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6 Precise multipole method for calculating hydrodynamic interactions between spherical particles in the Stokes flow

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1. Introduction

Dynamics of micro-particles in water-based fluids and macroscopic properties of such dispersive systems are of fundamental importance for numerous biological and industrial applications [1-7]. Typically, for such systems fluid inertia is irrelevant, and the mechanisms of locomotion as well as basic features of the particle dynamics differ significantly from those valid at the macroscopic scale. Fluid motion at the micro-scale usually satisfies

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the stationary Stokes equations, which have to be supplemented by appropriate boundary conditions at the particle surfaces and at the interfaces, which confine the fluid [6, 7, 8]. Efficient and accurate methods of solving these equations are necessary to investigate Stokesian dynamics of micro-particles and to determine structure and effective transport properties of dispersive systems. For spherical non-deformable micro-particles, the method of interest is the spherical-multipole expansion, corrected for lubrication [9, 10]. Its main advantage is the high accuracy, which is controlled by the choice of the multipole order of the truncation. Moreover, the method has been implemented numerically, and the HYDROMULTIPOLE codes have been extensively tested and applied to many physical systems, with various types of particles and interfaces confining the fluid.

In this paper, an outline of the spherical-multipole expansion applied to the stationary Stokes equations is given, based on the algorithm developed by Cichocki, Felderhof, Jones, Schmitz and collaborators. In Sec. 2, geometry of the system is specified, as well as boundary conditions at the particle surfaces and at the interfaces, which confine the fluid. The generalized friction and mobility problems for the particles are formulated. The Stokes problem for the fluid flow is transformed into a set of boundary integral equations for the force density at the particle surfaces. In Sec. 3, these equations are solved by projecting onto a complete set of multipole functions, and truncating at a certain multipole order L, with the details explained in the Appendices. For close particles in relative motion, the multipole expansion is slowly convergent with the increasing L. Therefore in Sec. 4, the multipole expansion is corrected for lubrication and some estimates of the precision are given. In Sec. 5, modifications needed to describe motion of particle conglomerates are pointed out. Finally, Sec. 6 contains examples of applications.

2. Fluid, particles and boundaries

Consider N particles immersed in a viscous fluid. Assume that the particles are non-deformable. Imagine that external (non-hydrodynamic) forces $\mathcal{F}_i$ and torques $\mathcal{T}_i$ are applied to each particle $i = 1, ..., N$, and there exist an ambient fluid flow $\mathbf{v}_\infty(\mathbf{r})$. Each particle $i = 1, ..., N$ has a spherical shape of radius a_i, and moves with a translational and rotational velocity, $\mathbf{U}_i$ and $\mathbf{\Omega}_i$. The resulting fluid flow is characterized by a very small Reynolds number Re $\ll 1$ and the fluid inertia effects are negligible [6, 7]. Here Re is the product of a particle velocity and its radius, divided by the fluid kinematic viscosity. The Stokes number Sk is also much smaller than unity and the particle inertia is also irrelevant. Here Sk is the product of the particle velocity, its radius, and the larger of the particle and the fluid densities, divided by the fluid dynamic viscosity [11]. The Peclet number is large, $\mathrm{Pe} \gg 1$, and the Brownian motion is irrelevant [1]. Here Pe is the product of a particle velocity and its radius, divided by the diffusion constant. Moreover, the Strouhal number St is not much larger than unity, and the fluid flow is stationary [3]. Here St is the ratio of the characteristic frequency of the fluid velocity variations and the fluid velocity, multiplied by the characteristic dimension.

For such a system, the hydrodynamic friction forces and torques exerted by the moving particles on the fluid are equal to the external forces $\mathcal{F}_i$ and torques $\mathcal{T}_i$ imposed on the particles. In the generalized friction problem, the question is what are $\mathcal{F}_i$ and $\mathcal{T}_i$, if the particle translational $\mathbf{U}_j$ and rotational $\mathbf{\Omega}_j$ velocities and the ambient flow $\mathbf{v}_\infty(\mathbf{r})$ are given. In the generalized mobility problem, $\mathcal{F}_i$, $\mathcal{T}_i$ and $\mathbf{v}_\infty(\mathbf{r})$ are known while $\mathbf{U}_j$ and $\mathbf{\Omega}_j$ are searched for. Solving one of these problems (or a mixed one) corresponds to evaluation of hydrodynamic interactions between the particles.

In this paper, it will be outlined how to apply the spherical-multipole method to solve the generalized friction and mobility problems [9, 10] and evaluate the fluid flow.

2.1. Basic equations for the fluid flow

For the system specified above, the fluid velocity $\mathbf{v}$ and pressure p satisfy the stationary Stokes equations [6, 7],

$$\eta\nabla^2\mathbf{v} - \nabla p = 0, \tag{1}$$

$$\nabla\cdot\mathbf{v} = 0, \tag{2}$$

where η is the fluid dynamic viscosity. The above set of partial differential equations has to be supplemented by the corresponding boundary conditions at the surface S_i of each spherical particle $i = 1, ..., N$, and at the interfaces, which confine the fluid.

The ambient-flow velocity $\mathbf{v}_\infty$ and pressure p_∞ satisfy the Stokes equations (1) in the absence of particles. When the group of N particles is immersed, it affects the surrounding fluid, but there is no change far away from the particles. For an unbounded fluid, the corresponding boundary condition at infinity reads,

$$\mathbf{v}(\mathbf{r}) - \mathbf{v}_\infty(\mathbf{r}) \to \mathbf{0}, \quad \text{for } |\mathbf{r}| \to \infty. \tag{3}$$

For a confined fluid, both the ambient and the actual flows, $\mathbf{v}_\infty(\mathbf{r})$ and $\mathbf{v}(\mathbf{r})$, have to satisfy the proper boundary conditions at the interfaces. The multipole method has been developed and applied for various geometries: 3D or 2D-periodic boundary conditions [12-18] and for a fluid limited by one [19] or two parallel flat interfaces [20]. Such an interface may be a hard (solid) wall [21, 22], a free surface [23, 24], or a fluid-fluid boundary, with or without a surfactant [25, 26].

These boundary conditions are explicitly listed in the next section. All of them can be also applied at the particle surfaces, and the multipole method has been developed to describe non-deformable spherical particles made of solid, fluid or gas, with the clean surface or covered with a surfactant [27-29]. For clarity of presentation of the basic concepts, this paper is mainly focused on solid particles and the stick boundary conditions on their surfaces,

$$\mathbf{v}(\mathbf{r}) = \mathbf{w}_i(\mathbf{r}) \equiv \mathbf{U}_i + \boldsymbol{\Omega}_i \times (\mathbf{r} - \mathbf{R}_i), \quad \text{for } \mathbf{r} \in S_i, \quad \mathbf{i} = 1, ..., N, \tag{4}$$

where $\mathbf{R}_i$ stands for the position of the center of particle i. Modifications needed to describe hydrodynamic interactions between other types of particles and interfaces will be mentioned; the full treatment can be found e.g. in Refs. [27-29].

2.2. Boundary conditions

The multipole method has been developed for the following boundary conditions at the surface I confining the fluid and at the particle surfaces S_i, $i = 1...N$.

If the fluid is in contact with a smooth solid surface, the stick (or no-slip) boundary conditions apply. The fluid velocity at the surface S_i of a solid spherical particle i is equal to its rigid velocity, $\mathbf{v}(\mathbf{r}) = \mathbf{w}_i(\mathbf{r})$, as in Eq. (4). If the fluid in a half-space $z > 0$ is limited at $z = 0$ by a flat surface I, which is the motionless hard wall, the stick boundary conditions at I have the form,

$$\mathbf{v}(\mathbf{r}) = 0, \quad \text{for } \mathbf{r} = (x, y, 0). \tag{5}$$

The above model can be generalized, allowing for a slip at the boundary. The mixed stick-slip boundary conditions at the particle i have the form [30],

$$\mathbf{n}_i \cdot \mathbf{v}(\mathbf{r}) = \mathbf{n}_i \cdot \mathbf{w}_i(\mathbf{r}), \quad \text{for} \quad \mathbf{r} \in S_i, \tag{6}$$

$$\mathbf{t}_i \cdot (\mathbf{v}(\mathbf{r}) - \mathbf{w}_i(\mathbf{r})) = (\lambda_i/\eta)\, \mathbf{t}_i \cdot \sigma(\mathbf{r}) \cdot \mathbf{n}_i, \quad \text{for} \quad \mathbf{r} \in S_i, \tag{7}$$

where $\mathbf{n}_i = (\mathbf{r} - \mathbf{R}_i)/|\mathbf{r} - \mathbf{R}_i|$ and $\mathbf{t}_i$ are the unit vectors normal and tangential to the particle surface S_i. The first condition, Eq. (6), expresses the fact that no fluid passes through the spherical surface. The second one, Eq. (7), states that the tangential component of the force exerted by the fluid on the unit surface of the sphere is proportional to the slip of the local tangential velocity, i.e. to the difference between the fluid-flow and the particle-surface velocities. The Cartesian components of the fluid stress tensors $\boldsymbol{\sigma}$ are given by the relation,

$$\sigma_{\alpha\beta} = \eta(\partial_\alpha v_\beta + \partial_\beta v_\alpha) - p\delta_{\alpha\beta}. \tag{8}$$

The value of the slip parameter $\lambda_i = 0$ in Eq. (7) corresponds to the stick boundary conditions, the value $\lambda_i = \infty$ to the perfect slip.

We consider now the boundary conditions at an interface between two fluids with different viscosities [3]. We assume that the interface is not deformable owing to a very high surface tension.

The spherical surface of a droplet with an internal viscosity η' different from the viscosity of the host fluid η is described by the condition that the normal components of the flows $\mathbf{v}(\mathbf{r})$ and $\mathbf{v}'(\mathbf{r})$ outside and inside the droplet are the same and equal to the normal component of the droplet velocity $\mathbf{U}_i$,

$$\mathbf{n}_i \cdot \mathbf{v}(\mathbf{r}) = \mathbf{n}_i \cdot \mathbf{U}_i, \text{ for } \mathbf{r} \in \mathbf{S}_i, \tag{9}$$

$$\mathbf{n}_i \cdot \mathbf{v}'(\mathbf{r}) = \mathbf{n}_i \cdot \mathbf{U}_i, \text{ for } \mathbf{r} \in \mathbf{S}_i. \tag{10}$$

This condition expresses the fact that no fluid passes through the droplet non-deformable surface. Moreover, the tangential velocity and tangential stress are continuous,

$$\mathbf{t}_i \cdot \mathbf{v}(\mathbf{r}) = \mathbf{t}_i \cdot \mathbf{v}'(\mathbf{r}), \quad \text{for} \quad \mathbf{r} \in S_\mathrm{i}, \tag{11}$$

$$\mathbf{t}_i \cdot \sigma(\mathbf{r}) \cdot \mathbf{n}_i = \mathbf{t}_i \cdot \sigma'(\mathbf{r}) \cdot \mathbf{n}_i, \quad \text{for} \quad \mathbf{r} \in S_\mathrm{i}, \tag{12}$$

where the Cartesian components of the fluid stress tensors σ' inside the droplet are given by

$$\sigma'_{\alpha\beta} = \eta'(\partial_\alpha v'_\beta + \partial_\beta v'_\alpha) - p'\delta_{\alpha\beta}. \tag{13}$$

Notice that the rotational velocity $\boldsymbol{\Omega}$ is not a relevant variable in the description of the droplet motion, and it has to be excluded from the friction and mobility problems, formulated in Sec. 2.3.

The boundary conditions at the droplet surface can be easily modified to describe a flat fluid-fluid interface at $z = 0$, when the fluid in a half-space

$z > 0$ has the viscosity η, and the fluid on the other side of the interface has the viscosity η',

$$\mathbf{n} \cdot \mathbf{v}(\mathbf{r}) = \mathbf{n} \cdot \mathbf{v}'(\mathbf{r}) = 0, \quad \text{for } \mathbf{r} = (x, y, 0), \tag{14}$$

$$\mathbf{t} \cdot \mathbf{v}(\mathbf{r}) = \mathbf{t} \cdot \mathbf{v}'(\mathbf{r}), \quad \text{for } \mathbf{r} = (x, y, 0), \tag{15}$$

$$\mathbf{t} \cdot \boldsymbol{\sigma}(\mathbf{r}) \cdot \mathbf{n} = \mathbf{t} \cdot \boldsymbol{\sigma}'(\mathbf{r}) \cdot \mathbf{n}, \quad \text{for } \mathbf{r} = (x, y, 0). \tag{16}$$

where $\mathbf{v}(\mathbf{r})$ and $\mathbf{v}'(\mathbf{r})$ are the flows on the $z > 0$ and $z < 0$ sides of the interface.

For a fluid-fluid interface covered with an incompressible surfactant the tangential stress continuity relations (12) and (16) are replaced by the condition that the flow along the interface is incompressible,

$$\nabla_s \cdot \mathbf{v}_s = 0, \tag{17}$$

where ∇_s is the gradient operator along the interface and $\mathbf{v}_s$ is the tangential component of the flow velocity.

It is of special interest to consider a gas-liquid interface (free surface). The corresponding boundary conditions can be obtained form those specified above in the limit $\eta' = 0$. Across a free surface between a gas and a liquid, there is no liquid flow and the tangential stress is equal to zero at the interface. For a bubble,

$$\mathbf{n}_i \cdot \mathbf{v}(\mathbf{r}) = \mathbf{n}_i \cdot \mathbf{U}_i, \quad \text{for } \mathbf{r} \in S_i, \tag{18}$$

$$\mathbf{t}_i \cdot \boldsymbol{\sigma}(\mathbf{r}) \cdot \mathbf{n}_i = 0, \quad \text{for } \mathbf{r} \in S_i \tag{19}$$

For a fluid in a half-space $z > 0$, limited by a flat free surface at $z = 0$, the free boundary conditions have the form,

$$\mathbf{n} \cdot \mathbf{v}(\mathbf{r}) = 0, \quad \text{for } \mathbf{r} = (x, y, 0), \tag{20}$$

$$\mathbf{t} \cdot \boldsymbol{\sigma}(\mathbf{r}) \cdot \mathbf{n} = 0, \quad \text{for } \mathbf{r} = (x, y, 0). \tag{21}$$

2.3. Friction and mobility of the particles

Alternatively to the boundary conditions for the (unknown) fluid velocity, the effect of the suspended particles on the surrounding fluid can be also described in terms of the distribution of the (unknown) induced forces

$\mathbf{f} = (\mathbf{f}_1, ..., \mathbf{f}_N)$, exerted by the particles on the fluid [30-32]. For the stick boundary conditions, $\mathbf{f}_i(\mathbf{r}) = -\delta(|\mathbf{r} - \mathbf{R}_i| - a_i)\, \boldsymbol{\sigma}(\mathbf{r}) \cdot \mathbf{n}$, where $\boldsymbol{\sigma}$ is the stress tensor and $\mathbf{n}$ is the unit vector normal to the particle surface, pointing into the fluid. The subsequent moments (integrals) of the force density are the forces $\mathcal{F}_i$, torques $\mathcal{T}_i$, stresslets $\mathbf{S}_i$ exerted by the sphere i on the fluid,

$$\mathcal{F}_i = \int \mathbf{f}_i(\mathbf{r})\, d^3\mathbf{r}, \quad \mathcal{T}_i = \int (\mathbf{r} - \mathbf{R}_i) \times \mathbf{f}_i(\mathbf{r})\, d^3\mathbf{r}, \quad \mathbf{S}_i = \int \overline{(\mathbf{r} - \mathbf{R}_i)\mathbf{f}_i(\mathbf{r})}\, d^3\mathbf{r}, \ ... \tag{22}$$

where the bar over a tensor denotes its symmetric traceless part, and the higher moments are indicated by the dots. For a spherical particle, the Cartesian components of the stresslet are given as,

$$S_{i,\alpha\beta} = \frac{1}{2} \int [(r_\alpha - R_{i,\alpha})\mathrm{f}_{i,\beta} + \mathrm{f}_{i,\alpha}(r_\beta - R_{i,\beta})]\, d^3\mathbf{r}. \tag{23}$$

In the following, we combine the forces, torques and stresslets into $\mathcal{F} = (\mathcal{F}_1, ..., \mathcal{F}_N)$, $\mathcal{T} = (\mathcal{T}_1, ..., \mathcal{T}_N)$ and $\mathbf{S} = (\mathbf{S}_1, ..., \mathbf{S}_N)$, respectively. In analogy, we represent the particle translational and rotational velocities as $\mathbf{U} = (\mathbf{U}_1, ..., \mathbf{U}_N)$ and $\boldsymbol{\Omega} = (\boldsymbol{\Omega}_1, ..., \boldsymbol{\Omega}_N)$, respectively. In a similar way, $\mathbf{v}_\infty = (\mathbf{v}_{\infty 1}, ..., \mathbf{v}_{\infty N})$, $\boldsymbol{\omega}_\infty = (\boldsymbol{\omega}_{\infty 1}, ..., \boldsymbol{\omega}_{\infty N})$, $\mathbf{g}_\infty = (\mathbf{g}_{\infty 1}, ..., \mathbf{g}_{\infty N})$, ... denote the ambient flow velocities, their gradients and higher derivatives, taken at the center of each sphere, with $\mathbf{v}_{\infty i} = \mathbf{v}_\infty(\mathbf{R}_i)$, $\boldsymbol{\omega}_{\infty i} = \frac{1}{2} \nabla \times \mathbf{v}_\infty(\mathbf{r})|_{\mathbf{r}=\mathbf{R}_i}$ and $g_{\infty i,\alpha\beta} = \frac{1}{2}[\nabla_\alpha v_{\infty\beta}(\mathbf{r}) + \nabla_\beta v_{\infty\alpha}(\mathbf{r})]_{\mathbf{r}=\mathbf{R}_i}$.

Owing to linearity of the Stokes equations (1)-(2), $\mathcal{F}$, $\mathcal{T}$ and $\mathbf{S}$ depend linearly on $\mathbf{v}_\infty - \mathbf{U}$, $\boldsymbol{\omega}_\infty - \boldsymbol{\Omega}$ and $\mathbf{g}_\infty$,

$$\begin{pmatrix} \mathcal{F} \\ \mathcal{T} \\ \mathbf{S} \\ \vdots \end{pmatrix} = - \begin{pmatrix} \zeta^{tt} & \zeta^{tr} & \zeta^{td} & .. \\ \zeta^{rt} & \zeta^{rr} & \zeta^{rd} & .. \\ \zeta^{dt} & \zeta^{dr} & \zeta^{dd} & .. \\ \vdots & \vdots & \vdots & \vdots \end{pmatrix} \cdot \begin{pmatrix} \mathbf{v}_\infty - \mathbf{U} \\ \omega_\infty - \boldsymbol{\Omega} \\ \mathbf{g}_\infty \\ \vdots \end{pmatrix}, \tag{24}$$

where the components $\boldsymbol{\zeta}^{pq}$ with p, $q = t, r, d, ...$ form the *generalized friction* (*or grand resistance*) *symmetric tensor* [7, 33, 34, 35]. Each component $\boldsymbol{\zeta}^{pq}$ consists of the elements ζ^{pq}_{ij} with $i, j = 1...N$. Each of them depends on the configuration of all the particles. In particular, the elements with p, $q = t$, r form the 6N×6N *friction tensor* for N spherical particles, in brief denoted as $\boldsymbol{\zeta}$,

$$\zeta = \begin{pmatrix} \zeta^{tt} & \zeta^{tr} \\ \zeta^{rt} & \zeta^{rr} \end{pmatrix}. \tag{25}$$

Evaluation of the generalized friction tensor is essential to solve the generalized friction problem [6, 34], i.e. to determine the hydrodynamic friction forces and torques exerted by the particles on the fluid, if their motion and the ambient fluid flow are known.

On the other hand, if the hydrodynamic friction forces and torques exerted by the particles on the flow and the ambient fluid flow are known, the particle motion is determined by solving the generalized mobility problem [6, 34],

$$\begin{pmatrix} \mathbf{v}_\infty - \mathbf{U} \\ \omega_\infty - \mathbf{\Omega} \\ \mathbf{S} \\ \vdots \end{pmatrix} = - \begin{pmatrix} \boldsymbol{\mu}^{tt} & \boldsymbol{\mu}^{tr} & \boldsymbol{\mu}^{td} & .. \\ \boldsymbol{\mu}^{rt} & \boldsymbol{\mu}^{rr} & \boldsymbol{\mu}^{rd} & .. \\ \boldsymbol{\mu}^{dt} & \boldsymbol{\mu}^{dr} & \boldsymbol{\mu}^{dd} & .. \\ \vdots & \vdots & \vdots & \vdots \end{pmatrix} \cdot \begin{pmatrix} \mathcal{F} \\ \mathcal{T} \\ \mathbf{g}_\infty \\ \vdots \end{pmatrix}, \tag{26}$$

with the use of the *generalized* (*grand*) *mobility tensor*, which consists of the elements $\boldsymbol{\mu}^{pq}$, $p,\ q = t,\ r,\ d,\ \ldots$ and depends on the configuration of all the particles. The elements with $p,\ q = t,\ r$ only form the mobility tensor for N spherical particles, denoted as $\boldsymbol{\mu}$,

$$\mu = \begin{pmatrix} \mu^{tt} & \mu^{tr} \\ \mu^{rt} & \mu^{rr} \end{pmatrix}. \tag{27}$$

Note that $\boldsymbol{\mu}$ is the inverse of $\boldsymbol{\zeta}$,

$$\mu = \zeta^{-1}, \tag{28}$$

but the generalized mobility tensor is obtained by only a partial inverse of the generalized friction tensor.

In many applications, the question is what is the particle motion under given external forces $\mathcal{F}$ and torques $\mathcal{T}$, and an ambient flow $\mathbf{v}_\infty(\mathbf{r})$. Solution of this problem is constructed from Eq. (24), which is now rewritten as,

$$\begin{pmatrix} \mathcal{F} \\ \mathcal{T} \end{pmatrix} = \begin{pmatrix} \mathcal{F}_\infty \\ \mathcal{T}_\infty \end{pmatrix} + \zeta \cdot \begin{pmatrix} \mathbf{U} \\ \Omega \end{pmatrix}, \tag{29}$$

where $\mathcal{F}_\infty$ and $\mathcal{T}_\infty$ are hydrodynamic forces and torques exerted by the motionless particles on the fluid in the presence of an ambient flow, but the absence of external forces,

$$\begin{pmatrix} \mathcal{F}_\infty \\ \mathcal{T}_\infty \end{pmatrix} = -\begin{pmatrix} \zeta^{tt} & \zeta^{tr} & \zeta^{td} & .. \\ \zeta^{rt} & \zeta^{rr} & \zeta^{rd} & .. \end{pmatrix} \cdot \begin{pmatrix} \mathbf{v}_\infty \\ \omega_\infty \\ \mathbf{g}_\infty \\ \vdots \end{pmatrix}. \tag{30}$$

Eq. (29) is now solved for the particle translational and angular velocities,

$$\begin{pmatrix} \mathbf{U} \\ \boldsymbol{\Omega} \end{pmatrix} = \boldsymbol{\mu} \cdot \begin{pmatrix} \mathcal{F} - \mathcal{F}_\infty \\ \mathcal{T} - \mathcal{T}_\infty \end{pmatrix} = \boldsymbol{\mu} \cdot \begin{pmatrix} \mathcal{F} \\ \mathcal{T} \end{pmatrix} + \boldsymbol{C} \cdot \begin{pmatrix} \mathbf{v}_\infty \\ \omega_\infty \\ \mathbf{g}_\infty \\ \vdots \end{pmatrix}, \tag{31}$$

where $\boldsymbol{C}$ is the convection operator [36],

$$\boldsymbol{C} = \boldsymbol{\mu} \cdot \begin{pmatrix} \zeta^{tt} & \zeta^{tr} & \zeta^{td} & .. \\ \zeta^{rt} & \zeta^{rr} & \zeta^{rd} & .. \end{pmatrix}. \tag{32}$$

Evaluation of the generalized friction and mobility tensors is based on solving the boundary integral equation by the multipole expansion. These procedure will be outlined in the following sections.

2.4. Boundary integral equation

With the use of the induced forces [30-32], the set of partial differential equations (1)-(2) for the fluid velocity and pressure can be transformed into a set of boundary integral equations for the density of the induced forces $\mathbf{f_i}$. This procedure will be outlined below.

The fluid flow field outside the particles can be represented as [9]

$$\mathbf{v}(\mathbf{r}) = \mathbf{v}_\infty(\mathbf{r}) + \sum_{j=1}^{N} \int \mathbf{T}(\mathbf{r}, \overline{\mathbf{r}}) \cdot \mathbf{f}_j(\overline{\mathbf{r}})\, d^3\overline{\mathbf{r}}, \tag{33}$$

In the above equation $\mathbf{v}_\infty$ denotes the imposed ambient flow and the integral term describes the flow generated by the induced forces. Here

$\mathbf{T}(\mathbf{r},\overline{\mathbf{r}})$ is the Green function for the Stokes flow in the presence of the boundaries. It is convenient to write is as

$$\mathbf{T}(\mathbf{r},\overline{\mathbf{r}}) = \mathbf{T}_0(\mathbf{r}-\overline{\mathbf{r}}) + \tilde{\mathbf{T}}(\mathbf{r},\overline{\mathbf{r}}), \tag{34}$$

where the Oseen tensor,

$$\mathbf{T}_0(\mathbf{r}) = \frac{1}{8\pi\eta}\frac{\mathbf{1}+\hat{\mathbf{r}}\hat{\mathbf{r}}}{r}, \tag{35}$$

is the Green function for the Stokes flow in the unbounded space. In case of the unbounded space, $\tilde{\mathbf{T}}(\mathbf{r},\overline{\mathbf{r}}) = 0$. Otherwise $\tilde{\mathbf{T}}(\mathbf{r},\overline{\mathbf{r}})$ describes the flow reflected from the interfaces, which confine the fluid.

For a fluid in a half-space $z > 0$, limited by a flat free surface at $z = 0$, i.e. for the boundary conditions given by Eqs. (18)-(19), the tensor $\tilde{\mathbf{T}}$ has the form [23, 21],

$$\tilde{\mathbf{T}}(\mathbf{r},\overline{\mathbf{r}}) = \mathbf{T}_0(\mathbf{r}-\overline{\mathbf{r}}') \cdot \mathbf{P}, \tag{36}$$

where

$$\mathbf{P} = 1 - 2\mathbf{nn}, \tag{37}$$

$$\overline{\mathbf{r}}' = \mathbf{P}\cdot\overline{\mathbf{r}} = (\overline{x},\overline{y},-\overline{z}), \tag{38}$$

with $\overline{\mathbf{r}} = (\overline{x},\overline{y},\overline{z})$ and the unit vector $\mathbf{n}$ normal to the interface pointing into the fluid.

For a fluid in a half-space $z > 0$, limited by a hard wall at $z = 0$, i.e. for the stick boundary conditions given by Eq. (5), the tensor $\tilde{\mathbf{T}}$ has the form [6],

$$\tilde{\mathbf{T}}(\mathbf{r},\overline{\mathbf{r}}) = -\mathbf{T}_0(\mathbf{r}-\overline{\mathbf{r}}') - 2\overline{z}\,\mathbf{n}\cdot\mathbf{T}_0(\mathbf{r}-\overline{\mathbf{r}}')\overleftarrow{\nabla}_{\mathbf{r}} \cdot \mathbf{P} + \overline{z}^2\nabla_{\mathbf{r}}^2\mathbf{T}_0(\mathbf{r}-\overline{\mathbf{r}}') \cdot \mathbf{P}, \tag{39}$$

where

$$[\mathbf{W}(\mathbf{r})\overleftarrow{\nabla}_{\mathbf{r}}]_{\alpha\beta} = \frac{\partial}{\partial \mathbf{r}_\beta}[\mathbf{W}(\mathbf{r})]_\alpha. \tag{40}$$

To obtain the boundary integral equation for the force density $\mathbf{f}_j$, the integral representation (33) will now be combined with the boundary conditions.

In particular, for the stick boundary conditions (4) on the surface of particle i,

$$\mathbf{w}_i(\mathbf{r}) = \mathbf{v}_\infty(\mathbf{r}) + \sum_{j=1}^{N} \int \mathbf{T}(\mathbf{r}-\overline{\mathbf{r}}) \cdot \mathbf{f}_j(\overline{\mathbf{r}})\, d^3\overline{\mathbf{r}}, \quad \mathbf{r} \in S_i. \tag{41}$$

Other boundary conditions on the sphere require more detailed analysis. In general, we decompose the flow around particle i into two flows

$$\mathbf{v}(\mathbf{r}) = \mathbf{v}_i^{\text{in}}(\mathbf{r}) + \mathbf{v}_i^{\text{out}}(\mathbf{r}), \tag{42}$$

where $\mathbf{v}_i^{\text{in}}$ is the incident (regular) and $\mathbf{v}_i^{\text{out}}$ the scattered (singular) part of the total flow $\mathbf{v}(\mathbf{r})$ around the particle i. The singular flow is given by

$$\mathbf{v}_i^{\text{out}}(\mathbf{r}) = \int \mathbf{T}_0(\mathbf{r}-\overline{\mathbf{r}}) \cdot \mathbf{f}_j(\overline{\mathbf{r}})\, d^3\overline{\mathbf{r}}, \tag{43}$$

and it represents the flow scattered by the considered particle.

The induced force distribution $\mathbf{f}_i$ on the surface of the particle i and the flow $\mathbf{v}_i^{\text{in}}$ incident to this particle are linearly related. The relation can be expressed in the form

$$\mathbf{f}_i(\mathbf{r}) = -\int \mathbf{Z}_i(\mathbf{r}-\mathbf{R}_i, \overline{\mathbf{r}}-\mathbf{R}_i) \cdot [\mathbf{v}_i^{\text{in}}(\overline{\mathbf{r}}) - \mathbf{w}_i(\overline{\mathbf{r}})]\, d\overline{\mathbf{r}}, \tag{44}$$

where the single-particle friction operator $\mathbf{Z}_i$ depends on the specific boundary conditions only at the particle i, and is explicitly obtained by solving the Stokes equations for an isolated particle subject to an external flow [28, 37]. Examples of such explicit expressions are given in Refs. [27-29].

In Eq. (44), the expression $\mathbf{v}_i^{\text{in}} - \mathbf{w}_i$ denotes the incident flow in the frame of reference moving with the particle. The Stokes flow $\mathbf{v}_i^{\text{in}} - \mathbf{w}_i$ is fully determined by its boundary value on the particle surface S_i and the condition that it is nonsingular in the region occupied by the particle. Thus Eq. (44) can be interpreted as a linear functional relation between the force vector field $\mathbf{f}_i$ and the incident flow $\mathbf{v}_i^{\text{in}} - \mathbf{w}_i$ on the surface S_i. Since a non-zero incident flow always produces a non-zero force distribution $\mathbf{f}_i$, the relation (44) can be inverted,

$$\mathbf{v}_i^{\mathrm{in}}(\mathbf{r})-\mathbf{w}_i(\mathbf{r})=-\int \mathbf{Z}_i^{-1}(\mathbf{r}-\mathbf{R}_i,\overline{\mathbf{r}}-\mathbf{R}_i)\cdot\mathbf{f}_i(\overline{\mathbf{r}})\,d^3\overline{\mathbf{r}},\quad \mathbf{r}\in S_i. \tag{45}$$

By collecting relations (42), (43) and (45) we obtain the expression

$$\mathbf{v}(\mathbf{r})=\mathbf{w}_i(\mathbf{r})-\int \mathbf{Z}_i^{-1}(\mathbf{r}-\mathbf{R}_i,\overline{\mathbf{r}}-\mathbf{R}_i)\cdot\mathbf{f}_i(\overline{\mathbf{r}})\,d^3\overline{\mathbf{r}}+\int \mathbf{T}_0(\mathbf{r}-\overline{\mathbf{r}})\cdot\mathbf{f}_i(\overline{\mathbf{r}})\,d^3\overline{\mathbf{r}},\ \mathbf{r}\in S_i, \tag{46}$$

for the flow $\mathbf{v}(\mathbf{r})$ at the surface S_i of the particle i.

Using the integral representation (33) at the surface S_i of the particle i, and applying the boundary condition (46), we obtain the set of the boundary-integral equations for the induced force densities $\mathbf{f}_i$ [22],

$$\begin{aligned}\mathbf{w}_i(\mathbf{r})-\mathbf{v}_\infty(\mathbf{r})=&\int \mathbf{Z}_i^{-1}(\mathbf{r}-\mathbf{R}_i,\overline{\mathbf{r}}-\mathbf{R}_i)\cdot\mathbf{f}_i(\overline{\mathbf{r}})\,d^3\overline{\mathbf{r}}+\\ &\sum_{j=1}^{N}\int[(1-\delta_{ij})\mathbf{T}_0(\mathbf{r}-\overline{\mathbf{r}})+\tilde{\mathbf{T}}(\mathbf{r},\overline{\mathbf{r}})]\cdot\mathbf{f}_j(\overline{\mathbf{r}})\,d^3\overline{\mathbf{r}},\quad \mathbf{r}\in S_i.\end{aligned} \tag{47}$$

For the rigid spheres, the boundary condition (46) reduces to the no-slip requirement, $\mathbf{v}(\mathbf{r})=\mathbf{w}_i(\mathbf{r})$ for $\mathbf{r}\in S_i$, and $\mathbf{Z}_i^{-1}(\mathbf{r}-\mathbf{R}_i,\overline{\mathbf{r}}-\mathbf{R}_i)=\mathbf{T}_0(\mathbf{r}-\overline{\mathbf{r}})$, if $\mathbf{r},\overline{\mathbf{r}}\in S_i$. In this case, the boundary integral equation (47) has the simple form given in Eq. (41).

In the following section, the method of solving Eq. (47) will be outlined. The set of the boundary integral equations will be transformed into an infinite set of algebraic equations for the force multipoles.

3. Multipole expansion

In this section, the basic idea of the spherical-multipole expansion [9, 34, 35, 38] will be outlined. We apply this expansion to N spherical particles in a fluid under a general ambient flow $\mathbf{v}_\infty(\mathbf{r})$. The procedure may be applied to different geometries of the boundaries, if the corresponding Green function is known. The forces, torques and velocities are projected on a basic set of multipole functions, and represented by the coefficients of this expansion (the so-called force and velocity multipoles), see Appendix A for the details. Here we use the real multipole vector functions $\boldsymbol{u}_{lm\sigma}^{+}(\mathbf{r}-\mathbf{R}_i)$, with $\boldsymbol{R}_i$ denoting the center of sphere i. The subscripts are the multipole indices $l = 1, 2, ...,$ $m = 0, \pm 1, .., \pm l$ and $\sigma = 0, 1, 2$. The definitions of all $\boldsymbol{u}^{+}_{lm\sigma}$ are given in Ref. [21] and also in Appendix A. The projection casts the integral equation (47) into an infinite set of algebraic equations,

$$c(ilm\sigma) = \sum_{j=1}^{N} \sum_{l'=1}^{\infty} \sum_{m'=-l'}^{l'} \sum_{\sigma'=0}^{2} M(ilm\sigma, jl'm'\sigma')\ f(jl'm'\sigma'), \tag{48}$$

which relate the force multipoles,

$$f(jlm\sigma) = \int \boldsymbol{u}^{+}_{lm\sigma}(\mathbf{r}-\mathbf{R}_i) \cdot \mathbf{f}_j(\mathbf{r})\, d^3\mathbf{r}, \tag{49}$$

to the velocity multipoles $c(ilm\sigma)$, which are defined in terms of two contributions,

$$c(ilm\sigma) = c_w(ilm\sigma) - c_\infty(ilm\sigma), \tag{50}$$

where c_w and c_∞ are, respectively, the expansion coefficients of the particle velocity $\mathbf{w}_i(\mathbf{r}) = \mathbf{U}_i + \mathbf{\Omega}_i \times (\mathbf{r} - \boldsymbol{R}_i)$ at a point $\mathbf{r}$ on the surface of sphere i, and of the ambient flow velocity $\mathbf{v}_\infty(\mathbf{r})$,

$$\mathbf{U}_i + \mathbf{\Omega}_i \times (\mathbf{r} - \boldsymbol{R}_i) = \sum_{m=-1}^{1} \sum_{\sigma=0}^{1} c_w(i1m\sigma)\ \boldsymbol{u}^{+}_{1m\sigma}(\mathbf{r} - \boldsymbol{R}_i), \tag{51}$$

$$\mathbf{v}_\infty(\mathbf{r}) = \sum_{l=1}^{\infty} \sum_{m=-1}^{l} \sum_{\sigma=0}^{2} c_\infty(ilm\sigma)\ \boldsymbol{u}^{+}_{lm\sigma}(\mathbf{r} - \boldsymbol{R}_i). \tag{52}$$

Note that the only non-vanishing $c_w(ilm\sigma)$ are those with $l = 1$, $\sigma = 0, 1$. Each of them is proportional to a component of the translational or the angular velocity of the sphere. Similarly, the force multipoles with $(l, \sigma) = (1, 0)$ and $(l, \sigma) = (1, 1)$ are proportional to components of the force and the torque exerted by the sphere on the fluid, with the coefficients given in Appendix B.

The multipole matrix elements $M(ilm\sigma, jl'm'\sigma')$ in Eq. (48) are determined by the corresponding elements of $\mathbf{Z}_i, \mathbf{T}_0$ and $\tilde{\mathbf{T}}$ (see Appendix A for the details),

$$M(ilm\sigma, jl'm'\sigma') = \delta_{ij} Z_i^{-1}(ilm\sigma, il'm'\sigma') + (1-\delta_{ij})[T_0(ilm\sigma, jl'm'\sigma') + \tilde{T}(ilm\sigma, jl'm'\sigma')]. \tag{53}$$

After truncating of the expansion at order L, i.e. neglecting the terms with l, $l' > L$, equation (48) reduces to a finite set of linear algebraic equations. These equations are solved for the force multipoles by inverting the large matrix **M** formed by the coefficients $M(ilm\sigma, jl'm'\sigma')$ with l, $l' \leq L$,

$$f(ilm\sigma) = \sum_{j=1}^{N} \sum_{l'=1}^{L} \sum_{m'=-l'}^{l'} \sum_{\sigma'=0}^{2} Z_L(ilm\sigma, jl'm'\sigma')\, c(jl'm'\sigma'), \tag{54}$$

where $\mathbf{Z}_L = \mathbf{M}^{-1}$ is called the *spherical generalized friction* (*or grand resistance*) matrix. The coefficients $Z_L(ilm\sigma, jlm'\sigma')$ depend on the multipole order L of the truncation. In Appendix A, their properties are discussed and the references are given to their explicit form for specific types of the particles. In Appendix B, the Cartesian generalized friction tensors $\boldsymbol{\zeta}^{pq}$, introduced in Sec. 2.3, are expressed in terms of the coefficients $Z_L(ilm\sigma, jlm'\sigma')$ of the spherical generalized friction matrix. In this way the generalized friction problem is solved.

In particular, in the absence of an ambient flow, for given translational and angular velocities of the sphere, the force and the torque are determined by Eq. (54) for the L-dependent force multipoles with $l = 1$, $m = 0, \pm 1$ and $\sigma = 0, 1$ only,

$$f_{w,L}(i1m\sigma) = \sum_{m'=-1}^{1} \sum_{\sigma'=0}^{1} Z_L(i1m\sigma, j1m'\sigma')\, c_w(j1m'\sigma'). \tag{55}$$

The multipole elements $Z_L(i1m\sigma, j1m'\sigma')$, which enter Eq. (55), form the *spherical friction matrix*, which solves the friction problem.

On the other hand, for a given ambient flow $\mathbf{v}_\infty$, it is of interest to evaluate the force and the torque exerted by a system of motionless spheres on the fluid. In the multipole expansion, it means that we search the force multipoles with $l = 1$, $m = 0, \pm 1$ and $\sigma = 0, 1$. We denote them as $f_{\infty,L}(i1m\sigma)$, with the subscript ∞ specifying the problem (the sphere is fixed and the force multipoles are determined by the coefficients $c_\infty(il'm'\sigma')$ only). Note that all the force multipoles depend on L, as indicated by the second subscript L. They are evaluated from Eq. (54), which now takes the form,

$$f_{\infty,L}(i1m\sigma) \;=\; -\sum_{j=1}^{N}\sum_{l'=1}^{L}\sum_{m'=-l'}^{l'}\sum_{\sigma'=0}^{2} Z_L(i1m\sigma, jl'm'\sigma')\, c_\infty(jl'm'\sigma'). \qquad (56)$$

In Appendix C, it is described how to project an ambient flow onto the multipole functions, and evaluate the velocity multipoles $c_\infty(ilm\sigma)$. In many practical applications, the ambient flow is a combination of a small number of the multipole functions only, e.g. for the shear or Poiseuille flows. In such cases, it is possible to truncate the expansion at such a multipole order L, that all the coefficients $c_\infty(jl'm'\sigma') \neq 0$ are included in Eq. (56).

Assume now that the spherical particles are moving in an ambient flow, under given external forces and torques, which determine $f(ilm\sigma)$ for $i = 1, ..., N$, $l = 1$, $m = 0, \pm 1$ and $\sigma = 0, 1$. The goal is to evaluate the particle translational and rotational velocities. As explained in Appendix B, these velocities are expressed by the velocity multipoles $c_{w,L}$ with $l = 1$, $m = 0, \pm 1$ and $\sigma = 0, 1$, where the index L reminds the dependence on the truncation order. By virtue of Eqs. (50) and (54), one obtains

$$c_{w,L}(i1m\sigma) \;=\; \sum_{j=1}^{N}\sum_{m'=-1}^{1}\sum_{\sigma'=0}^{1} \mu_L(i1m\sigma, j1m'\sigma')\,[f(j1m'\sigma') - f_{\infty,L}(j1m'\sigma')], \quad (57)$$

with $f_{\infty,L}$ already calculated in (56). Here $\mu_L(ilm\sigma, jl'm'\sigma')$ denote coefficients of the *spherical mobility matrix*, which is the inverse of the corresponding spherical friction matrix. The spherical-multipole mobility matrix can be easily transformed into the corresponding Cartesian mobility tensor. The explicit transformation from the spherical to the Cartesian representation is given in Appendix B.

The algorithm described above has been implemented in a numerical FORTRAN code called HYDROMULTIPOLE, and calculations have been carried out with both double and quadruple precision. The accuracy is controlled by changing the multipole order L of the truncation, and even extrapolating to $L \to \infty$. For systems of solid particles with no relative motion of close surfaces, the method presented above is sufficient for precise calculations already for very low multipole order L [39]. However, if close solid surfaces move with respect to each other, the appropriate treatment of the lubrication effects is needed for accurate and efficient computations. Such a modification of the algorithm will be discussed in the next section.

4. Multipole expansion corrected for lubrication

4.1. Lubrication between two close solid surfaces

If a solid sphere (labeled i) moves with respect to another solid sphere (labeled j), almost touching it, the fluid strongly resists the movement; the friction force and torque diverge when the size of the gap between the surfaces tends to zero, and the motion of the particles is fixed [6, 40].

For very small distances between the sphere surfaces,

$$\xi = R_{ij}/a - 2 \ll 1, \tag{58}$$

with $R_{ij} = |\boldsymbol{R}_i - \boldsymbol{R}_j|$ and $a = (a_i + a_j)/2$, the two-particle friction tensors $\boldsymbol{\zeta}(ij)$ have the following asymptotic form,

$$\boldsymbol{\zeta}^{lub}(ij) = \frac{\mathbf{A}(ij)}{\xi} + \mathbf{B}(ij)\ln\xi + \mathbf{C}(ij) + \mathbf{D}(ij)\,\xi\ln\xi + \mathcal{O}(\xi), \tag{59}$$

The constant matrices **A**, **B**, **C**, **D** are specified explicitly e.g. in Refs. [6, 40]. The only non-zero elements of the matrix **A** correspond to the forces caused by the relative motion of the spheres along their line of centers.

The expression Eq. (59) for the lubrication singularities is general. It applies to the generalized friction tensor for two particles of an arbitrary shape [41].

Hydrodynamic interactions described by Eq. (59) are called *lubrication* interactions [6]. They require a special attention in numerical calculations [42]. In particular, for very close spheres with $\xi \ll 1$ in relative motion, they cause a very slow convergence of the multipole expansion with the increasing multipole order L. Therefore the asymptotic expressions (59) have been used to construct the so-called *lubrication correction* [41, 43, 44] for many-particle hydrodynamic interactions. This procedure will be outlined in the next sections.

4.2. Accurate friction tensor for two spherical particles

For two spheres, labeled i and j, the friction tensor $\boldsymbol{\zeta}(ij)$ was first evaluated in Ref. [40], and then recalculated with an improved precision and generalized for various ambient flows with the use of several different techniques, including bispherical coordinates (extensively discussed in this book) and the multipole expansion [28]. Within the multipole method, any friction coefficient can be represented as a sum of multiple scattering sequences, each proportional to a given power k of the inverse interparticle distance $x = 2a/R_{ij}$. Next, the multiple scattering sequences with the same k may be collected, to obtain the friction coefficient as a power series, so that $\zeta(ij) = \sum_k \mathbf{C}_k(ij)\, x^k$. For very close sphere surfaces, however, it is essential to speed up the convergence rate of the series, in a similar way as it was proposed by Jeffrey and Onishi [40]. This is achieved by subtracting from the friction tensor the corresponding asymptotic expressions (59), non-analytic and divergent when $\xi \to 0$,

$$\mathcal{G}(ij) = \zeta(ij) - \zeta^{lub}(ij), \tag{60}$$

Then, *the difference* $\mathcal{G}$ is represented as a power series of x,

$$\mathcal{G}_n(ij) = \sum_{k=0}^{n} \mathbf{D}_k(ij)\, x^k. \tag{61}$$

The matrix $\mathbf{D}_k$ differs from $\mathbf{C}_k$ by the series expansion of ζ^{lub}. As the result, the series (61) is fast convergent and its truncation leads to a high accuracy of the friction coefficients [40]. Typically, in the spherical-multipole numerical codes, $n = 300$ has been used [10], with the pre-calculated tables of all the coefficients of $\mathbf{D}_k$. This procedure is sufficient to reach the $2 \cdot 10^{-5}$ absolute precision of the two-particle friction coefficients even at the contact.

4.3. Lubrication correction for many-particle hydrodynamic interactions

4.3.1. Standard approach

Consider first such a system of N spherical particles, where a particle 1 moves with respect to another very close particle labeled 2, and the other spheres are well-separated from each other and from spheres 1 and 2. The total hydrodynamic force exerted by the fluid flow on sphere 1 is practically caused by the motion of the fluid in the lubrication gap between the particles 1 and 2. Indeed, in this case the lubrication expression $\zeta^{lub}(12)$, given by Eq. (59), is large and dominates all the other contributions to the N-particle friction tensor $\zeta(12...N)$. This lubrication contribution is independent of the other particles. If the sphere 1 is surrounded by several spheres, which almost touch it, one may expect that the total friction forces and torques exerted on sphere 1 by the fluid flow are approximated by the superposition of the two-particle contributions (59) corresponding to lubrication gaps between the surfaces of spheres 1 and j.

This property has been used to construct a modified multipole expansion, fast-convergent even for ξ ≪ 1, and valid for a general configuration of N spheres [43, 44]. In this procedure, a superposition of the two-particle friction tensors is formed in the following way,

$$\zeta_{ii}^{sup}(1...N) = \sum_{j\neq i}^{N} \zeta_{ii}(ij), \tag{62}$$

$$\zeta_{ii}^{sup}(1...N) = \zeta_{ij}(ij) \qquad \text{for } i \neq j, \tag{63}$$

where the lower indices ij label the 6×6 tensor components of the $6N\times 6N$ tensors. The two-particle friction tensor $\boldsymbol{\zeta}(ij)$ is evaluated by the procedure described in Sec. 4.2, with a high precision even for extremely small distances between the sphere surfaces. In the following, the arguments $(1...N)$ will be omitted wherever this does not interfere with clarity of the presentation.

The idea introduced in Refs [43, 44] is to "correct" the slow convergence rate of $\boldsymbol{\zeta}_L$, replacing it by another fast-convergent expression,

$$\bar{\zeta}_L = \zeta_L + \Delta_L, \tag{64}$$

where the lubrication correction,

$$\Delta_L = \zeta^{sup} - \zeta_L^{sup} \tag{65}$$

is defined as the difference between the accurate pairwise-additive expressions (62)-(63) and their multipole approximations of the order L,

$$\zeta_{L,ii}^{sup}(1...N) = \sum_{j\neq i}^{N} \zeta_{L,ii}(ij), \tag{66}$$

$$\zeta_{L,ij}^{sup}(1...N) = \zeta_{L,ij}(ij), \qquad \text{for } i \neq j, \tag{67}$$

with $\zeta_L(ij)$ denoting the multipole approximation with the order L of the two-particle friction tensor $\boldsymbol{\zeta}(ij)$.

Both $\bar{\zeta}_L$ and ζ_L approach the same limit $\bar{\zeta}_\infty = \zeta_\infty$ when $L \to \infty$, and keep the same long-distance asymptotics, because for well-separated particles the lubrication correction is negligible. The key point of Eqs. (64)-(65) is that $\bar{\zeta}_L = \zeta^{sup} + (\zeta_L - \zeta_L^{sup})$ is fast-convergent even if some of the particles are close and move with respect to each other. Indeed, the first term, ζ^{sup}, is independent of the multipole order, and the second one, $\zeta_L - \zeta_L^{sup}$, does not contain any lubrication singularities and therefore is fast-convergent with the increasing L. It is essential that $\bar{\zeta}_L$ is an accurate approximation of ζ_∞ already for a low multipole order.

Lubrication correction of the other generalized friction coefficients is constructed by the same reasoning. Then, the corrected generalized mobility tensor follows from Eq. (26). In particular, correcting the L-order mobility tensor $\mu_L = \zeta_L^{-1}$ results in

$$\bar{\mu}_L = \bar{\zeta}_L^{-1} = \mu_L \cdot [1 + \Delta_L \cdot \mu_L]^{-1} = \mu_L - \mu_L \cdot \Delta_L \cdot \mu_L + \ldots . \tag{68}$$

The standard lubrication correction allows for accurate evaluation of the particle dynamics. However, a refined treatment is needed if the cluster expansion is performed and three-particle hydrodynamic interactions are calculated separately [10, 47, 48]. In particular, the three-particle contribution to the translational self-diffusion coefficient is infinite if evaluated with the standard lubrication correction [10]. The spurious divergence is caused by the incorrect asymptotics of the three-particle mobility $\boldsymbol{\mu}^{tt}$ for such a configuration in which a single sphere (e.g. labeled 3) is far away from the other two (with labels 1 and 2), with $R_{13} \gg R_{12}$ and $R_{23} \gg R_{12}$. In this case, the dominant three-body contribution to $\mu_{33}^{tt}(123)$ scales as $1/R_{12}^4$. However, the standard lubrication correction adds to $\mu_{33}^{tt}(123)$ a small artificial term, which scales as $1/R_{12}^2$ and therefore is non-integrable. This paradox will be solved in the next section.

4.3.2. Improved lubrication correction

In general, the standard lubrication correction changes the total hydrodynamic force and torque exerted on the fluid by a pair of spheres in relative motion if a third particle is present. The lubrication correction adds a very small spurious singlet contribution, a source of a $1/r$ flow, which dominates at large distances r if the real flow is proportional to $1/r^2$. Therefore in Ref. [10] an improved lubrication correction was constructed, which does neither modify the total force nor the torque exerted on the fluid by a pair of spheres in relative motion. The procedure described in the previous section was repeated, but with the friction tensor $\boldsymbol{\zeta}$ replaced by a tensor **s**, which contains the same singular terms and satisfies the same symmetries (translational, rotational and Lorentz invariance). Moreover, **s** applied to an arbitrary rigid motion of the pair of spheres has to give vanishing forces and torques.

These conditions are satisfied if

$$\mathbf{s}(ij) \equiv \mathbf{q}^T \cdot \zeta(ij) \cdot \mathbf{q}, \tag{69}$$

and the 6×6 matrix $\mathbf{q}$ projects onto relative motion of the spheres, with $\mathbf{q} = \mathbf{q}^2$. In practice, the operator $\mathbf{q}$ is constructed from the requirement that $\mathbf{c} = \mathbf{1} - \mathbf{q}$, applied to $(\mathbf{U}_1, \mathbf{U}_2, \boldsymbol{\Omega}_1, \boldsymbol{\Omega}_2)$, results in a rigid motion of both spheres, and $\mathbf{c} = \mathbf{c}^2$. The rigid motion of the spheres is not uniquely defined, although of course it should be "close" to the motion of both spheres. For example, a choice of the rigid motion is the translation of the center of mass system, superposed with the rotation around the center of mass with the angular velocity $(\boldsymbol{\Omega}_1 + \boldsymbol{\Omega}_2)/2$. In particular, for identical spheres, the center of mass is located at $(\mathbf{R}_1 + \mathbf{R}_2)/2$ and translates with velocity $(\mathbf{U}_1 + \mathbf{U}_2)/2$. The projection $\mathbf{c}$ on this rigid motion corresponds to the following operator $\mathbf{I} - \mathbf{c} = \mathbf{q}$, which projects on relative motion [10],

$$\mathbf{q} = \frac{1}{2}\begin{pmatrix} \boldsymbol{I} & -\boldsymbol{I} & -\boldsymbol{A} & -\boldsymbol{A} \\ -\boldsymbol{I} & \boldsymbol{I} & \boldsymbol{A} & \boldsymbol{A} \\ \mathbf{0} & \mathbf{0} & \boldsymbol{I} & -\boldsymbol{I} \\ \mathbf{0} & \mathbf{0} & -\boldsymbol{I} & \boldsymbol{I} \end{pmatrix}, \tag{70}$$

where $\boldsymbol{I}$ is the 3×3 unit matrix, and $\boldsymbol{A}$ depends on relative position of the sphere centers, $\boldsymbol{R} = \boldsymbol{R}_2 - \boldsymbol{R}_1$,

$$A_{\alpha\beta} = -\varepsilon_{\alpha\beta\gamma} R_\gamma / 2, \tag{71}$$

with the Cartesian components labeled by α, β, γ, and the summation over γ.

In general, the flow inside the small gap between the sphere surfaces moves with respect to the center-of-mass system, and if the slip is large, a rigid motion can be taken which is closer to the fluid motion in the gap. For example, the operator **c** can project on a rigid motion which is the average of the individual rigid motions $\mathbf{w}_1(\mathbf{r})$ and $\mathbf{w}_2(\mathbf{r})$ of spheres 1 and 2, respectively, see Eq. (4). This choice results in the operator **q** independent of the sphere radii,

$$\mathbf{q} = \frac{1}{2}\begin{pmatrix} \boldsymbol{I} & -\boldsymbol{I} & 0 & -2\boldsymbol{A} \\ -\boldsymbol{I} & \boldsymbol{I} & 2\boldsymbol{A} & 0 \\ \mathbf{0} & \mathbf{0} & \boldsymbol{I} & -\boldsymbol{I} \\ \mathbf{0} & \mathbf{0} & -\boldsymbol{I} & \boldsymbol{I} \end{pmatrix}. \tag{72}$$

This operator **q** projects on such a relative motion that corresponds to the opposite velocity field inside both spheres, $(\mathbf{w}_1(\mathbf{r}) - \mathbf{w}_2(\mathbf{r}))/2$ and $-(\mathbf{w}_1(\mathbf{r}) - \mathbf{w}_2(\mathbf{r}))/2$, respectively. Moreover, inside the gap and in the limit of a small gap size, the relative velocities of the sphere surfaces are almost opposite, and the rigid-motion velocity is almost equal to the averaged velocity of the closest points of both surfaces.

When the projection **q** on the relative motions is specified, the expression **s** in Eq. (69) is known. The improved lubrication correction is now constructed by the procedure described in the previous section, but with $\boldsymbol{\zeta}$ replaced by **s**. We obtain the modified pairwise-additive expressions,

$$\mathbf{s}_{ii}^{sup}(1...N) = \sum_{j\neq i}^{N} \mathbf{s}_{ii}(ij), \tag{73}$$

$$\mathbf{s}_{ij}^{sup}(1...N) = \mathbf{s}_{ij}(ij) \qquad \text{for } i \neq j, \tag{74}$$

and their approximations of the order L,

$$\mathbf{s}_{L,ii}^{sup}(1...N) = \sum_{j\neq i}^{N} \mathbf{s}_{L,ii}(ij), \tag{75}$$

$$\mathbf{s}_{L,ij}^{sup}(1...N) = \mathbf{s}_{L,ij}(ij) \qquad \text{for } i \neq j, \tag{76}$$

with

$$\mathbf{s}_L(ij) \equiv \mathbf{q}^T \cdot \boldsymbol{\zeta}_L(ij) \cdot \mathbf{q}. \tag{77}$$

Now, the corrected N-particle friction tensor has the form,

$$\bar{\zeta}_L = \zeta_L + \delta_L \tag{78}$$

with the improved lubrication correction,

$$\delta_L = \mathbf{s}^{sup} - \mathbf{s}_L^{sup}. \tag{79}$$

Let us comment now that if the improved lubrication correction is used, then the self-diffusion coefficient is finite. Indeed, with Δ in Eq. (68) replaced by $\boldsymbol{\delta}$, the dominant three-body contribution to μ_{33}^{tt} (123) scales as $1/R_{12}^4$ with no spurious extra terms. [10]

Finally, we briefly discuss accuracy of the multipole expansion. With this procedure, the convergence of the multipole expansion is fast, and truncation at a relatively small L results in a high accuracy [9, 17, 45, 46]. In Ref. [9], friction and mobility coefficients were evaluated for a number of particle configurations and the accuracy of the results was estimated. For groups of rigidly moving particles, truncation at $L = 4$ typically leads to extremely high 0.1% relative precision of the drag coefficients, because the collective motion does not involve lubrication interactions. The relative motion of particles results in a lower precision, which has not been extensively discussed in Ref. [9]. Below we study an example of a simple particle configuration, and we estimate the accuracy of the friction and mobility tensors, evaluated by the spherical-multipole method with the standard and the improved lubrication corrections.

We consider two test configurations of three identical close spheres, with their centers located at vertexes of an isosceles right triangle. In the first case, the smallest gap size is equal to 0.01 diameter, and in the second case to 0.0001 diameter. The friction and mobility tensors have been evaluated with the standard and both improved lubrication corrections, for the multipole order $L = 4$ and for $L = 25$. Relative precision of the results with $L = 4$ has been estimated by evaluating the differences of the tensor elements corresponding to $L = 4$ and $L = 25$, and calculating the square root of the sum of the squared differences, normalized by the square root of the sum of all the squared elements. The resulting accuracy is equal to $7 \cdot 10^{-5} - 3 \cdot 10^{-3}$. The relative difference between the results for $L = 4$, evaluated with different lubrication corrections, is smaller than the above precision. The accuracy rapidly improves if the distance between the sphere surfaces is increased.

5. Conglomerates of particles

5.1. Friction and mobility

In this section we consider hydrodynamic interactions between rigid arrays of particles. We solve the friction and mobility problems, formulated in Sec. 2.3, but now for the conglomerates rather than for the individual particles.

We consider K conglomerates of particles. Each conglomerate, labeled with

$$k = 1, \ldots, K, \tag{80}$$

consists of $N^{(k)}$ spherical particles (in general with different radii) labeled with i_k,

$$i_k = \sum_{s=1}^{k-1} N^{(s)} + 1, \; \ldots, \; \sum_{s=1}^{k} N^{(s)}. \tag{81}$$

The total number of particles is equal to N,

$$\sum_{k=1}^{K} N^{(k)} = N. \tag{82}$$

The position in space of a conglomerate k is defined by the position of an arbitrary reference point $\mathbf{R}_0^{(k)}$ of this conglomerate (often it is the geometrical center of this conglomerate) and the Euler angles. In general, there are three such angles; for the conglomerates with axial symmetry (e.g. linear polymers) two angles are sufficient. The reference point $\mathbf{R}_0^{(k)}$ and the corresponding Euler angles determine the positions $\mathbf{R}_{i_k}$ of all the sphere centers in the conglomerate k.

Conglomerates move collectively like rigid bodies. Therefore the motion of a conglomerate k is characterized by the translational collective velocity $\mathbf{U}^{(k)}$ of the reference point $\mathbf{R}_0^{(k)}$ and the rotational collective velocity $\mathbf{\Omega}^{(k)}$ of this conglomerate. The translational and rotational velocities $\mathbf{U}_{i_k}$ and $\mathbf{\Omega}_{i_k}$ of all the $N^{(k)}$ particles of this conglomerate follow as linear functions of $\mathbf{U}^{(k)}$ and $\mathbf{\Omega}^{(k)}$,

$$\mathbf{U}_{i_k} = \mathbf{U}^{(k)} + \mathbf{\Omega}^{(k)} \times (\mathbf{R}_{i_k} - \mathbf{R}_0^{(k)}), \tag{83}$$

$$\boldsymbol{\Omega}_{i_k} \;=\; \boldsymbol{\Omega}^{(k)}, \tag{84}$$

with the range of k and i_k given by Eqs. (80) and (81). Relations (83) and (84) can be written in short,

$$\begin{pmatrix} \mathbf{U} \\ \boldsymbol{\Omega} \end{pmatrix} = \mathbf{C}\cdot\begin{pmatrix} \mathbf{U}^{\mathrm{C}} \\ \boldsymbol{\Omega}^{\mathrm{C}} \end{pmatrix}, \tag{85}$$

with the abbreviated notation for the particle velocities $\mathbf{U}$ and $\boldsymbol{\Omega}$ defined in Section 2.3. In analogy, we have arranged the conglomerate velocities into 3K-dimensional vectors $\mathbf{U}^{\mathrm{C}}=(\mathbf{U}^{(1)},\ldots,\mathbf{U}^{(K)})$ and $\boldsymbol{\Omega}^{\mathrm{C}}=(\boldsymbol{\Omega}^{(1)},\ldots,\boldsymbol{\Omega}^{(K)})$. The $6N\times 6K$ rectangular matrix $\mathbf{C}$ can be read out explicitly from Eqs. (83-84). The matrix $\mathbf{C}$ is a function of the positions $\mathbf{R}_{i_k}$ of all the N sphere centers.

The total force and $\mathcal{F}^{(k)}$ and total torque $\mathcal{T}^{(k)}$ exerted by the conglomerate k on the fluid are given as superpositions of the individual forces and torques, respectively,

$$\mathcal{F}^{(k)} \;=\; \sum_{i_k}\mathcal{F}_{i_k}, \tag{86}$$

$$\mathcal{T}^{(k)} \;=\; \sum_{i_k}[\mathcal{T}_{i_k}+(\mathbf{R}_{i_k}-\mathbf{R}_0^{(k)})\times\mathcal{F}_{i_k}], \tag{87}$$

with the range of k given by Eq. (80) and the range of the summation given by Eq. (81). Following Sec. 2.3, we use the abbreviated notation $\mathcal{F}$ and $\mathcal{T}$ for the individual forces and torques. We also represent the total forces and torques exerted by the conglomerates on the fluid as $3K$-dimensional vectors $\mathcal{F}^{\mathrm{C}}=(\mathcal{F}^{(1)},\ldots,\mathcal{F}^{(K)})$ and $\mathcal{T}^{\mathrm{C}}=(\mathcal{T}^{(1)},\ldots,\mathcal{T}^{(K)})$. Now the relations (86) and (87) can be rewritten in short,

$$\begin{pmatrix} \mathcal{F}^{\mathrm{C}} \\ \mathcal{T}^{\mathrm{C}} \end{pmatrix} = \mathbf{C}^{\mathrm{T}}\cdot\begin{pmatrix} \mathcal{F} \\ \mathcal{T} \end{pmatrix}. \tag{88}$$

From Eqs. (86-87) it follows that the $6K\times 6N$ rectangular matrix of the linear transformation is just the transposed matrix $\mathbf{C}$. In general, the external forces $\mathcal{F}^{\mathrm{C}}$ and torques $\mathcal{T}^{\mathrm{C}}$ imposed on the conglomerates are controlled. This is not the case for individual particles, which undergo reaction forces in addition to the imposed ones. Therefore for conglomerates the friction relation reads,

$$\begin{pmatrix} \mathcal{F}^{\mathrm{C}} \\ \mathcal{T}^{\mathrm{C}} \end{pmatrix} = \boldsymbol{\zeta}^{\mathbf{C}} \cdot \begin{pmatrix} \mathbf{U}^{\mathrm{C}} \\ \boldsymbol{\Omega}^{\mathrm{C}} \end{pmatrix}, \tag{89}$$

where the $6K \times 6K$ many-conglomerate friction tensor,

$$\boldsymbol{\zeta}^{\mathbf{C}} = \mathbf{C}^{\mathrm{T}} \cdot \boldsymbol{\zeta} \cdot \mathbf{C}, \tag{90}$$

has been obtained from the many-particle friction tensor, defined in Eq. (25), with the use of Eqs. (85) and (88).

As explained in Sec. 4.3, the multipole algorithm used for evaluating the N-particle friction tensor $\boldsymbol{\zeta}$ is in general corrected for lubrication. While evaluating $\boldsymbol{\zeta}^{\mathrm{C}}$ form Eq. (90), the lubrication correction is included in $\boldsymbol{\zeta}$ only for the pairs of spheres belonging to different conglomerates. The pairs of spheres belonging to the same conglomerate move collectively and therefore the multipole expansion without a lubrication correction is fast convergent, as explained in Sec. 4.3. With the lubrication correction switched off, spheres in a single conglomerate may touch each other.

Finally, we define the conglomerate mobility tensor by the relation,

$$\boldsymbol{\mu}^{\mathrm{C}} = (\boldsymbol{\zeta}^{\mathrm{C}})^{-1}. \tag{91}$$

The mobility tensor allows to evaluate the collective velocities and then integrate the trajectories of all the K conglomerates, which are subject to external forces and torques, e.g. sedimenting under gravity.

5.2. Motion in ambient flow

In this section we evaluate the translational and rotational velocities of conglomerates which are subject to external forces and torques and to an external flow.

This task is similar to the motion of particles under external forces, torques and an ambient flow, solved in Sec. 2.3. We first apply $\mathbf{C}^{\mathrm{T}}$ to the l.h.s. of Eq. (29), then use Eqs. (85), (88) and (90),

$$\begin{pmatrix} \mathcal{F}^{\mathrm{C}} \\ \mathcal{T}^{\mathrm{C}} \end{pmatrix} = \boldsymbol{\zeta}^{\mathrm{C}} \cdot \begin{pmatrix} \mathbf{U}^{\mathrm{C}} \\ \boldsymbol{\Omega}^{\mathrm{C}} \end{pmatrix} + \mathbf{C}^{\mathrm{T}} \cdot \begin{pmatrix} \mathcal{F}_{\infty} \\ \mathcal{T}_{\infty} \end{pmatrix}, \tag{92}$$

and finally use Eq. (91) to evaluate the collective velocities of the conglomerates,

$$\begin{pmatrix} \mathbf{U}^{\mathrm{C}} \\ \boldsymbol{\Omega}^{\mathrm{C}} \end{pmatrix} = \boldsymbol{\mu}^{\mathrm{C}} \cdot \begin{pmatrix} \mathcal{F}^{\mathrm{C}} - \mathcal{F}^{\mathrm{C}}_{\infty} \\ \mathcal{T}^{\mathrm{C}} - \mathcal{T}^{\mathrm{C}}_{\infty} \end{pmatrix}, \tag{93}$$

where

$$\begin{pmatrix} \mathcal{F}^{\mathrm{C}}_{\infty} \\ \mathcal{T}^{\mathrm{C}}_{\infty} \end{pmatrix} = \mathbf{C}^{\mathrm{T}} \begin{pmatrix} \mathcal{F}_{\infty} \\ \mathcal{T}_{\infty} \end{pmatrix}. \tag{94}$$

Here $\mathcal{F}_{\infty}$ and $\mathcal{T}_{\infty}$ are the forces and torques exerted by the motionless particles on the flow, evaluated from Eq. (30), where, in the absence of relative motion of the particles, the friction tensors are determined without lubrication corrections. On the other hand, $\boldsymbol{\mu}^{\mathrm{C}}$ is calculated with the use of lubrication correction between particles belonging to different conglomerates.

6. Conclusions

An efficient procedure [9, 10, 21] with a controlled high accuracy, the spherical-multipole method, was presented, adequate for evaluating Stokesian dynamics of non-deformable spherical particles suspended in a fluid, or hydrodynamic resistance of moving or motionless systems of such particles under low-Reynolds-number flows. Below we present a few examples of specific applications of this method. Different versions of the multipole expansion were used in the literature by many authors in numerous physical contexts [14, 15, 17, 43, 45], and listing all the results would require a separate review. Therefore we concentrate mainly on the results obtained by the accurate spherical-multipole method. In this procedure, the relative motion of particles is corrected for lubrication, to achieve fast convergence with the multipole order of the truncation. The main advantage of this algorithm, even in comparison with the Cartesian-multipole formulation [49], is that it is possible to perform computations with a very high multipole order of the truncation, controlling the accuracy. Moreover, the method is applicable to systems of various types of the particles in a fluid bounded by one or two parallel flat interfaces.

Friction and mobility problems for groups of particles in an unbounded fluid were solved. Drag coefficients of conglomerates of particles have been calculated and shown to agree with the experimental data [50]. Dynamics of symmetric configurations of three spherical solid particles was analyzed and an attracting equilibrium configuration was found, non-existent in the point-particle approximation [51]. A model of mechanical-contact and hydrodynamic interactions between rough spherical particles was constructed and shown to account well for the experimentally measured relative translation and rotation [52, 53]. In Ref. [27], the Stokes equations were solved for a single surfactant-covered drop in an arbitrary incident flow, and then the pair hydrodynamic interactions of surfactant covered bubbles were computed from the one-particle solution using a multiple-scattering expansion.

Statistical properties of particulate systems were also determined. Virial expansion of suspension effective transport coefficients was performed. Two-particle and three-particle contributions to the short-time self-diffusion, sedimentation velocity and high-frequency viscosity were evaluated [10, 47, 48, 54-58]. The short time self-diffusion coefficient of a sphere in a suspension of rigid rods was calculated in the first order in the rod volume fraction [59]. Two-particle correlation function for non-Brownian suspension in a stationary state was determined and used to evaluate the virial expansion of the sedimentation coefficient. In the stationary state, the term proportional to the volume fraction was shown to be larger than in the equilibrium, owing to the excess of close particle pairs in comparison to the equilibrium [60].

Dynamics of particles close to interfaces was also analyzed. The effect of a planar hard wall on the motion of particle clusters under external forces, shear or Poiseuille flow was determined [21, 61, 62]. Hydrodynamic interactions between solid particles touching a free surface and moving along it (a quasi-two-dimensional system) were evaluated, and the range of validity of the long-distance pairwise asymptotics and the point-particle approximation was given [24, 63].

The spherical-multipole method was also used for theoretical and numerical studies of hydrodynamic interactions of spherical particles confined between two parallel planar solid walls. A new efficient algorithm for evaluating many-particle friction and mobility matrices for such a system was developed [20, 22]. Numerical implementation of this algorithm was used to evaluate the hydrodynamic friction and mobility for a single particle, a pair of particles, and a system of many particles confined between two planar walls. The results show that the standard single-wall-superposition approximation is insufficient for problems when the particles are laterally separated by many inter-wall distances [64]. In Ref. [65], the effect of confining walls on the dynamics of a dilute suspension of noninteracting, elongated axisymmetric particles undergoing a steady shear flow in a parallel-wall rheometer was presented. It was found that the particle motion in the two-wall system qualitatively resembles the Jeffrey's orbits in the unbounded space [66], but, unlike in the unbounded space, the period of the motion and the evolution depend on the initial position of the particle. In Ref. [64], the effect of the walls on the hydrodynamic interactions in ambient Poiseuille flow in a narrow channel were studied. In Ref. [67], the crossover behavior between near-field flow and far-field asymptotic Hele-Shaw flow [68], was analyzed. It was shown that for a few inter-wall distances from the particle, the flow assumes the asymptotic form. This facilitates significantly the numerical evaluation of the Green tensor for the two-wall system. In Ref. [69], the new class of binary trajectories that result in cross-streamline particle migration in a wall bounded shear flow was identified.

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Appendix A. Multipole functions and multipole matrix elements

In Section 3 the boundary integral equation (47) has been projected onto multipole functions, resulting in Eq. (48). In this Appendix, we explain in details how this procedure has been carried out and we provide the explicit expressions for the velocity multipoles $c(ilm\sigma)$, the force multipoles $f(ilm\sigma)$ and the matrix elements $M(ilm\sigma, jl'm'\sigma')$.

The complete set of elementary flows $\mathbf{u}^{+}_{lm\sigma}$ has been constructed [28] in terms of the regular solid harmonics, [37], [70]. The regular solid harmonics are the following solutions of the Laplace equation,

$$\Phi_{lm}(\mathbf{r}) = r^l Y_{lm}(\hat{\mathbf{r}}), \tag{95}$$

where the normalized complex spherical harmonics are given in terms of the associated Legendre polynomials, [71]

$$Y_{lm}(\hat{\mathbf{r}}) = \frac{1}{n_{lm}}(-1)^m P_{lm}(\cos\theta)e^{im\varphi}, \tag{96}$$

with $|m| \leq l$ and the normalization coefficients,

$$n_{lm} = \left[\frac{4\pi(l+m)!}{(2l+1)(l-m)!}\right]^{1/2}. \tag{97}$$

In the numerical calculations, the real spherical harmonics are used,

$$Y_{l0}^{(R)} = Y_{l0}, \tag{98}$$

$$Y_{lm}^{(R)} = \sqrt{2}\mathrm{Re}(Y_{lm}) = \frac{Y_{lm} + (-1)^m Y_{l,-m}}{\sqrt{2}}, \text{ for } m \geq 1, \tag{99}$$

$$Y_{l,-m}^{(R)} = \sqrt{2}\mathrm{Im}(Y_{lm}) = \frac{Y_{lm} - (-1)^m Y_{l,-m}}{i\sqrt{2}}, \text{ for } m \geq 1, \tag{100}$$

and the corresponding real solid harmonics,

$$\Phi_{lm}^{(R)}(\mathbf{r}) = r^l Y_{lm}^{(R)}(\hat{\mathbf{r}}). \tag{101}$$

The complete set of the elementary flows $\mathbf{u}_{lm\sigma}^{+}$ is given in terms of gradients of the real solid harmonics, and the pressure fields $p_{lm\sigma}^{+}$ in terms of the real solid harmonics [28],

$$\begin{aligned} \mathbf{u}_{lm0}^{+}(\mathbf{r}) &= \nabla\Phi_{lm}^{(R)}(\mathbf{r}), \\ p_{lm0}^{+} &= 0, \end{aligned} \tag{102}$$

$$\begin{aligned} \mathbf{u}_{lm1}^{+}(\mathbf{r}) &= \mathbf{u}_{lm0}^{+}(\mathbf{r}) \times \mathbf{r}, \\ p_{lm1}^{+} &= 0, \end{aligned} \tag{103}$$

$$\begin{aligned} \mathbf{u}_{lm2}^{+}(\mathbf{r}) &= \frac{(2l+1)}{l}\left[\frac{l+3}{2} r^2 \mathbf{u}_{lm0}^{+}(\mathbf{r}) - \mathbf{r}\cdot\mathbf{u}_{lm0}^{+}(\mathbf{r})\,\mathbf{r}\right], \\ p_{lm2}^{+} &= \eta\frac{(l+1)(2l+1)(2l+3)}{l}\Phi_{lm}^{(R)}(\mathbf{r}). \end{aligned} \tag{104}$$

The following scalar product of vector fields $\mathbf{A}(\mathbf{r})$ and $\mathbf{B}(\mathbf{r})$ is defined

$$\langle \mathbf{A} | \mathbf{B} \rangle = \int \mathbf{A}(\mathbf{r}) \cdot \mathbf{B}(\mathbf{r})\, d\mathbf{r}, \tag{105}$$

with $\mathbf{A}$ or $\mathbf{B}$ containing the factor $\delta(|\mathbf{r}| - a_i)$, because the integral is restricted to the boundary surface of a particle with radius a_i. The elementary flows $\mathbf{u}_{lm\sigma}^{+}$ are not orthogonal to each other with the scalar product (105), therefore the adjoint basic set of functions $\omega_{lm\sigma}^{+}$ is introduced according to the relation

$$\left\langle \delta_b \boldsymbol{\omega}^{+}_{lm\sigma} \middle| \mathbf{u}^{+}_{l'm'\sigma'} \right\rangle = \delta_{ll'}\delta_{mm'}\delta_{\sigma\sigma'}, \tag{106}$$

for all values of the parameter $b > 0$, where

$$\delta_b(\mathbf{r}) = b^{-1}\delta(|\mathbf{r}| - b). \tag{107}$$

The adjoint functions $\boldsymbol{\omega}^{+}_{lm\sigma}$ are [28],

$$\boldsymbol{\omega}^{+}_{lm0}(\mathbf{r}) = \frac{1}{2l(2l+1)} r^{-2l+1} \left[-(2l+1)\mathbf{1} + \frac{(2l+1)(2l+3)}{l}\hat{\mathbf{r}}\hat{\mathbf{r}} \right] \cdot \mathbf{u}^{+}_{lm0}(\mathbf{r}), \tag{108}$$

$$\boldsymbol{\omega}^{+}_{lm1}(\mathbf{r}) = \frac{1}{l(l+1)} r^{-2l} \mathbf{u}^{+}_{lm0}(\mathbf{r}) \times \hat{\mathbf{r}}, \tag{109}$$

$$\boldsymbol{\omega}^{+}_{lm2}(\mathbf{r}) = \frac{1}{(l+1)(2l+1)} r^{-2l-1} \left[1 - \frac{2l+1}{l}\hat{\mathbf{r}}\hat{\mathbf{r}} \right] \cdot \mathbf{u}^{+}_{lm0}(\mathbf{r}). \tag{110}$$

The force and velocity multipoles, introduced in section 3, are projections onto the corresponding multipole functions $\mathbf{u}^{+}_{lm\sigma}$ and $\boldsymbol{\omega}^{+}_{lm\sigma}$, respectively. The force multipoles were given in Eq. (49),

$$f(ilm\sigma) = \left\langle \mathbf{u}^{+}_{lm\sigma}(i) \middle| \mathbf{f}_i \right\rangle. \tag{111}$$

The velocity multipoles, defined in Eqs. (51), (52), are now expressed in terms of $\boldsymbol{\omega}^{+}_{lm\sigma}$,

$$c_w(ilm\sigma) \;=\; \left\langle \delta_{ai}(i)\boldsymbol{\omega}^{+}_{lm\sigma}(i) \middle| \mathbf{w}_i \right\rangle, \tag{112}$$

$$c_\infty(ilm\sigma) \;=\; \left\langle \delta_{ai}(i)\boldsymbol{\omega}^{+}_{lm\sigma}(i) \middle| \mathbf{v}_\infty \right\rangle. \tag{113}$$

In the above equations, the standard bra-ket notation is used. Moreover, $|\mathbf{A}\rangle$ denotes the vector field $\mathbf{A}(\mathbf{r})$ and $|\mathbf{A}(i)\rangle$ represents the vector field $\mathbf{A}(\mathbf{r} - \mathbf{R}_i)$.

The multipole expansion of the particle and ambient flow velocity fields in terms of $\mathbf{u}^{+}_{lm\sigma}$ was given in Eqs. (51) and (52). In analogy, the multipole expansion of the induced force distribution $\mathbf{f}_i$ is given by

$$\mathbf{f}_i(\mathbf{r}) = \sum_{lm\sigma} f(ilm\sigma)\,\delta_{a_i}(\mathbf{r}-\mathbf{R}_i)\,\boldsymbol{\omega}^{+}_{lm\sigma}(\mathbf{r}-\mathbf{R}_i). \tag{114}$$

Substituting the multipole expansion (114) of $\mathbf{f}_i$ into the boundary-integral equation (47), applying the bra vector $\left\langle \delta_{a_i}(i)\,\boldsymbol{\omega}^{+}_{lm\sigma}(i) \right|$ and using Eq.(112) and (113), one obtains the set of algebraic equations (48) for $f(ilm\sigma)$, with multipole matrix elements,

$$\begin{aligned} M(ilm\sigma, jl'm'\sigma') = \delta_{ij} Z_i^{-1}(ilm\sigma, il'm'\sigma') + \\ (1-\delta_{ij})[T_0(ilm\sigma, jl'm'\sigma') + \tilde{T}(ilm\sigma, jl'm'\sigma')], \end{aligned} \tag{115}$$

where

$$Z_i^{-1}(ilm\sigma, il'm'\sigma') = \left\langle \delta_{a_i}(i)\,\boldsymbol{\omega}^{+}_{lm\sigma}(i) \right| \mathbf{Z}_i^{-1} \left| \delta_{a_i}(i)\,\boldsymbol{\omega}^{+}_{l'm'\sigma'}(i) \right\rangle, \tag{116}$$

$$T_0(ilm\sigma, jl'm'\sigma') = \left\langle \delta_{a_i}(i)\,\boldsymbol{\omega}^{+}_{lm\sigma}(i) \right| \mathbf{T}_0 \left| \delta_{a_j}(j)\,\boldsymbol{\omega}^{+}_{l'm'\sigma'}(j) \right\rangle, \tag{117}$$

$$\tilde{T}(ilm\sigma, jl'm'\sigma') = \left\langle \delta_{a_i}(i)\,\boldsymbol{\omega}^{+}_{lm\sigma}(i) \right| \tilde{\mathbf{T}} \left| \delta_{a_j}(j)\,\boldsymbol{\omega}^{+}_{l'm'\sigma'}(j) \right\rangle. \tag{118}$$

From now on we will use a shorthand notation $\left\langle \boldsymbol{\omega}^{+} \right| \mathbf{A} \left| \boldsymbol{\omega}^{+} \right\rangle$ for the matrices with the elements $A(ilm\sigma,\ jl'm'\sigma')$ given in the above equations with $\mathbf{A} = \mathbf{Z}_i^{-1}$, $\mathbf{T}_0$ or $\tilde{\mathbf{T}}$. Expressions for the above matrices can be found in the literature, see e.g. [21]. The technical difficulty is that, historically, in many papers, complex rather than real elementary flows $\mathbf{v}^{+}_{lm\sigma}$ and their adjoints $\mathbf{w}^{+}_{lm\sigma}$, have been used [21, 28]. As a consequence, the explicit expressions have been derived for the matrices $\left\langle \mathbf{w}^{+} \right| \mathbf{A} \left| \mathbf{w}^{+} \right\rangle$ rather than for $\left\langle \boldsymbol{\omega}^{+} \right| \mathbf{A} \left| \boldsymbol{\omega}^{+} \right\rangle$. The transformation between the real and the complex basic sets of the multipole functions $\boldsymbol{\omega}^{+}$ and $\mathbf{w}^{+}$ is the same as between $\mathbf{u}^{+}$ and $\mathbf{v}^{+}$, [21]

$$\mathbf{u}^{+}_{l0\sigma} = \frac{\mathbf{v}^{+}_{l0\sigma}}{j(\sigma)}, \tag{119}$$

$$\mathbf{u}^{+}_{lm\sigma} = \sqrt{2}\mathrm{Re}\frac{\mathbf{v}^{+}_{lm\sigma}}{j(\sigma)} = \frac{\mathbf{v}^{+}_{lm\sigma} + (-1)^{m}\,\mathbf{v}^{+}_{l,-m\sigma}}{j(\sigma)\sqrt{2}}, \qquad m \geq 1, \tag{120}$$

$$\mathbf{u}^{+}_{l-m\sigma} = \sqrt{2}\mathrm{Im}\frac{\mathbf{v}^{+}_{lm\sigma}}{j(\sigma)} = \frac{\mathbf{v}^{+}_{lm\sigma} - (-1)^{m}\,\mathbf{v}^{+}_{l,-m\sigma}}{ij(\sigma)\sqrt{2}}, \qquad m \geq 1, \tag{121}$$

where

$$j(\sigma) = \begin{cases} 1 & \text{for}\,\sigma = 0,\ 2, \\ i & \text{for}\,\sigma = 1. \end{cases} \tag{122}$$

Alternatively, using the transformation matrix **X** in the many-particle space, [21]

$$\mathbf{u}^{+}_{lm\sigma}(\mathbf{r}-\mathbf{R}_i) = \sum_{jl'm'\sigma'} \mathbf{v}^{+}_{l'm'\sigma'}(\mathbf{r}-\mathbf{R}_j)X^{\dagger}(jl'm'\sigma', ilm\sigma), \tag{123}$$

$$\omega^{+}_{lm\sigma}(\mathbf{r}-\mathbf{R}_i) = \sum_{jl'm'\sigma'} \mathbf{w}^{+}_{l'm'\sigma'}(\mathbf{r}-\mathbf{R}_j)X^{\dagger}(jl'm'\sigma', ilm\sigma), \tag{124}$$

with

$$\begin{aligned} X^{\dagger}(jl'm'\sigma', ilm\sigma) = \delta_{ji}\delta_{|m'||m|}\delta_{l'l}\delta_{\sigma'\sigma}\,\frac{1}{j(\sigma)}[\delta_{|m'|0} + \\ (1-\delta_{|m'|0})C_{\mathrm{sgn}(m'),\mathrm{sgn}(m)}(m)], \end{aligned} \tag{125}$$

where the dagger denotes the Hermitian adjoint, and $C_{\pm 1,\pm 1}(m)$ are the elements of the 2×2 unitary matrix

$$\{C_{\mu',\mu}(m)\}_{\mu',\mu=+1,-1} = \frac{1}{\sqrt{2}}\begin{pmatrix} 1 & -i \\ (-1)^{m} & i(-1)^{m} \end{pmatrix}.$$

In short, Eqs. (123) and (124) can be written as

$$\mathbf{u}^{+} = \mathbf{v}^{+} \cdot \mathbf{X}^{\dagger}, \tag{126}$$

$$\omega^{+} = \mathbf{w}^{+} \cdot \mathbf{X}^{\dagger}. \tag{127}$$

The matrix elements transform according to

$$\left\langle \mathbf{u}^+ \middle| \mathbf{A} \middle| \mathbf{u}^+ \right\rangle = \mathbf{X} \cdot \left\langle \mathbf{v}^+ \middle| \mathbf{A} \middle| \mathbf{v}^+ \right\rangle \cdot \mathbf{X}^\dagger, \tag{128}$$

$$\left\langle \boldsymbol{\omega}^+ \middle| \mathbf{A} \middle| \boldsymbol{\omega}^+ \right\rangle = \mathbf{X} \cdot \left\langle \mathbf{w}^+ \middle| \mathbf{A} \middle| \mathbf{w}^+ \right\rangle \cdot \mathbf{X}^\dagger. \tag{129}$$

With the use of the above equation the expressions $\left\langle \boldsymbol{\omega}^+ \middle| \mathbf{A} \middle| \boldsymbol{\omega}^+ \right\rangle$, appearing in (116)-(118), are evaluated in terms of the corresponding $\left\langle \mathbf{w}^+ \middle| \mathbf{A} \middle| \mathbf{w}^+ \right\rangle$ matrices, which in turn can be found in the literature. A brief outline of the available results will now be given.

We start from the matrix elements of the operator $\mathbf{Z}_i^{-1}$. Note that $\left\langle \mathbf{w}^+ \middle| \mathbf{Z}_i^{-1} \middle| \mathbf{w}^+ \right\rangle = \left\langle \mathbf{v}^+ \middle| \mathbf{Z}_i \middle| \mathbf{v}^+ \right\rangle^{-1}$. Therefore, the elements $Z_i^{-1}(ilm\sigma,\ il'm'\sigma')$ are obtained by inversion of the matrix $\left\langle \mathbf{v}^+ \middle| \mathbf{Z}_i \middle| \mathbf{v}^+ \right\rangle^{-1}$. These procedure can be carried out rigorously because, due to the spherical symmetry, $\left\langle \mathbf{v}^+ \middle| \mathbf{Z}_i \middle| \mathbf{v}^+ \right\rangle^{-1}$ is diagonal in the index l.

The task is to evaluate the matrix elements of the friction operator $\mathbf{Z}_i$ of a single spherical particle. This is achieved by solving the Stokes equations for an isolated particle subject to an external flow. The matrix $\left\langle \mathbf{v}^+ \middle| \mathbf{Z}_i \middle| \mathbf{v}^+ \right\rangle^{-1}$ is diagonal in the particle labels i and j. Due to the spherical symmetry, it is also diagonal in the azimuthal number m. Moreover it does not mix the even and odd σ indices, which correspond to vector and pseudovector components respectively. Owing to these properties, $\left\langle \mathbf{u}^+ \middle| \mathbf{Z}_i \middle| \mathbf{u}^+ \right\rangle = \left\langle \mathbf{v}^+ \middle| \mathbf{Z}_i \middle| \mathbf{v}^+ \right\rangle$. The matrix elements of the operator $\mathbf{Z}_i$ have the form [47],

$$Z_i(ilm\sigma, jl'm'\sigma') = \eta(2a_i)^{2l+\sigma+\sigma'-1}\delta_{ij}\delta_{ll'}\delta_{mm'}\begin{pmatrix} z_{l,00} & 0 & z_{l,02} \\ 0 & z_{l,11} & 0 \\ z_{l,02} & 0 & z_{l,22} \end{pmatrix}, \tag{130}$$

where the coefficients $z_{l,\sigma\sigma'}$ are specific for given boundary conditions imposed on the spherical particles. They can be found e.g. in Refs. [27, 28, 29].

Now we are going to outline how to obtain the elements $T_0(ilm\sigma,\ jl'm'\sigma')$ and $\tilde{T}(ilm\sigma, jl'm'\sigma')$ of the real matrices $\langle \boldsymbol{\omega}^+ | \mathbf{T}_0 | \boldsymbol{\omega}^+ \rangle$ and $\langle \boldsymbol{\omega}^+ | \tilde{\mathbf{T}} | \boldsymbol{\omega}^+ \rangle$, respectively. To this goal, the transformation rule (129) is applied to the complex matrices $\langle \mathbf{w}^+ | \mathbf{T}_0 | \mathbf{w}^+ \rangle$ and $\langle \mathbf{w}^+ | \tilde{\mathbf{T}} | \mathbf{w}^+ \rangle$; their elements are denoted as $G_0(ilm\sigma, jl'm'\sigma')$ and $\tilde{G}(ilm\sigma, jl'm'\sigma')$, respectively. In particular,

$$G_0(ilm\sigma, jl'm'\sigma') = (1-\delta_{ij})\frac{n_{lm}}{\eta n_{l'm'}} S^{+-}(\mathbf{R}_i - \mathbf{R}_j; lm\sigma, l'm'\sigma'), \tag{131}$$

where S^{+-} is given explicitly in Ref. [72]. It scales with the interparticle distance $R_{ij} = |\mathbf{R}_i - \mathbf{R}_j|$ as,

$$S^{+-}(\mathbf{R}_i - \mathbf{R}_j; lm\sigma, l'm'\sigma') \sim \frac{1}{R_{ij}^{l+\sigma+l'+\sigma'-1}}. \tag{132}$$

The multipole matrix elements of the operator $\tilde{\mathbf{T}}$ depend on the boundaries of the system. In particular, when the fluid occupies the halfspace $z > 0$ with $z = 0$ corresponding to an interface between two fluids of different viscosity, the operator $\tilde{\mathbf{T}}$ can be constructed with the method of images, [25, 27, 73]. The resulting multipole matrix elements read,

$$\tilde{G}(ilm\sigma, jl'm'\sigma') = \sum_{l_1 m_1 \sigma_1} \frac{n_{lm}}{\eta n_{l_1 m_1}} S^{+-}(\mathbf{R} - \mathbf{P}\cdot\mathbf{R}_j; lm\sigma, l_1 m_1 \sigma_1) \boldsymbol{\mathcal{R}}_{\mathbf{A}}(h_j; l_1 m_1 \sigma_1, l'm'\sigma'), \tag{133}$$

with the operator $\mathbf{P} = \mathbf{1} - 2\mathbf{nn}$, which reflects a vector in the $z = 0$ plane. The unit vector $\mathbf{n}$ is normal to the wall pointing into the fluid and $h_i = \mathbf{R}_i \cdot \mathbf{n}$. Due to the axial symmetry with respect to $\mathbf{n}$, the matrix $\boldsymbol{\mathcal{R}}_{\mathbf{A}}$ is diagonal in the azimuthal number m.

The formula (133) includes the limiting cases of free surface, [21, 23] and a hard wall, [19, 74, 75]. For a free surface, the matrix $\boldsymbol{\mathcal{R}}_{\mathbf{A}} = \boldsymbol{\mathcal{R}}_{\mathbf{F}}$ is given by, see [19]

$$\boldsymbol{\mathcal{R}}_{\mathbf{F}}(h_j; lm\sigma, l'm'\sigma') = (-1)^{l+m+\sigma} \delta_{mm'} \delta_{ll'} \delta_{\sigma\sigma'}. \tag{134}$$

For a hard wall the corresponding operator $\boldsymbol{\mathcal{R}}_{\mathbf{A}} = \boldsymbol{\mathcal{R}}_{\mathbf{H}}$ is not diagonal in l and σ and its matrix elements are given in Ref. [21].

For a fluid-fluid interface, when the particle is placed in a fluid with viscosity η, which occupies the halfspace $z > 0$, and the fluid on the other side of the interface, at $z < 0$, has the viscosity η', the corresponding operator $\boldsymbol{\mathcal{R}}_{\mathbf{A}}$ reads [5, 26],

$$\boldsymbol{\mathcal{R}}_{\mathbf{A}} = \frac{1}{1+\lambda}\boldsymbol{\mathcal{R}}_{\mathbf{H}} + \frac{\lambda}{1+\lambda}\boldsymbol{\mathcal{R}}_{\mathbf{F}}, \tag{135}$$

where $\lambda = \eta/\eta'$.

Recently, an efficient algorithm for accurate evaluation of the operator $\tilde{\mathbf{T}}$ for the hydrodynamic interactions of the particles confined between two parallel planar hard walls has been proposed [20, 22]. This approach involves expanding the fluid velocity field into spherical and Cartesian fundamental sets of Stokes flows. The interaction of the fluid with the particles is described using the spherical basis fields, the flow scattered by the walls is expressed in terms of the Cartesian fundamental solutions. At the core of the method are transformation relations between the spherical and the Cartesian basis sets.

Appendix B. How do Cartesian tensors relate the multipole matrix elements

Physical quantities, such as translational and angular velocities, force and torque or stresslet, are Cartesian tensors. In this Appendix it will be explained how they are related to the corresponding velocity and force spherical-multipoles. Also, the Cartesian N-particle friction tensor $\boldsymbol{\zeta}(1...N)$, with the components ζ_{ij}^{pq} $(1...N)$, will be related to the spherical-multipole matrix $\left\langle \mathbf{v}^+ \middle| \mathbf{Z} \middle| \mathbf{v}^+ \right\rangle$, with the elements $Z(ilm\sigma, jl'm'\sigma')$.

In general, transformation from the multipole to the Cartesian representation employs a set of irreducible [76, 77], i.e. completely symmetric and traceless constant tensors given by the formula [79],

$$\mathbf{y}_{lm}^{(R)} = \frac{1}{\gamma_l}\frac{1}{l!}\nabla^l \Phi_{lm}^{(R)}(\mathbf{r}) = \frac{1}{\gamma_l}\frac{1}{l!}\underbrace{\nabla\nabla...\nabla}_{l\ times}\Phi_{lm}^{(R)}(\mathbf{r}), \quad m = -l,...,l, \tag{136}$$

where

$$\gamma_l = \sqrt{\frac{(2l+1)!!}{4\pi l!}}. \tag{137}$$

The tensors $\mathbf{y}_{lm}^{(R)}$ are constant, i.e. independent of **r**, because the real solid harmonics $\Phi_{lm}^{(R)}$ are the $l-th$ order polynomials in the Cartesian components x, y, z of the position **r**, i.e. $\mathbf{y}_{lm}^{(R)}$ are combinations of terms $x^{l_1}y^{l_2}z^{l_3}$ with $l_1 + l_2 + l_3 = l$. For a given l, the tensors $\mathbf{y}_{lm}^{(R)}$ constitute the $(2l+1)$-element basis in the space of irreducible $l-th$ rank tensors, labeled by l Cartesian indices.

For a given l and $m = -l, \ldots, l$, the tensors $\mathbf{y}_{lm}^{(R)}$ are orthonormal,

$$\mathbf{y}_{lm}^{(R)} \overset{l}{\odot} \mathbf{y}_{lm'}^{(R)} = \delta_{mm'}, \quad m, m' = -l, \ldots, l, \tag{138}$$

where $\overset{l}{\odot}$ denotes l-fold contraction between the l last Cartesian indices of the tensor **A** and the first l indices of the tensor **B** [77, 78],

$$(A \overset{l}{\odot} B)_{\beta\gamma} = \sum_{\alpha_1=1}^{3} \ldots \sum_{\alpha_l=1}^{3} A_{\beta\alpha_1\ldots\alpha_l} B_{\alpha_1\ldots\alpha_l\gamma}. \tag{139}$$

In the following, $\mathbf{r}^l$ denotes l-rank tensor product of a vector **r**,

$$\mathbf{r}^l = \underbrace{\mathbf{r}\mathbf{r}\ldots\mathbf{r}}_{l\ times}. \tag{140}$$

With the use of this notation, the solid harmonics $\Phi_{lm}^{(R)}(\mathbf{r})$ may be written as

$$\Phi_{lm}^{(R)}(\mathbf{r}) = \gamma_l\, \mathbf{y}_{lm}^{(R)} \overset{l}{\odot} \mathbf{r}^l, \tag{141}$$

and their gradients are given by

$$\nabla\Phi_{lm}^{(R)}(\mathbf{r}) = \Gamma_l\, \mathbf{y}_{lm}^{(R)} \overset{l-1}{\odot} \mathbf{r}^{l-1}, \tag{142}$$

where

$$\Gamma_l = l\gamma_l = \sqrt{\frac{l(2l+1)!!}{4\pi(l-1)!}}. \tag{143}$$

The set of tensors $\mathbf{y}_{lm}^{(R)}$ allows to rewrite the formulas for the elementary flows $\mathbf{u}_{lm\sigma}^{+}$. To this goal we substitute the formula (142) for gradient $\nabla\Phi_{lm}^{(R)}$ into the definition (102) of the elementary flow $\mathbf{u}_{lm0}^{+}$,

$$\mathbf{u}_{lm0}^{+}(\mathbf{r}) = \Gamma_l\, \mathbf{y}_{lm}^{(R)} \overset{l-1}{\odot} \mathbf{r}^{l-1}. \tag{144}$$

The other flows with $\sigma = 1, 2$ follow from Eqs. (103-104). In particular,

$$\mathbf{u}_{1m0}^{+}(\mathbf{r}) = \Gamma_1\, \mathbf{y}_{1m}^{(R)}, \tag{145}$$

$$\mathbf{u}_{1m1}^{+}(\mathbf{r}) = \Gamma_1\, \mathbf{y}_{1m}^{(R)} \times \mathbf{r}, \tag{146}$$

$$\mathbf{u}_{2m0}^{+}(\mathbf{r}) = \Gamma_2\, \mathbf{y}_{2m}^{(R)} \cdot \mathbf{r}, \tag{147}$$

where

$$\mathbf{y}_{1-1}^{(R)} = -\mathbf{e}_y = \begin{pmatrix} 0 \\ -1 \\ 0 \end{pmatrix}, \quad \mathbf{y}_{10}^{(R)} = \mathbf{e}_z = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}, \quad \mathbf{y}_{11}^{(R)} = -\mathbf{e}_x = \begin{pmatrix} -1 \\ 0 \\ 0 \end{pmatrix}, \tag{148}$$

$$\mathbf{y}_{2-2}^{(R)} = \begin{pmatrix} 0 & \frac{1}{\sqrt{2}} & 0 \\ \frac{1}{\sqrt{2}} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \mathbf{y}_{2-1}^{(R)} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -\frac{1}{\sqrt{2}} \\ 0 & -\frac{1}{\sqrt{2}} & 0 \end{pmatrix}, \tag{149}$$

$$\mathbf{y}_{20}^{(R)} = \begin{pmatrix} -\frac{1}{\sqrt{6}} & 0 & 0 \\ 0 & -\frac{1}{\sqrt{6}} & 0 \\ 0 & 0 & \sqrt{\frac{2}{3}} \end{pmatrix}, \quad \mathbf{y}_{21}^{(R)} = \begin{pmatrix} 0 & 0 & -\frac{1}{\sqrt{2}} \\ 0 & 0 & 0 \\ -\frac{1}{\sqrt{2}} & 0 & 0 \end{pmatrix}, \tag{150}$$

$$\mathbf{y}_{22}^{(R)} = \begin{pmatrix} \frac{1}{\sqrt{2}} & 0 & 0 \\ 0 & -\frac{1}{\sqrt{2}} & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{151}$$

and

$$\Gamma_1 = \sqrt{\frac{3}{4\pi}}, \qquad \Gamma_2 = \sqrt{\frac{15}{2\pi}}. \tag{152}$$

To obtain the force multipoles, the expressions (145)-(147) for the flows $\mathbf{u}^{+}_{lm\sigma}$ are now integrated with the force density, according to Eq. (49). The tensors $\mathbf{y}^{(R)}_{lm}$ separate out from the integrals, which result in forces, torques and stresslets, respectively, in agreement with Eqs. (22),

$$f(i1m0) \;=\; \Gamma_1\, \mathbf{y}^{(R)}_{1m} \cdot \mathcal{F}_i, \qquad m = 0, \pm 1, \tag{153}$$

$$f(i1m1) \;=\; \Gamma_1\, \mathbf{y}^{(R)}_{1m} \cdot \mathcal{T}_i, \qquad m = 0, \pm 1, \tag{154}$$

$$f(i2m0) \;=\; \Gamma_2\, \mathbf{y}^{(R)}_{2m} \overset{2}{\odot} \mathbf{S}_i, \qquad m = 0, \pm 1, \pm 2. \tag{155}$$

Using the above equations and the orthonormality property, Eq. (138), we express the force, torque and stresslet in terms of the force multipoles as

$$\mathcal{F}_i = \frac{1}{\Gamma_1} \sum_{m=-1}^{1} f(i1m0)\mathbf{y}^{(R)}_{1m}, \tag{156}$$

$$\mathcal{T}_i = \frac{1}{\Gamma_1} \sum_{m=-1}^{1} f(i1m1)\mathbf{y}^{(R)}_{1m}, \tag{157}$$

$$\mathbf{S}_i = \frac{1}{\Gamma_2} \sum_{m=-2}^{2} f(i2m0)\mathbf{y}^{(R)}_{2m}. \tag{158}$$

With the use of Eq. (148) and (152), the forces and torques are now written down explicitly,

$$\begin{aligned} \mathcal{F}_i &= \begin{pmatrix} F_x \\ F_y \\ F_z \end{pmatrix} = \sqrt{\frac{4\pi}{3}} \begin{pmatrix} -f(i110) \\ -f(i1{-1}0) \\ f(i100) \end{pmatrix}, \\ \mathcal{T}_i &= \begin{pmatrix} T_x \\ T_y \\ T_z \end{pmatrix} = \sqrt{\frac{4\pi}{3}} \begin{pmatrix} -f(i111) \\ -f(i1{-1}1) \\ f(i101) \end{pmatrix}. \end{aligned} \tag{159}$$

Note that each Cartesian component of the force and torque is proportional to a single force multipole only.

The translational and rotational velocities of the spheres will also be expressed in terms of the velocity multipoles. To this goal, the expressions (145) and (146) for $\mathbf{u}^{+}_{1m0}(\mathbf{r}-\mathbf{R}_i)$ and $\mathbf{u}^{+}_{1m1}(\mathbf{r}-\mathbf{R}_i)$, respectively, are now substituted to Eq. (51) defining the rigid velocity $\mathbf{w}_i(\mathbf{r})$. We obtain,

$$\mathbf{U}_i = \Gamma_1 \sum_{m=-1}^{1} c_w(i1m0)\mathbf{y}^{(R)}_{1m}, \tag{160}$$

$$\mathbf{\Omega}_i = \Gamma_1 \sum_{m=-1}^{1} c_w(i1m1)\mathbf{y}^{(R)}_{1m}, \tag{161}$$

or explicitly,

$$\begin{aligned}
\mathbf{U}_i &= \begin{pmatrix} U_x \\ U_y \\ U_z \end{pmatrix} = \sqrt{\frac{3}{4\pi}} \begin{pmatrix} -c_w(i110) \\ -c_w(i1-10) \\ c_w(i100) \end{pmatrix}, \\
\mathbf{\Omega}_i &= \begin{pmatrix} \Omega_x \\ \Omega_y \\ \Omega_z \end{pmatrix} = \sqrt{\frac{3}{4\pi}} \begin{pmatrix} -c_w(i111) \\ -c_w(i1-11) \\ c_w(i101) \end{pmatrix}.
\end{aligned} \tag{162}$$

The ambient flow and its derivatives at the center of the sphere also can be expressed by the corresponding velocity multipoles. To this end the expansion (52) of the ambient flow $\mathbf{v}_\infty$ is used. In analogy to Eq. (162),

$$\begin{aligned}
\mathbf{v}_\infty(\mathbf{R}_i) &= \sqrt{\frac{3}{4\pi}} \begin{pmatrix} -c_\infty(i110) \\ -c_\infty(i1-10) \\ c_\infty(i100) \end{pmatrix}, \\
\frac{1}{2}\nabla\times\mathbf{v}_\infty(\mathbf{R}_i) &= \sqrt{\frac{3}{4\pi}} \begin{pmatrix} -c_\infty(i111) \\ -c_\infty(i1-11) \\ c_\infty(i101) \end{pmatrix}.
\end{aligned} \tag{163}$$

Evaluating the gradients of Eqs. (52) and (144) we obtain the rate of strain,

$$\mathbf{g}_{\infty i} = \sqrt{\frac{15}{2\pi}} \sum_{m=-2}^{2} c_\infty(i2m0)\, \mathbf{y}_{2m}^{(R)}. \tag{164}$$

The velocity multipoles are now explicitly evaluated, using the orthonormality property (138),

$$c_w(i1m0) = \frac{1}{\Gamma_1} \mathbf{y}_{1m}^{(R)} \cdot \mathbf{U}_i, \tag{165}$$

$$c_w(i1m1) = \frac{1}{\Gamma_1} \mathbf{y}_{1m}^{(R)} \cdot \mathbf{\Omega}_i, \tag{166}$$

$$c_\infty(i1m0) = \frac{1}{\Gamma_1} \mathbf{y}_{1m}^{(R)} \cdot \mathbf{v}_{\infty i}, \tag{167}$$

$$c_\infty(i1m1) = \frac{1}{\Gamma_1} \mathbf{y}_{1m}^{(R)} \cdot \boldsymbol{\omega}_{\infty i}, \tag{168}$$

$$c_\infty(i2m0) = \frac{1}{\Gamma_2} \mathbf{y}_{2m}^{(R)} \overset{2}{\odot} \mathbf{g}_{\infty i}. \tag{169}$$

Finally, the Cartesian generalized friction tensors ζ_{ij}^{pq} with p, $q = t$, r, d will be related to the correspondingmultipole elements $Z(ilm\sigma, jl'm'\sigma')$. To this end, Eqs. (156-158), (165)-(169) and (54) are combined. In particular,

$$\zeta_{ij}^{\mathrm{tt}}(1...N) = \frac{4\pi}{3} \begin{pmatrix} Z(i110, j110) & Z(i110, j1-10) & -Z(i110, j100) \\ Z(i1-10, j110) & Z(i1-10, j1-10) & -Z(i1-10, j100) \\ -Z(i100, j110) & -Z(i100, j1-10) & Z(i100, j100) \end{pmatrix}, \tag{170}$$

and in general,

$$\zeta_{ij}^{pq}(1...N) = \frac{1}{\Gamma_{\mathrm{p}}\Gamma_{\mathrm{q}}} \sum_{m=-l_{\mathrm{p}}}^{l_{\mathrm{p}}} \sum_{m'=-l_{\mathrm{q}}}^{l_{\mathrm{q}}} \mathbf{y}_{l_{\mathrm{p}}m}^{(R)} Z(il_{\mathrm{p}}m\sigma_{\mathrm{p}}, jl_{\mathrm{q}}m'\sigma_{\mathrm{q}})\, \mathbf{y}_{l_{\mathrm{q}}m'}^{(R)}, \tag{171}$$

where

$$l_{\mathrm{p}} = \begin{cases} 1 & \text{for } p = t, r, \\ 2 & \text{for } p = d, \end{cases} \tag{172}$$

and

$$\sigma_{\mathrm{p}} = \begin{cases} 0 & \text{for } p = t, d, \\ 1 & \text{for } p = r. \end{cases} \tag{173}$$

Appendix C. Ambient flows as combinations of the elementary flows

In this section, we outline the procedure to evaluate the expansion coefficients $c_{\infty}(ilm\sigma)$ of an ambient flow $\mathbf{v}_{\infty}$, appearing in Eq. (52),

$$\mathbf{v}_{\infty}(\mathbf{r}) \;=\; \sum_{l=1}^{\infty} \sum_{m=-1}^{l} \sum_{\sigma=0}^{2} c_{\infty}(ilm\sigma)\, \boldsymbol{u}^{+}_{lm\sigma}(\mathbf{r}-\mathbf{R}_i). \tag{174}$$

It is not convenient to compute them by integration of the product of external flow $\mathbf{v}_{\infty}(\mathbf{r})$ and the basis fields $\boldsymbol{\omega}^{+}_{lm\sigma}(\mathbf{r}-\mathbf{R}_i)$ over the surface of sphere i, as in Eq. (113). Here we propose an alternative method for evaluating the coefficients $c_{\infty}(ilm\sigma)$ for arbitrary external flow $\mathbf{v}_{\infty}$. This method requires some algebraic manipulations and evaluation of the ambient flow derivatives only and no integration is involved. The basic idea is to decompose $\mathbf{v}_{\infty}$ into three families of solutions to the Stokes equations constructed by Lamb [37].

An arbitrary regular ambient flow $\mathbf{v}_{\infty}(\mathbf{r})$ satisfying the Stokes equations can be expanded as the Taylor series about the center of particle i,

$$\mathbf{v}_{\infty}(\mathbf{r}) = \sum_{l=0}^{\infty} (\mathbf{r}-\mathbf{R}_i)^l \overset{l}{\odot} \mathbf{c}^{(l+1)}, \tag{175}$$

where the $(l+1)$-th rank tensors $\mathbf{c}^{(l+1)}$ are given by

$$\mathbf{c}^{(l+1)} = \frac{1}{l!} \nabla^{l} \mathbf{v}_{\infty}(\mathbf{R}_i), \quad l = 0, ..., \infty. \tag{176}$$

The coefficients $\mathbf{c}^{(l+1)}$ depend on the choice of the particle i. Each component $(\mathbf{r}-\mathbf{R}_i)^l \overset{l}{\odot} \mathbf{c}^{(l+1)}$ appearing in Eq. (175) satisfies the Stokes equations, with the appropriately chosen pressure. Our goal is to represent it as a linear combination of the elementary flows $\mathbf{u}^{+}(\mathbf{r}-\mathbf{R}_i)$. To this end, three families, labeled by σ = 0, 1, 2, of the irreducible tensors $\mathbf{c}_{l,\sigma}$ have been constructed [50, 80, 81] in terms of the tensors defined in Eq. (176),

$$\mathbf{c}_{l+1,0} = \overline{\mathbf{c}^{(l+1)}}, \tag{177}$$

$$(\mathbf{c}_{l,1})_{\gamma_1\ldots\gamma_l} = \frac{l}{l+1}\overline{\varepsilon_{\gamma_1\lambda\mu} c^{(l+1)}_{\lambda\gamma_2\ldots\gamma_l\mu}}, \tag{178}$$

$$(\mathbf{c}_{l-1,2})_{\gamma_1\ldots\gamma_{l-1}} = \frac{(l-1)^2}{(2l-1)(2l+1)}\overline{\delta_{\lambda\mu} c^{(l+1)}_{\lambda\mu\gamma_1\ldots\gamma_{l-1}}}, \tag{179}$$

or equivalently

$$\mathbf{c}_{l+1,0} = \frac{1}{l!}\overline{\nabla^{l}\mathbf{v}_\infty(\mathbf{R}_i)} \tag{180}$$

$$\mathbf{c}_{l,1} = \frac{1}{l+1}\frac{1}{l!}\overline{\nabla^{l-1}\,[\nabla\times\mathbf{v}_\infty(\mathbf{R}_i)]}, \tag{181}$$

$$\mathbf{c}_{l-1,2} = \frac{(l-1)^2}{(2l-1)(2l+1)}\frac{1}{l!}\overline{\nabla^{l-2}\,[\nabla^2\mathbf{v}_\infty(\mathbf{R}_i)]}, \tag{182}$$

where $\varepsilon_{\gamma_1\lambda\mu}$ is the completely antisymmetric Levi-Civita tensor and $\delta_{\lambda\mu}$ is the Kronecker symbol and $\overline{\mathbf{a}}$ indicates the irreducible (completely symmetric and traceless) part of a tensor $\mathbf{a}$, evaluated by the procedure described in Refs. [76, 77]. The first index of the tensor $\mathbf{c}_{l,\sigma}$ denotes its rank and the second is the label of the family, σ = 0, 1, 2.

Now we define the linear operators $\mathbf{V}_{l\sigma}$, $P_{l\sigma}$, σ = 0, 1, 2, which map irreducible l − th rank tensors $\mathbf{d}_l$ into regular solutions of the Stokes equations,

$$\mathbf{d}_l \rightarrow (\mathbf{V}_{l\sigma}[\mathbf{d}_l],\, P_{l\sigma}[\mathbf{d}_l]), \quad \sigma = 0,1,2, \tag{183}$$

where

$$\begin{aligned} \{\mathbf{V}_{l0}[\mathbf{d}_l]\}(\mathbf{r}) &= \mathbf{r}^{l-1} \overset{l-1}{\odot} \mathbf{d}_l, \\ \{P_{l0}[\mathbf{d}_l]\}(\mathbf{r}) &= 0, \end{aligned} \tag{184}$$

$$\begin{aligned}\{\mathbf{V}_{l1}[\mathbf{d}_l]\}(\mathbf{r}) &= \{\mathbf{V}_{l0}[\mathbf{d}_l]\}(\mathbf{r})\times\mathbf{r}\\ \{P_{l1}[\mathbf{d}_l]\}(\mathbf{r}) &= 0,\end{aligned}\tag{185}$$

$$\begin{aligned}\{\mathbf{V}_{l2}[\mathbf{d}_l]\}(\mathbf{r}) &= \frac{(2l+1)}{l}\left[\frac{(l+3)}{2}r^2\{\mathbf{V}_{l0}[\mathbf{d}_l]\}(\mathbf{r})-\mathbf{r}\cdot\{\mathbf{V}_{l0}[\mathbf{d}_l]\}(\mathbf{r})\mathbf{r}\right],\\ \{P_{l2}[\mathbf{d}_l]\}(\mathbf{r}) &= \eta\frac{(l+1)(2l+1)(2l+3)}{l^2}\mathbf{r}^l\overset{l}{\odot}\mathbf{d}_l.\end{aligned}\tag{186}$$

In terms of these operators the elementary flows $\mathbf{u}^+_{lm\sigma}$ and the pressure fields $p^+_{lm\sigma}$ (102)-(104) are given by

$$\mathbf{u}^+_{lm\sigma} = \Gamma_l\mathbf{V}_{l\sigma}[\mathbf{y}^{(R)}_{lm}],\tag{187}$$

$$p^+_{lm\sigma} = \Gamma_l P_{l\sigma}[\mathbf{y}^{(R)}_{lm}].\tag{188}$$

Each partial flow $(\mathbf{r}-\mathbf{R}_i)^l\overset{l}{\odot}\mathbf{c}^{(l+1)}$ appearing in Eq. (175) can be in turn decomposed into three flows from the three families, see Refs. [80], [81],

$$\begin{aligned}(\mathbf{r}-\mathbf{R}_i)^l\overset{l}{\odot}\mathbf{c}^{(l+1)} = {}&\{\mathbf{V}_{l+1,0}[\mathbf{c}_{l+1,0}]\}(\mathbf{r}-\mathbf{R}_i)+\\ &\{\mathbf{V}_{l,1}[\mathbf{c}_{l,1}]\}(\mathbf{r}-\mathbf{R}_i)+\{\mathbf{V}_{l-1,2}[\mathbf{c}_{l-1,2}]\}(\mathbf{r}-\mathbf{R}_i).\end{aligned}\tag{189}$$

In the above expression, we represent each tensor $\mathbf{c}_{l,\sigma}$ as a linear combination of the tensors $\mathbf{y}^{(R)}_{lm}$,

$$\mathbf{c}_{l\sigma} = \sum_{m=-1}^{l}\left(\mathbf{c}_{l,\sigma}\overset{l}{\odot}\mathbf{y}^{(R)}_{lm}\right)\mathbf{y}^{(R)}_{lm}.\tag{190}$$

Then we use Eq. (187) to write each partial flow $(\mathbf{r}-\mathbf{R}_i)^l\overset{l}{\odot}\mathbf{c}^{(l+1)}$ as a combination of $\mathbf{u}^+_{lm\sigma}$.

Finally, summing up the partial flows, c.f. Eq. (175), we obtain an arbitrary regular flow $\mathbf{v}_\infty(\mathbf{r})$ as a combination of the elementary flows

$$\mathbf{v}_\infty(\mathbf{r}) = \sum_{l=1}^{\infty}\frac{1}{\Gamma_l}\sum_{m=-1}^{l}\sum_{\sigma=0}^{2}\left(\mathbf{c}_{l,\sigma}\overset{l}{\odot}\mathbf{y}^{(R)}_{l,m}\right)\mathbf{u}^+_{lm\sigma}(\mathbf{r}-\mathbf{R}_i).\tag{191}$$

Therefore,

$$\mathbf{c}_{\infty}(ilm\sigma) = \frac{1}{\Gamma_l}\left(\mathbf{c}_{l,\sigma} \overset{l}{\odot} \mathbf{y}^{(R)}_{l,m}\right). \tag{192}$$

Note that the tensors $\mathbf{c}_{l,\sigma}$ depend on i, c.f. Eq. (176), but $\mathbf{y}^{(R)}_{l,m}$ do not. Explicit expressions for the coefficients $c_{\infty}(ilm\sigma)$ for shear and Poiseuille flows were also given in Refs. [61] and [62], respectively.

Nonpolynomial regular flows can by expanded approximately by retaining in Eq. (191) terms with $l \leq l_{\max}$ only for an arbitrary $l_{\max}$. Polynomial flows can be expanded rigorously. Indeed, in this case, $\mathbf{c}_{l,\sigma} = 0$ for $l > l_{\max}$, where $l_{\max} - 1$ is the order of the polynomial, see (180), (181) and (182).

The algorithm described in this Appendix has been implemented in MATHEMATICA, used in Ref. [39] to decompose shear, modulated shear, quadratic and Poiseuille flows, and applied to analyze particle dynamics in Refs. [64, 65, 69].

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