Physica **120A** (1983) 77–102 *North-Holland Publishing Co.*

MANY-SPHERE HYDRODYNAMIC INTERACTIONS

II. MOBILITIES AT FINITE FREQUENCIES

W. VAN SAARLOOS* and P. MAZUR

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

Received 17 January 1983

We extend our previously developed scheme to evaluate the static mobility tensors of an arbitrary number of spheres in a viscous fluid, to the case of finite frequencies.

1. Introduction

A well-known problem in low Reynolds number hydrodynamics[1]) is to calculate the hydrodynamic interactions between spherical particles moving in a fluid, as its solution is necessary for understanding the properties of suspensions. While the first treatment of such hydrodynamic interactions dates from 1911 when Smoluchowski[2]) analysed the hydrodynamic friction between two spherical particles, there has recently been a revival of interest in this problem[3-10])†. This renewed interest is greatly motivated by recent advances in experimental light-scattering techniques in which multiple scattering is reduced, either by using scattering cells of different sizes[11,12]), or by using a solvent and solute which have refractive indices that are not too different[13]), or still by using a two-beam, two-detector light scattering spectrometer[14]). These techniques enable one to single out the effects due to hydrodynamic interactions in suspensions, so that a comparison between theory and experiment can be made. As an example, we note that the experiments by Knops–Werkhoven and Fijnaut[13]) on the mutual diffusion coefficient of dilute silica dispersions were in good agreement with the values obtained theoretically by Batchelor[5]) and Felderhof[15]).

In general, correction terms to e.g. the diffusion constant can, for dilute suspensions, be obtained by taking only hydrodynamic pair interactions into account. Since it was doubtful that this is a reasonable approximation for suspensions which are not dilute, we recently developed a systematic expansion to treat the full many sphere problem[10]). This work extended the original analysis

*Present address: Bell Laboratories, 600 Mountain Avenue, Murray Hill New Jersey 07974, USA.
†See refs. 1 and 10 for earlier references.

of Kynch[3]) of many sphere hydrodynamic interactions. From our results, Beenakker and Mazur[16]) determined the concentration dependence of the self-diffusion coefficient to second order in the density, and showed that contributions from two- and three-sphere interactions were of comparable size*. Their result is in reasonable agreement with the values measured by Bauer[12]).

Recent experiments by Pusey and Van Megen[27]) also lead to the conclusion that many sphere hydrodynamic interactions cannot be neglected in moderately dense suspensions.

The analysis of ref. 10 (hereafter to be referred to as paper I) was performed in the static case (frequency zero). In view of the above it seems worthwhile to extend the analysis to the case of finite frequencies. This is the purpose of the present paper. In considering hydrodynamic interactions at finite frequencies ω and low Reynolds numbers, two dimensionless quantities play a role, namely the parameters a/R and $a\sqrt{2\omega/\nu}$. Here a is a typical radius of a sphere, R the typical distance between spheres and ν the kinematic viscosity. As is well known[18]), the quantity $(2\omega/\nu)^{1/2}$ is the inverse penetration depth of transverse waves. Our scheme will yield an expansion in these two dimensionless parameters and is therefore a low frequency ($\omega \ll a^2/\nu$) expansion (the parameter a/R is of course always smaller than $\frac{1}{2}$; for dilute suspensions $a/R \ll \frac{1}{2}$). For the frequency range of interest in most experiments, one has indeed $\omega \ll a^2/\nu$. However, even if a/R and $a(2\omega/\nu)^{1/2}$ are small, there are two different regimes, distinguished by the magnitude of the ratio of these parameters, $R(2\omega/\nu)^{1/2}$. To see this, consider the solution of the flow field of a single sphere oscillating with frequency ω and amplitude u_0, which has the form[18])

$$\boldsymbol{v} = \mathrm{e}^{-\mathrm{i}\omega t}\nabla \wedge \nabla \wedge (f(r)\boldsymbol{u}_0), \tag{1.1}$$

where

$$\frac{\partial f}{\partial r} = \{c_1\,\mathrm{e}^{\mathrm{i}r\sqrt{\mathrm{i}\omega/\nu}}(r + \mathrm{i}/(\mathrm{i}\omega/\nu)^{1/2}) + c_2\}/r^2\,. \tag{1.2}$$

Here r is the distance from the centre of the sphere and c_1 and c_2 are constants that can be obtained from the boundary conditions[18]). Since the imaginary part of $\sqrt{\mathrm{i}\omega/\nu}$ is taken positive, the first term in the expression for $\partial f/\partial r$ is exponentially damped. Consequently, for large distances the flow field is of order r^{-3} and not of order r^{-1}, as in the static case ($\omega = 0$). It is essentially for this reason that hydrodynamic interactions between two spheres at finite frequencies will be of order $(a/R)^3$ if $R(\omega/\nu)^{1/2} \gg 1$ and of order a/R if $R(\omega/\nu)^{1/2} \lesssim 1$. As we will discuss, the first regime is relevant for dilute suspensions, the second for colloidal crystals.

*Recently 3-sphere hydrodynamic interactions were also taken into account by Phillies[17]).

In section 2, we review the equations of motion within the context of the method of induced forces[19,20,8,10]). In section 3 we derive, along the lines of paper I, equations for the velocities and angular velocities of the spheres up to third order in our expansion parameters. Up to this order, the mobility tensors, the expressions of which are given in section 4, contain only two sphere interactions. In a higher order approximation however, they will contain three sphere interactions. Some general results, useful to derive such higher order approximations, are given in appendices.

2. Equations of motion

As in paper I, we consider N macroscopic spheres of masses m_j and radii a_j ($j = 1, \ldots, N$) immersed in an otherwise unbounded incompressible fluid. The centres of the spheres have positions $\boldsymbol{R}_j(t)$ 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.

Contrary to the case considered in paper I, where the analysis was based on the quasistatic Stokes equation for the fluid, we shall now consider the more general case where the fluid obeys the time-dependent linearized Navier–Stokes equation for an incompressible fluid. Within the context of the induced force method[19,20,8,10]), this equation reads, for all $\boldsymbol{r}$

$$\rho \frac{\partial \boldsymbol{v}(\boldsymbol{r}, t)}{\partial t} + \boldsymbol{\nabla} \cdot \boldsymbol{P}(\boldsymbol{r}, t) = \sum_j \boldsymbol{F}_j(\boldsymbol{r}, t) \tag{2.1}$$

with

$$\boldsymbol{\nabla} \cdot \boldsymbol{v}(\boldsymbol{r}, t) = 0 \tag{2.2}$$

and

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

The notations are essentially those of paper I: $\boldsymbol{v}$ is the velocity field, $\boldsymbol{P}$ the pressure tensor, p the hydrostatic pressure and η the viscosity of the fluid. In addition, ρ is the constant fluid density. The index j runs from 1 to N and labels the spheres (so do the indices k and l, to be used later), and Greek indices run from 1 to 3 and denote Cartesian components. The induced forces are defined in such a way that

$$\boldsymbol{F}_j(\boldsymbol{r}, t) \equiv 0 \quad \text{for} \left|\boldsymbol{r} - \boldsymbol{R}_j(t)\right| > a_j\,, \tag{2.4}$$

so that eq. (2.1) reduces to the linearized Navier–Stokes equation within the fluid.

In addition, in the induced force method it is required that

$$\boldsymbol{v}(\boldsymbol{r}, t) = \boldsymbol{u}_j(t) + \boldsymbol{\Omega}_j(t) \wedge [\boldsymbol{r} - \boldsymbol{R}_j(t)] \quad \text{for } |\boldsymbol{r} - \boldsymbol{R}_j(t)| \leqslant a_j, \tag{2.5}$$

so that stick boundary conditions hold at the surfaces of the spheres, and that

$$p(\boldsymbol{r}, t) = 0, \quad \text{for } |\boldsymbol{r} - \boldsymbol{R}_j(t)| < a_j. \tag{2.6}$$

The velocity $\boldsymbol{u}_j$ and the angular velocity $\boldsymbol{\Omega}_j$ of the jth sphere* obey the equations of motion

$$m_j \frac{\mathrm{d}\boldsymbol{u}_j(t)}{\mathrm{d}t} = - \int\limits_{S_j(t)} \mathrm{d}S \boldsymbol{P}(\boldsymbol{r}, t) \cdot \hat{n}_j + \boldsymbol{K}_j^{\mathrm{ext}}(t) \equiv \boldsymbol{K}_j(t) + \boldsymbol{K}_j^{\mathrm{ext}}(t), \tag{2.7}$$

$$I_j \frac{\mathrm{d}\boldsymbol{\Omega}_j(t)}{\mathrm{d}t} = - \int\limits_{S_j(t)} \mathrm{d}S [\boldsymbol{r} - \boldsymbol{R}_j(t)] \wedge \boldsymbol{P} \cdot \hat{n}_j + \boldsymbol{T}_j^{\mathrm{ext}} \equiv \boldsymbol{T}_j^{\mathrm{ext}}(t). \tag{2.8}$$

Here, $\boldsymbol{K}_j$, $\boldsymbol{T}_j$, $\boldsymbol{K}_j^{\mathrm{ext}}$, $\boldsymbol{T}_j^{\mathrm{ext}}$ are, respectively, the force and torque exerted by the fluid on sphere j, and the external force and torque on this sphere. $S_j(t)$ is the surface of sphere j at time t, $\hat{n}_j$ a unit vector normal to this surface and pointing in the outward direction, and $I_j = 2m_j a_j^2/5$ its moment of inertia (where a homogeneous mass distribution has been assumed).

For one sphere, one may neglect within the fully linearized scheme, the time dependence of its surface S and its position $\boldsymbol{R}$**. We shall make the same approximation here, i.e. we will also neglect for the case of many spheres the time dependences of the surfaces S_j and the positions $\boldsymbol{R}_j$ of sphere j. In doing so, we assume that the sphere-positions do not change appreciably over time intervals of interest, or, alternatively, that the spheres perform harmonic motion with small amplitudes around arbitrary equilibrium positions. It is then convenient to introduce the Fourier-transform with respect to time of the various quantities, e.g.

$$\boldsymbol{u}_j(\omega) = \int \mathrm{d}t \, \mathrm{e}^{\mathrm{i}\omega t} \boldsymbol{u}_j(t). \tag{2.9}$$

Eqs. (2.1), (2.7) and (2.8) then become

$$- \mathrm{i}\omega\rho \boldsymbol{v}(\boldsymbol{r}, \omega) + \boldsymbol{\nabla} \cdot \boldsymbol{P}(\boldsymbol{r}, \omega) = \sum_j \boldsymbol{F}_j(\boldsymbol{r}, \omega), \tag{2.10}$$

$$- \mathrm{i}\omega m_j \boldsymbol{u}_j(\omega) = - \int\limits_{S_j} \mathrm{d}S \, \boldsymbol{P}(\boldsymbol{r}, \omega) \cdot \hat{n}_j + \boldsymbol{K}_j^{\mathrm{ext}} = \boldsymbol{K}_j(\omega) + \boldsymbol{K}_j^{\mathrm{ext}}(\omega), \tag{2.11}$$

*Here the notation deviates from the one in paper I: $\boldsymbol{\Omega}$ (*and not* $\boldsymbol{\omega}$) denotes here the angular velocity.

** Including the time dependence of S_j and $\boldsymbol{R}_j$ in the case of stationary motion of a single sphere, amounts in fact to analysing this motion in the Oseen approximation[21]).

$$-\mathrm{i}\omega I_j \boldsymbol{\Omega}_j(\omega) = -\int_{S_j} \mathrm{d}S[\boldsymbol{r}-\boldsymbol{R}_j] \wedge \boldsymbol{P}(\boldsymbol{r},\omega)\cdot\hat{n}_j + \boldsymbol{T}_j^{\mathrm{ext}}(\omega) \equiv \boldsymbol{T}_j(\omega) + \boldsymbol{T}_j^{\mathrm{ext}}(\omega)\,. \tag{2.12}$$

If one now uses eqs. (2.4)–(2.6), in which the time dependence of $\boldsymbol{R}_j$ is neglected, it follows from eq. (2.10) that $\boldsymbol{F}_j(\boldsymbol{r},\omega)$ is of the form

$$\boldsymbol{F}_j(\boldsymbol{r},\omega) = a_j^{-2}\boldsymbol{f}_j(\hat{n}_j,\omega)\delta(|\boldsymbol{r}-\boldsymbol{R}_j|-a_j) - \mathrm{i}\omega\rho[\boldsymbol{u}_j(\omega) + \boldsymbol{\Omega}_j(\omega)\wedge(\boldsymbol{r}-\boldsymbol{R}_j)]\Theta(a_j-|\boldsymbol{r}-\boldsymbol{R}_j|) \equiv \boldsymbol{F}_j^{\mathrm{s}}(\boldsymbol{r},\omega) + \boldsymbol{F}_j^{\mathrm{v}}(\boldsymbol{r},\omega)\,. \tag{2.13}$$

Here $\Theta(x)$ is the Heaviside function, and $\boldsymbol{F}_j^{\mathrm{s}}(\boldsymbol{r},\omega)$ and $\boldsymbol{F}_j^{\mathrm{v}}(\boldsymbol{r},\omega)$ are the force densities induced on the surface of and within the sphere j, respectively.

From eqs. (2.11)–(2.13) one obtains along the same lines as in paper I, section 2, relations between on the one side the hydrodynamic force $\boldsymbol{K}_j(\omega)$ and torque $\boldsymbol{T}_j(\omega)$ exerted on the jth sphere, and on the other side the component of the force $\boldsymbol{F}_j^{\mathrm{s}}$ which is induced at the surface of sphere j. These relations are

$$\boldsymbol{K}_j(\omega) = -\int_{S_j} \mathrm{d}S\boldsymbol{P}(\boldsymbol{r},\omega)\cdot\hat{n}_j = -\int_{|\boldsymbol{r}-\boldsymbol{R}_j(t)|\leqslant a_j} \mathrm{d}\boldsymbol{r}\,\nabla\cdot\boldsymbol{P}(\boldsymbol{r},\omega) = -\int \mathrm{d}\boldsymbol{r}\,\boldsymbol{F}_j^{\mathrm{s}}(\boldsymbol{r},\omega)\,, \tag{2.14}$$

$$\boldsymbol{T}_j(\omega) = -\int_{S_j} \mathrm{d}S[\boldsymbol{r}-\boldsymbol{R}_j]\wedge\boldsymbol{P}(\boldsymbol{r},\omega)\cdot\hat{n}_j = -\int \mathrm{d}\boldsymbol{r}[\boldsymbol{r}-\boldsymbol{R}_j]\wedge\boldsymbol{F}_j^{\mathrm{s}}(\omega)\,. \tag{2.15}$$

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

$$\boldsymbol{v}(\boldsymbol{k},\omega) = \int \mathrm{d}\boldsymbol{r}\;\mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}\boldsymbol{v}(\boldsymbol{r},\omega)\,. \tag{2.16}$$

We also define the Fourier transform of the induced force density $\boldsymbol{F}_j$ in a reference frame in which sphere j is at rest at the origin,

$$\boldsymbol{F}_j(\boldsymbol{k},\omega) = \int \mathrm{d}\boldsymbol{r}\;\mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{r}-\boldsymbol{R}_j)}\boldsymbol{F}_j(\boldsymbol{r},\omega)\,. \tag{2.17}$$

The equations of motion (2.10) and (2.2) then become in wave-vector representation

$$(-\mathrm{i}\omega\rho + \eta k^2)\boldsymbol{v}(\boldsymbol{k},\omega) + \mathrm{i}\boldsymbol{k}p(\boldsymbol{k},\omega) = \sum_j \mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_j}\boldsymbol{F}_j(\boldsymbol{k},\omega)\,, \tag{2.18}$$

$$\boldsymbol{k}\cdot\boldsymbol{v}(\boldsymbol{k},\omega) = 0\,. \tag{2.19}$$

If one applies the operator $\mathbb{1} - \hat{k}\hat{k}$ (where $\mathbb{1}$ is the unit tensor and $\hat{k}$ a unit vector in the direction of $\boldsymbol{k}$) to both sides of eq. (2.18), one obtains with eq. (2.19)

$$(-\mathrm{i}\omega\rho + \eta k^2)\boldsymbol{v}(\boldsymbol{k}, \omega) = \sum_j (\mathbb{1} - \hat{k}\hat{k}) \cdot \boldsymbol{F}_j(\boldsymbol{k}, \omega)\,. \tag{2.20}$$

If the fluid would be at rest if unperturbed by the motion of the spheres, the formal solution of eq. (2.20) is

$$\begin{aligned}\boldsymbol{v}(\boldsymbol{k}, \omega) &= \sum_j (-\mathrm{i}\omega\rho + \eta k^2)^{-1}\, \mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_j}(\mathbb{1} - \hat{k}\hat{k}) \cdot \boldsymbol{F}_j(\boldsymbol{k}, \omega) \\ &= \sum_j \eta^{-1}(\alpha^2 + k^2)^{-1}\, \mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_j}(\mathbb{1} - \hat{k}\hat{k}) \cdot \boldsymbol{F}_j(\boldsymbol{k}, \omega)\,.\end{aligned} \tag{2.21}$$

Here the parameter α is defined as

$$\alpha \equiv \sqrt{-\mathrm{i}\omega\rho/\eta}, \quad \mathrm{Re}\,\alpha > 0\,. \tag{2.22}$$

Its real part represents the inverse penetration length $\sqrt{2\eta/\rho\omega}$ of planar waves of frequency ω[18]. It is eq. (2.21) which we shall use to calculate the frequency-dependent forces and torques exerted by the fluid on the spheres.

We will also need an expansion of $\boldsymbol{F}_j^{\mathrm{s}}(\boldsymbol{k}, \omega)$ in terms of irreducible multipoles $\boldsymbol{F}_j^{(p+1)}(\omega)$, defined by

$$\boldsymbol{F}_j^{(p+1)}(\omega) = (p!)^{-1} \int \mathrm{d}\hat{n}_j\, \overline{\hat{n}_j^p} \boldsymbol{f}_j(\hat{n}_j, \omega) = (\mathrm{i}/a_j)^p (p!)^{-1} \left[\frac{\overline{\partial^p}}{\partial \boldsymbol{k}^p} \boldsymbol{F}_j^{\mathrm{s}}(\boldsymbol{k}, \omega) \right]_{k=0} \quad (p \geqslant 0)\,. \tag{2.23}$$

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 $\boldsymbol{b}$ (cf. paper I, section 3, or ref. 22). Note also that one has according to eq. (2.14) and (2.15)

$$\boldsymbol{F}_j^{(1)}(\omega) = -\boldsymbol{K}_j(\omega), \quad \boldsymbol{F}_j^{(2a)}(\omega) = -(2a_j)^{-1} \boldsymbol{\epsilon} \cdot \boldsymbol{T}_j(\omega)\,, \tag{2.24}$$

where $\boldsymbol{F}^{(2a)}$ is the antisymmetric part of $\boldsymbol{F}^{(2)}$ and $\boldsymbol{\epsilon}$ the Levi-Civita tensor (cf. paper I, eqs. (3.10)–(3.12)).

In paper I (cf. eq. (3.14) and appendix A), we have shown that $\boldsymbol{F}_j^{\mathrm{s}}$ can be expanded as

$$\boldsymbol{F}_j^{\mathrm{s}}(\boldsymbol{k}, \omega) = \sum_{p=0}^{\infty} (2p+1)!!(\mathrm{i}/a_j)^p \left(\frac{\overline{\partial^p}}{\partial \boldsymbol{k}^p} \frac{\sin k a_j}{k a_j} \right) \odot \boldsymbol{F}_j^{(p+1)}(\omega)\,. \tag{2.25}$$

Here, the dot $\odot$ denotes a full contraction over the first p indices of $\boldsymbol{F}^{(p+1)}$ and the p indices of the term between brackets. This expansion, on which our analysis of many-sphere hydrodynamics in the static case was based, will be employed here for finite frequencies too.

3. Equations for $\boldsymbol{u}_j(\omega)$ and $\boldsymbol{\Omega}_j(\omega)$

We have shown in paper I that one obtains a hierarchy of equations for the force multipoles induced in the spheres by analyzing the so-called velocity surface moments. We shall follow here in principle the same procedure. However, if we restrict ourselves to contributions to the mobility-tensors up to third order in αa and a/R, where a and R are a typical sphere radius and intersphere distance respectively, it will be sufficient, as we shall see, to consider only the first two moments. This is done in the next subsections. The exact equation resulting from the zeroth surface moments is given in eq. (3.3). It is then argued that up to the order considered in this paper, this equation reduces to relation (3.24). Similarly, in subsection 3.2 it is shown that the exact result (3.26) reduces to eq. (3.39) in the same approximation. Eqs. (3.24) and (3.39) yield the expressions for the mobilities.

3.1. *Zeroth order surface moment*

We first consider the zeroth order surface moment, defined as

$$\overline{\boldsymbol{v}(\boldsymbol{r},\omega)}^{S_j} \equiv \frac{1}{4\pi a_j^2}\int \mathrm{d}\boldsymbol{r}\, \boldsymbol{v}(\boldsymbol{r},\omega)\delta(|\boldsymbol{r}-\boldsymbol{R}_j| - a_j)$$
$$= \frac{1}{(2\pi)^3}\int \mathrm{d}\boldsymbol{k}\, \frac{\sin ka_j}{ka_j}\, \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_j}\boldsymbol{v}(\boldsymbol{k},\omega)\,. \tag{3.1}$$

Applying the boundary conditions (2.5) and substituting the formal solution (2.21) at the right hand side, one obtains the following set of N equations

$$6\pi\eta a_j \boldsymbol{u}_j(\omega) = \frac{3}{8\pi^2}\int \mathrm{d}\hat{k}\int_{-\infty}^{+\infty} \mathrm{d}k\, \frac{k\sin ka_j}{k^2+\alpha^2}(\mathbb{1}-\hat{k}\hat{k})\cdot \boldsymbol{F}_j(\boldsymbol{k},\omega)$$
$$+ \sum_{k\neq j}\frac{3}{8\pi^2}\int \mathrm{d}\hat{k}\int_{-\infty}^{+\infty} \mathrm{d}k\, \frac{k\sin ka_j}{k^2+\alpha^2}\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_j-\boldsymbol{R}_k)}(\mathbb{1}-\hat{k}\hat{k})\cdot \boldsymbol{F}_k(\boldsymbol{k},\omega)\,. \tag{3.2}$$

One may now use the decomposition (2.13) as well as the series expansion (2.25) in the integrals on the r.h.s. One then gets

$$6\pi\eta a_j\left(\boldsymbol{u}_j(\omega) + \sum_k \alpha^2 a_k^2 \boldsymbol{M}_{jk}^{(1,1)}(\omega)\cdot \boldsymbol{u}_k(\omega) + \sum_k \alpha^2 a_k^2 \boldsymbol{M}_{jk}^{(1,2)}(\omega)\cdot \boldsymbol{\Omega}(\omega)\right)$$
$$= -\sum_{m=1}^{\infty} \boldsymbol{B}_j^{(1,m)}(\omega)\odot \boldsymbol{F}_j^{(m)}(\omega) + \sum_{k\neq j}\sum_{m=1}^{\infty} \boldsymbol{A}_{jk}^{(1,m)}(\omega)\odot \boldsymbol{F}_k^{(m)}(\omega)\,. \tag{3.3}$$

This equation is the analogue of eq. (4.9) of paper I for finite frequencies. The connectors $\boldsymbol{B}^{(1,m)}(\omega)$, $\boldsymbol{A}^{(1,m)}(\omega)$, $\boldsymbol{M}^{(1,1)}(\omega)$ and $\boldsymbol{M}^{(1,2)}(\omega)$ are defined as

$$\boldsymbol{B}_j^{(1,m)}(\omega) = -(2m-1)!!\left(\frac{\mathrm{i}}{a_j}\right)^{m-1} \frac{3}{8\pi^2} \int \mathrm{d}\hat{k} \int_{-\infty}^{+\infty} \mathrm{d}k \frac{k \sin ka_j}{k^2+\alpha^2} \times (\mathbb{1} - \hat{k}\hat{k})\left(\overline{\frac{\partial^{m-1}}{\partial \boldsymbol{k}^{m-1}}} \frac{\sin ka_j}{ka_j}\right), \tag{3.4}$$

$$\boldsymbol{A}_{jk}^{(1,m)}(\omega) = (2m-1)!!\left(\frac{\mathrm{i}}{a_k}\right)^{m-1} \frac{3}{8\pi^2} \int \mathrm{d}\hat{k} \int_{-\infty}^{+\infty} \mathrm{d}k \frac{k \sin ka_j}{k^2+\alpha^2} \times (\mathbb{1} - \hat{k}\hat{k})\left(\overline{\frac{\partial^{m-1}}{\partial \boldsymbol{k}^{m-1}}} \frac{\sin ka_k}{ka_k}\right) \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_j - \boldsymbol{R}_k)}, \tag{3.5}$$

$$\boldsymbol{M}_{jk}^{(1,1)}(\omega) = -\frac{a_k}{4\pi^2 a_j} \int \mathrm{d}\hat{k} \int_{-\infty}^{+\infty} \mathrm{d}k \frac{k \sin ka_j}{k^2+\alpha^2} \left[\frac{\sin ka_k - ka_k \cos ka_k}{(ka_k)^3}\right] \times e^{\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_j - \boldsymbol{R}_k)}(\mathbb{1} - \hat{k}\hat{k}), \tag{3.6}$$

$$\boldsymbol{M}_{jk}^{(1,2)}(\omega) = -\frac{\mathrm{i}a_k}{4\pi^2 a_j} \int \mathrm{d}\hat{k} \int_{-\infty}^{+\infty} \mathrm{d}k \frac{k \sin ka_j}{k^2+\alpha^2} \frac{\partial}{\partial k} \left[\frac{\sin ka_k - ka_k \cos ka_k}{(ka_k)^3}\right] \times \hat{k} \cdot \boldsymbol{\epsilon}\, \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_j - \boldsymbol{R}_k)}. \tag{3.7}$$

In eqs. (3.6) and (3.7), the function between square brackets arises from the Fourier transform of the Θ-function in eq. (2.13); $\boldsymbol{\epsilon}$ denotes, as before, the Levi–Civita tensor. Note also that the expressions for $\boldsymbol{B}_j^{(1,m)}(\omega)$ and $\boldsymbol{A}_{jk}^{(1,m)}(\omega)$ reduce for $\omega = 0$ to those given in paper I for the static case (cf. eqs. (C.1) and (4.32)).

We shall now successively discuss the behaviour of the various connectors.

(a) According to eq. (3.3) the connectors $\boldsymbol{M}^{(1,1)}$ and $\boldsymbol{M}^{(1,2)}$ must in our approximation be taken into account to first order in αa and a/R. In appendix A, we show that one has up to this order

$$\boldsymbol{M}_{jk}^{(1,1)}(\omega) = \begin{cases} -\frac{2}{9}\mathbb{1}(1 - \alpha a + \text{h.o.}) & \text{for } j = k \\ -\frac{1}{6}\frac{a_k}{R_{jk}} \mathrm{e}^{-\alpha R_{jk}}(\mathbb{1} + \hat{r}_{jk}\hat{r}_{jk}) & \\ -\frac{1}{2}\frac{a_k}{R_{jk}}\left(\frac{2}{\alpha^2 R_{jk}^2} - \mathrm{e}^{-\alpha R_{jk}}\left(1 + \frac{2}{\alpha R_{jk}} + \frac{2}{\alpha^2 R_{jk}^2}\right)\right)(\hat{r}_{jk}\hat{r}_{jk} - \tfrac{1}{3}\mathbb{1}), & \text{for } j \neq k, \end{cases} \tag{3.8}$$

$$\boldsymbol{M}_{jk}^{(1,2)}(\omega) = \begin{cases} 0 & \text{for } j = k, \\ \mathcal{O}((\alpha a)^{2-p}(a/R)^p), \quad p = 0, 1, 2 & \text{for } j \neq k. \end{cases} \tag{3.9}$$

Here $R_{jk} \equiv |\boldsymbol{R}_k - \boldsymbol{R}_j|$ is the distance between the centres of sphere j and k, and $\hat{r}_{jk} \equiv (\boldsymbol{R}_k - \boldsymbol{R}_j)/R_{jk}$ is a unit vector pointing from sphere j to sphere k. In eq. (3.8), we have denoted terms which are of higher order in αa and a/R by h.o. The lowest order term for $j \neq k$ has a coefficient proportional to $\exp -\alpha R_{jk}$ due to the exponential damping of velocity perturbations which decay as $1/r$ (cf. the introduction). That $\boldsymbol{M}_{jj}^{(1,2)}(\omega)$ vanishes, simply expresses the well known fact that translation and rotation of a single sphere do not couple.

(b) For the connectors $\boldsymbol{B}^{(1,m)}(\omega)$, we show in appendix B that*

$$\boldsymbol{B}_j^{(1,m)}(\omega) = \begin{cases} 0 & \text{for } m \neq 1 \text{ and } m \neq 3\,, \\ \mathcal{O}(1) & \text{for } m = 1\,, \\ \mathcal{O}(\alpha a_j)^2 & \text{for } m = 3\,. \end{cases} \tag{3.10}$$

$\boldsymbol{B}^{(1,3)}(\omega)$ multiplies the multipole moment $\boldsymbol{F}^{(3)}(\omega)$ which is of third order in the parameters in which we expand. This is best illustrated by the results of paper I, where we have shown that at zero frequency $\boldsymbol{F}^{(3)}(0)$ is of order $(a/R)^3$. The extension to the case of non-zero frequencies follows from the result, to be discussed sub (c), that the ordering of the frequency dependent connectors in a/R and αa is essentially the same as for the static ones. To the order retained here, we therefore only need to calculate $\boldsymbol{B}^{(1,1)}(\omega)$ up to order $\alpha^3 a^3$. One finds (see appendix B)

$$\boldsymbol{B}_j^{(1,1)}(\omega) = -\mathbb{1}(1 - \alpha a_j + \tfrac{2}{3}\alpha^2 a_j^2 - \tfrac{1}{3}\alpha^3 a_j^3 + \dots)\,. \tag{3.11}$$

(c) Finally, we turn to the behaviour of the connectors $\boldsymbol{A}^{(1,m)}(\omega)$. We show in appendix C that $\boldsymbol{A}^{(1,m)}(\omega)$ is of the form

$$\boldsymbol{A}_{jk}^{(1,m)}(\omega) = \boldsymbol{G}_{jk}^{(1,m)}(\omega)/R_{jk}^m + \boldsymbol{H}_{jk}^{(1,m)}(\omega)/R_{jk}^{m+2} + \boldsymbol{L}_{jk}^{(1,m)}(\omega, R_{jk})\,. \tag{3.12}$$

The frequency dependent tensors $\boldsymbol{G}_{jk}^{(1,m)}(\omega)$ and $\boldsymbol{H}_{jk}^{(1,m)}(\omega)$, which do not depend on the interparticle distance R_{jk}, reduce in the limit $\omega \to 0$ to the corresponding tensors $\boldsymbol{G}_{jk}^{(1,m)}$ and $\boldsymbol{H}_{jk}^{(1,m)}$ calculated in paper I (eqs. (5.10)–(5.12)). These tensors are proportional to a^m and a^{m+2}, respectively, and have, as is discussed in appendix C, a power series expansion in the parameter $\alpha^2 a^2$, so that for small frequencies

$$R_{jk}^{-m}\boldsymbol{G}_{jk}^{(1,m)}(\omega) = R_{jk}^{-m}\boldsymbol{G}_{jk}^{(1,m)}(\omega = 0)(1 + \mathcal{O}(\alpha a)^2) = \mathcal{O}(a/R)^m(1 + \mathcal{O}(\alpha a)^2)\,, \tag{3.13}$$

$$\begin{aligned} R_{jk}^{-m-2}\boldsymbol{H}_{jk}^{(1,m)}(\omega) &= R_{jk}^{-m-2}\boldsymbol{H}_{jk}^{(1,m)}(\omega = 0)(1 + \mathcal{O}(\alpha a)^2) \\ &= \mathcal{O}(a/R)^{m+2}(1 + \mathcal{O}(\alpha a)^2)\,. \end{aligned} \tag{3.14}$$

* It follows from eq. (3.10) that in the case of one sphere, $\boldsymbol{F}^{(1)}$ only couples to $\boldsymbol{F}^{(3)}$. This result is implicit in the work of Mazur and Van der Zwan[20].

The behaviour of the tensor $\boldsymbol{L}_{jk}^{(1,m)}(\omega, R_{jk})$ strongly depends on the parameter αR_{jk}, which is the ratio of the two parameters αa and a/R, in which we expand. The behaviour in the regime $\alpha R < 1$ is essentially determined by the limit (cf. appendix C)

$$\lim_{\omega\to 0}(\alpha a)^{-1}\boldsymbol{L}_{jk}^{(1,1)}(\omega, R_{jk}) = \text{constant}\,,$$

$$\lim_{\omega\to 0}(\alpha a)^{-2}\boldsymbol{L}_{jk}^{(1,m)}(\omega, R_{jk}) = \mathcal{O}(a/R_{jk})^{m-2}, \quad m \geqslant 2\,, \tag{3.15}$$

while for $\alpha R > 1$ the behaviour of $\boldsymbol{L}^{(1,m)}$ is governed by the limit

$$\lim_{R_{jk}\to\infty} R_{jk}^{m}\boldsymbol{L}_{jk}^{(1,m)}(\omega, R_{jk}) = -\,\boldsymbol{G}_{jk}^{(1,m)}(\omega)\,. \tag{3.16}$$

To summarize, it follows from eqs. (3.12)–(3.16) that the connector $\boldsymbol{A}_{jk}^{(1,m)}(\omega)$ is of the mth order in our expansion parameters if $\alpha R < 1$, while it is even smaller for $\alpha R \gg 1$, since the most dominant term of $\boldsymbol{L}^{(1,m)}(\omega)$ then cancels $\boldsymbol{G}^{(1,m)}(\omega)$. Thus, one may again neglect, for the same reasons as given in sub (b), all terms involving connectors $\boldsymbol{A}^{(1,m)}(\omega)$ with $m \geqslant 3$, as well as the term involving $\boldsymbol{A}^{(1,2s)}(\omega)$, which is that part of $\boldsymbol{A}^{(1,2)}(\omega)$ which is symmetric in its last two indices.* For $\boldsymbol{A}^{(1,1)}(\omega)$ which couples to the force, we have up to third order (cf. appendix C)

$$\boldsymbol{G}_{jk}^{(1,1)}(\omega)/R_{jk} = \frac{3}{4}\frac{a_j}{R_{jk}}\left(1 + \frac{1}{6}(a_j^2 + a_k^2)\alpha^2\right)(\mathbb{1} + \hat{\boldsymbol{r}}_{jk}\hat{\boldsymbol{r}}_{jk})\,, \tag{3.17}$$

$$\boldsymbol{H}_{jk}^{(1,1)}(\omega)/R_{jk} = -\frac{3}{4}\frac{a_j(a_j^2 + a_k^2)}{R_{jk}^3}\left(\hat{\boldsymbol{r}}_{jk}\hat{\boldsymbol{r}}_{jk} - \frac{1}{3}\mathbb{1}\right), \tag{3.18}$$

$$\begin{aligned}\boldsymbol{L}^{(1,1)}(\omega, R_{jk}) = {} & -\frac{3}{4}\frac{a_j}{R_{jk}}\left(1 + \frac{1}{6}(a_j^2 + a_k^2)\alpha^2\right)(1 - \mathrm{e}^{-\alpha R_{jk}})(\mathbb{1} + \hat{\boldsymbol{r}}_{jk}\hat{\boldsymbol{r}}_{jk}) \\ & + \frac{9}{4}\alpha a_j\left(1 + \frac{1}{6}(a_j^2 + a_k^2)\alpha^2\right)\left(\frac{2}{\alpha^3 R_{jk}^3}\right. \\ & \left.- \mathrm{e}^{-\alpha R_{jk}}\left(\frac{1}{\alpha R_{jk}} + \frac{2}{\alpha^2 R_{jk}^2} + \frac{2}{\alpha^3 R_{jk}^3}\right)\right)\left(\hat{\boldsymbol{r}}_{jk}\hat{\boldsymbol{r}}_{jk} - \frac{1}{3}\mathbb{1}\right).\end{aligned} \tag{3.19}$$

For $\boldsymbol{A}^{(1,2a)}(\omega)$, that part of $\boldsymbol{A}^{(1,2)}(\omega)$ which is antisymmetric in its last two indices and which couples to the torque, we only need to evaluate $\boldsymbol{G}^{(1,2a)}(\omega)$ and $\boldsymbol{L}^{(1,2a)}(\omega)$,

* According to table I of paper I the connectors $\boldsymbol{A}^{(1,m)}(\omega = 0)$ with $m \geqslant 3$ and $\boldsymbol{A}^{(1,2s)}(\omega = 0)$ give contributions to the mobilities of fourth and higher order in a/R. Since the ordering of the frequency dependent connectors in a/R and αa is essentially the same as for the static ones, we only need to retain $\boldsymbol{A}^{(1,1)}(\omega)$ and $\boldsymbol{A}^{(1,2a)}(\omega)$ in eq. (3.3) in an expansion up to third order.

since the term involving $\boldsymbol{H}^{(1,2)}(\omega)$ is of fourth order*. We find up to third order

$$(\boldsymbol{G}_{jk}^{(1,2a)}(\omega))_{\alpha\beta\gamma} = \tfrac{3}{4}\, a_j a_k (r_{jk\beta}\delta_{\alpha\gamma} - r_{jk\gamma}\delta_{\alpha\beta}) , \tag{3.20}$$

$$(\boldsymbol{L}_{jk}^{(1,2a)}(\omega, \boldsymbol{R}_{jk}))_{\alpha\beta\gamma} = -\tfrac{3}{4}\frac{a_j a_k}{R_{jk}^2}(1 - \mathrm{e}^{-\alpha R_{jk}}(1 + \alpha R_{jk}))(r_{jk\beta}\delta_{\alpha\gamma} - r_{jk\gamma}\delta_{\alpha\beta}) . \tag{3.21}$$

From the above discussion, sub(a), (b) and (c), it follows that eq. (3.3) reduces to third order to

$$\begin{aligned} &6\pi\eta a_j\Big[(1 - \tfrac{2}{9}\alpha^2 a_j^2 + \tfrac{2}{9}\alpha^3 a_j^3)\boldsymbol{u}_j(\omega) + \sum_{k\neq j} \alpha^2 a_k^2 \boldsymbol{M}_{jk}^{(1,1)}(\omega)\cdot \boldsymbol{u}_k(\omega)\Big] \\ &\quad = -(1 - \alpha a_j + \tfrac{2}{3}\alpha^2 a_j^2 - \tfrac{1}{3}\alpha^3 a_j^3)\boldsymbol{K}_j(\omega) \\ &\qquad - \sum_{k\neq j} \boldsymbol{A}_{jk}^{(1,1)}(\omega)\cdot \boldsymbol{K}_k(\omega) - \sum_{k\neq j}\frac{1}{2a_k}\boldsymbol{A}_{jk}^{(1,2a)}(\omega):\boldsymbol{\epsilon}\cdot \boldsymbol{T}_j(\omega) . \end{aligned} \tag{3.22}$$

Here, use has been made of eq. (2.24). The connectors appearing in this equation are given in eqs. (3.8), (3.12), (3.17)–(3.21).

As the term involving $\boldsymbol{M}^{(1,1)}$ in this equation is already of third order in our expansion parameters (cf. eq. (3.8)), we may eliminate $\boldsymbol{u}_k(\omega)$ in this term in favour of $\boldsymbol{K}_k(\omega)$ to zeroth order,

$$\boldsymbol{u}_k(\omega) = -(6\pi\eta a_k)^{-1}\boldsymbol{K}_k(\omega) + \text{h.o.} . \tag{3.23}$$

Eq. (3.22) then becomes

$$\begin{aligned} &6\pi\eta a_j(1 - \tfrac{2}{9}\alpha^2 a_j^2 + \tfrac{2}{9}\alpha^3 a_j^3)\boldsymbol{u}_j(\omega) \\ &\quad = -(1 - \alpha a_j + \tfrac{2}{3}\alpha^2 a_j^2 - \tfrac{1}{3}\alpha^3 a_j^3)\boldsymbol{K}_j(\omega) + \sum_{k\neq j}\alpha^2 a_k a_j \boldsymbol{M}_{jk}^{(1,1)}(\omega)\cdot \boldsymbol{K}_k(\omega) \\ &\qquad - \sum_{k\neq j} \boldsymbol{A}_{jk}^{(1,1)}(\omega)\cdot \boldsymbol{K}_k(\omega) - \sum_{k\neq j}\frac{1}{2a_k}\boldsymbol{A}_{jk}^{(1,2a)}(\omega):\boldsymbol{\epsilon}\cdot \boldsymbol{T}_j(\omega) . \end{aligned} \tag{3.24}$$

3.2. *First order surface moment*

We now turn to the evaluation of the first order surface moment

$$\begin{aligned} 3\overline{\hat{n}_j \boldsymbol{v}(\boldsymbol{r}, \omega)}^{S_j} &\equiv \frac{3}{4\pi a_j^2}\int \mathrm{d}\boldsymbol{r}\left(\frac{\boldsymbol{r} - \boldsymbol{R}_j}{a_j}\right)\boldsymbol{v}(\boldsymbol{r}, \omega)\delta(|\boldsymbol{r} - \boldsymbol{R}_j| - a_j) \\ &= -\frac{3\mathrm{i}}{8\pi^3 a_j}\int \mathrm{d}\boldsymbol{k}\, \hat{\boldsymbol{k}}\boldsymbol{v}(\boldsymbol{k}, \omega)\left(\frac{\partial}{\partial k}\frac{\sin k a_j}{k a_j}\right) . \end{aligned} \tag{3.25}$$

Along the same lines as for the zeroth order surface moment (cf. eqs. (3.2) and

* Moreover, it turns out that $\boldsymbol{H}^{(1,2)}(\omega)$ is fully symmetric, cf. paper I, eq. (5.13), so that $\boldsymbol{H}^{(1,2a)}(\omega)$ in fact vanishes identically.

(3.3)), we now obtain

$$6\pi\eta a_j^2\left(\boldsymbol{\epsilon}\cdot\boldsymbol{\Omega}_j(\omega)+\sum_k \alpha^2 a_k^2 \boldsymbol{M}_{jk}^{(2,1)}(\omega)\cdot\boldsymbol{u}_j(\omega)+\sum_k \alpha^2 a_k^2 \boldsymbol{M}_{jk}^{(2,2)}(\omega)\cdot\boldsymbol{\Omega}_k(\omega)\right)$$
$$=-\sum_{m=1}^{\infty}\boldsymbol{B}_j^{(2,m)}(\omega)\odot\boldsymbol{F}_j^{(m)}(\omega)+\sum_{m=1}^{\infty}\sum_{k\neq j}\boldsymbol{A}_{jk}^{(2,m)}(\omega)\odot\boldsymbol{F}_k^{(m)}(\omega) \tag{3.26}$$

with

$$\boldsymbol{M}_{jk}^{(2,1)}(\omega)\equiv\frac{3\mathrm{i}a_k}{4\pi^2 a_j^2}\int \mathrm{d}\hat{k}\int_{-\infty}^{+\infty}\mathrm{d}k\,\hat{k}(\mathbb{1}-\hat{k}\hat{k})\frac{k^2}{k^2+\alpha^2}\left(\frac{\partial}{\partial k}\frac{\sin ka_j}{ka_j}\right)$$
$$\times\left[\frac{\sin ka_k-ka_k\cos ka_k}{k^3a_k^3}\right]\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_j-\boldsymbol{R}_k)}, \tag{3.27}$$

$$\boldsymbol{M}_{jk}^{(2,2)}(\omega)\equiv-\frac{3a_k}{4\pi^2 a_j}\int \mathrm{d}\hat{k}\int_{-\infty}^{+\infty}\mathrm{d}k\,\hat{k}\hat{k}\cdot\boldsymbol{\epsilon}\frac{k^2}{k^2+\alpha^2}\left(\frac{\partial}{\partial k}\frac{\sin ka_j}{ka_j}\right)$$
$$\times\frac{\partial}{\partial k}\left[\frac{\sin ka_k-ka_k\cos ka_k}{k^3a_k^3}\right]\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_j-\boldsymbol{R}_k)}, \tag{3.28}$$

$$\boldsymbol{B}_j^{(2,m)}(\omega)\equiv\frac{9a_j^{1-m}(2m-1)!!\mathrm{i}^m}{8\pi^2}\int \mathrm{d}\hat{k}\int_{-\infty}^{+\infty}\mathrm{d}k\,\hat{k}(\mathbb{1}-\hat{k}\hat{k})\left(\overline{\frac{\partial^{m-1}}{\partial\boldsymbol{k}}}\frac{\sin ka_j}{ka_j}\right)$$
$$\times\left(\frac{\partial}{\partial k}\frac{\sin ka_j}{ka_j}\right)\frac{k^2}{k^2+\alpha^2}, \tag{3.29}$$

$$\boldsymbol{A}_{jk}^{(2,m)}(\omega)\equiv-\frac{9a_k^{m-1}\mathrm{i}^m(2m-1)!!}{8\pi^2}\int \mathrm{d}\hat{k}\int_{-\infty}^{+\infty}\mathrm{d}k\,\hat{k}(\mathbb{1}-\hat{k}\hat{k})\left(\overline{\frac{\partial^{m-1}}{\partial\boldsymbol{k}}}\frac{\sin ka_k}{ka_k}\right)$$
$$\times\frac{k^2}{k^2+\alpha^2}\left(\frac{\partial}{\partial k}\frac{\sin ka_j}{ka_j}\right)\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot(\boldsymbol{R}_j-\boldsymbol{R}_k)}. \tag{3.30}$$

We briefly discuss the behaviour of these connectors in a similar way as in the previous subsection:

(a) For $\boldsymbol{M}^{(2,1)}(\omega)$ and $\boldsymbol{M}^{(2,2)}(\omega)$ one finds (cf. appendix A)

$$\boldsymbol{M}_{jk}^{(2,1)}(\omega)=\begin{cases}0 & \text{for } j=k,\\ \mathcal{O}((\alpha a)^{2-p}(a/R)^p), \quad p=0,1,2 & \text{for } j\neq k,\end{cases} \tag{3.31}$$

$$\boldsymbol{M}_{jk}^{(2,2)}(\omega)=\begin{cases}\frac{1}{15}\boldsymbol{\epsilon}+\mathcal{O}(\alpha a_j)^2 & \text{for } j=k,\\ \mathcal{O}((\alpha a)^{3-p}(a/R)^p), \quad p=0,1,2,3 & \text{for } j\neq k.\end{cases} \tag{3.32}$$

(b) The result for $\boldsymbol{B}^{(2,m)}(\omega)$, analogous to the result (3.10) for $\boldsymbol{B}^{(1,m)}(\omega)$ is

$$\boldsymbol{B}^{(2,m)}(\omega)=\begin{cases}0 & \text{for } m\neq 2 \text{ and } m\neq 4\,,\\ \mathcal{O}(1) & \text{for } m=2\,,\\ \mathcal{O}(\alpha a_j)^2 & \text{for } m=4\,.\end{cases} \tag{3.33}$$

In this case, we may neglect all terms with $m \geqslant 4$, so that the only connector to be retained is $\boldsymbol{B}^{(2,2)}(\omega)$, which is evaluated in appendix B. The result is, up to order $(\alpha a_j)^3$,

$$\boldsymbol{B}^{(2,2)}(\omega)=\boldsymbol{B}^{(2s,2s)}(\omega)+\boldsymbol{B}^{(2a,2a)}(\omega)\,, \tag{3.34}$$

where

$$(\boldsymbol{B}^{(2s,2s)}(\omega))_{\alpha\beta\gamma\delta}\equiv-\tfrac{9}{10}(\tfrac{1}{2}\delta_{\alpha\delta}\delta_{\beta\gamma}+\tfrac{1}{2}\delta_{\alpha\gamma}\delta_{\beta\delta}-\tfrac{1}{3}\delta_{\alpha\beta}\delta_{\delta\gamma})(1-\tfrac{2}{5}\alpha^2a_j^2+\tfrac{1}{3}\alpha^3a_j^3)\,, \tag{3.35}$$

$$(\boldsymbol{B}^{(2a,2a)}(\omega))_{\alpha\beta\gamma\delta}\equiv-\tfrac{3}{2}(\tfrac{1}{2}\delta_{\alpha\delta}\delta_{\beta\gamma}-\tfrac{1}{2}\delta_{\alpha\gamma}\delta_{\beta\delta})(1-\tfrac{2}{5}\alpha^2a_j^2+\tfrac{1}{3}\alpha^3a_j^3)\,. \tag{3.36}$$

(c) Finally, we turn to the connectors $\boldsymbol{A}^{(2,m)}(\omega)$. For these connectors, it may be shown that they are of order $m+1$ in our expansion parameters αa and a/R. (This follows from the general analysis of appendix A). An analogous discussion as in the previous subsection, sub (c) then leads to the conclusion that only the connectors $\boldsymbol{A}^{(2,1)}(\omega)$ and $\boldsymbol{A}^{(2,2a)}(\omega)$ need be retained. Due to the symmetry of the connectors (cf. paper I, eq. (4.35)), $\boldsymbol{A}^{(2,1)}$ obeys the relation

$$(\boldsymbol{A}_{jk}^{(2,1)}(\omega))_{\alpha\beta\gamma}=a_j/a_k(\boldsymbol{A}_{kj}^{(1,2)}(\omega))_{\gamma\beta\alpha}\,. \tag{3.37}$$

The expression for $\boldsymbol{A}^{(2,2a)}(\omega)$ is given later.

The above mentioned properties of the connectors $\boldsymbol{A}^{(2,m)}(\omega)$ imply that eq. (3.26) reduces to, using also eq. (2.24),

$$\begin{aligned}6\pi\eta a_j^2(1-\tfrac{1}{15}\alpha^2a_j^2)\boldsymbol{\epsilon}\cdot\boldsymbol{\Omega}_j(\omega)&=\tfrac{9}{10}(1-\tfrac{2}{5}\alpha^2a_j^2+\tfrac{1}{3}\alpha^3a_j^3)\boldsymbol{F}_j^{(2s)}(\omega)\\&-\frac{3}{4a_j}\left(1-\frac{2}{5}\alpha^2a_j^2+\frac{1}{3}\alpha^3a_j^3\right)\boldsymbol{\epsilon}\cdot\boldsymbol{T}_j(\omega)-\sum_{k\neq j}\frac{1}{2a_k}\boldsymbol{A}_{jk}^{(2,2a)}(\omega):\boldsymbol{\epsilon}\cdot\boldsymbol{T}_k(\omega)\\&-\sum_{k\neq j}\boldsymbol{A}_{jk}^{(2,1)}(\omega)\cdot\boldsymbol{K}_k(\omega)\,.\end{aligned} \tag{3.38}$$

This is an equation for a tensor of rank 2. Its symmetric part yields an equation for $\boldsymbol{F}^{(2s)}(\omega)$, which explicitly shows, as was indicated before (cf. after eq. (3.10)), that $\boldsymbol{F}^{(2s)}(\omega)$ is of second order in our expansion parameters. Since $\boldsymbol{F}^{(2s)}(\omega)$ appears in eq. (3.3) multiplied by a connector of second order, it need not be considered here. We obtain the antisymmetric part of eq. (3.38) by contracting with $\boldsymbol{\epsilon}$ and using the fact that $\boldsymbol{\epsilon}:\boldsymbol{\epsilon}=-2\mathbb{1}$. We get

$$\begin{aligned}8\pi\eta a_j^3(1-\tfrac{1}{15}\alpha^2a_j^2)\boldsymbol{\Omega}_j(\omega)&=-(1-\tfrac{2}{5}\alpha^2a_j^2+\tfrac{1}{3}\alpha^3a_j^3)\boldsymbol{T}_j(\omega)\\&+\sum_{k\neq j}\frac{1}{3}\frac{a_j}{a_k}\boldsymbol{\epsilon}:\boldsymbol{A}_{jk}^{(2a,2a)}(\omega):\boldsymbol{\epsilon}\cdot\boldsymbol{T}_k(\omega)+\sum_{k\neq j}\frac{2}{3}a_j\boldsymbol{\epsilon}:\boldsymbol{A}_{jk}^{(2a,1)}(\omega)\cdot\boldsymbol{K}_k(\omega)\,.\end{aligned} \tag{3.39}$$

For the connector $\boldsymbol{A}^{(2a,2a)}(\omega)$, we find in appendix C up to the order considered

$$\epsilon:\boldsymbol{A}_{jk}^{(2a,2a)}(\omega):\epsilon = -\frac{9}{4}\frac{\alpha^2 a_j^2 a_k}{R_{jk}}\,\mathrm{e}^{-\alpha R_{jk}}\left(1+\frac{2}{\alpha R_{jk}}+\frac{2}{\alpha^2 R_{jk}^2}\right)\left(\hat{r}_{jk}\hat{r}_{jk}-\frac{1}{3}\mathbb{1}\right) + \frac{3}{2}\frac{\alpha^2 a_j^2 a_k}{R_{jk}}\,\mathrm{e}^{-\alpha R_{jk}}(\mathbb{1}-\hat{r}_{jk}\hat{r}_{jk})\,. \tag{3.40}$$

The two equations (3.24) and (3.39) yield to third order the frequency dependent mobility tensors which are further analyzed within the next section.

4. Frequency-dependent mobilities

Eqs. (3.24) and (3.39) may be written in the form

$$\boldsymbol{u}_j(\omega) = -\sum_k \boldsymbol{\mu}_{jk}^{\mathrm{TT}}(\omega)\cdot\boldsymbol{K}_k(\omega) - \sum_k \boldsymbol{\mu}_{jk}^{\mathrm{TR}}(\omega)\cdot\boldsymbol{T}_j(\omega)\,, \tag{4.1}$$

$$\boldsymbol{\Omega}_j(\omega) = -\sum_k \boldsymbol{\mu}_{jk}^{\mathrm{RT}}(\omega)\cdot\boldsymbol{K}_k(\omega) - \sum_k \boldsymbol{\mu}_{jk}^{\mathrm{RR}}(\omega)\cdot\boldsymbol{T}_j(\omega)\,. \tag{4.2}$$

One finds to third order the following expressions for the various mobility tensors

$$\begin{aligned}6\pi\eta a_j\boldsymbol{\mu}_{jk}^{\mathrm{TT}}(\omega) &= \delta_{jk}\mathbb{1}(1-\alpha a_j+\tfrac{8}{9}\alpha^2a_j^2-\tfrac{7}{9}\alpha^3a_j^3)+(1-\delta_{jk})\\ &\quad\times\left[\frac{3}{4}a_j\left(1+\alpha^2\left(\frac{7a_j^2}{18}+\frac{a_k^2}{6}\right)\right)+\alpha^2\frac{a_k^3}{6}\right]\frac{\mathrm{e}^{-\alpha R_{jk}}}{R_{jk}}(\mathbb{1}+\hat{r}_{jk}\hat{r}_{jk})\\ &\quad+(1-\delta_{jk})\left[\frac{9}{2}\frac{a_j}{\alpha^2R_{jk}^3}+\frac{a_k^3}{R_{jk}^3}-\left\{\frac{9}{4}\frac{a_j}{R_{jk}}\left(1+\frac{\alpha^2(a_j^2+a_k^2)}{6}\right)+\frac{1}{2}\frac{\alpha^2a_k^3}{R_{jk}}\right\}\right.\\ &\quad\left.\times\,\mathrm{e}^{-\alpha R_{jk}}\left(1+\frac{2}{\alpha R_{jk}}+\frac{2}{\alpha^2R_{jk}^2}\right)\right](\hat{r}_{jk}\hat{r}_{jk}-\tfrac{1}{3}\mathbb{1})\,,\end{aligned} \tag{4.3}$$

$$8\pi\eta a_j^2\boldsymbol{\mu}_{jk}^{\mathrm{TR}}(\omega) = 8\pi\eta a_j^2\tilde{\boldsymbol{\mu}}_{kj}^{\mathrm{RT}}(\omega) = -(1-\delta_{jk})(1+\alpha R_{jk})\frac{a_j^2}{R_{jk}^2}\,\mathrm{e}^{-\alpha R_{jk}}\epsilon\cdot\hat{r}_{jk}\,, \tag{4.4}$$

$$\begin{aligned}8\pi\eta a_j^3\boldsymbol{\mu}_{jk}^{\mathrm{RR}}(\omega) &= \delta_{jk}(1-\tfrac{1}{3}\alpha^2a_j^2+\tfrac{1}{3}\alpha^3a_j^3)\mathbb{1}+(1-\delta_{jk})\frac{3}{4}\frac{\alpha^2a_j^3}{R_{jk}}\,\mathrm{e}^{-\alpha R_{jk}}\\ &\quad\times\left(1+\frac{2}{\alpha R_{jk}}+\frac{2}{\alpha^2R_{jk}^2}\right)\left(\hat{r}_{jk}\hat{r}_{jk}-\frac{1}{3}\mathbb{1}\right)-\frac{1}{2}\frac{\alpha^2a_j^3}{R_{jk}}\,\mathrm{e}^{-\alpha R_{jk}}(\mathbb{1}-\hat{r}_{jk}\hat{r}_{jk})\,.\end{aligned} \tag{4.5}$$

Here $\tilde{\boldsymbol{\mu}}^{\mathrm{RT}}(\omega)$ denotes the transposed of $\boldsymbol{\mu}^{\mathrm{RT}}(\omega)$. The expressions for the diagonal terms ($j=k$) of these mobilities are in agreement with the well-known results for one sphere[19,23]).

The above results greatly simplify in the two regimes $|\alpha|R_{jk} \gg 1$ and $|\alpha|R_{jk} \ll 1$. Let us first analyze the case $|\alpha|R_{jk} \gg 1$, which is the case at sufficiently large particle separations. In this regime, the mobility tensors reduce to

$$6\pi\eta a_j \boldsymbol{\mu}_{jk}^{\mathrm{TT}}(\omega) = \left(1 - \alpha a_j + \frac{8}{9}\alpha^2 a_j^2 - \frac{7}{9}\alpha^3 a_j^3\right)\delta_{jk}\mathbb{1} + (1 - \delta_{jk})\left(\frac{9}{2}\frac{a_j}{\alpha^2 R_{jk}^3} + \frac{a_k^3}{R_{jk}^3}\right)\left(\hat{r}_{jk}\hat{r}_{jk} - \frac{1}{3}\mathbb{1}\right), \quad |\alpha|R_{jk} \gg 1\,, \tag{4.6}$$

$$\boldsymbol{\mu}_{jk}^{\mathrm{TR}}(\omega) = \tilde{\boldsymbol{\mu}}_{kj}^{\mathrm{RT}}(\omega) \simeq 0, \quad |\alpha|R_{jk} \gg 1\,, \tag{4.7}$$

$$8\pi\eta a_j^3 \boldsymbol{\mu}_{jk}^{\mathrm{RR}}(\omega) = \delta_{jk}(1 - \tfrac{1}{3}\alpha^2 a_j^2 + \tfrac{1}{3}\alpha^3 a_j^3)\mathbb{1}, \quad |\alpha|R_{jk} \gg 1\,. \tag{4.8}$$

In this regime, therefore, translational and rotational motion do not couple to third order, nor do the rotational motions of different particles. Note that the first of the two two-particle interaction terms in $\boldsymbol{\mu}^{\mathrm{TT}}(\omega)$ is smaller than a term of first order in a/R, since $1/\alpha^2 R_{jk}^2 \ll 1$ in the regime considered, but larger than the term of order $(a/R_{jk})^3$, since $1/\alpha^2 R_{jk}^2 = (a/R_{jk})^2 \times 1/\alpha^2 a^2$ and we have assumed that $|\alpha|a < 1$.

If, on the other hand $|\alpha|R_{jk} \ll 1$ and if one wants to retain terms up to third order in the expansion coefficients, one has to take into account terms like $\alpha^3 a R_{jk}^2$ since $R_{jk} \geqslant a_j + a_k$ so that such contributions are larger than a term $\alpha^3 a^3$. For simplicity, we will only give the resulting expressions to first order in αa and a/R_{jk}. To this order, $\boldsymbol{\mu}_{jk}^{\mathrm{TR}}$, $\boldsymbol{\mu}_{jk}^{\mathrm{RT}}$ and $\boldsymbol{\mu}_{jk}^{\mathrm{RR}}$ for $j \neq k$ all vanish; for $\boldsymbol{\mu}_{jk}^{\mathrm{TT}}$ we get

$$6\pi\eta a_j \boldsymbol{\mu}_{jk}^{\mathrm{TT}}(\omega) = \frac{3}{4}\frac{a_j}{R_{jk}}(\mathbb{1} + \hat{r}_{jk}\hat{r}_{jk}) - \alpha a_j \mathbb{1}, \quad j \neq k, |\alpha|R_{jk} \ll 1\,. \tag{4.9}$$

This expression shows that at sufficiently small frequencies the mutual mobilities $\boldsymbol{\mu}_{jk}^{\mathrm{TT}}$, $j \neq k$, have a contribution proportional to $\omega^{1/2}$, just as the direct mobilities $\boldsymbol{\mu}_{jj}^{\mathrm{TT}}$. Consequently the velocity correlation function of two different particles also possesses a $t^{-3/2}$ long-time tail.

It should be clear from the above analysis that, whether effects connected with the derived small frequency behaviour will manifest themselves, strongly depends on the experimental conditions, in particular the particle concentration and the frequency range under investigation. In recent experiments on the long time tail by Paul and Pusey[24]), the frequency interval is such that $3 \times 10^2\,\mathrm{cm}^{-1} \lesssim |\alpha| \lesssim 3 \times 10^3\,\mathrm{cm}^{-1}$, while the suspension is such that the mean distance R_m between particles is approximately $7 \times 10^{-3}\,\mathrm{cm}$. In these experiments therefore, one has for the parameter $|\alpha|R_m$

$$2 \lesssim |\alpha|R_m \lesssim 20\,.$$

Thus, their measurements are typically in the regime where the hydrodynamic interactions are a factor $1/\alpha^2 R_{\mathrm{m}}^2$ smaller than the static interactions would be at the same mean particle separation (cf. eq. (4.6) and the discussion thereafter). Moreover, the typical particle radius in the experiments of Paul and Pusey is 1.7×10^{-4} cm, so that their suspension is extremely dilute. Therefore, our analysis supports Paul and Pusey's[24]) statement that "it seems extremely unlikely that interparticle interactions would have a significant effect in such a dilute suspension". On the other hand, when studying the dynamics of colloidal crystals[25,26]) for which the distance between neighbouring particles is about 1.5×10^{-4} cm, and for the same values of α as in the experiments of Paul and Pusey, one has for the parameter αR

$$0.05 \lesssim |\alpha| R \lesssim 0.5 .$$

Under these circumstances, one is therefore in the opposite regime where eq. (4.9) holds to lowest approximation, and where effects due to the long time tail in the velocity cross correlation should play a role.

As a final remark we stress that the mobility tensor matrix contains up to third order only two-body-hydrodynamic interactions. This is *not* the case for the friction tensor matrix, which is its inverse, and which to third order already contains three and four sphere contributions[8]).

Acknowledgement

One of us, (WvS), is grateful to Dr. A.J. Hurd for stimulating correspondence and discussions.

Appendix A. General results for the connectors $\boldsymbol{A}_{jk}^{(n,m)}(\omega)$

Though the general connector $\boldsymbol{A}_{jk}^{(n,m)}(\omega)$ has not been given in the text, comparison with eq. (4.32) of paper I shows that the general expression for this connector reads

$$\boldsymbol{A}_{jk}^{(n,m)}(\omega) = \frac{3a_j(2n-1)!!(2m-1)!!}{8\pi^2} \left(\frac{-\mathrm{i}}{a_j}\right)^{n-1} \left(\frac{\mathrm{i}}{a_k}\right)^{m-1} \int \mathrm{d}\hat{k} \int_{-\infty}^{+\infty} \mathrm{d}k \, \frac{k^2}{k^2+\alpha^2}$$
$$\times \mathrm{e}^{-\mathrm{i}k\xi_{jk}R_{jk}} \left(\overline{\frac{\partial^{n-1}}{\partial k^{n-1}}} \frac{\sin ka_j}{ka_j}\right)(\mathbb{1} - \hat{k}\hat{k}) \left(\overline{\frac{\partial^{m-1}}{\partial k^{m-1}}} \frac{\sin ka_k}{ka_k}\right). \qquad \text{(A.1)}$$

Here, as in paper *I*, $\xi_{jk} \equiv \hat{r}_{jk} \cdot \hat{k}$ is the cosine of the angle between the unit vector

$\hat{r}_{jk}$ and $\hat{k}$. In paper I, cf. eq. (3.16) and appendix A, we have shown that

$$(2n+1)!!(-a_j)^{-n}\left(\overline{\frac{\partial^n}{\partial k^n}}\frac{\sin ka_j}{ka_j}\right)=\overline{\hat{k}^n}a_j^n k^n\left(1-\frac{a_j^2k^2}{4n+6}+\mathcal{O}(a_jk)^4\right)$$
$$\equiv \overline{\hat{k}^n}S_n(a_jk),\quad n\geqslant 0\,. \tag{A.2}$$

Thus, the $S_n(a_jk)$ are essentially polynomials in $a_j^2k^2$. Rewriting eq. (A.1) with the aid of eq. (A.2) yields

$$\boldsymbol{A}_{jk}^{(n,m)}(\omega)=\frac{3a_j}{4\pi}\mathrm{i}^{n-1}(-\mathrm{i})^{m-1}\int \mathrm{d}\hat{k}\,\overline{\hat{k}^{n-1}}(\mathbb{1}-\hat{k}\hat{k})\overline{\hat{k}^{m-1}}$$
$$\times\frac{1}{2\pi}\int_{-\infty}^{+\infty}\mathrm{d}k\;\mathrm{e}^{-\mathrm{i}k\xi_{jk}R_{jk}}S_{n-1}(a_jk)S_{m-1}(a_kk)\frac{k^2}{k^2+\alpha^2}\,. \tag{A.3}$$

The following analysis is based on an extension of the arguments given in section 5 of paper I. According to eq. (A.2), the product $S_n(a_jk)S_m(a_kk)$ has an expansion in powers of k of the form

$$S_n(a_jk)S_m(a_kk)=\sum_{p=0}^{\infty}K_{jk}^{(2p)}k^{n+m+2p}\,, \tag{A.4}$$

where $K_{jk}^{(0)}=a_j^na_k^m$ and $K_{jk}^{(2)}=-a_j^na_k^m(a_j^2/(4n+6)+a_k^2/(4m+6))$. For the k-integral in eq. (A.3), we obtain with the aid of this expansion*

$$I_{jk}(\xi_{jk})\equiv\frac{1}{2\pi}\int_{-\infty}^{+\infty}\mathrm{d}k\;\mathrm{e}^{-\mathrm{i}k\xi_{jk}R_{jk}}S_{n-1}(a_jk)S_{m-1}(a_kk)\frac{k^2}{k^2+\alpha^2}$$
$$=\sum_{p=0}^{\infty}K_{jk}^{(2p)}\left(\frac{\mathrm{i}}{R_{jk}}\right)^{n+m+2p}\frac{\partial^{n+m+2p}}{\partial\xi_{jk}^{n+m+2p}}\cdot\frac{1}{2\pi}\int_{-\infty}^{+\infty}\mathrm{d}k\,\frac{\mathrm{e}^{-\mathrm{i}k\xi_{jk}R_{jk}}}{k^2+\alpha^2}$$
$$=\sum_{p=0}^{\infty}K_{jk}^{(2p)}\left(\frac{\mathrm{i}}{R_{jk}}\right)^{n+m+2p}\frac{\partial^{n+m+2p}}{\partial\xi_{jk}^{n+m+2p}}\frac{1}{2\alpha}\,\mathrm{e}^{-\alpha R_{jk}|\xi_{jk}|}\,. \tag{A.5}$$

Using the fact that $\partial\Theta(x)/\partial x=\delta(x)$, one readily shows that

$$\frac{\partial^2\mathrm{e}^{-\beta|x|}}{\partial x^2}=\beta^2\,\mathrm{e}^{-\beta|x|}-2\beta\delta(x)\,, \tag{A.6}$$

so that for $p\geqslant 1$

$$\frac{\partial^{2p+2}}{\partial x^{2p+2}}\mathrm{e}^{-\beta|x|}=\beta^{2p+2}\,\mathrm{e}^{-\beta|x|}-2\beta^{2p+1}\delta(x)-2\beta^{2p-1}\frac{\partial^2}{\partial x^2}\delta(x)+\text{h.o.}\,. \tag{A.7}$$

* The intermediate step of eliminating the powers of k by taking derivatives with respect to ξ_{jk} is necessary in order to arrive at an integral which is convergent for all values of ξ_{jk}. The integrals in the first line of eq. (A.5) that are obtained by substituting eq. (A.4) are divergent for $\xi_{jk}=0$.

Here, h.o. stands for higher order derivatives of the delta function. Using this result to work out the $2p+2$ derivatives with respect to ξ_{jk} in eq. (A.5), one gets

$$
\begin{aligned}
I_{jk}(\xi_{jk}) &= \sum_{p=0}^{\infty} K_{jk}^{(2p)}(\mathrm{i}\alpha)^{2p} \frac{\mathrm{i}^{n+m}}{R_{jk}^{n+m-1}} \frac{\partial^{n+m-2}}{\partial \xi_{jk}^{n+m-2}} \left[\frac{1}{2} \alpha R_{jk}\, \mathrm{e}^{-\alpha R_{jk}|\xi_{jk}|} - \delta(\xi_{jk}) \right] \\
&\quad - \sum_{p=1}^{\infty} K_{jk}^{(2p)}(\mathrm{i}\alpha)^{2p} \frac{\mathrm{i}^{n+m}}{\alpha^2 R_{jk}^{n+m+1}} \frac{\partial^{n+m}}{\partial \xi_{jk}^{n+m}} \delta(\xi_{jk}) + \text{h.o.} \\
&= \frac{\mathrm{i}^{n+m}}{R_{jk}^{n+m-1}} \frac{S_{n-1}(\mathrm{i}\alpha a_j) S_{m-1}(\mathrm{i}\alpha a_k)}{(\mathrm{i}\alpha)^{n+m-2}} \frac{\partial^{n+m-2}}{\partial \xi_{jk}^{n+m-2}} \left[\frac{1}{2} \alpha R_{jk}\, \mathrm{e}^{-\alpha R_{jk}|\xi_{jk}|} - \delta(\xi_{jk}) \right] \\
&\quad - \frac{\mathrm{i}^{n+m}}{\alpha^2 R_{jk}^{n+m+1}} \left(\frac{S_{n-1}(\mathrm{i}\alpha a_j) S_{m-1}(\mathrm{i}\alpha a_k)}{(\mathrm{i}\alpha)^{n+m-2}} - a_j^{n-1} a_k^{m-1} \right) \frac{\partial^{n+m}}{\partial \xi_{jk}^{n+m}} \delta(\xi_{jk}) + \text{h.o.}\,.
\end{aligned}
\tag{A.8}
$$

In the second line, use has been made of eq. (A.4) and the fact that $K_{jk}^{(0)} = a_j^n a_k^m$. If we substitute this result into eq. (A.3), the terms indicated by h.o. in the above equation do not contribute to $\boldsymbol{A}^{(n,m)}(\omega)$, for reasons also discussed in paper I after eq. (5.9). For, in a reference frame in which the z-axis is parallel to the unit vector $\hat{r}_{jk}$, we may write $\mathrm{d}\hat{k} = -\,\mathrm{d}\xi_{jk}\,\mathrm{d}\phi_{jk}$, where ϕ_{jk} is the other polar angle. After integration over ϕ_{jk} in eq. (A.3), any element of $\overline{\hat{k}^{n-1}}(\mathbb{1} - \hat{k}\hat{k})\overline{\hat{k}^{m-1}}$ will be a polynomial in ξ_{jk} of which the highest order term is proportional to ξ_{jk}^{n+m}. Consequently terms containing higher derivatives of $\delta(\xi_{jk})$ then the $(n+m)$th, do not contribute. We therefore have the *exact* result

$$
\boldsymbol{A}_{jk}^{(n,m)}(\omega) = \boldsymbol{G}_{jk}^{(n,m)}(\omega)/R_{jk}^{n+m-1} + \boldsymbol{H}_{jk}^{(n,m)}(\omega)/R_{jk}^{n+m+1} + \boldsymbol{L}_{jk}^{(n,m)}(\omega, R_{jk})\,, \tag{A.9}
$$

with

$$
\begin{aligned}
\boldsymbol{G}_{jk}^{(n,m)}(\omega) &\equiv \frac{S_{n-1}(\mathrm{i}\alpha a_j) S_{m-1}(\mathrm{i}\alpha a_k)(-1)^{n-1}}{(\mathrm{i}\alpha a_j)^{n-1}(\mathrm{i}\alpha a_k)^{m-1}} \frac{3}{4\pi} a_j^n a_k^{m-1} \\
&\quad \times \int \mathrm{d}\hat{k}\, \overline{\hat{k}^{n-1}}(\mathbb{1} - \hat{k}\hat{k})\overline{\hat{k}^{m-1}} \frac{\partial^{n+m-2}}{\partial \xi_{jk}^{n+m-2}} \delta(\xi_{jk}) \\
&= \frac{S_{n-1}(\mathrm{i}\alpha a_j) S_{m-1}(\mathrm{i}\alpha a_k)}{(\mathrm{i}\alpha a_j)^{n-1}(\mathrm{i}\alpha a_k)^{m-1}} \boldsymbol{G}_{jk}^{(n,m)}\,,
\end{aligned}
\tag{A.10}
$$

$$
\begin{aligned}
\boldsymbol{H}_{jk}^{(n,m)}(\omega) &\equiv \left(\frac{S_{n-1}(\mathrm{i}\alpha a_j) S_{m-1}(\mathrm{i}\alpha a_k)}{(\mathrm{i}\alpha a_j)^{n-1}(\mathrm{i}\alpha a_k)^{m-1}} - 1 \right)(-1)^{n-1} \frac{3}{4\pi} a_j^n a_k^{m-1} \alpha^{-2} \\
&\quad \times \int \mathrm{d}\hat{k}\, \overline{\hat{k}^{n-1}}(\mathbb{1} - \hat{k}\hat{k})\overline{\hat{k}^{m-1}} \frac{\partial^{n+m}}{\partial \xi_{jk}} \delta(\xi_{jk}) \\
&= \left(\frac{S_{n-1}(\mathrm{i}\alpha a_j) S_{m-1}(\mathrm{i}\alpha a_k)}{(\mathrm{i}\alpha a_j)^{n-1}(\mathrm{i}\alpha a_k)^{m-1}} - 1 \right) \left[\left(\frac{a_j^2}{4n+2} + \frac{a_k^2}{4m+2} \right) \right]^{-1} \alpha^{-2} \boldsymbol{H}_{jk}^{(n,m)}\,,
\end{aligned}
\tag{A.11}
$$

$$\boldsymbol{L}_{jk}^{(n,m)}(\omega, R_{jk}) \equiv (-1)^n \frac{3}{4\pi} \frac{a_j^n a_k^{m-1}}{R_{jk}^{n+m-1}} \frac{S_{n-1}(\mathrm{i}\alpha a_j) S_{m-1}(\mathrm{i}\alpha a_k)}{(\mathrm{i}\alpha a_j)^{n-1} (\mathrm{i}\alpha a_k)^{m-1}}$$
$$\int \mathrm{d}\hat{k}\, \overline{\hat{k}^{n-1}} (\mathbb{1} - \hat{k}\hat{k})\, \overline{\hat{k}^{m-1}} \frac{\partial^{n+m-2}}{\partial \xi_{jk}^{n+m-2}} \left(\frac{1}{2} \alpha R_{jk}\, \mathrm{e}^{-\alpha R_{jk} |\xi_{jk}|} \right). \quad \text{(A.12)}$$

Here, $\boldsymbol{G}^{(n,m)}$ and $\boldsymbol{H}^{(n,m)}$ are the frequency independent connectors on which the analysis of the static case was based (cf. paper I, eqs. (5.11) and (5.12)). As was asserted for the case $n = 1$ in section 3, sub c, eqs. (A.10) and (A.11) show that the tensors $\boldsymbol{G}^{(n,m)}(\omega)$ and $\boldsymbol{H}^{(n,m)}(\omega)$ are just the frequency independent connectors $\boldsymbol{G}^{(n,m)}$ and $\boldsymbol{H}^{(n,m)}$ multiplied by a frequency dependent forefactor. As can easily be checked with the aid of the properties of the polynomials S_n (cf. eq. (A.3)), these forefactors go to 1 in the limit $\omega \to 0$ ($\alpha \to 0$), as it should. Moreover, $\boldsymbol{G}^{(n,m)}(\omega)$ and $\boldsymbol{H}^{(n,m)}(\omega)$ have an expansion in powers of $(\alpha a)^2$, not in powers of αa.

In the limit of large $|\alpha| R_{jk}$, we may use for $\boldsymbol{L}^{(n,m)}(\omega, R_{jk})$ the (asymptotic) expansion

$$\tfrac{1}{2}\alpha R_{jk}\, \mathrm{e}^{-\alpha R_{jk}|\xi_{jk}|} = \delta(\xi_{jk}) + \frac{1}{\alpha^2 R_{jk}^2} \frac{\partial^2}{\partial \xi_{jk}^2} \delta(\xi_{jk}) + \dots \quad \text{(A.13)}$$

Upon substitution of this expansion into the expression for $\boldsymbol{L}^{(n,m)}(\omega, R_{jk})$, one immediately obtains

$$\lim_{R_{jk} \to \infty} R_{jk}^{n+m-1} \boldsymbol{L}_{jk}^{(n,m)}(\omega, R_{jk}) = -\boldsymbol{G}_{jk}^{(n,m)}(\omega). \quad \text{(A.14)}$$

For $n = 1$, eq. (A.14) reduces to eq. (3.16). The next term in the expansion (A.13) yields for large $|\alpha| R_{jk}$, together with eqs. (A.9)–(A.14) and the explicit result for $\boldsymbol{H}_{jk}^{(n,m)}$ (paper I, eq. (5.12)),

$$\boldsymbol{A}_{jk}^{(n,m)}(\omega) \simeq (-1)^{m-1} \frac{3}{2} (2n + 2m - 1)!! \frac{a_j^n a_k^{m-1}}{\alpha^2 R_{jk}^{m+n+1}} \overline{\hat{r}_{jk}^{n+m}} \quad (|\alpha| R_{jk} \gg 1). \quad \text{(A.15)}$$

For $n = m = 1$, one recovers in this regime the correction to the translational mobility, given in the second line of eq. (4.6). Eq. (A.15) also shows that corrections to eq. (4.6) in the regime $|\alpha| R_{jk} \gg 1$ are extremely small, since then terms of the form $\boldsymbol{A}^{(1,2)} \odot \boldsymbol{A}^{(2,1)}$, which yield fourth order contributions in the static case (cf. paper I, table I), are proportional to $a^4/\alpha^4 R^8$.

If $|\alpha| R_{jk} \ll 1$, the integrand in the expression for $\boldsymbol{L}^{(n,m)}(\omega, R_{jk})$ may be expanded in powers of α. For $m = n = 1$, one finds in a straight-forward way that $\boldsymbol{L}^{(1,1)}(\omega, R_{jk})$ becomes of order αa, whereas it becomes of order $\alpha^2 a^{n+m-1}/R_{jk}^{n+m-3}$ if $m \neq 1$ or $n \neq 1$. In both cases, $\boldsymbol{L}^{(n,m)}(\omega, R_{jk})$ is of order $n + m - 1$ in our expansion parameters, as indicated in eq. (3.15).

Appendix B. Evaluation of the connectors $\boldsymbol{M}(\omega)$

We first consider $\boldsymbol{M}_{jk}^{(1,1)}(\omega)$, given by eq. (3.6), for $j = k$. In this case, the angular integration can be readily performed (cf. appendix B of paper I). the k-integration can be done by splitting $\sin ka = (\mathrm{e}^{ika} - \mathrm{e}^{-ika})/2\mathrm{i}$, and closing the contour involving e^{ika} in the upper half-plane and the other one in the lower half-plane. Since the term between square brackets has a regular expansion around $k = 0$, there are only poles at $\pm\,\mathrm{i}\alpha$, so that

$$\begin{aligned} \boldsymbol{M}_{jj}^{(1,1)}(\omega) &= -\frac{2}{3}\mathbb{1}\cdot 2 \operatorname*{Res}_{k\to \mathrm{i}\alpha}\left(\frac{k}{k^2+\alpha^2}\,\mathrm{e}^{ika_j}\left[\frac{\sin ka_j - ka_j\cos ka_j}{(ka_j)^3}\right]\right) \\ &= -\frac{1}{3}\mathbb{1}\,\frac{1}{(\alpha a_j)^3}\left(1-\alpha a_j - \mathrm{e}^{-2\alpha a_j}(1+\alpha a_j)\right) \\ &= -\frac{2}{9}\mathbb{1}(1-\alpha a_j + \dots)\,. \end{aligned} \tag{B.1}$$

To calculate $\boldsymbol{M}^{(1,1)}(\omega)$, we write

$$\boldsymbol{M}_{jk}^{(1,1)}(\omega) = -\frac{a_k}{6\pi}\int \mathrm{d}\hat{k}(\mathbb{1}-\hat{k}\hat{k})\,\frac{1}{2\pi}\int_{-\infty}^{+\infty}\mathrm{d}k\;\mathrm{e}^{-ik\xi_{jk}R_{jk}}\,S_0(a_jk)S'(a_kk)\,\frac{k^2}{k^2\alpha^2}\,. \tag{B.2}$$

Here, we have employed the notation of appendix A, and we have defined

$$S'(a_kk) \equiv 3\,\frac{\sin ka_k - ka_k\cos ka_k}{(ka_k)^3} = 1 - \frac{1}{10}a_k^2k^2 + \dots\,. \tag{B.3}$$

Obviously, $\boldsymbol{M}_{jk}^{(1,1)}(\omega)$ is of a similar structure as $\boldsymbol{A}_{jk}^{(1,1)}(\omega)$, cf. eq. (A.3). Therefore, the analysis of the k-integral in appendix A also applies to the one appearing in eq. (B.2), if we identify S' in eq. (B.2) with S_{m-1} for $m = 1$ in eq. (A.3). Accordingly, to lowest order one may neglect the terms arising in the second line of eq. (A.8), and take $S_0(\mathrm{i}\alpha a_j)S'(\mathrm{i}\alpha a_k) \simeq 1$. Upon combining the results, one then gets up to the order considered

$$\boldsymbol{M}_{jk}^{(1,1)}(\omega) = \frac{a_k}{6\pi R_{jk}}\int \mathrm{d}\hat{k}(\mathbb{1}-\hat{k}\hat{k})\left(\frac{1}{2}\alpha R_{jk}\,\mathrm{e}^{-\alpha R_{jk}|\xi_{jk}|} - \delta(\xi_{jk})\right). \tag{B.4}$$

This tensor must be of the form

$$\boldsymbol{M}_{jk}^{(1,1)}(\omega) = \frac{a_k}{6R_{jk}}(a\mathbb{1} + b\hat{r}_{jk}\hat{r}_{jk})\,. \tag{B.5}$$

Contraction with the unit tensor gives

$$3a + b = \frac{2}{\pi} \int_0^{2\pi} \mathrm{d}\phi_{jk} \int_{-1}^{+1} \mathrm{d}\xi_{jk} \left(\frac{1}{2} \alpha R_{jk} \, \mathrm{e}^{-\alpha R_{jk} |\xi_{jk}|} - \delta(\xi_{jk}) \right) = -4\mathrm{e}^{-\alpha R_{jk}} , \tag{B.6}$$

while contraction with $\hat{r}_{jk}\hat{r}_{jk}$ yields

$$a + b = -2\mathrm{e}^{-\alpha R_{jk}} - 2\alpha R_{jk} \int_0^1 \mathrm{d}\xi_{jk} \xi_{jk}^2 \, \mathrm{e}^{-\alpha R_{jk} \xi_{jk}} . \tag{B.7}$$

For the integral, one finds

$$\alpha R_{jk} \int_0^1 \mathrm{d}\xi_{jk} \xi_{jk}^2 \, \mathrm{e}^{-\alpha R_{jk} \xi_{jk}} = \frac{2}{\alpha^2 R_{jk}^2} - \mathrm{e}^{-\alpha R_{jk}} \left(1 + \frac{2}{\alpha R_{jk}} + \frac{2}{\alpha^2 R_{jk}^2} \right) \equiv J(\alpha R_{jk}) . \tag{B.8}$$

The solution of eqs. (B.6), together with eq. (B.8), is $a = -\exp(-\alpha R_{jk}) + J(\alpha R_{jk})$, $b = -\exp(-\alpha R_{jk}) - 3J(\alpha R_{jk})$. Substitution of these values into eq. (B.5) leads to the result (3.8).

Next, we consider $\boldsymbol{M}^{(1,2)}(\omega)$ and $\boldsymbol{M}^{(2,1)}(\omega)$, given by eqs. (3.7) and (3.27), respectively. Since $\boldsymbol{M}^{(2,1)}(\omega)$ is of a similar structure as $\boldsymbol{M}^{(1,2)}(\omega)$, its analysis is analogous to the one of $\boldsymbol{M}^{(1,2)}(\omega)$, and we will therefore only discuss this connector. For $j = k$, eq. (3.7) shows that the angular integration and the k-integration decouple and that the angular integration is of the form $\int \mathrm{d}\hat{k}\, \hat{k}$. Obviously, this integral vanishes and thus $\boldsymbol{M}_{jj}^{(1,2)}(\omega) = 0$. To evaluate the connector in the case $j \neq k$, we may again use the result of appendix A. If we now define the polynomial

$$S''(\alpha_k k) \equiv -15 \frac{\partial}{\partial k} \left(\frac{\sin a_k k - k a_k \cos k a_k}{k^3 a_k^3} \right) = a_k k \left(1 - \frac{1}{28} a_k^2 k^2 + \dots \right), \tag{B.9}$$

the analysis of the k-integral in appendix A applies if we identify S'' with S_{m-1} for $m = 2$ and take $n = 1$. The result in the second line of eq. (3.9) then follows directly from eq. (A.8).

Since the discussion of $\boldsymbol{M}_{jk}^{(2,2)}(\omega)$ for $j \neq k$ again parallels the one of $\boldsymbol{M}_{jk}^{(1,2)}(\omega)$ for $j \neq k$, we will only evaluate this connector for $j = k$. In this case we obtain from eq. (3.28)

$$\begin{aligned} \boldsymbol{M}_{jj}^{(2,2)}(\omega) = & -\frac{3}{4\pi} \int \mathrm{d}\hat{k}\, \hat{k}\hat{k} \cdot \epsilon \frac{1}{\pi} \int_{-\infty}^{+\infty} \mathrm{d}k \frac{1}{k^2 + \alpha^2} \left\{ -\frac{\sin k a_j}{a_j} + k \cos k a_j \right\} \\ & \times \frac{\partial}{\partial k} \left[\frac{\sin k a_k - k a_k \cos k a_k}{k^3 a_k^3} \right] . \end{aligned} \tag{B.10}$$

The angular integration is easily performed. Moreover, if we split the sine and cosine in the term between curly brackets into exponentials, the contour for the terms $\exp(\mathrm{i}a_k k)$ may be closed in the upper half plane, whereas for the terms with $\exp(-\mathrm{i}ka_k)$, the contour may be closed in the lower half-plane. Since the term between square brackets has a regular expansion around $k=0$, there are only poles at $\pm\mathrm{i}\alpha$. One then gets with the aid of eq. (B.9)

$$\begin{aligned} \boldsymbol{M}_{jj}^{(2,2)}(\omega) &= \epsilon\cdot\frac{2}{a_j}\operatorname*{Res}_{k\to\mathrm{i}\alpha}\left[\frac{1}{k^2+\alpha^2}\mathrm{e}^{\mathrm{i}ka_j}(1-\mathrm{i}a_jk)\left(-\frac{1}{15}a_jk+\mathcal{O}(a_jk^2)\right)\right] \\ &= -\frac{1}{15}\epsilon+\mathcal{O}(\alpha^2a_j)^2\,, \end{aligned} \tag{B.11}$$

which is the desired result.

Appendix C. The connectors $\boldsymbol{B}(\omega)$

Employing the functions S_n defined in eq. (A.2), we may write for $\boldsymbol{B}^{(1,m)}(\omega)$, given in eq. (3.3)

$$\boldsymbol{B}^{(1,m)}(\omega) = -(-\mathrm{i})^{m-1}\frac{3}{8\pi}\int \mathrm{d}\hat{k}\left(\frac{2}{3}\overline{\mathbb{1}\,\overline{\hat{k}\hat{k}}}\right)\overline{\hat{k}^{m-1}}\,\frac{1}{\pi}\int_{-\infty}^{+\infty}\mathrm{d}k\,\frac{k\sin ka_j}{k^2+\alpha^2}S_{m-1}(a_jk)\,. \tag{C.1}$$

Here, we have used the fact that $\overline{\hat{k}\hat{k}}=\hat{k}\hat{k}-\frac{1}{3}\mathbb{1}$. According to eq. (5.4) of Hess and Köhler[22]) (cf. also eq. (A.8) of paper I, where their explicit expression for the case $l=m$ is also quoted), one has

$$\frac{1}{4\pi}\int \mathrm{d}\hat{k}\,\overline{\hat{k}^l}\,\overline{\hat{k}^m}=0 \quad \text{if } l\neq m\,. \tag{C.2}$$

This result immediately implies that $\boldsymbol{B}_j^{(1,m)}(\omega)$ vanishes for $m\neq 1$ and $m\neq 3$. Furthermore, by closing the contour for the k integral in the complex plane in the same way as was done in appendix B, one has

$$\frac{1}{\pi}\int_{-\infty}^{+\infty}\mathrm{d}k\,\frac{k\sin ka_j}{k^2+\alpha^2}S_{m-1}(a_jk)=\mathrm{e}^{-\alpha a_j}S_{m-1}(\mathrm{i}\alpha a_j)\,. \tag{C.3}$$

In view of the fact that $S_2(\mathrm{i}\alpha a_j)$ is of order $\alpha^2a_j^2$ (cf. eq. (A.2)), we immediately find that $\boldsymbol{B}^{(1,3)}(\omega)$ is of order $\alpha^2a_j^2$. For $m=1$, we obtain, using the fact that $S_0(k)=\sin k/k$

$$\boldsymbol{B}^{(1,1)}(\omega) = -\mathbb{1}\,\mathrm{e}^{-\alpha a_j}\frac{\sin \mathrm{i}\alpha a_j}{\mathrm{i}\alpha a_j} = -\mathbb{1}\left(1-\alpha a_j+\frac{2}{3}\alpha^2a_j^2-\frac{1}{3}\alpha^3a_j^3+\cdots\right). \tag{C.4}$$

This concludes the evaluation of $\boldsymbol{B}^{(1,m)}(\omega)$.

Eq. (3.32) for $\boldsymbol{B}^{(2,m)}(\omega)$ may be derived along similar lines as above. After writing $\hat{k}(\mathbb{1}-\hat{k}\hat{k})$ in terms of $\overline{\hat{k}\hat{k}\hat{k}}$ and combinations of $\hat{k}$ and the unit tensor, it immediately follows from eq. (C.2) that $\boldsymbol{B}^{(2,m)}(\omega)$ is only non-zero for $m=2$ or $m=4$. Furthermore, it is easy to demonstrate by using complex integration that $\boldsymbol{B}^{(2,4)}(\omega)$ is of order $\alpha^2 a^2$. As for $\boldsymbol{B}^{(2,2)}(\omega)$, we have

$$\boldsymbol{B}^{(2,2)}(\omega)=\frac{-9}{8\pi}\int \mathrm{d}\hat{k}\,\hat{k}(\mathbb{1}-\hat{k}\hat{k})\hat{k}$$

$$\times\frac{3}{\pi a_j^2}\int_{-\infty}^{+\infty}\mathrm{d}k\,\frac{1}{k^2+\alpha^2}(-\sin ka_j+ka_j\cos ka_j)\left(\frac{\partial}{\partial k}\frac{\sin ka_j}{ka_j}\right). \tag{C.5}$$

The angular integration in the first line has been calculated in paper I, cf. eqs. (4.13), (4.16)–(4.20). For the k integral, we get

$$\begin{aligned}&\frac{3}{\pi a_j^2}\int_{-\infty}^{+\infty}\mathrm{d}k\,\frac{1}{k^2+\alpha^2}(-\sin ka_j+ka_j\cos ka_j)\left(\frac{\partial}{\partial k}\frac{\sin ka_j}{ka_j}\right)\\&=\frac{6}{a_j}\operatorname*{Res}_{k\to i\alpha}\left[\frac{\mathrm{e}^{ika_j}}{k^2+\alpha^2}(-1+\mathrm{i}ka_j)\left(-\frac{1}{3}ka_j+\frac{1}{30}a_j^3k^3-\dots\right)\right]\\&=1-\frac{2}{5}\alpha^2a_j^2+\frac{1}{3}\alpha^3a_j^3\,.\end{aligned} \tag{C.6}$$

Eqs. (C.5) and (C.6) together with the results of paper I for the angular integral, lead to eqs. (3.34)–(3.36).

Appendix D. Explicit evaluation of some connectors $\boldsymbol{A}(\omega)$

In this appendix we calculate with the aid of the results of appendix A, the connectors $\boldsymbol{A}^{(1,1)}(\omega)$, $\boldsymbol{A}^{(1,2a)}(\omega)$ and $\boldsymbol{A}^{(2a,2a)}(\omega)$ explicitly up to the third order.

a) We first consider $\boldsymbol{A}_{jk}^{(1,1)}(\omega)$, using the splitting (3.12) (cf. eq. (A.9)). As shown in eq. (A.10), the tensor $\boldsymbol{G}^{(1,1)}(\omega)$ is proportional to $\boldsymbol{G}^{(1,1)}$ given in paper I, eq. (6.5). Upon expansion of the α dependent forefactor in eq. (A.10) up to second order, one then immediately arrives at eq. (3.17). Similarly, eq. (A.11) shows that up to the order considered, $\boldsymbol{H}^{(1,1)}(\omega)$ may be approximated by $\boldsymbol{H}^{(1,1)}$, which may be found in eq. (6.20) of paper I. Finally, for $\boldsymbol{L}^{(1,1)}(\omega)$, we have according to eqs. (A.13) and (A.2)

$$\boldsymbol{L}_{jk}^{(1,1)}(\omega)=-\frac{3a_j}{4R_{jk}}\left(1+\frac{\alpha^2}{6}(a_j^2+a_k^2)\right)\cdot\frac{1}{2\pi}\int\mathrm{d}\hat{k}(\mathbb{1}-\hat{k}\hat{k})\alpha R_{jk}\,\mathrm{e}^{-\alpha R_{jk}|\xi_{jk}|}\,. \tag{D.1}$$

To calculate the angular integral, we note that it must be of the form $a\mathbb{1}+b\hat{r}_{jk}\hat{r}_{jk}$.

Contraction with the unit tensor then gives

$$3a + b = 4\alpha R_{jk} \int_0^1 d\xi_{jk}\, e^{-\alpha R_{jk}\xi_{jk}} = 4(1 - e^{-\alpha R_{jk}}) , \tag{D.2}$$

whereas contraction with $\hat{r}_{jk}\hat{r}_{jk}$ yields

$$a + b = 2\alpha R_{jk} \int_0^1 d\xi_{jk}(1 - \xi_{jk}^2)\, e^{-\alpha R_{jk}\xi_{jk}} = 2(1 - e^{-\alpha R_{jk}}) - 2J(\alpha R_{jk}) , \tag{D.3}$$

where the function J is defined in eq. (B.8). The solution of eqs. (D.2) and (D.3) is $a = 1 - \exp(-\alpha R_{jk}) + J(\alpha R_{jk})$, $b = 1 - \exp(-\alpha R_{jk}) - 3J(\alpha R_{jk})$. Taking all these results together, one arrives at eq. (3.19).

b) Next, we consider $\boldsymbol{A}^{(1,2a)}(\omega)$. Since $\boldsymbol{G}^{(1,2a)}(\omega)$, has, as discussed in section 3 sub c and in appendix A, an expansion in powers of $\alpha^2 a^2$, and since the term involving $\boldsymbol{G}^{(1,2a)}(\omega = 0)$ is already of second order in our expansion parameters, it is sufficient to approximate $\boldsymbol{G}^{(1,2a)}(\omega)$ by $\boldsymbol{G}^{(1,2a)}$, as given in eq. (6.11) of paper I. For $\boldsymbol{L}^{(1,2a)}(\omega, R_{jk})$, one has according to eq. (A.12) up to third order

$$(\boldsymbol{L}_{jk}^{(1,2a)}(\omega, R_{jk}))_{\alpha\beta\gamma} = -\frac{a_j a_k}{R_{jk}^2}\frac{3}{16\pi} \int d\hat{k}(\delta_{\alpha\beta}k_\gamma - \delta_{\alpha\gamma}k_\beta)\frac{\partial}{\partial \xi_{jk}}(\alpha R_{jk}\, e^{-\alpha R_{jk}|\xi_{jk}|}) . \tag{D.4}$$

Obviously this element of $\boldsymbol{L}^{(1,2a)}(\omega, R_{jk})$ must be of the form $c(\delta_{\alpha\beta}\hat{r}_{jk\gamma} - \delta_{\alpha\gamma}\hat{r}_{jk\beta})$. Contraction with $\delta_{\alpha\beta}\hat{r}_{jk\gamma}$ then yields

$$\begin{aligned} 2c &= -\frac{3}{2}\frac{a_j a_k}{R_{jk}^2} \int_0^1 d\xi_{jk}\xi_{jk}\frac{\partial}{\partial \xi_{jk}}\alpha R_{jk}\, e^{-\alpha R_{jk}\xi_{jk}} \\ &= -\frac{3}{4}\frac{a_j a_k}{R_{jk}^2}(\alpha R_{jk}\, e^{-\alpha R_{jk}} + e^{-\alpha R_{jk}} - 1) , \end{aligned} \tag{D.5}$$

in agreement with eq. (3.21).

c) Finally, we evaluate $\boldsymbol{A}^{(2a,2a)}(\omega)$. Also here $\boldsymbol{G}^{(2a2s)}(\omega)$ may be approximated by the static value, which can be found in eq. (6.33) of paper I. For $\boldsymbol{L}^{(2a,2a)}(\omega, R_{jk})$, one finds from eq. (A.12) to lowest order

$$\begin{aligned} \epsilon : \boldsymbol{L}_{jk}^{(2a,2a)}(\omega, R_{jk}) : \epsilon &= -\frac{a_j^2 a_k}{R_{jk}^3}\frac{3}{8\pi} \int d\hat{k}\epsilon : \hat{k}\mathbb{1}\hat{k} : \epsilon \frac{\partial^2}{\partial \xi_{jk}^2}\alpha R_{jk}\, e^{-\alpha R_{jk}|\xi_{jk}|} \\ &= \frac{a_j^2 a_k}{R_{jk}^2}\frac{3}{8\pi} \int d\hat{k}(\mathbb{1} - \hat{k}\hat{k})\frac{\partial^2}{\partial \xi_{jk}^2}\alpha R_{jk}\, e^{-\alpha R_{jk}|\xi_{jk}|} \\ &= \alpha^2 a_j^2 a_k \frac{3}{8\pi} \int d\hat{k}(\mathbb{1} - \hat{k}\hat{k})(\alpha R_{jk}\, e^{-\alpha R_{jk}|\xi_{jk}|} - 2\delta(\xi_{jk})) . \end{aligned} \tag{D.6}$$

In the last line, use has been made of eq. (A.6). Let us write

$$\frac{1}{2\pi}\int \mathrm{d}\hat{k}(\mathbb{1} - \hat{k}\hat{k})(\alpha R_{jk}\,\mathrm{e}^{-\alpha R_{jk}|\xi_{jk}|} - 2\delta(\xi_{jk})) = d(\mathbb{1} - \hat{r}_{jk}\hat{r}_{jk}) + e(\hat{r}_{jk}\hat{r}_{jk} - \tfrac{1}{3}\mathbb{1})\,. \tag{D.7}$$

By taking the trace of the last equation, one gets

$$2d = 2\int_{-1}^{1} \mathrm{d}\xi_{jk}(\alpha R_{jk}\,\mathrm{e}^{-\alpha R_{jk}|\xi_{jk}|} - 2\delta(\xi_{jk})) = 4\mathrm{e}^{-\alpha R_{jk}}\,, \tag{D.8}$$

while contraction with $\hat{r}_{jk}\hat{r}_{jk}$ yields

$$\frac{2}{3}e = \int_{-1}^{+1} \mathrm{d}\xi_{jk}(1 - \xi_{jk}^2)(\alpha R_{jk}\,\mathrm{e}^{-\alpha R_{jk}|\xi_{jk}|} - 2\delta(\xi_{jk})) = 2\mathrm{e}^{-\alpha R_{jk}} - 2J(\alpha R_{jk})\,, \tag{D.9}$$

with, as before, J given by eq. (B.8). Upon substitution of eqs. (D.7)–(D.9) into eq. (D.6), one arrives, together with the result for $\boldsymbol{G}^{(2a,2a)}$ of paper I, at eq. (3.40).

Note added in proof

After having submitted this paper to Physica, we learned that I. Pieńkowska in a recent article in Archives of Mechanics **33** No. 3 (1982) has evaluated frequency-dependent friction tensors, taking into account many-sphere hydrodynamic interactions. Her analysis was based on an extension of a method for the calculation of many-sphere friction tensors, developed by Oshizaki and Yamakawa (J. Chem. Phys. **73** (1980) 578). In as far as a comparison could be made, Pieńkowska's explicit result agrees with our formula (4.9).

References

1) J. Happel and H. Brenner, Low Reynolds Number Hydrodynamics (Noordhoff, Leiden, 1973).
2) M. Smoluchowski, Bull. Int. Acad. Polonaise Sci. Lett. **1A** (1911) 28.
3) G.J. Kynch, J. Fluid Mech. **5** (1959) 193.
4) G.K. Batchelor and J.T. Green, J. Fluid Mech. **56** (1972) 375.
5) G.K. Batchelor, J. Fluid Mech. **74** (1976) 1.
6) B.U. Felderhof, Physica **89A** (1977) 373.
7) P. Reuland, B.U. Felderhof and R.B. Jones, Physica **93A** (1978) 465.
8) P. Mazur, Physica **110A** (1982) 128.
9) R. Schmitz and B.U. Felderhof, Physica **113A** (1982) 90 and 103.
10) P. Mazur and W. van Saarloos, Physica **115A** (1982) 21.
11) D.R. Bauer, in: Polymer Colloids II, R.M. Fitch ed. (Plenum, New York, 1980).
12) D.R. Bauer, J. Phys. Chem. **84** (1980) 1592.
13) M.M. Kops-Werkhoven and H.M. Fijnaut, J. Chem. Phys. **74** (1981) 1618; **77** (1982) 2242.

14) G.D.J. Phillies, Phys. Rev. **A24** (1984) 1939.
15) B.U. Felderhof, J. Phys. **A11** (1978) 929.
16) C.W.J. Beenakker and P. Mazur, Phys. Lett. **91A** (1982) 290.
17) G.D.J. Phillies, J. Chem. Phys. **77** (1982) 2623.
18) L.D. Landau and E.M. Lifshitz, Fluid Mechanics (Pergamon, New York, 1978).
19) P. Mazur and D. Bedeaux, Physica **76** (1974) 235.
20) P. Mazur and G. van der Zwan, Physica **92A** (1978) 483.
21) I. Proudman and J.R.A. Pearson, J. Fluid Mech. **2** (1957) 237.
22) S. Hess and W. Köhler, Formeln zur Tensor-Rechnung (Palm und Enke, Erlangen, 1980).
23) B.P. Hills, Physica **80A** (1975) 360.
24) G.L. Paul and P.N. Pusey, J. Phys. **A14** (1981) 3301.
25) A.J. Hurd, The Lattice Dynamics of Colloidal Crystals (thesis Univ. of Colorado, Boulder, 1981).
26) A.J. Hurd, N.A. Clark, R.C. Mockler, W.J. Sullivan, Phys. Rev. **A26** (1982) 2869.
27) P.N. Pusey and W. van Megen, J. Physique **44** (1983) 285.