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Scattering series in the mobility problem for suspensions

Karol Makuch

Institute of Theoretical Physics, Faculty of Physics, University of Warsaw,
Hoża 69, 00-681 Warsaw, Poland
E-mail: Karol.Makuch@fuw.edu.pl

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Abstract. The mobility problem for suspensions of spherical particles immersed in an arbitrary flow of a viscous, incompressible fluid is considered in the regime of low Reynolds numbers. The scattering series which appears in the mobility problem is simplified. The simplification relies on the reduction of the number of types of single-particle scattering operators appearing in the scattering series. In our formulation there is only one type of single-particle scattering operator.

Keywords: heterogeneous materials (theory), colloids, bio-colloids and nano-colloids

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1. Introduction

Various types of suspension of spherical particles can be found in nature and industry. The diversity follows from the fact that systems may be composed of many sorts of particles, such as hard spheres or spherical polymers [1], and many types of forces may act between particles [2, 3]. This variety in a structure implies a wide range of phenomena exhibited by suspensions. To understand them it is often crucial to consider hydrodynamic interactions between particles, that is their mutual influence through movement of the surrounding liquid (which is different from the influence through direct forces, such as magnetic or van der Waals force). In many physical situations examination of the hydrodynamic interactions amounts to the friction problem or the mobility problem [4]. In both cases the particles are immersed in a flow of the surrounding liquid (ambient flow). In the friction problem the velocities of particles are assumed and the hydrodynamic forces on the fluid produced by the particles are calculated. This problem appears, for example, in the determination of Stokes coefficient for a polymer modeled as an agglomerate of spherical particles [5]–[7]. In the mobility problem one determines the velocities of freely moving particles and also the hydrodynamic forces acting on the fluid. The particles are assumed to be immersed in the ambient flow and the external forces may act on them. Among the situations in which the mobility problem appears, the determination of the sedimentation coefficient of the suspension can be mentioned [8, 9].

To solve both the friction and the mobility problem one starts with the equations which govern the dynamics of the suspension. Here we assume linear, stationary Stokes equations for an incompressible fluid with stick boundary conditions on the surface of particles. One of the possible approaches to the Stokes equations is the method of successive approximations [10], also known as the reflection method, developed by Smoluchowski [11]. It is based on linearity of the Stokes equations. The method consists in successive superpositions of the single-particle solutions of the Stokes equations chosen to fulfil the boundary conditions with increasing accuracy. The above procedure leads

to the solution of the friction problem in a form of the superpositions of multiple reflected flows. The resulting structure of the solution is called the scattering series. In the series the single-particle operator plays an important role. In the case of the friction problem the scattering series has a simple form because there is only one type of single-particle operator. The situation is more complicated in the mobility problem. Here four types of single-particle operators are considered [12]. Due to the difference in the number of single-particle operators, statistical physics considerations are easier in the case of the friction problem than in the case of the mobility problem. For the latter, many formulas of the same structure can be found in the literature [12]–[19].

The aim of the present paper is the reformulation of the mobility problem. The reformulation results in a scattering series in which only one type of single-particle operator appears. It enables simplification of the statistical physics considerations relevant to the mobility problem.

2. Governing equations

The system under consideration consists of N identical hard spherical particles of radius a immersed in an incompressible, infinite Newtonian liquid with kinematic viscosity η . The inertia of particles and the inertia terms in the incompressible Navier–Stokes equations are assumed to be negligible. As a consequence, the fluid is governed by the steady Stokes equations [4]. We supplement the Stokes equations with stick (no-slip) boundary conditions on the surface of the particles [20]. Next, following an idea of Mazur and Bedeaux [21] we extend the Stokes equations inside the particles in the following way:

$$\nabla p(\mathbf{r}) - \eta \Delta \mathbf{v}(\mathbf{r}) = \mathbf{f}(\mathbf{r}), \quad (1a)$$

$$\nabla \cdot \mathbf{v}(\mathbf{r}) = 0, \quad (1b)$$

introducing induced force densities $\mathbf{f}(\mathbf{r})$ [22, 23]. Here $p(\mathbf{r})$ and $\mathbf{v}(\mathbf{r})$ are the pressure field and the velocity field of the suspension. The induced force densities are determined [24] by the condition that the flow of the suspension inside the particles reproduces their hard-sphere velocity field,

$$\mathbf{v}(\mathbf{r}) = \mathbf{U}_i(\mathbf{r}) = \mathbf{V}_i + \boldsymbol{\Omega}_i \times (\mathbf{r} - \mathbf{R}_i) \quad \text{for } |\mathbf{r} - \mathbf{R}_i| \leq a, \quad (2)$$

where \mathbf{V}_i and $\boldsymbol{\Omega}_i$ are translational and rotational velocities of the i th particle, which is located at the position \mathbf{R}_i .

For the case under consideration the force densities $\mathbf{f}(\mathbf{r})$ are localized only on the surface of particles [15, 25], that is

$$\mathbf{f}(\mathbf{r}) = \sum_{i=1}^N \mathbf{f}_i(\mathbf{r}), \quad (3)$$

with $\mathbf{f}_i(\mathbf{r})$ localized only on the surface of the i th particle,

$$\mathbf{f}_i(\mathbf{r}) = -\boldsymbol{\sigma} \cdot \mathbf{n}_i \delta(|\mathbf{r} - \mathbf{R}_i| - a), \quad (4)$$

where $\boldsymbol{\sigma}$ is the stress tensor for the fluid [4], \mathbf{n}_i is a vector normal to the surface of the particle i at point \mathbf{r} , and $\delta(x)$ is the Dirac delta function.

The above extension of the Stokes equations allows one to use the Green function method. The method leads to the expression for the flow of suspension in the whole space [26],

$$\mathbf{v}(\mathbf{r}) = \int d^3\mathbf{r}' \mathbf{G}_0(\mathbf{r} - \mathbf{r}') \cdot \mathbf{f}(\mathbf{r}'), \quad (5)$$

where the Oseen tensor \mathbf{G}_0 has the following form [27]

$$\mathbf{G}_0(\mathbf{r}) = \frac{1}{8\pi\eta} \frac{\mathbf{1} + \hat{\mathbf{r}}\hat{\mathbf{r}}}{|\mathbf{r}|}, \quad \hat{\mathbf{r}} = \frac{\mathbf{r}}{|\mathbf{r}|}. \quad (6)$$

3. Friction problem

In the friction problem the particles are immersed in a liquid in which initially ambient flow $\mathbf{v}_0(\mathbf{r})$ is present, and their translational \mathbf{V}_i and rotational velocities $\mathbf{\Omega}_i$ are assumed to be known. The aim of the friction problem is a determination of the force densities $\mathbf{f}_i(\mathbf{r})$ induced on the surface of the particles.

For a single particle, the friction problem for a particular type of ambient flow was solved more than a century ago [10]. It was later generalized for an arbitrary ambient flow [24, 28]. The solution of the Stokes equations (1) in this case has the form of the following linear relation [28]

$$\mathbf{f}_1(\mathbf{r}) = \int d^3\mathbf{r}' \mathbf{Z}_0(\mathbf{R}_1, \mathbf{r}, \mathbf{r}') \cdot (\mathbf{U}_1(\mathbf{r}') - \mathbf{v}_0(\mathbf{r}')). \quad (7)$$

The single-particle resistance operator \mathbf{Z}_0 is localized on the surface of a particle. It means that the force density $\mathbf{f}_1(\mathbf{r})$ is localized on the particle surface—consistently with the equation (4)—and its value depends only on the velocity field $\mathbf{U}_1(\mathbf{r}) - \mathbf{v}_0(\mathbf{r})$ at points $|\mathbf{r} - \mathbf{R}_1| = a$. The details of the resistance operator \mathbf{Z}_0 can be found in the appendix. Equation (7) will have the following form in shorthand notation

$$\mathbf{f}_1 = \mathbf{Z}_0(1) \cdot (\mathbf{U}_1 - \mathbf{v}_0), \quad (8)$$

in which the integral variables are omitted and the position of a particle is denoted by its index. The notation will be extensively used further on.

It is worth mentioning that the single-particle friction problem for a spherical shape has been solved not only for a hard sphere with stick boundary conditions. Many other physical situations have also been considered in the literature, e.g. different boundary conditions [29], permeable particles [30], spherical polymers [31], immiscible droplets [32, 33] or more complex cases [34, 35]. In general, the relation (8) is still valid but with a modified \mathbf{Z}_0 operator.

The concept of the induced force densities and linearity of the Stokes equations allows one to use the single-particle friction problem to find the solution of the friction problem for a suspension. In fact, the i th particle in a suspension is surrounded by a flow which is a superposition of the ambient flow $\mathbf{v}_0(\mathbf{r})$ and the flow induced by the other particles, $\sum_{j \neq i} \int d\mathbf{r}' \mathbf{G}_0(\mathbf{r}, \mathbf{r}') \mathbf{f}_j(\mathbf{r}')$. Applying this modified ambient flow to equation (8) leads to

the following expression

$$\mathbf{f}_i = \mathbf{Z}_0(i) \cdot \left(\mathbf{U}_i - \mathbf{v}_0 - \sum_{j \neq i} \mathbf{G}_0 \mathbf{f}_j \right) \quad (9)$$

written in the shorthand notation. This is the formula where one can directly implement the reflection method by successive iterations. It yields

$$\mathbf{f}_i = \sum_{j=1}^N \mathbf{Z}_{ij}(1 \cdots N) (\mathbf{U}_j - \mathbf{v}_0), \quad (10)$$

where the friction operator \mathbf{Z}_{ij} has the form of the scattering series

$$\begin{aligned} \mathbf{Z}_{ij}(1 \cdots N) &= \delta_{ij} \mathbf{Z}_0(i) - (1 - \delta_{ij}) \mathbf{Z}_0(i) \mathbf{G}_0(ij) \mathbf{Z}_0(j) \\ &+ \sum_k^{\prime} \mathbf{Z}_0(i) \mathbf{G}_0(ik) \mathbf{Z}_0(k) \mathbf{G}_0(kj) \mathbf{Z}_0(j) + \cdots \end{aligned} \quad (11)$$

The different terms in the equation (11) correspond to scattering sequences. The prime symbol indicates summation over k different than neighboring particle indices in the scattering sequence.

4. Mobility problem

In the mobility problem one considers freely moving particles immersed in an ambient flow of the fluid $\mathbf{v}_0(\mathbf{r})$ and subjected to the action of the external forces. The aim of the problem is to calculate the velocity fields of the particles $\mathbf{U}_i(\mathbf{r})$ and also the induced force densities $\mathbf{f}_i(\mathbf{r})$ [4].

In order to obtain the solution for a suspension we first analyze the case of a single particle. Before going into the details, it should be noticed that linearity of the Stokes equations implies a linear relation between the response of the particle and the source of a disturbance. Therefore the velocity field of the particle $\mathbf{U}_i(\mathbf{r})$ and the induced forces $\mathbf{f}_i(\mathbf{r})$ on its surface are linear in the ambient flow $\mathbf{v}_0(\mathbf{r})$ and the external forces $\mathbf{f}_{\text{ext}}(\mathbf{r})$,

$$\mathbf{U}_1(\mathbf{r}) = \int d\mathbf{r}' \mathbf{M}_0(\mathbf{R}_1, \mathbf{r}, \mathbf{r}') \mathbf{f}_{\text{ext}}(\mathbf{r}') + \int d\mathbf{r}' \mathbf{M}_<(\mathbf{R}_1, \mathbf{r}, \mathbf{r}') \mathbf{v}_0(\mathbf{r}'), \quad (12)$$

$$\mathbf{f}_1(\mathbf{r}) = \int d\mathbf{r}' \mathbf{M}_>(\mathbf{R}_1, \mathbf{r}, \mathbf{r}') \mathbf{f}_{\text{ext}}(\mathbf{r}') + \int d\mathbf{r}' \hat{\mathbf{M}}(\mathbf{R}_1, \mathbf{r}, \mathbf{r}') \mathbf{v}_0(\mathbf{r}'). \quad (13)$$

Since the above statement is based only on the linearity of the governing equations it is correct for the different particles and boundary conditions mentioned in section 3. The single-particle operators \mathbf{M}_0 , $\mathbf{M}_<$, $\mathbf{M}_>$ and $\hat{\mathbf{M}}$ need to be determined for every particular case. In what follows we discuss hard spheres with stick boundary conditions [12, 16]. From the papers cited here, one can easily infer the form of the $\mathbf{M}_0 \equiv \boldsymbol{\mu}_0$ operator which is explicitly given in the appendix. The remaining operators are expressed with the following equations

$$\mathbf{M}_<(1) = \boldsymbol{\mu}_0(1) \mathbf{Z}_0(1), \quad (14)$$

$$\mathbf{M}_{>}(1) = \mathbf{Z}_0(1)\boldsymbol{\mu}_0(1), \quad (15)$$

$$\hat{\mathbf{M}}(1) = -\mathbf{Z}_0(1) + \mathbf{Z}_0(1)\boldsymbol{\mu}_0(1)\mathbf{Z}_0(1) \quad (16)$$

which were written in the shorthand notation.

Due to linearity of the Stokes equations the above single-particle solution of the mobility problem can be used to solve the case of a suspension. In the suspension, the i th particle is immersed in the flow given by a superposition of the ambient flow $\mathbf{v}_0(\mathbf{r})$ and the flow generated by other particles, $\sum_{j \neq i} \int d\mathbf{r}' \mathbf{G}_0(\mathbf{r}, \mathbf{r}') \mathbf{f}_j(\mathbf{r}')$. Assuming this modified ambient flow in equations (12) and (13) leads to the expressions for the velocity of particles

$$\mathbf{U}_i = \mathbf{M}_0(i)\mathbf{f}_{\text{ext}} + \mathbf{M}_{<}(i) \left(\mathbf{v}_0 + \sum_{j \neq i} \mathbf{G}_0 \mathbf{f}_j \right) \quad (17)$$

and the induced force densities in suspension

$$\mathbf{f}_i = \mathbf{M}_{>}(i)\mathbf{f}_{\text{ext}} + \hat{\mathbf{M}}(i) \left(\mathbf{v}_0 + \sum_{j \neq i} \mathbf{G}_0 \mathbf{f}_j \right). \quad (18)$$

In what follows we rewrite the equations (17) and (18) in the following concise form

$$\mathbf{s}_i = \mathbf{M}(i) \left(\boldsymbol{\psi}_0 + \sum_{j \neq i} \mathbf{G} \mathbf{s}_j \right), \quad (19)$$

where the response of the i th particle \mathbf{s}_i , single-particle mobility operator $\mathbf{M}(i)$, and external field $\boldsymbol{\psi}_0$ are defined respectively below:

$$\mathbf{s}_i = \begin{bmatrix} \mathbf{U}_i \\ \mathbf{f}_i \end{bmatrix}, \quad (20)$$

$$\boldsymbol{\psi}_0 = \begin{bmatrix} \mathbf{F}_{\text{ext}} \\ \mathbf{v}_0 \end{bmatrix}, \quad (21)$$

$$\mathbf{M}(i) = \begin{bmatrix} \mathbf{M}_0(i) & \mathbf{M}_{<}(i) \\ \mathbf{M}_{>}(i) & \hat{\mathbf{M}}(i) \end{bmatrix}, \quad (22)$$

whereas \mathbf{G} is generalized Oseen tensor \mathbf{G}_0 :

$$\mathbf{G} = \begin{bmatrix} 0 & 0 \\ 0 & \mathbf{G}_0 \end{bmatrix}. \quad (23)$$

The method of iterations applied to the equations (19) leads to the following solution of the mobility problem

$$\mathbf{s}_i(\mathbf{R}_1, \dots, \mathbf{R}_N) = \sum_{j=1}^N \mathbf{T}_{ij}(\mathbf{R}_1, \dots, \mathbf{R}_N) \boldsymbol{\psi}_0, \quad (24)$$

where \mathbf{T}_{ij} is given by the scattering series as follows

$$\begin{aligned} \mathbf{T}_{ij}(\mathbf{R}_1, \dots, \mathbf{R}_N) = & \delta_{ij} \mathbf{M}(\mathbf{R}_i) + (1 - \delta_{ij}) \mathbf{M}(\mathbf{R}_i) \mathbf{GM}(\mathbf{R}_j) \\ & + \sum_k^l \mathbf{M}(\mathbf{R}_i) \mathbf{GM}(\mathbf{R}_k) \mathbf{GM}(\mathbf{R}_j) + \dots \end{aligned} \quad (25)$$

It is worth comparing the scattering series given by expression (25) to the scattering series

$$\delta_{ij} \mathbf{M}_0(\mathbf{R}_i) + (1 - \delta_{ij}) \mathbf{M}_<(\mathbf{R}_i) \mathbf{GM}_>(\mathbf{R}_j) + \sum_k^l \mathbf{M}_<(\mathbf{R}_i) \hat{\mathbf{M}}(\mathbf{R}_k) \mathbf{GM}_>(\mathbf{R}_j) + \dots \quad (26)$$

which is considered in the literature [12]. Notice that here four types of single-particle operators \mathbf{M}_0 , $\mathbf{M}_<$, $\mathbf{M}_>$, and $\hat{\mathbf{M}}$ appear. With respect to the number of types of single-particle operators, the formulation of the scattering series (25) introduced in the present paper is simpler than the series given by expression (26).

It is worth mentioning that no approximation was made in the above analysis. In particular, equation (19) is a proper starting point to analyze hydrodynamic interactions of particles in close contact.

5. Discussion

In the present paper the scattering series for the mobility problem has been reformulated, which results in the simple form given by equation (25). The simplification relies on the fact that in expression (25) there is only one type of single-particle operator, \mathbf{M} . In the formulation hitherto used in the literature [12], in the scattering series there were four types of single-particle operator \mathbf{M}_0 , $\mathbf{M}_<$, $\mathbf{M}_>$, and $\hat{\mathbf{M}}$, as shown by the expression (26).

At first sight the difference between both formulations may not seem to be significant. However, the mobility problem plays a crucial role in, for example, calculations of transport coefficients of suspensions. In this context statistical physics considerations contain many different formulas of the same structure [12]–[14], [16]. A striking example of cumbersomeness following from a lack of a simple formulation of the mobility problem is the Beenakker–Mazur method [17]–[19], which is used to calculate short-time dynamic properties of suspensions. A multitude of expressions occurring in cited articles obscures the essence of the method, which, on the other hand, is the most comprehensive analytical scheme available so far [36]. Using the reformulated scattering sequence the Beenakker–Mazur method can be presented in a simple form, as we will show in another paper [37]. The simple formulation of the mobility problem also allows one to carry out advanced analysis of scattering series in a clear and simple way. In this context the formulation will also be used in our subsequent work on macroscopic characteristics of suspensions.

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Appendix A. Single-particle operators and multiple picture

Here we give the explicit form of the \mathbf{Z}_0 and $\boldsymbol{\mu}_0$ operators. Based on [25] the \mathbf{Z}_0 operator can be expressed by the following formula

$$\begin{aligned} \mathbf{Z}_0(\mathbf{R}, \mathbf{r}, \mathbf{r}') &= \sum_{l,l'=1}^{\infty} \sum_{m'=-l'}^{l'} \sum_{m=-l}^l \sum_{\sigma,\sigma'=0}^2 \delta_a(\mathbf{r} - \mathbf{R}) \mathbf{w}_{lm\sigma}^+(\mathbf{r} - \mathbf{R}) \\ &\times [Z_0(\mathbf{R})]_{lm\sigma,l'm'\sigma'} \delta_a(\mathbf{r}' - \mathbf{R}) \mathbf{w}_{l'm'\sigma'}^{+*}(\mathbf{r}' - \mathbf{R}), \end{aligned} \quad (\text{A.1})$$

where $[Z_0(\mathbf{R})]_{lm\sigma,l'm'\sigma'}$ stands for the multipole matrix with indices $l = 1, \dots, \infty$; $m = -l, \dots, l$; $\sigma = 0, 1, 2$. Its matrix elements are explicitly given e.g. in [38]. A set of multipole functions $\mathbf{v}_{lm\sigma}^+(\mathbf{r})$ and $\mathbf{w}_{lm\sigma}^+(\mathbf{r})$ is defined e.g. in [39] or [25]. Every solution of the homogeneous Stokes equations may be expressed as a combination of the multipole functions $\mathbf{v}_{lm\sigma}^+(\mathbf{r})$

$$\mathbf{v}_0(\mathbf{r}) = \sum_{l=1}^{\infty} \sum_{m=-l}^l \sum_{\sigma=0}^2 [v_0(\mathbf{R})]_{lm\sigma} \mathbf{v}_{lm\sigma}^+(\mathbf{r} - \mathbf{R}), \quad (\text{A.2})$$

whereas the induced surface force $\mathbf{f}_i(\mathbf{r})$ is a combination of multipole functions $\mathbf{w}_{lm\sigma}^+(\mathbf{r})$:

$$\mathbf{f}_i(\mathbf{r}) = \sum_{l=1}^{\infty} \sum_{m=-l}^l \sum_{\sigma=0}^2 [f_i]_{lm\sigma} \delta_a(\mathbf{r} - \mathbf{R}_i) \mathbf{w}_{lm\sigma}^+(\mathbf{r} - \mathbf{R}_i). \quad (\text{A.3})$$

The multipole functions $\mathbf{w}_{lm\sigma}^+(\mathbf{r})$ are defined as orthonormal to the $\mathbf{v}_{lm\sigma}^+(\mathbf{r})$ functions. This orthonormality is expressed in the following way

$$\langle \delta_a \mathbf{w}_{lm\sigma}^+ | \mathbf{v}_{l'm'\sigma'}^+ \rangle = \delta_{ll'} \delta_{mm'} \delta_{\sigma\sigma'}, \quad (\text{A.4})$$

with the Dirac notation [40] for the scalar product of two vector fields $\mathbf{A}(\mathbf{r})$ and $\mathbf{B}(\mathbf{r})$:

$$\langle \mathbf{A} | \mathbf{B} \rangle = \int d^3\mathbf{r} \mathbf{A}^*(\mathbf{r}) \cdot \mathbf{B}(\mathbf{r}), \quad (\text{A.5})$$

and a scalar function $\delta_a(\mathbf{r})$ of the form

$$\delta_a(\mathbf{r}) = a^{-1} \delta(|\mathbf{r}| - a)$$

which confines the integration area to a sphere of radius a .

Operator $\boldsymbol{\mu}_0$ is given by the expression

$$\begin{aligned} \boldsymbol{\mu}_0(\mathbf{R}, \mathbf{r}, \mathbf{r}') &= \sum_{l,l'=1}^{\infty} \sum_{m'=-l'}^{l'} \sum_{m=-l}^l \sum_{\sigma,\sigma'=0}^2 \Theta_a(\mathbf{r} - \mathbf{R}) \mathbf{v}_{lm\sigma}^+(\mathbf{r} - \mathbf{R}) \\ &\times [\boldsymbol{\mu}_0(\mathbf{R})]_{lm\sigma,l'm'\sigma'} \Theta_a(\mathbf{r}' - \mathbf{R}) \mathbf{v}_{l'm'\sigma'}^{+*}(\mathbf{r}' - \mathbf{R}) \end{aligned} \quad (\text{A.6})$$

with the multipole matrix $[\boldsymbol{\mu}_0(\mathbf{R})]_{lm\sigma,l'm'\sigma'}$ explicitly given in [38]. Here $\Theta_a(\mathbf{r} - \mathbf{R})$ is a characteristic function of the particle at position \mathbf{R} : it equals 0 whenever \mathbf{r} points outside the particle, and equals 1 otherwise.

Finally, we represent equations (17) and (18) in the multipole expansion formalism. To pass on to the multipole picture we put the expressions (A.1) and (A.6) into these equations and multiply them by $\langle \mathbf{w}_{lm\sigma}^+(i) \delta_a(i) |$ and $\langle \mathbf{v}_{lm\sigma}^+(i) \delta_a(i) |$ respectively from the left side. Simple algebra yields

$$U_i = \mu_0(i) f_{\text{ext}}(i) + \mu_0(i) Z_0(i) \left(v_0(i) + \sum_{j \neq i} G_0(ij) f_j \right), \quad (\text{A.7a})$$

$$f_i = Z_0(i) \mu_0(i) f_{\text{ext}}(i) - \hat{Z}_0(i) \left(v_0(i) + \sum_{j \neq i} G_0(ij) f_j \right), \quad (\text{A.7b})$$

where the multipole vector of the ambient flow at point \mathbf{R}

$$[v_0(\mathbf{R})]_{lm\sigma} = \langle \mathbf{w}_{lm\sigma}^+(\mathbf{R}) \delta_a(\mathbf{R}) | \mathbf{v}_0 \rangle, \quad (\text{A.8})$$

multipole velocity field U_i for the i th particle

$$[U_i]_{lm\sigma} = \langle \mathbf{w}_{lm\sigma}^+(i) \delta_a(i) | \mathbf{U}_i \rangle, \quad (\text{A.9})$$

induced surface force multipole f_i for the i th particle

$$[f_i]_{lm\sigma} = \langle \mathbf{v}_{lm\sigma}^+(i) | \mathbf{F}_i \rangle, \quad (\text{A.10})$$

and external force multipole field $f_{\text{ext}}(\mathbf{R})$ at point \mathbf{R}

$$[f_{\text{ext}}(\mathbf{R})]_{lm\sigma} = \langle \mathbf{v}_{lm\sigma}^+(\mathbf{R}) \Theta_a(\mathbf{R}) | \mathbf{F}_{\text{ext}} \rangle. \quad (\text{A.11})$$

In the above formulas $|\mathbf{A}(i)\rangle$ or $|\mathbf{A}(\mathbf{R})\rangle$ denote vector fields $\mathbf{A}(\mathbf{r} - \mathbf{R}_i)$ or $\mathbf{A}(\mathbf{r} - \mathbf{R})$ respectively. Moreover matrix $G_0(\mathbf{R}, \mathbf{R}')$ is defined with the formula

$$[G_0(\mathbf{R}, \mathbf{R}')]_{lm\sigma,l'm'\sigma'} = \langle \mathbf{w}_{lm\sigma}^+(\mathbf{R}) \delta_a(\mathbf{R}) | \mathbf{G}_0 | \mathbf{w}_{l'm'\sigma'}^+(\mathbf{R}') \delta_a(\mathbf{R}') \rangle. \quad (\text{A.12})$$

Its matrix elements may be found in [38, 41]. It is worth mentioning that $[G_0(\mathbf{R}, \mathbf{R}')]_{lm\sigma,l'm'\sigma'}$ depends on the difference of positions $\mathbf{R} - \mathbf{R}'$ and for nonoverlapping configurations, i.e. $|\mathbf{R} - \mathbf{R}'| \geq 2a$, it scales as $1/|\mathbf{R} - \mathbf{R}'|^{l+l'+\sigma+\sigma'-1}$. To obtain equations (A.7) we also used the following definition [16]

$$\hat{Z}_0(i) = Z_0(i) - Z_0(i) \mu_0(i) Z_0(i). \quad (\text{A.13})$$

In the multipole formalism the integral equations (A.7) may be easily reformulated into equation

$$s_i = M(\mathbf{R}_i) \left(\psi_0(\mathbf{R}_i) + \sum_{j \neq i} G(\mathbf{R}_i, \mathbf{R}_j) s_j \right) \quad (\text{A.14})$$

in the same way as equations (17) and (18) were transformed into equation (19). Moreover the definitions of s_i , ψ_0 and M are similar to the definitions (20)–(22):

$$s_i = \begin{bmatrix} U_i \\ F_i \end{bmatrix}, \quad (\text{A.15})$$

$$\psi_0(\mathbf{R}) = \begin{bmatrix} f_{\text{ext}}(\mathbf{R}) \\ v_0(\mathbf{R}) \end{bmatrix}, \quad (\text{A.16})$$

$$M(i) = \begin{bmatrix} \mu_0(i) & \mu_0(i) Z_0(i) \\ Z_0(i) \mu_0(i) & -\hat{Z}_0(i) \end{bmatrix} \quad (\text{A.17})$$

and the Green function G in extended multipole space has the following form:

$$G = \begin{bmatrix} 0 & 0 \\ 0 & G_0 \end{bmatrix}. \quad (\text{A.18})$$

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