{G}_{ij}^{(1,2s)} = g_3\hat{r}_{ij}\overline{\hat{r}_{ij}\hat{r}_{ij}}. \tag{6.6}$$

It follows therefore using eq. (5.11) that

$$\hat{r}_{ij}\hat{r}_{ij}\hat{r}_{ij} \vdots \boldsymbol{G}_{ij}^{(1,2s)} = \tfrac{2}{3}g_3 = \frac{3}{4\pi}a_ia_j \int_0^{2\pi} \mathrm{d}\phi_{ij} \int_{-1}^{+1} \mathrm{d}\xi_{ij}(\xi_{ij} - \xi_{ij}^3)\frac{\partial}{\partial\xi_{ij}}\delta(\xi_{ij})$$
$$= -\tfrac{3}{2}a_ia_j. \tag{6.7}$$

Hence

$$\boldsymbol{G}_{ij}^{(1,2s)} = -\tfrac{9}{4}a_ia_j\hat{r}_{ij}\overline{\hat{r}_{ij}\hat{r}_{ij}}. \tag{6.8}$$

Next we consider the tensor $\boldsymbol{G}^{(1,2a)}$. The only tensor of rank 3 which is antisymmetric in its last two indices and which can be constructed with the vector $\hat{r}_{ij}$ is the tensor with components

$$(\boldsymbol{G}_{ij}^{(1,2a)})_{\alpha\beta\gamma} = g_4(r_{ij\beta}\delta_{\alpha\gamma} - r_{ij\gamma}\delta_{\alpha\beta}). \tag{6.9}$$

From eqs. (5.11) and (6.9), we obtain

$$\hat{r}_{ij}\boldsymbol{1} \vdots \boldsymbol{G}_{ij}^{(1,2a)} = -2g_4 = \tfrac{3}{4}a_ia_j \int_0^{2\pi} \mathrm{d}\phi_{ij} \int_{-1}^{+1} \mathrm{d}\xi_{ij}\xi_{ij}\frac{\partial}{\partial\xi_{ij}}\delta(\xi_{ij}) = -\tfrac{3}{2}a_ia_j, \tag{6.10}$$

and therefore

$$(\boldsymbol{G}^{(1,2a)})_{\alpha\beta\gamma} = \tfrac{3}{4}a_ia_j(r_{ij\beta}\delta_{\alpha\gamma} - r_{ij\gamma}\delta_{\alpha\beta}). \tag{6.11}$$

In a similar way, cf. appendix E, one obtains for the tensors of higher rank

$$\boldsymbol{G}_{ij}^{(1,3)} = \tfrac{3}{4}a_ia_j^2[2\boldsymbol{\Delta} + 3(\boldsymbol{1} + \hat{r}_{ij}\hat{r}_{ij})\overline{\hat{r}_{ij}\hat{r}_{ij}} + 6\boldsymbol{D}_{ij}], \tag{6.12}$$

$$\boldsymbol{G}_{ij}^{(2s,2s)} = -\tfrac{9}{4}a_i^2a_j[3\overline{\hat{r}_{ij}\hat{r}_{ij}}\,\overline{\hat{r}_{ij}\hat{r}_{ij}} + \boldsymbol{D}_{ij}], \tag{6.13}$$

$$(\boldsymbol{G}_{ij}^{(2a,2a)})_{\alpha\beta\gamma\delta} = -\tfrac{3}{4}a_i^2a_j[\delta_{\alpha\gamma}\delta_{\beta\delta} - \delta_{\alpha\delta}\delta_{\beta\gamma} + \tfrac{3}{2}(r_{ij\alpha}r_{ij\delta}\delta_{\beta\gamma}$$
$$- r_{ij\beta}r_{ij\delta}\delta_{\alpha\gamma} - r_{ij\alpha}r_{ij\gamma}\delta_{\beta\delta} + r_{ij\beta}r_{ij\gamma}\delta_{\alpha\delta})], \tag{6.14}$$

$$(\boldsymbol{G}_{ij}^{(2s,2a)})_{\alpha\beta\gamma\delta} = -\tfrac{9}{8}a_i^2a_j[r_{ij\alpha}r_{ij\delta}\delta_{\beta\gamma} + r_{ij\beta}r_{ij\delta}\delta_{\alpha\gamma} - r_{ij\alpha}r_{ij\gamma}\delta_{\beta\delta} - r_{ij\beta}r_{ij\gamma}\delta_{\alpha\delta}], \tag{6.15}$$

$$(\boldsymbol{G}_{ij}^{(2a,3)})_{\alpha\beta\gamma\delta\epsilon} = \tfrac{45}{4}a_i^2a_j^2[\delta_{\beta\gamma}\overline{r_{ij\alpha}r_{ij\delta}r_{ij\epsilon}} - \delta_{\alpha\gamma}\overline{r_{ij\beta}r_{ij\delta}r_{ij\epsilon}}]. \tag{6.16}$$

The tensor $\boldsymbol{\Delta}$ appearing here was defined in eq. (4.18); the tensor $\boldsymbol{D}$ is traceless and symmetric in its first and last two indices, and defined by

$$(\boldsymbol{D}_{ij})_{\alpha\beta\gamma\delta} = 2r_{ij\alpha}r_{ij\beta}r_{ij\gamma}r_{ij\delta} - \tfrac{1}{2}(r_{ij\alpha}r_{ij\gamma}\delta_{\beta\delta} + r_{ij\alpha}r_{ij\delta}\delta_{\beta\gamma} + r_{ij\beta}r_{ij\delta}\delta_{\alpha\gamma} + r_{ij\beta}r_{ij\gamma}\delta_{\alpha\delta}). \tag{6.17}$$

In order to calculate the mobility tensors, we also need $\boldsymbol{B}^{(3,3)^{-1}}$. It is shown in appendix F that one has

$$\boldsymbol{B}^{(3,3)^{-1}} : \boldsymbol{G}_{ij}^{(3,1)} = -\tfrac{1}{16}a_i^3[2\boldsymbol{\Delta} + 3\overline{\hat{r}_{ij}\hat{r}_{ij}}(\boldsymbol{1} + 5\hat{r}_{ij}\hat{r}_{ij}) + 10\boldsymbol{D}_{ij}]. \tag{6.18}$$

We can now list the various products of tensors $\boldsymbol{G}$ and $\boldsymbol{H}$ occurring in the expressions for the mobilities given in table II.

I. For the translational mobilities $\boldsymbol{\mu}_{ij}^{\mathrm{TT}}$ one has the successive terms*

$$\boldsymbol{G}_{ij}^{(1,1)} = \tfrac{3}{4}a_i(\boldsymbol{1} + \hat{r}_{ij}\hat{r}_{ij}), \tag{6.19}$$

TABLE II

Expansions of the mobility tensors to order R^{-7}. Together with eqs. (6.19)–(6.29), (6.33) and (6.34) these yield the corresponding explicit expressions

$6\pi\eta a_i \boldsymbol{\mu}_{ij}^{\mathrm{TT}}$

$$= \delta_{ij}\boldsymbol{1} + R_{ij}^{-1}\boldsymbol{G}_{ij}^{(1,1)} + R_{ij}^{-3}\boldsymbol{H}_{ij}^{(1,1)} + \sum_{k\neq i,j} R_{ik}^{-2}R_{kj}^{-2}\left(\frac{-10}{9}\right)\boldsymbol{G}_{ik}^{(1,2s)}:\boldsymbol{G}_{kj}^{(2s,1)}$$

$$+ \sum_{k\neq i,j}\left\{R_{ik}^{-2}R_{kj}^{-4}\left(\frac{-10}{9}\right)\boldsymbol{G}_{ik}^{(1,2s)}:\boldsymbol{H}_{kj}^{(2,1)} + R_{ik}^{-4}R_{kj}^{-2}\left(\frac{-10}{9}\right)\boldsymbol{H}_{ik}^{(1,2)}:\boldsymbol{G}_{kj}^{(2s,1)} + R_{ik}^{-3}R_{kj}^{-3}\boldsymbol{G}_{ik}^{(1,3)} : \boldsymbol{B}^{(3,3)^{-1}} : \boldsymbol{G}_{kj}^{(3,1)}\right\}$$

$$+ \sum_{k\neq i,l}\sum_{l\neq j} R_{ik}^{-2}R_{kl}^{-3}R_{lj}^{-2}\left(\frac{-10}{9}\right)^2 \boldsymbol{G}_{ik}^{(1,2s)}:\boldsymbol{G}_{kl}^{(2s,2s)}:\boldsymbol{G}_{lj}^{(2s,1)}$$

$12\pi\eta a_i^2\boldsymbol{\mu}_{ij}^{\mathrm{RT}}$

$$= -R_{ij}^{-2}\boldsymbol{\epsilon}:\boldsymbol{G}_{ij}^{(2a,1)} + \sum_{k\neq i,j} R_{ik}^{-3}R_{kj}^{-2}\frac{10}{9}\boldsymbol{\epsilon}:\boldsymbol{G}_{ik}^{(2a,2s)}:\boldsymbol{G}_{kj}^{(2s,1)}$$

$$+ \sum_{k\neq i,j}\left\{R_{ik}^{-3}R_{kj}^{-4}\frac{10}{9}\boldsymbol{\epsilon}:\boldsymbol{G}_{ik}^{(2a,2s)}:\boldsymbol{H}_{kj}^{(2,1)} - R_{ik}^{-4}R_{kj}^{-3}\boldsymbol{\epsilon}:\boldsymbol{G}_{ik}^{(2a,3)} : \boldsymbol{B}^{(3,3)^{-1}} : \boldsymbol{G}_{kj}^{(3,1)}\right\}$$

$8\pi\eta a_i^3\boldsymbol{\mu}_{ij}^{\mathrm{RR}}$

$$= \delta_{ij}\boldsymbol{1} - R_{ij}^{-3}\frac{a_i}{3a_j}\boldsymbol{\epsilon}:\boldsymbol{G}_{ij}^{(2a,2a)}:\boldsymbol{\epsilon} + \sum_{k\neq i,j} R_{ik}^{-3}R_{kj}^{-3}\frac{10}{9}\frac{a_i}{3a_j}\boldsymbol{\epsilon}:\boldsymbol{G}_{ik}^{(2a,2s)}:\boldsymbol{G}_{kj}^{(2s,2a)}:\boldsymbol{\epsilon}$$

*Since by convention $\boldsymbol{G}_{ii}^{(n,m)} = \boldsymbol{H}_{ii}^{(n,m)} = 0$, eqs. (6.19) and (6.20) are only valid for $j\neq i$, eqs. (6.20)–(6.24) only if both $k\neq i$ and $k\neq j$, etc.

$$\boldsymbol{H}_{ij}^{(1,1)} = -\tfrac{3}{4}a_i(a_i^2 + a_j^2)(\hat{\boldsymbol{r}}_{ij}\hat{\boldsymbol{r}}_{ij} - \tfrac{1}{3}\boldsymbol{1}), \tag{6.20}$$

$$-\tfrac{10}{9}\boldsymbol{G}_{ik}^{(1,2s)} : \boldsymbol{G}_{kj}^{(2s,1)} = -\tfrac{15}{8}a_i a_k^3(1 - 3(\hat{\boldsymbol{r}}_{ik}\cdot\hat{\boldsymbol{r}}_{kj})^2)\hat{\boldsymbol{r}}_{ik}\hat{\boldsymbol{r}}_{kj}, \tag{6.21}$$

$$-\tfrac{10}{9}\boldsymbol{G}_{ik}^{(1,2s)} : \boldsymbol{H}_{kj}^{(2,1)} = \tfrac{3}{8}a_i a_k^3(5a_j^2 + 3a_k^2) \times [(1 - 5(\hat{\boldsymbol{r}}_{ik}\cdot\hat{\boldsymbol{r}}_{kj})^2)\hat{\boldsymbol{r}}_{ik}\hat{\boldsymbol{r}}_{kj} + 2(\hat{\boldsymbol{r}}_{ik}\cdot\hat{\boldsymbol{r}}_{kj})\hat{\boldsymbol{r}}_{ik}\hat{\boldsymbol{r}}_{ik}], \tag{6.22}$$

$$-\tfrac{10}{9}\boldsymbol{H}_{ik}^{(1,2)} : \boldsymbol{G}_{kj}^{(2s,1)} = \tfrac{3}{8}a_i a_k^3(5a_i^2 + 3a_k^2) \times [(1 - 5(\hat{\boldsymbol{r}}_{ik}\cdot\hat{\boldsymbol{r}}_{kj})^2)\hat{\boldsymbol{r}}_{ik}\hat{\boldsymbol{r}}_{kj} + 2(\hat{\boldsymbol{r}}_{ik}\cdot\hat{\boldsymbol{r}}_{kj})\hat{\boldsymbol{r}}_{kj}\hat{\boldsymbol{r}}_{kj}], \tag{6.23}$$

$$\boldsymbol{G}_{ik}^{(1,3)} : \boldsymbol{B}^{(3,3)^{-1}} : \boldsymbol{G}_{kj}^{(3,1)} = \tfrac{1}{64}a_i a_k^5[(49 - 117(\hat{\boldsymbol{r}}_{ik}\cdot\hat{\boldsymbol{r}}_{kj})^2)\boldsymbol{1} - (93 - 315(\hat{\boldsymbol{r}}_{ik}\cdot\hat{\boldsymbol{r}}_{kj})^2)(\hat{\boldsymbol{r}}_{ik}\hat{\boldsymbol{r}}_{ik} + \hat{\boldsymbol{r}}_{kj}\hat{\boldsymbol{r}}_{kj}) + 54(\hat{\boldsymbol{r}}_{ik}\cdot\hat{\boldsymbol{r}}_{kj})\hat{\boldsymbol{r}}_{kj}\hat{\boldsymbol{r}}_{ik} + (729 - 1575(\hat{\boldsymbol{r}}_{ik}\cdot\hat{\boldsymbol{r}}_{kj})^2)(\hat{\boldsymbol{r}}_{ik}\cdot\hat{\boldsymbol{r}}_{kj})\hat{\boldsymbol{r}}_{ik}\hat{\boldsymbol{r}}_{kj}], \tag{6.24}$$

$$(-\tfrac{10}{9})^2\boldsymbol{G}_{ik}^{(1,2s)} : \boldsymbol{G}_{kl}^{(2s,2s)} : \boldsymbol{G}_{lj}^{(2s,1)} = \tfrac{75}{16}a_i a_k^3 a_l^3[(1 - 3(\hat{\boldsymbol{r}}_{ik}\cdot\hat{\boldsymbol{r}}_{kl})^2)(1 - 3(\hat{\boldsymbol{r}}_{kl}\cdot\hat{\boldsymbol{r}}_{lj})^2) + 6(\hat{\boldsymbol{r}}_{ik}\cdot\hat{\boldsymbol{r}}_{kl})^2(\hat{\boldsymbol{r}}_{kl}\cdot\hat{\boldsymbol{r}}_{lj})^2 - 6(\hat{\boldsymbol{r}}_{ik}\cdot\hat{\boldsymbol{r}}_{kl})(\hat{\boldsymbol{r}}_{kl}\cdot\hat{\boldsymbol{r}}_{lj})(\hat{\boldsymbol{r}}_{lj}\cdot\hat{\boldsymbol{r}}_{ik})]\hat{\boldsymbol{r}}_{ik}\hat{\boldsymbol{r}}_{lj}. \tag{6.25}$$

Formulae (6.19) and (6.20), which give the coefficients of the terms proportional to R_{ij}^{-1} and R_{ij}^{-3} respectively, are the well-known[5]) expressions for two-sphere interactions to that order. Formulae (6.21)–(6.23) are expressions for the three-sphere contributions to the mobilities $\boldsymbol{\mu}_{ij}^{TT}$ for $j \neq i$ due to an induced force dipole on sphere k. They represent two-sphere contributions to the mobilities $\boldsymbol{\mu}_{ii}^{TT}$ to order R^{-4} and R^{-6}. The term (6.24) also represents three-sphere contributions to $\boldsymbol{\mu}_{ij}^{TT}$ for $j \neq i$, but is due to an induced force quadrupole on sphere k (for $j = i$, it represents a two-sphere interaction of order R^{-6}). Finally for $j \neq i$, the term (6.25) accounts for contributions due to two-(for $l = i$, $k = j$), three-(for $l = i$, $k \neq j$ or $l \neq i$, $k = j$) and four-($l \neq i$, $k \neq j$) sphere interactions; for $j = i$ this term represents a three-sphere interaction only. All these contributions from the term (6.25) are due to induced dipoles.

It should be noted that these expressions agree with those previously given by Kynch[8]) on the basis of a different method*.

The above expressions may also be applied to a system of two spheres only. In that case, we obtain the translational mobilities $\boldsymbol{\mu}_{11}^{TT}$, $\boldsymbol{\mu}_{22}^{TT}$ and $\boldsymbol{\mu}_{12}^{TT} = \tilde{\boldsymbol{\mu}}_{21}^{TT}$ calculated by Batchelor[6]) and Felderhof[7]) in their analyses of the two-sphere problem in the case of free rotation ($\boldsymbol{T}_1 = \boldsymbol{T}_2 = 0$) to the order R_{12}^{-5} and R_{12}^{-7} respectively. In turn, their expressions are consistent with the results given by Happel and Brenner[1]) for the two-sphere friction tensors to order R_{12}^{-5}. It is clear from the above expressions (6.19)–(6.25) that up to the order considered $\boldsymbol{\mu}_{ii}^{TT}$ contains only contributions of order R_{12}^{-4} and R_{12}^{-6} for the case of two spheres; for more than two spheres, however, $\boldsymbol{\mu}_{ii}^{TT}$ also contains terms

*As is clear from Kynch's eqs. (5.6) and (5.7), there is an obvious misprint in Kynch's formula (5.5): one should multiply the first line by $1/(8R^2S^4)$ and the second line by $1/(8R^4S^2)$.

of order R^{-7}. Similarly, in the two-sphere problem $\boldsymbol{\mu}_{ij}^{\mathrm{TT}}$ for $j \neq i$ contains only terms of order R_{12}^{-1}, R_{12}^{-3} and R_{12}^{-7}, whereas for more than two spheres, terms of order R^{-4} and R^{-6} also occur.

II. We now consider the mobility tensor $\boldsymbol{\mu}_{ij}^{\mathrm{RT}}$. The successive terms appearing in the expression for this mobility tensor are, according to table II, to order R^{-7}

$$-\boldsymbol{\epsilon} : \boldsymbol{G}_{ij}^{(2a,1)} = -\tfrac{3}{2} a_i^2 \boldsymbol{\epsilon} \cdot \hat{\boldsymbol{r}}_{ij}, \tag{6.26}$$

$$\tfrac{10}{9} \boldsymbol{\epsilon} : \boldsymbol{G}_{ik}^{(2a,2s)} : \boldsymbol{G}_{kj}^{(2s,1)} = \tfrac{45}{4} a_i^2 a_k^3 (\hat{\boldsymbol{r}}_{ik} \cdot \hat{\boldsymbol{r}}_{kj})(\hat{\boldsymbol{r}}_{ik} \wedge \hat{\boldsymbol{r}}_{kj}) \hat{\boldsymbol{r}}_{kj}, \tag{6.27}$$

$$\begin{aligned} \tfrac{10}{9} \boldsymbol{\epsilon} : \boldsymbol{G}_{ik}^{(2a,2s)} : \boldsymbol{H}_{kj}^{(2s,1)} &= -\tfrac{3}{4} a_i^2 a_k^3 (3a_k^2 + 5a_j^2) \\ &\times [5(\hat{\boldsymbol{r}}_{ik} \cdot \hat{\boldsymbol{r}}_{kj})(\hat{\boldsymbol{r}}_{ik} \wedge \hat{\boldsymbol{r}}_{kj}) \hat{\boldsymbol{r}}_{kj} - (\hat{\boldsymbol{r}}_{ik} \wedge \hat{\boldsymbol{r}}_{kj}) \hat{\boldsymbol{r}}_{ik} + (\hat{\boldsymbol{r}}_{ik} \cdot \hat{\boldsymbol{r}}_{kj}) \boldsymbol{\epsilon} \cdot \hat{\boldsymbol{r}}_{ik}], \end{aligned} \tag{6.28}$$

$$\begin{aligned} -\boldsymbol{\epsilon} : \boldsymbol{G}_{ik}^{(2a,3)} : \boldsymbol{B}^{(3,3)^{-1}} : \boldsymbol{G}_{kj}^{(3,1)} &= \tfrac{9}{32} a_i^2 a_k^5 [25(1 - 7(\hat{\boldsymbol{r}}_{ik} \cdot \hat{\boldsymbol{r}}_{kj})^2)(\hat{\boldsymbol{r}}_{ik} \wedge \hat{\boldsymbol{r}}_{kj}) \hat{\boldsymbol{r}}_{kj} \\ &+ 50(\hat{\boldsymbol{r}}_{ik} \cdot \hat{\boldsymbol{r}}_{kj})(\hat{\boldsymbol{r}}_{ik} \wedge \hat{\boldsymbol{r}}_{kj}) \hat{\boldsymbol{r}}_{ik} - 16(\hat{\boldsymbol{r}}_{ik} \cdot \hat{\boldsymbol{r}}_{kj}) \boldsymbol{\epsilon} \cdot \hat{\boldsymbol{r}}_{kj} - 3(1 - 5(\hat{\boldsymbol{r}}_{ik} \cdot \hat{\boldsymbol{r}}_{kj})^2) \boldsymbol{\epsilon} \cdot \hat{\boldsymbol{r}}_{ik}]. \end{aligned} \tag{6.29}$$

Since there is in this case no term of order unity (no coupling between translation and rotation for a single sphere), formula (6.26) represents the dominant contribution to $\boldsymbol{\mu}_{ij}^{\mathrm{RT}}$, which only exists if $j \neq i$. It is of order R_{ij}^{-2} and is due to two-sphere interactions only. Expression (6.27) represents the contributions of order R^{-5} to $\boldsymbol{\mu}_{ij}^{\mathrm{RT}}$, $j \neq i$, due to three-sphere interactions; this expression vanishes however for $j = i$, since $\hat{\boldsymbol{r}}_{ik} \wedge \hat{\boldsymbol{r}}_{ki} = 0$, so that $\boldsymbol{\mu}_{ii}^{\mathrm{RT}}$ does not contain contributions of order R^{-5}. Formulae (6.28) and (6.29) account for three-sphere contributions of order R^{-7} to $\boldsymbol{\mu}_{ij}^{\mathrm{RT}}$ for $j \neq i$ and for two sphere contributions of order R^{-7} to $\boldsymbol{\mu}_{ii}^{\mathrm{RT}}$. For a system of two spheres, the expressions for $\boldsymbol{\mu}_{ij}^{\mathrm{RT}}$ reduce to

$$\boldsymbol{\mu}_{11}^{\mathrm{RT}} = (32\pi\eta)^{-1} R_{12}^{-7} a_2^3 (3a_2^2 + 10a_1^2) \boldsymbol{\epsilon} \cdot \hat{\boldsymbol{r}}_{12} + \mathcal{O}(R_{12}^{-9}), \tag{6.30}$$

$$\boldsymbol{\mu}_{12}^{\mathrm{RT}} = -\boldsymbol{\mu}_{21}^{\mathrm{TR}} = -(8\pi\eta)^{-1} R_{12}^{-2} \boldsymbol{\epsilon} \cdot \hat{\boldsymbol{r}}_{12} + \mathcal{O}(R_{12}^{-8}). \tag{6.31}$$

The tensor $\boldsymbol{\mu}_{22}^{\mathrm{RT}}$ can be found by interchanging the indices 1 and 2 in eq. (6.30). Note that e.g. $\boldsymbol{\epsilon} : \hat{\boldsymbol{r}}_{12} \boldsymbol{K}_2 = -\hat{\boldsymbol{r}}_{12} \wedge \boldsymbol{K}_2$, which means that in the case of free rotation ($\boldsymbol{T}_1 = \boldsymbol{T}_2 = 0$) and under the influence of forces $\boldsymbol{K}_1$ and $\boldsymbol{K}_2$, e.g. sphere 1 will have an angular velocity

$$\boldsymbol{\omega}_1 = -(8\pi\eta)^{-1} R_{12}^{-2} \hat{\boldsymbol{r}}_{12} \wedge \boldsymbol{K}_2 + (32\pi\eta)^{-1} R_{12}^{-7} a_2^3 (3a_2^2 + 10a_1^2) \hat{\boldsymbol{r}}_{12} \wedge \boldsymbol{K}_1. \tag{6.32}$$

The results (6.30) and (6.31) are consistent with the expressions given by Happel and Brenner[1]), which relate for the case of free rotation the angular velocities of two spheres to their velocities up to order R_{12}^{-5}.

III. The successive terms in the expressions of table II for the rotational mobilities $\boldsymbol{\mu}_{ij}^{RR}$ are

$$\frac{-a_i}{3a_j}\boldsymbol{\epsilon}:\boldsymbol{G}_{ij}^{(2a,2a)}:\boldsymbol{\epsilon}=\tfrac{3}{2}a_i^3(\hat{r}_{ij}\hat{r}_{ij}-\tfrac{1}{3}\mathbf{1}), \tag{6.33}$$

$$\frac{10}{9}\frac{a_i}{3a_j}\boldsymbol{\epsilon}:\boldsymbol{G}_{ik}^{(2a,2s)}:\boldsymbol{G}_{kj}^{(2s,2a)}:\boldsymbol{\epsilon}=\tfrac{15}{4}a_i^3a_k^3[(\hat{r}_{ik}\wedge\hat{r}_{kj})(\hat{r}_{ik}\wedge\hat{r}_{kj}) + (\hat{r}_{ik}\cdot\hat{r}_{kj})\hat{r}_{kj}\hat{r}_{ik}-(\hat{r}_{ik}\cdot\hat{r}_{kj})^2\mathbf{1}). \tag{6.34}$$

Formula (6.33) represents a contribution to the rotational mobility $\boldsymbol{\mu}_{ij}^{RR}$ for $j\neq i$ of order R_{ij}^{-3} due to two-sphere interactions. Expression (6.34) is a contribution of order R^{-6} which is due to three-sphere interactions for $j\neq i$ and to two-sphere interactions for $j=i$. For a two-sphere system, one finds from eqs. (6.33), (6.34) and table II

$$\boldsymbol{\mu}_{ii}^{RR}=(8\pi\eta a_i^3)^{-1}[\mathbf{1}-\tfrac{15}{4}a_1^3a_2^3R_{12}^{-6}(\mathbf{1}-\hat{r}_{12}\hat{r}_{12})]+\mathcal{O}(R^{-8}),\quad (i=1,2) \tag{6.35}$$

$$\boldsymbol{\mu}_{12}^{RR}=\boldsymbol{\mu}_{21}^{RR}=3(16\pi\eta)^{-1}R_{12}^{-3}(\hat{r}_{12}\hat{r}_{12}-\tfrac{1}{3}\mathbf{1})+\mathcal{O}(R^{-9}). \tag{6.36}$$

Expression (6.36) is consistent with the results derived to order R^{-6} for hindered rotation of two spheres by Happel and Brenner[1])*. Note that if the spheres experience no force ($\boldsymbol{K}_1=\boldsymbol{K}_2=0$), and if the applied torque $\boldsymbol{T}_1$ acting on sphere 1 is parallel to $\hat{r}_{12}$ while $\boldsymbol{T}_2=0$, $\boldsymbol{\omega}_1=-(8\pi\eta a_1^3)^{-1}\boldsymbol{T}_1+\mathcal{O}(R^{-8})$ while $\boldsymbol{\omega}_2=-(8\pi\eta R_{12}^3)^{-1}\boldsymbol{T}_1+\mathcal{O}(R^{-8})$. If, on the other hand, $\hat{r}_{12}\cdot\boldsymbol{T}_1=0$, then

$$\boldsymbol{\omega}_1=-(8\pi\eta a_1^3)^{-1}(1-\tfrac{15}{4}a_1^3a_2^3R_{12}^{-6})\boldsymbol{T}_1+\mathcal{O}(R^{-8}), \tag{6.37}$$

$$\boldsymbol{\omega}_2=(16\pi\eta R_{12}^3)^{-1}\boldsymbol{T}_1+\mathcal{O}(R^{-9}). \tag{6.38}$$

We see that in the second case, the sphere 2 will rotate under the influence of the first one in the opposite direction, as it should. Expressions for $\boldsymbol{\mu}^{TR}$ and $\boldsymbol{\mu}^{RR}$ for the case of two spheres have also been derived by Reuland et. al[19]). Our results agree with theirs, with the exception of a difference in sign in eq. (6.30).

7. Concluding remarks

We have developed in the preceding sections a consistent scheme to evaluate the mobility tensors for an arbitrary number of spheres in an unbounded fluid, as a power series in their reciprocal distances, and derived explicit expressions up to order R^{-7}. To conclude, we wish to make several remarks:

(i) Within the present scheme, higher order contributions to the mobilities can be calculated in a straightforward manner. Thus, after evaluation of the

*The terms of order R^{-6} in eq. (6.35) can not be compared with Happel and Brenner's expressions as it contributes in higher order only to their results.

integral expressions for the connector $\mathbf{A}_{ij}^{(1,4)}$ and the tensor $\mathbf{B}^{(4,4)}$, one has all the ingredients to calculate $\boldsymbol{\mu}_{ij}^{\mathrm{TT}}$ to order R^{-8} by forming the necessary products of connectors.

(ii) The friction tensors $\boldsymbol{\xi}_{ij}^{\mathrm{TT}}$, $\boldsymbol{\xi}_{ij}^{\mathrm{RT}}$ and $\boldsymbol{\xi}_{ij}^{\mathrm{RR}}$ can be found by inversion of the mobility tensor matrix. This has been done for $\boldsymbol{\xi}_{ij}^{\mathrm{TT}}$ to order R^{-3} in paper I. *It turns out that already to this order $\boldsymbol{\xi}_{ij}^{\mathrm{TT}}$ contains three- and four-sphere contributions, whereas the mobility tensor $\boldsymbol{\mu}_{ij}^{\mathrm{TT}}$ contains to order R^{-3} only two-sphere contributions.* The structure of the mobility tensors is also simpler than the one of the friction tensors for another reason: as we have seen in section 5, certain inverse powers of R do not occur in the expressions for $\boldsymbol{\mu}_{ij}^{\mathrm{TT}}$, $\boldsymbol{\mu}_{ij}^{\mathrm{TR}}$ and $\boldsymbol{\mu}_{ij}^{\mathrm{RR}}$. Moreover, as noted also by Felderhof[7]) for the case of two spheres, the series expansion for the mobility tensor converges much more rapidly than the one for the friction tensor.

(iii) In our treatment we have assumed that the unperturbed fluid is at rest (cf. eq. (2.20)). The present formalism may however by extended to the case that $\boldsymbol{v}_0$ is an arbitrary non-vanishing solution of the quasi-static Stokes equations. In that case, one obtains instead of eqs. (4.9), (4.12), (4.25) and (4.30)

$$\boldsymbol{u}_i = \overline{\boldsymbol{v}^0}^{S_i} + (6\pi\eta a_i)^{-1}\boldsymbol{F}_i^{(1)} + (6\pi\eta a_i)^{-1} \sum_{m=1}^{\infty} \sum_{j\neq i} \mathbf{A}_{ij}^{(1,m)} \odot \boldsymbol{F}_j^{(m)}, \tag{7.1}$$

$$a_i \boldsymbol{\epsilon} \cdot \boldsymbol{\omega}_i = 3!!\overline{\hat{n}_i \boldsymbol{v}^0}^{S_i} - (6\pi\eta a_i)^{-1}\boldsymbol{B}^{(2,2)} : \boldsymbol{F}_i^{(2)} + (6\pi\eta a_i)^{-1} \sum_{m=1}^{\infty} \sum_{j\neq i} \mathbf{A}_{ij}^{(2,m)} \odot \boldsymbol{F}_j^{(m)}, \tag{7.2}$$

$$\boldsymbol{B}^{(p,p)} \odot \boldsymbol{F}_i^{(p)} = 6\pi\eta a_i (2p-1)!!\overline{\overbrace{\hat{n}_i^{p-1}} \boldsymbol{v}^0}^{S_i} + \sum_{m=1}^{\infty} \sum_{j\neq i} \mathbf{A}_{ij}^{(p,m)} \odot \boldsymbol{F}_j^{(m)}, \quad (p \geq 3). \tag{7.3}$$

By elimination of the higher order induced force multipoles, one finds e.g. for the case of free rotation ($\boldsymbol{T}_i = 0$)

$$\boldsymbol{u}_i = -\sum_{j\neq i} \boldsymbol{\mu}_{ij}^{\mathrm{TT}} \cdot \boldsymbol{K}_j + \overline{\boldsymbol{v}^0}^{S_i} + {\sum_{m=2}^{\infty}}' \sum_{j\neq i} \left((2m-1)!!\mathbf{A}_{ij}^{(1,m)} \odot \boldsymbol{B}^{(m,m)^{-1}} + \cdots\right) \odot \overline{\overbrace{\hat{n}_j^{m-1}} \boldsymbol{v}^0}^{S_j}. \tag{7.4}$$

Here, $\boldsymbol{\mu}_{ij}^{\mathrm{TT}}$ is the same translational mobility tensor as calculated before for the case $\boldsymbol{v}^0 = 0$; the restricted sum Σ' has been defined after eq. (5.5). In the term between brackets in eq. (7.4), contributions containing products of connectors have not been written out explicitly.

By inverting these equations, one obtains a generalization of Faxén's theorem[12]) for the case of an arbitrary number of spheres (cf. also paper I, where such a generalization of Faxén's theorem was given up to order R^{-3}, and where the last term in eq. (7.4) was neglected). The applicability of such generalizations will depend on the problem under consideration. If e.g. the gradient of the fluid velocity field $\boldsymbol{v}^0$ is approximately constant over the linear dimension of the spheres, it follows from eq. (4.29) that the terms with $m \geq 3$ in the restricted summation in eq. (7.4) may be neglected.

Finally it should be noted that the above mentioned generalization of Faxén's theorem can serve as a starting point for the discussion of the Brownian motion of interacting spherical particles (cf. paper I and also refs. 15-17, 7 and 13 for other approaches to this problem).

Appendix A

Expansion of $\boldsymbol{F}_i(\boldsymbol{k})$ in irreducible multipoles

We first derive the expansion (3.14). From the fact that the Legendre polynomials $P_l(x)$ form a complete orthogonal set of functions on the interval $[-1, 1]$, it follows that $\delta(\hat{r} - \hat{r}')$ can be expanded as

$$\delta(\hat{r} - \hat{r}') = \sum_{l=0}^{\infty} \frac{2l+1}{4\pi} P_l(\hat{r} \cdot \hat{r}'). \tag{A.1}$$

(See e.g. Jackson[18]), eqs. (3.62) and (3.117)). According to eq. (4.21) of Hess and Köhler[14]), one also has

$$P_l(\hat{r} \cdot \hat{r}') = \frac{(2l-1)!!}{l!} \overline{\hat{r}^l} \odot \overline{\hat{r}'^l}. \tag{A.2}$$

Combination of eqs. (A.1) and (A.2) yields

$$\delta(\hat{r} - \hat{r}') = \frac{1}{4\pi} \sum_{l=0}^{\infty} \frac{(2l+1)!!}{l!} \overline{\hat{r}^l} \odot \overline{\hat{r}'^l}. \tag{A.3}$$

One thus obtains from eqs. (2.15), (2.11) and (A.3)

$$\begin{aligned} \boldsymbol{F}_i(\boldsymbol{k}) &= \int \mathrm{d}\hat{r}\, \mathrm{e}^{-\mathrm{i}a_i \boldsymbol{k} \cdot \hat{r}} \boldsymbol{f}_i(\hat{r}) = \int \mathrm{d}\hat{r} \int \mathrm{d}\hat{r}' \delta(\hat{r} - \hat{r}')\, \mathrm{e}^{-\mathrm{i}a_i \boldsymbol{k} \cdot \hat{r}'} \boldsymbol{f}_i(\hat{r}), \\ &= \sum_{l=0}^{\infty} \frac{(2l+1)!!}{l!} \frac{1}{4\pi} \int \mathrm{d}\hat{r}'\, \mathrm{e}^{-\mathrm{i}a_i \boldsymbol{k} \cdot \hat{r}'} \overline{\hat{r}'^l} \odot \int \mathrm{d}\hat{r} \overline{\hat{r}^l} \boldsymbol{f}_i(\hat{r}), \\ &= \sum_{l=0}^{\infty} (2l+1)!!(\mathrm{i}/a_i)^l \left(\overline{\frac{\partial^l}{\partial \boldsymbol{k}^l}} \frac{1}{4\pi} \int \mathrm{d}\hat{r}'\, \mathrm{e}^{-\mathrm{i}a_i \boldsymbol{k} \cdot \hat{r}'} \right) \odot \boldsymbol{F}_i^{(l+1)}, \\ &= \sum_{l=0}^{\infty} (2l+1)!!(\mathrm{i}/a_i)^l \left(\overline{\frac{\partial^l}{\partial \boldsymbol{k}^l}} \frac{\sin k a_i}{k a_i} \right) \odot \boldsymbol{F}_i^{(l+1)}. \end{aligned} \tag{A.4}$$

Here, use was made of the definition (3.7). Eq. (A.4) constitutes the desired expansion.

Next, we derive the result (3.15). One has (cf. the derivation of eq. (A.4))

$$(2l+1)!!(\mathrm{i}/a_i)^l \,\overline{\frac{\partial^l}{\partial \boldsymbol{k}^l}}\, \frac{\sin ka_i}{ka_i} = (2l+1)!!\,\frac{1}{4\pi}\int \mathrm{d}\hat{r}\,\overline{\hat{r}^l}\,\mathrm{e}^{-\mathrm{i}a_i\boldsymbol{k}\cdot\hat{r}},$$
$$= (2l+1)!!\,\frac{1}{4\pi}\int \mathrm{d}\hat{r}\,\overline{\hat{r}^l}\sum_{m=0}^{\infty}\frac{(-\mathrm{i}a_i)^m}{m!}\,\hat{r}^m \odot \boldsymbol{k}^m. \tag{A.5}$$

The tensor $\hat{r}_m$ can be expressed in the irreducible tensors $\overline{\hat{r}^p}$ of rank $p \leqslant m$. This expansion must be of the form

$$\hat{r}_{\alpha_1}\ldots\hat{r}_{\alpha_m} = \overline{\hat{r}_{\alpha_1}\ldots\hat{r}_{\alpha_m}} + b_m \sum_{\substack{\text{pairs}\\ ij}} \delta_{\alpha_i\alpha_j}\overline{\hat{r}_{\alpha_1}\ldots\hat{r}_{\alpha_{i-1}}\hat{r}_{\alpha_{i+1}}\ldots\hat{r}_{\alpha_{j-1}}\hat{r}_{\alpha_{j+1}}\ldots\hat{r}_{\alpha_m}}$$
$$+\ \text{terms of order } m-4. \tag{A.6}$$

Here, the summation runs over all distinct pairs ij; "terms of order $m-4$" stand for terms involving components of irreducible tensors of rank $\leqslant m-4$. The coefficient b_m can be determined as follows. If we contract this equation over α_1 and α_2, the left hand side becomes a component of the tensor $\hat{r}^{m-2}$. The first term in the right-hand side vanishes, by definition. All terms in the sum on the right-hand side with $i \neq 1, 2$ and $j \neq 1, 2$ also vanish; the term with $i = 1$, $j = 2$ gives a factor $3b_m\overline{\hat{r}_{\alpha_3}\ldots\hat{r}_{\alpha_m}}$, and each of the $2(m-2)$ terms with $i = 1, 2$ $j \geqslant 3$ contribute a factor $b_m\overline{\hat{r}_{\alpha_3}\ldots\hat{r}_{\alpha_m}}$. Thus, after contraction over α_1 and α_2, we obtain from eq. (A.6)

$$\hat{r}_{\alpha_3}\ldots\hat{r}_{\alpha_m} = (2m-1)b_m\overline{\hat{r}_{\alpha_3}\ldots\hat{r}_{\alpha_m}} + \text{terms of order } m-4. \tag{A.7}$$

In view of the normalisation explained in the first footnote of section 3, this equation implies that $b_m = 1/(2m-1)$. To proceed, we also note that according to eq. (5.4) of Hess and Köhler[14]), one has

$$\frac{1}{4\pi}\int \mathrm{d}\hat{r}\,\overline{\hat{r}^l}\,\overline{\hat{r}^m} = \frac{l!}{(2l+1)!!}\,\delta_{lm}\boldsymbol{\Delta}^{(l,l)}. \tag{A.8}$$

Here $\boldsymbol{\Delta}^{(l,l)}$ is a tensor of rank $2l$ which projects out the irreducible part of a tensor of rank l. (Note that $\boldsymbol{\Delta}^{(2,2)}$ is just the tensor $\boldsymbol{\Delta}$ defined in eq. (4.18)). Hence

$$\boldsymbol{\Delta}^{(l,l)} \odot \boldsymbol{k}^l = \overline{\boldsymbol{k}^l}. \tag{A.9}$$

If the expansion (A.6) is substituted into eq. (A.5), eq. (A.8) shows that the terms with $m < l$ all vanish. For $m = l$ only the first term in the expansion (A.6) contributes, while the other terms again vanish. For $m = l + 2$, all $\frac{1}{2}(l+1)(l+2)$ distinct pairs arising from the second term in the expansion (A.6), give non-vanishing contributions. One thus obtains from eq. (A.5) with the aid of eq. (A.9) and the result $b_m = 1/(2m-1)$

$$
\begin{aligned}
&(2l+1)!!(\mathrm{i}/a_i)^l \overline{\frac{\partial^l}{\partial \boldsymbol{k}^l}} \frac{\sin ka_i}{ka_i} \\
&= (-\mathrm{i}a_i)^l \left(\frac{(2l+1)!!}{4\pi l!} \int \mathrm{d}\hat{r}\overline{\hat{r}^l}\,\overline{\hat{r}^l}\right) \odot \boldsymbol{k}^l \left(1 - \frac{l!(l+1)(l+2)}{2(l+2)!(2l+3)} a_i^2 k^2 + \mathcal{O}(a_i k)^4\right) \\
&= (-\mathrm{i}a_i)^l \overline{\boldsymbol{k}^l}(1 - a_i^2 k^2/(4l+6) + \mathcal{O}(a_i k)^4),
\end{aligned}
\tag{A.10}
$$

which is the desired result.

Appendix B

Integrals over the unit sphere

In this appendix we evaluate integrals of the form

$$\frac{1}{4\pi} \int \mathrm{d}\boldsymbol{\Omega} \Omega_{\alpha_1} \ldots \Omega_{\alpha_m}. \tag{B.1}$$

This integral must be equal to a component of the tensor of rank m which can be constructed from the isotropic tensor $\delta_{\alpha\beta}$ and which is symmetric in any pair of its indices. Obviously, for m odd, the expression (B.1) always vanishes. For m even, the only tensor of this form is

$$\frac{1}{4\pi} \int \mathrm{d}\boldsymbol{\Omega} \Omega_{\alpha_1} \ldots \Omega_{\alpha_m} = c_m \sum_p \delta_{\alpha_1\alpha_2}\delta_{\alpha_3\alpha_4} \ldots \delta_{\alpha_{m-1}\alpha_m}, \quad (m \text{ even}). \tag{B.2}$$

Here Σ_p stands for a sum over all permutations of indices which yield distinct pairs of indices. The coefficient c_m can most easily be determined by putting $\alpha_1 = \alpha_2 = \cdots = \alpha_m$. The integral on the left-hand side is then equal to $1/(m+1)$; since there are $(m-1)!!$ terms in the summation on the right-hand side, one obtains in that case $c_m = 1/(m+1)!!$, so that

$$\frac{1}{4\pi} \int \mathrm{d}\boldsymbol{\Omega} \Omega_{\alpha_1} \ldots \Omega_{\alpha_m} = \frac{1}{(m+1)!!} \sum_p \delta_{\alpha_1\alpha_2}\delta_{\alpha_3\alpha_4} \ldots \delta_{\alpha_{m-1}\alpha_m}, \quad (m \text{ even}). \tag{B.3}$$

Hence, for $m = 2$ and 4

$$\frac{1}{4\pi} \int \mathrm{d}\boldsymbol{\Omega} \Omega_\alpha \Omega_\beta = \tfrac{1}{3}\delta_{\alpha\beta}, \tag{B.4}$$

$$\frac{1}{4\pi}\int \mathrm{d}\boldsymbol{\Omega}\Omega_\alpha\Omega_\beta\Omega_\gamma\Omega_\delta = \tfrac{1}{15}(\delta_{\alpha\beta}\delta_{\gamma\delta} + \delta_{\alpha\gamma}\delta_{\beta\delta} + \delta_{\alpha\delta}\delta_{\beta\gamma}). \tag{B.5}$$

From eqs. (B.4) and (B.5), eq. (4.16) is easily derived.

Appendix C

Justification of eq. (4.30)

In this appendix, we show that if we consider the nth surface moment (4.28) on sphere i, only the multipole $\boldsymbol{F}_i^{(n)}$ of sphere i appears on the left-hand side of eq. (4.30). This is in fact the case provided that $\boldsymbol{B}^{(n,m)} = 0$ for $n \neq m$, where $\boldsymbol{B}^{(n,m)}$ is defined by

$$\boldsymbol{B}^{(n,m)} = -\left(\frac{-\mathrm{i}}{a_i}\right)^{n-1}\left(\frac{\mathrm{i}}{a_i}\right)^{m-1}(2n-1)!!(2m-1)!!\frac{3}{8\pi^2}\int \mathrm{d}\boldsymbol{\Omega}\int_{-\infty}^{+\infty}\mathrm{d}k$$
$$\times\left(\frac{\overbrace{\partial^{n-1}}}{\partial\boldsymbol{k}^{n-1}}\frac{\sin ka_i}{ka_i}\right)(\mathbf{1} - \boldsymbol{\Omega}\boldsymbol{\Omega})\left(\frac{\overbrace{\partial^{m-1}}}{\partial\boldsymbol{k}^{m-1}}\frac{\sin ka_i}{ka_i}\right). \tag{C.1}$$

Note that for $n = m$, this equation reduces to eq. (4.31). Obviously, $\boldsymbol{B}^{(n,m)} = 0$ if n is even and m odd or the other way around, since in that case the integrand is an odd function of $\boldsymbol{k}$. If both m and n are even or odd, we may without loss of generality assume that m is larger than n, so that $m \geqslant n+2$. We then obtain after $n-1$ partial integrations

$$\boldsymbol{B}^{(n,m)} = \mathrm{i}^{n+m}\frac{(2n-1)!!(2m-1)!!}{a_i^{n+m-1}}\frac{3}{8\pi^2}\int \mathrm{d}\boldsymbol{\Omega}\int_{-\infty}^{+\infty}\mathrm{d}k$$
$$\times\frac{\sin ka_i}{ka_i}\left[\frac{\overbrace{\partial^{n-1}}}{\partial\boldsymbol{k}^{n-1}}(\mathbf{1} - \boldsymbol{\Omega}\boldsymbol{\Omega})\frac{\overbrace{\partial^{m-1}}}{\partial\boldsymbol{k}^{m-1}}\frac{\sin ka_i}{ka_i}\right]. \tag{C.2}$$

It follows from the expansion (3.15) that the term between square brackets is a tensor of rank $m+n$ constructed from $\boldsymbol{\Omega}$ multiplied by a polynomial in k^2 of which the lowest order term is of order k^{m-n}, with $m-n \geqslant 2$. All terms in the expansion therefore give k-integrals of the form

$$\int_{-\infty}^{+\infty}\mathrm{d}k k^{m-n-1}\sin a_i k, \quad m-n-1 \geqslant 1. \tag{C.3}$$

These integrals are all zero. Hence $\boldsymbol{B}^{(n,m)} = 0$ for $m \neq n$.

Appendix D

The tensor $\boldsymbol{H}^{(n,m)}$

According to eq. (5.12), the tensor $\boldsymbol{H}_{ij}^{(n,m)}$ is given by

$$\boldsymbol{H}_{ij}^{(n,m)} = (-1)^{n-1}\frac{3}{4\pi}a_i^n a_j^{m-1}\left(\frac{a_i^2}{4n+2}+\frac{a_j^2}{4m+2}\right) \times \int \mathrm{d}\boldsymbol{\Omega}[\overline{\boldsymbol{\Omega}^{n-1}}(\boldsymbol{1}-\boldsymbol{\Omega\Omega})\overline{\boldsymbol{\Omega}^{m-1}}]\frac{\partial^{n+m}}{\partial \xi_{ij}^{n+m}}\delta(\xi_{ij}). \tag{D.1}$$

Consider the tensor between brackets. If we write out the irreducible tensors in this expression, and use the normalization condition explained in the first footnote of section 3, we obtain

$$\overline{\boldsymbol{\Omega}^{n-1}}(\boldsymbol{1}-\boldsymbol{\Omega\Omega})\overline{\boldsymbol{\Omega}^{m-1}} = -\boldsymbol{\Omega}^{n+m} + \text{lower order terms}. \tag{D.2}$$

Here, "lower order terms" stands for tensors constructed from at least one isotropic tensor $\boldsymbol{1}$ and at most $n+m-2$ vectors $\boldsymbol{\Omega}$. However, all these "lower order terms" give vanishing contributions in eq. (D.1) since any of its components yields, after integration over ϕ_{ij}, a polynomial in ξ_{ij} of which the highest order term is of order ξ_{ij}^{n+m-2} (cf. the discussion after eq. (5.9)). We thus find that $\boldsymbol{H}_{ij}^{(n,m)}$ may be written as

$$\boldsymbol{H}_{ij}^{(n,m)} = (-1)^{n}\frac{3}{4\pi}a_i^n a_j^{m-1}\left(\frac{a_i^2}{4n+2}+\frac{a_j^2}{4m+2}\right)\int \mathrm{d}\boldsymbol{\Omega}\boldsymbol{\Omega}^{n+m}\frac{\partial^{n+m}}{\partial \xi_{ij}^{n+m}}\delta(\xi_{ij}). \tag{D.3}$$

This is an irreducible tensor: it is symmetric in any pair of its indices, and also traceless, since by taking a trace over any pair, we again obtain after the ϕ_{ij} integration a polynomial in ξ_{ij} of which the highest order term is of order ξ_{ij}^{n+m-2}. Thus $\boldsymbol{H}_{ij}^{(n,m)}$ must be of the form

$$\boldsymbol{H}_{ij}^{(n,m)} = h\overline{\hat{\boldsymbol{r}}_{ij}^{n+m}}. \tag{D.4}$$

The Legendre polynomials $P_l(x)$ have the property that $P_l(1) = 1$ for all l. It therefore follows from eq. (A.2) that

$$\overline{\hat{\boldsymbol{r}}_{ij}^{l}} \odot \overline{\hat{\boldsymbol{r}}_{ij}^{l}} = l!/(2l-1)!!. \tag{D.5}$$

Hence, we obtain from eqs. (D.3)–(D.5)

$$\overline{\hat{\boldsymbol{r}}_{ij}^{n+m}} \odot \boldsymbol{H}_{ij}^{(n,m)} = \frac{(n+m)!h}{(2n+2m-1)!!} = (-1)^m \tfrac{3}{2}a_i^n a_j^{m-1}\left(\frac{a_i^2}{4n+2}+\frac{a_j^2}{4m+2}\right)(n+m)! \tag{D.6}$$

Eqs. (D.4) and (D.6) yield eq. (5.13).

Appendix E

Derivation of eqs. (6.12)–(6.16)

(a) First, we consider the tensor $\boldsymbol{G}^{(1,3)}$. This tensor is symmetric in its first and last pair of indices, and traceless in its last pair as well. In general, one can only construct 10 different tensors of rank 4 with the isotropic tensor $\boldsymbol{1}$ and the vector $\hat{r}_{ij}$. The requirement that these tensors be symmetric in their first and last two indices reduces the number of different tensors to 6, and one finds that there are only 4 of these which are also traceless in the last two indices. Thus we may write quite generally

$$\boldsymbol{G}_{ij}^{(1,3)} = a_i a_j^2 [g_5 \boldsymbol{\Delta} + g_6 \boldsymbol{1}\overline{\hat{r}_{ij}\hat{r}_{ij}} + g_7 \hat{r}_{ij}\hat{r}_{ij}\overline{\hat{r}_{ij}\hat{r}_{ij}} + g_8 \boldsymbol{D}_{ij}]. \tag{E.1}$$

The tensors $\boldsymbol{\Delta}$ and $\boldsymbol{D}_{ij}$ were defined in eqs. (4.18) and (6.17). Using the summation convention for repeated indices, we obtain from eqs. (5.11) and (E.1)

$$(a_i a_j^2)^{-1} r_{ij,\alpha} r_{ij,\beta} \delta_{\gamma\delta} (\boldsymbol{G}_{ij}^{(1,3)})_{\delta\gamma\beta\alpha} = 2g_6 + \tfrac{2}{3} g_7 = 6, \tag{E.2}$$

$$(a_i a_j^2)^{-1} r_{ij,\alpha} r_{ij,\beta} r_{ij,\gamma} r_{ij,\delta} (\boldsymbol{G}_{ij}^{(1,3)})_{\delta\gamma\beta\alpha} = \tfrac{2}{3} g_5 + \tfrac{2}{3} g_6 + \tfrac{2}{3} g_7 = 4, \tag{E.3}$$

$$(a_i a_j^2)^{-1} \delta_{\beta\delta} r_{ij,\alpha} r_{ij,\gamma} (\boldsymbol{G}_{ij}^{(1,3)})_{\delta\gamma\beta\alpha} = \tfrac{5}{3} g_5 + \tfrac{2}{3} g_6 + \tfrac{2}{3} g_7 - g_8 = 1, \tag{E.4}$$

$$(a_i a_j^2)^{-1} \delta_{\beta\delta} \delta_{\alpha\gamma} (\boldsymbol{G}_{ij}^{(1,3)})_{\delta\gamma\beta\alpha} = 5g_5 + \tfrac{2}{3} g_7 - 2g_8 = 0. \tag{E.5}$$

The solution of these equations is $g_5 = \frac{3}{2}$, $g_6 = \frac{9}{4}$, $g_7 = \frac{9}{4}$, $g_8 = \frac{9}{2}$. Substitution of these values into eq. (E.1) yields the result (6.12).

(b) Next, consider $\boldsymbol{G}^{(2s,2s)}$. This tensor is of a similar structure as $\boldsymbol{G}^{(1,3)}$, apart from the fact that it is also traceless in its first two indices. Comparison with eq. (E.1) shows that it must therefore be of the form

$$\boldsymbol{G}_{ij}^{(2s,2s)} = a_i^2 a_j [g_9 \boldsymbol{\Delta} + g_{10} \overline{\hat{r}_{ij}\hat{r}_{ij}}\,\overline{\hat{r}_{ij}\hat{r}_{ij}} + g_{11} \boldsymbol{D}_{ij}]. \tag{E.6}$$

We now obtain from eq. (5.11)

$$(a_i^2 a_j)^{-1} r_{ij,\alpha} r_{ij,\beta} r_{ij,\gamma} r_{ij,\delta} (\boldsymbol{G}_{ij}^{(2s,2s)})_{\delta\gamma\beta\alpha} = \tfrac{2}{3} g_9 + \tfrac{4}{9} g_{10} = -3, \tag{E.7}$$

$$(a_i^2 a_j)^{-1} \delta_{\delta\beta} r_{ij,\alpha} r_{ij,\gamma} (\boldsymbol{G}_{ij}^{(2s,2s)})_{\delta\gamma\beta\alpha} = \tfrac{5}{3} g_9 + \tfrac{4}{9} g_{10} - g_{11} = -\tfrac{3}{4}, \tag{E.8}$$

$$(a_i^2 a_j)^{-1} \delta_{\delta\beta} \delta_{\alpha\gamma} (\boldsymbol{G}_{ij}^{(2s,2s)})_{\delta\gamma\beta\alpha} = 5g_9 + \tfrac{2}{3} g_{10} - 2g_{11} = 0, \tag{E.9}$$

with the solution $g_9 = 0$, $g_{10} = -\frac{27}{4}$ and $g_{11} = -\frac{9}{4}$. Substitution of these values into eq. (E.6) yields eq. (6.13).

(c) We now evaluate $\boldsymbol{G}^{(2a,2a)}$. The only tensor which is antisymmetric in the first and last two indices and which can be constructed from the isotropic tensor and the vector $\hat{r}_{ij}$, is the tensor with components

$$(\boldsymbol{G}_{ij}^{(2a,2a)})_{\alpha\beta\gamma\delta} = a_i^2 a_j g_{12}(\delta_{\alpha\gamma}\delta_{\beta\delta} - \delta_{\alpha\delta}\delta_{\beta\gamma}) + a_i^2 a_j g_{13} \times (r_{ij,\alpha} r_{ij,\delta}\delta_{\beta\gamma} + r_{ij,\beta} r_{ij,\gamma}\delta_{\alpha\delta} - r_{ij,\alpha} r_{ij,\gamma}\delta_{\beta\delta} - r_{ij,\beta} r_{ij,\delta}\delta_{\alpha\gamma}). \tag{E.10}$$

Eq. (5.11) yields

$$(a_i^2 a_j)^{-1}\delta_{\alpha\gamma} r_{ij,\beta} r_{ij,\delta}(\boldsymbol{G}_{ij}^{(2a,2s)})_{\alpha\beta\gamma\delta} = 2g_{12} - 2g_{13} = +\tfrac{3}{4}, \tag{E.11}$$

$$(a_i^2 a_j)^{-1}\delta_{\alpha\gamma}\delta_{\beta\delta}(\boldsymbol{G}_{ij}^{(2a,2a)})_{\alpha\beta\gamma\delta} = 6g_{12} - 4g_{13} = 0. \tag{E.12}$$

From these equations, one finds that $g_{12} = -\frac{3}{4}$ and $g_{13} = -\frac{9}{8}$; this result, together with eq. (E.10), leads to eq. (6.14).

(d) It turns out that $\boldsymbol{G}^{(2s,2a)}$ can only be of the form

$$(\boldsymbol{G}_{ij}^{(2s,2a)})_{\alpha\beta\gamma\delta} = a_i^2 a_j g_{14}(r_{ij,\alpha} r_{ij,\delta}\delta_{\beta\gamma} + r_{ij,\beta} r_{ij,\delta}\delta_{\alpha\gamma} - r_{ij,\alpha} r_{ij,\gamma}\delta_{\beta\delta} - r_{ij,\beta} r_{ij,\gamma}\delta_{\alpha\delta}). \tag{E.13}$$

In this case, it follows from eq. (5.11) that

$$(a_i^2 a_j)^{-1}\delta_{\alpha\gamma} r_{ij,\beta} r_{ij,\delta}(\boldsymbol{G}_{ij}^{(2s,2a)})_{\alpha\beta\gamma\delta} = 2g_{14} = \frac{-9}{4}. \tag{E.14}$$

By combining eqs. (E.13) and (E.14), one gets the result (6.15).

(e) According to eq. (5.11) the tensor $\boldsymbol{G}_{ij}^{(2,3)}$ is given by

$$\boldsymbol{G}_{ij}^{(2,3)} = -\frac{3}{4\pi} a_i^2 a_j^2 \int \mathrm{d}\boldsymbol{\Omega}\boldsymbol{\Omega}(\boldsymbol{1} - \boldsymbol{\Omega}\boldsymbol{\Omega})\overline{\boldsymbol{\Omega}^2}\frac{\partial^3}{\partial\xi^3}\delta(\xi_{ij}). \tag{E.15}$$

We therefore obtain for the antisymmetric part

$$(\boldsymbol{G}_{ij}^{(2a,3)})_{\alpha\beta\gamma\delta\epsilon} = -\frac{3}{8\pi} a_i^2 a_j^2 \left[\delta_{\beta\gamma}\int \mathrm{d}\boldsymbol{\Omega}\Omega_\alpha\overline{\Omega\Omega_\epsilon}\frac{\partial^3}{\partial\xi_{ij}^3}\delta(\xi_{ij}) - \delta_{\alpha\gamma}\int \mathrm{d}\boldsymbol{\Omega}\Omega_\beta\overline{\Omega_\delta\Omega_\epsilon}\frac{\partial^3}{\partial\xi_{ij}^3}\delta(\xi_{ij})\right]. \tag{E.16}$$

For similar reasons as discussed in appendix D, both integrals in this equation represent components of irreducible tensors. Using the fact that (cf. appendix D)

$$\frac{1}{4\pi}\int \mathrm{d}\boldsymbol{\Omega}\Omega_\alpha\overline{\Omega_\delta\Omega_\epsilon}\frac{\partial^3}{\partial\xi_{ij}^3}\delta(\xi_{ij}^3) = -\tfrac{15}{2}\overline{r_{ij,\alpha} r_{ij,\delta} r_{ij,\epsilon}}, \tag{E.17}$$

one immediately obtains eq. (6.16) from eqs. (E.16) and (E.17).

Appendix F

Determination of $\mathbf{B}^{(3,3)^{-1}}$

In this appendix we determine $\mathbf{B}^{(3,3)^{-1}} \odot \mathbf{A}_{ij}^{(3,1)}$. We first define a tensor $\mathbf{I}_i$ of rank 3 by

$$\mathbf{I}_i = \sum_{j\neq i} \sum_{m=1}^{\infty} \mathbf{A}_{ij}^{(3,m)} \odot \mathbf{F}_j^{(m)}. \tag{F.1}$$

This tensor is traceless and symmetric in its first two indices. The tensor $\mathbf{B}^{(3,3)}$, given in eq. (4.26) is easily evaluated with the aid of the results derived in appendix B. Eq. (4.25) then becomes explicitly (the index i which labels the spheres, is suppressed)

$$-\tfrac{9}{7}(6F^{(3)}_{\alpha\beta\gamma} - F^{(3)}_{\beta\gamma\alpha} - F^{(3)}_{\gamma\alpha\beta} - \delta_{\alpha\gamma}F^{(3)}_{\delta\beta\delta} - \delta_{\beta\gamma}F^{(3)}_{\delta\alpha\delta} + \tfrac{4}{3}\delta_{\alpha\beta}F^{(3)}_{\delta\gamma\delta}) = I_{\alpha\beta\gamma}. \tag{F.2}$$

Here, the summation convention for repeated indices has been employed. Note that if one contracts α and γ in this equation, and uses the fact that $F_{\alpha\beta\gamma} = F_{\beta\alpha\gamma}$ and $F_{\delta\delta\alpha} = 0$, one gets

$$-3F^{(3)}_{\delta\beta\delta} = I_{\delta\beta\delta}. \tag{F.3}$$

To invert eq. (F.2), we first consider the case $\alpha \neq \beta$, $\beta \neq \gamma$, $\gamma \neq \alpha$; then, eq. (F.2) reduces to

$$-\tfrac{9}{7}(6F^{(3)}_{\alpha\beta\gamma} - F^{(3)}_{\beta\gamma\alpha} - F^{(3)}_{\gamma\alpha\beta}) = I_{\alpha\beta\gamma}, \quad (\alpha \neq \beta, \beta \neq \gamma, \gamma \neq \alpha). \tag{F.4}$$

In view of the fact that both $\mathbf{F}^{(3)}$ and $\mathbf{I}$ are symmetric in their first two indices, this equation yields three equations, for $F^{(3)}_{123}(= F^{(3)}_{213})$, $F^{(3)}_{231}(= F^{(3)}_{321})$ and $F^{(3)}_{312}(= F^{(3)}_{132})$ in terms of I_{123}, I_{231} and I_{312}, which can readily be solved, so that

$$F^{(3)}_{\alpha\beta\gamma} = -\tfrac{1}{36}(5I_{\alpha\beta\gamma} + I_{\beta\gamma\alpha} + I_{\gamma\alpha\beta}), \quad (\alpha \neq \beta, \beta \neq \gamma, \gamma \neq \alpha). \tag{F.5}$$

The inverse of eq. (F.2) should reduce to eq. (F.5) in this special case. This is only possible if the inverse of eq. (F.2) is of the form

$$F^{(3)}_{\alpha\beta\gamma} = -\tfrac{1}{36}(5I_{\alpha\beta\gamma} + I_{\beta\gamma\alpha} + I_{\gamma\alpha\beta} + a\delta_{\alpha\beta}I_{\delta\gamma\delta} + b\delta_{\alpha\gamma}I_{\delta\beta\delta} + b\delta_{\beta\gamma}I_{\delta\alpha\delta}). \tag{F.6}$$

Here, use has been made of the fact that the right-hand side of eq. (F.6) must be symmetric in α and β, and also of the fact that the only non-zero contracted form that can be constructed from $\mathbf{I}$ is $I_{\delta\alpha\delta}(= I_{\alpha\delta\delta})$. If we contract α and β in eq. (F.6), we get, since $F^{(3)}_{\delta\delta\gamma} = 0$

$$0 = 2 + 3a + 2b. \tag{F.7}$$

Contraction of eq. (F.6) over α and γ yields with the aid of eq. (F.3)

$$F^{(3)}_{\delta\beta\delta} = -\tfrac{1}{36}(6 + a + 4b)I_{\delta\beta\delta} = \tfrac{1}{12}(6 + a + 4b)F^{(3)}_{\delta\beta\delta}. \tag{F.8}$$

The solution of eqs. (F.7) and (F.8) is $a=-2$, $b=2$. With these values, eq. (F.6) is eq. (5.15) written out explicitly for the case $n=3$. In particular, it follows from eq. (F.6) that

$$(\boldsymbol{B}^{(3,3)^{-1}} \odot \boldsymbol{G}_{ij}^{(3,1)})_{\alpha\beta\gamma\delta} = -\tfrac{1}{36}[5G^{(3,1)}_{ij,\alpha\beta\gamma\delta} + G^{(3,1)}_{ij,\beta\gamma\alpha\delta} + G^{(3,1)}_{ij,\gamma\alpha\beta\delta} - 2\delta_{\alpha\beta}G^{(3,1)}_{ij,\epsilon\gamma\epsilon\delta} + 2\delta_{\alpha\gamma}G^{(3,1)}_{ij,\epsilon\beta\epsilon\delta} + 2\delta_{\beta\gamma}G^{(3,1)}_{ij,\epsilon\alpha\epsilon\delta}]. \tag{F.9}$$

For reasons discussed in connection with $\boldsymbol{G}^{(1,3)}$, cf. appendix E sub a, we must have

$$\boldsymbol{B}^{(3,3)^{-1}} \odot \boldsymbol{G}_{ij}^{(3,1)} = a_i^3[c\boldsymbol{\Delta} + d\overline{\hat{\boldsymbol{r}}_{ij}\hat{\boldsymbol{r}}_{ij}\boldsymbol{1}} + e\overline{\hat{\boldsymbol{r}}_{ij}\hat{\boldsymbol{r}}_{ij}}\hat{\boldsymbol{r}}_{ij}\hat{\boldsymbol{r}}_{ij} + f\boldsymbol{D}_{ij}]. \tag{F.10}$$

From eqs. (F.9), (F.10), (6.12) and the symmetry relation

$$\boldsymbol{G}_{ij}^{(3,1)} = a_i/a_j \widetilde{\boldsymbol{G}_{ij}^{(1,3)}},$$

one gets (cf. also eqs. (E.1)–(E.5))

$$a_i^{-3} r_{ij,\alpha} r_{ij,\beta} \delta_{\gamma\delta} (\boldsymbol{B}^{(3,3)^{-1}} \odot \boldsymbol{G}_{ij}^{(3,1)})_{\alpha\beta\gamma\delta} = 2d + \tfrac{2}{3}e = -1, \tag{F.11}$$

$$a_i^{-3} r_{ij,\alpha} r_{ij,\beta} r_{ij,\gamma} r_{ij,\delta} (\boldsymbol{B}^{(3,3)^{-1}} \odot \boldsymbol{G}_{ij}^{(3,1)})_{\alpha\beta\gamma\delta} = \tfrac{2}{3}c + \tfrac{2}{3}d + \tfrac{2}{3}e = -\tfrac{5}{6}, \tag{F.12}$$

$$a_i^{-3} \delta_{\beta\delta} r_{ij,\alpha} r_{ij,\gamma} (\boldsymbol{B}^{(3,3)^{-1}} \odot \boldsymbol{G}_{ij}^{(3,1)})_{\alpha\beta\gamma\delta} = \tfrac{5}{3}c + \tfrac{2}{3}d + \tfrac{2}{3}e - f = -\tfrac{1}{3}, \tag{F.13}$$

$$a_i^{-3} \delta_{\alpha\gamma} \delta_{\beta\delta} (\boldsymbol{B}^{(3,3)^{-1}} \odot \boldsymbol{G}_{ij}^{(3,1)})_{\alpha\beta\gamma\delta} = 5c + \tfrac{2}{3}e - 2f = 0. \tag{F.14}$$

The solution of these equations is $c=-\frac{1}{8}$, $d=-\frac{3}{16}$, $e=-\frac{15}{16}$ and $f=-\frac{5}{8}$. After substitution of these values into eq. (F.10), one arrives at eq. (6.18).

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