J. Fluid Mech. (2013), *vol.* 731, R3, doi:10.1017/jfm.2013.402

JFM RAPIDS
journals.cambridge.org/rapids

Generalization of the Rotne–Prager–Yamakawa mobility and shear disturbance tensors

Eligiusz Wajnryb[1], Krzysztof A. Mizerski[2,†], Pawel J. Zuk[3] and Piotr Szymczak[3]

[1]Department of Mechanics and Physics of Fluids, Institute of Fundamental and Technological Research, Polish Academy of Sciences, Pawinskiego 5B, 02-106, Warsaw, Poland

[2]Department of Magnetism, Institute of Geophysics, Polish Academy of Sciences, ul. Ksiecia Janusza 64, 01-452 Warsaw, Poland

[3]Institute of Theoretical Physics, Faculty of Physics, University of Warsaw, Hoza 69, 00-681, Warsaw, Poland

(Received 12 June 2013; revised 22 July 2013; accepted 30 July 2013)

The Rotne–Prager–Yamakawa approximation is one of the most commonly used methods of including hydrodynamic interactions in modelling of colloidal suspensions and polymer solutions. The two main merits of this approximation are that it includes all long-range terms (i.e. decaying as R^{-3} or slower in interparticle distances) and that the diffusion matrix is positive definite, which is essential for Brownian dynamics modelling. Here, we extend the Rotne–Prager–Yamakawa approach to include both translational and rotational degrees of freedom, and derive the regularizing corrections to account for overlapping particles. Additionally, we show how the Rotne–Prager–Yamakawa approximation can be generalized for other geometries and boundary conditions.

Key words: computational methods, low-Reynolds-number flows, suspensions

1. Introduction

Particles moving in a viscous fluid induce a local flow field that affects other particles. These long-range, many-body interactions, mediated by the solvent are commonly called ‘hydrodynamic interactions’ (HI). The presence of HI is known to affect the dynamic properties of soft matter: they modify the values of diffusion coefficients in colloidal suspensions (Dhont 1996), affect the characteristics of the coil–stretch transition in polymers (Larson & Magda 1989), change the kinetic pathways of phase separation in binary mixtures (Tanaka 2001), alter the kinetics

† Email address for correspondence: krzysztof.mizerski@gmail.com

of macromolecule adsorption on surfaces (Wojtaszczyk & Avalos 1998) or cause the polymer migration in microchannels (Usta, Butler & Ladd 2007). They are also important in the dynamics of biological soft matter, such as DNA (Shaqfeh 2005), proteins (Frembgen-Kesner & Elcock 2009; Szymczak & Cieplak 2011) or lipid membranes (Ando & Skolnick 2013).

A proper account of hydrodynamic interactions is thus essential in simulation studies of soft matter flow. Unfortunately, HI depends in a complicated nonlinear way on the instantaneous positions of all particles in the system. For a system of spheres, exact explicit expressions for the hydrodynamic interaction tensors exist in the form of the power series in interparticle distances, which may be incorporated into the simulation scheme (Mazur & van Saarloos 1982; Brady & Bossis 1988; Felderhof 1988; Kim & Karrila 1991; Cichocki *et al.* 1994). These are however relatively expensive numerically, thus various approximations are resorted to in order to make the computations more tractable. The simplest one is based on the Oseen tensor, which assumes that the particles can be regarded as point force sources in the fluid. However, the diffusion matrix constructed in this way is not suitable for Brownian dynamics simulations, because it becomes non-positive definite when separations between the particles become small. This is not only unphysical (since the positivity of diffusion is a consequence of the second law of thermodynamics) but also leads to numerical problems in the Brownian dynamics simulations, where a square-root of the diffusion matrix is needed. Another commonly used approximation is the Rotne–Prager–Yamakawa (RPY) tensor (Rotne & Prager 1969; Yamakawa 1970), which takes into account all the HI terms up to $O(a/r_{ij})^3$ in the expansion in the inverse distance between the particles (where a is the particle radius). Nevertheless, if the particles overlap, $r_{ij} < 2a$, the RPY tensor again loses its positive definiteness. To avoid this, a regularization for $r_{ij} < 2a$ has been proposed by Rotne & Prager (1969), which is not singular at $r_{ij} = 0$ and is positive definite for all the particle configurations. The RPY tensor with this regularization is by far the most popular method of accounting for HI in soft matter modelling (Nägele 2006).

The present paper takes a close look at the Rotne–Prager–Yamakawa approximation and generalizes it in a number of ways. First, we re-derive the original RPY tensor using direct integration of force densities over the sphere surfaces. When the spheres overlap then this method gives us automatically the regularization correction. In this way we derive the RPY regularizations not only for the translational degrees of freedom (already obtained by Rotne & Prager 1969) but also for rotational degrees of freedom, as well as for the shear disturbance matrix $\boldsymbol{C}$ – another hydrodynamic tensor, which gives the response of the particles to the external shear flow. The mobility evaluated using our technique may be applied to calculate the diffusion tensor of complex molecules (de la Torre, del Rio Echenique & Ortega 2007; Adamczyk *et al.* 2012) using bead models which include overlapping spheres. Finally, we show how these results can be generalized for other boundary conditions and corresponding propagators.

2. The mobility problem under shear flow

We consider a suspension of N identical spherical particles of radius a, in an incompressible fluid of viscosity η at a low Reynolds number. The particles are immersed in a linear shear flow

$$\boldsymbol{v}_\infty(\boldsymbol{r}) = \boldsymbol{K}_\infty \cdot \boldsymbol{r}, \tag{2.1}$$

where $\boldsymbol{K}_\infty$ is the constant velocity gradient matrix, e.g. for a simple shear flow

$$\boldsymbol{K}_\infty = \begin{bmatrix} 0 & 0 & \dot{\gamma} \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \dot{\gamma} = \text{const.} \tag{2.2}$$

Due to the linearity of the Stokes equations, the forces and torques exerted by the fluid on the particles ($\boldsymbol{\mathcal{F}}_j$ and $\boldsymbol{\mathcal{T}}_j$) depend linearly on the translational and rotational velocities of the particles ($\boldsymbol{U}_i$, $\boldsymbol{\Omega}_i$). This relation defines the generalized friction matrix $\boldsymbol{\zeta}$:

$$\begin{pmatrix} \boldsymbol{\mathcal{F}}_j \\ \boldsymbol{\mathcal{T}}_j \end{pmatrix} = -\sum_i \begin{pmatrix} \boldsymbol{\zeta}^{tt}_{ji} & \boldsymbol{\zeta}^{tr}_{ji} & \boldsymbol{\zeta}^{td}_{ji} \\ \boldsymbol{\zeta}^{rt}_{ji} & \boldsymbol{\zeta}^{rr}_{ji} & \boldsymbol{\zeta}^{rd}_{ji} \end{pmatrix} \cdot \begin{pmatrix} \boldsymbol{v}_\infty(\boldsymbol{R}_i) - \boldsymbol{U}_i \\ \boldsymbol{\omega}_\infty(\boldsymbol{R}_i) - \boldsymbol{\Omega}_i \\ \boldsymbol{E}_\infty \end{pmatrix}, \tag{2.3}$$

where $\boldsymbol{\zeta}^{pq}$ (with $p = t, r$ and $q = t, r, d$) are the Cartesian tensors and the superscripts t, r and d correspond to the translational, rotational and dipolar components, respectively. The tensor $\boldsymbol{E}_\infty$ is the symmetric part of $\boldsymbol{K}_\infty$ in (2.2) and $\boldsymbol{\omega}_\infty = (\nabla \times \boldsymbol{v}_\infty(\boldsymbol{R}_i))/2 = \boldsymbol{\epsilon}/2 : \boldsymbol{K}_\infty$ is the vorticity of the incident flow. Finally $\boldsymbol{R}_i$ corresponds to the position of particle i. The reciprocal relation giving velocities of particles moving under external forces/torques in the external flow $\boldsymbol{v}_\infty$ is determined by the generalized mobility matrix $\boldsymbol{\mu}$ written, after Dhont (1996),

$$\begin{pmatrix} \boldsymbol{U}_i \\ \boldsymbol{\Omega}_i \end{pmatrix} = \begin{pmatrix} \boldsymbol{v}_\infty(\boldsymbol{R}_i) \\ \boldsymbol{\omega}_\infty(\boldsymbol{R}_i) \end{pmatrix} + \sum_j \left[\begin{pmatrix} \boldsymbol{\mu}^{tt}_{ij} & \boldsymbol{\mu}^{tr}_{ij} \\ \boldsymbol{\mu}^{rt}_{ij} & \boldsymbol{\mu}^{rr}_{ij} \end{pmatrix} \cdot \begin{pmatrix} \boldsymbol{\mathcal{F}}_j \\ \boldsymbol{\mathcal{T}}_j \end{pmatrix} \right] + \begin{pmatrix} \boldsymbol{C}^t_i \\ \boldsymbol{C}^r_i \end{pmatrix} : \boldsymbol{E}_\infty, \tag{2.4}$$

where the elements of the shear disturbance tensor $\boldsymbol{C}$ are defined as

$$\boldsymbol{C}^t_i = \sum_j \boldsymbol{\mu}^{td}_{ij}, \quad \boldsymbol{C}^r_i = \sum_j \boldsymbol{\mu}^{rd}_{ij}. \tag{2.5a}$$

In the case of a single particle the mobility matrices reduce to

$$\boldsymbol{\mu}^{tt}_{ii} = \frac{1}{\zeta^{tt}}\mathbf{1}, \quad \boldsymbol{\mu}^{rr}_{ii} = \frac{1}{\zeta^{rr}}\mathbf{1}, \quad \boldsymbol{\mu}^{tr}_{ii} = \boldsymbol{\mu}^{rt}_{ii} = 0, \tag{2.6}$$

where the friction coefficients for a spherical particle are given by $\zeta^{tt} = 6\pi\eta a$ and $\zeta^{rr} = 8\pi\eta a^3$.

Finding the mobility matrix (or the associated diffusion matrix, $\boldsymbol{D} = k_B T \boldsymbol{\mu}$, where k_B is the Boltzmann constant and T is the temperature) is the problem of a fundamental importance in constructing the numerical algorithms for tracking the motion of the particles in viscous fluid. The two main numerical methods used for this purpose are the *Stokesian Dynamics*, which corresponds to the numerical integration of (2.4) and the *Brownian dynamics*, used whenever the Brownian motion of the particles cannot be neglected (Nägele 2006). In the latter, the random displacements of the particles, $\boldsymbol{\Gamma}_i(\Delta t)$ need to be added to the deterministic displacements governed by (2.4). The fluctuation–dissipation theorem implies that the covariance of $\boldsymbol{\Gamma}$ is connected to the mobility matrix, e.g. for the translational displacements

$$\left\langle \boldsymbol{\Gamma}_i(\Delta t)\, \boldsymbol{\Gamma}_j(\Delta t) \right\rangle = 2 k_B T \boldsymbol{\mu}^{tt}_{ij} \Delta t. \tag{2.7}$$

Hence the calculation of $\boldsymbol{\Gamma}_i(\Delta t)$ requires finding a matrix $\boldsymbol{d}$ such that $\boldsymbol{\mu}^{tt} = \boldsymbol{d}\boldsymbol{d}^{\mathrm{T}}$. This is possible only when the mobility matrix is positively defined. Any valid approximation scheme for the hydrodynamic interactions should then not only correctly reproduce the particle mobilities but also guarantee the positive definiteness of the mobility tensors.

3. The Rotne–Prager–Yamakawa form of $\boldsymbol{\mu}$ and $\boldsymbol{C}$ for systems with shear

In principle the HI tensors can be calculated with arbitrary precision, following e.g. the multipole expansion or boundary integral method (Kim & Karrila 1991; Pozrikidis 1992). In practice, however, the exact approach turns out to be too demanding computationally, so various approximation procedures have to be resorted to. The most commonly used is the Rotne–Prager–Yamakawa approximation (Rotne & Prager 1969; Yamakawa 1970), based on the following idea: when a force (or torque) is applied to particle i, that particle begins to move, inducing flow in the bulk of the fluid. The extent to which this additional flow affects translational and rotational velocities of another particle (j) is then calculated using Faxen's laws (Kim & Karrila 1991). In that way one neglects not only the multi-body effects (involving three and more particles) but also the higher-order terms in two-particle interactions (e.g. we do not consider the impact of the movement of particle j back on particle i). Below, we follow this procedure to derive in a systematic way hydrodynamic tensors for both translational and rotational degrees of freedom.

3.1. *The mobility matrix* $\boldsymbol{\mu}$

The Stokes flow generated by a point force in the unbounded space is given by the Oseen tensor (Kim & Karrila 1991)

$$\boldsymbol{T}_0(\boldsymbol{r}) = \frac{1}{8\pi\eta r}\left(\mathbf{1} + \hat{\boldsymbol{r}}\hat{\boldsymbol{r}}\right). \tag{3.1}$$

Since $\boldsymbol{T}_0(\boldsymbol{r})$ is a Green function for Stokes equations, one can use it to calculate the translational $\boldsymbol{v}_0^t(\boldsymbol{r})$ and rotational $\boldsymbol{v}_0^r(\boldsymbol{r})$ flows generated by a sphere situated at $\boldsymbol{R}_j$, to which we apply force $\boldsymbol{\mathcal{F}}$ and/or torque $\boldsymbol{\mathcal{T}}$:

$$\begin{aligned}\boldsymbol{v}_0^t(\boldsymbol{r}) &= \int_{S_j} \boldsymbol{T}_0\left(\boldsymbol{r}-\boldsymbol{r}'\right)\cdot\frac{\boldsymbol{\mathcal{F}}}{4\pi a^2}\,\mathrm{d}\sigma' \\ &= \begin{cases}\left(1+\dfrac{a^2}{6}\nabla^2\right)\boldsymbol{T}_0\left(\boldsymbol{\rho}_j\right)\cdot\boldsymbol{\mathcal{F}} = \dfrac{1}{8\pi\eta\rho_j}\left[\left(1+\dfrac{a^2}{3\rho_j^2}\right)\mathbf{1}\right. \\ \qquad\left.+\left(1-\dfrac{a^2}{\rho_j^2}\right)\hat{\boldsymbol{\rho}}_j\hat{\boldsymbol{\rho}}_j\right]\cdot\boldsymbol{\mathcal{F}}, \quad \rho_j > a, \\ \dfrac{1}{\zeta^{tt}}\boldsymbol{\mathcal{F}}, \quad \rho_j \leqslant a,\end{cases}\end{aligned} \tag{3.2}$$

$$\begin{aligned}\boldsymbol{v}_0^r(\boldsymbol{r}) &= \int_{S_j} \boldsymbol{T}_0\left(\boldsymbol{r}-\boldsymbol{r}'\right)\cdot\frac{3}{8\pi a^3}\boldsymbol{\mathcal{T}}\times\boldsymbol{n}'\,\mathrm{d}\sigma' \\ &= \begin{cases}\dfrac{1}{2}\nabla\times\boldsymbol{T}_0\left(\boldsymbol{\rho}_j\right)\cdot\boldsymbol{\mathcal{T}} = \dfrac{1}{8\pi\eta\rho_j^3}\boldsymbol{\mathcal{T}}\times\boldsymbol{\rho}_j, & \rho_j > a, \\ \dfrac{1}{\zeta^{rr}}\boldsymbol{\mathcal{T}}\times\boldsymbol{\rho}_j, & \rho_j \leqslant a,\end{cases}\end{aligned} \tag{3.3}$$

where $\boldsymbol{\rho}_j = \boldsymbol{r} - \boldsymbol{R}_j$ is the distance from the sphere centre, $\boldsymbol{r}'$ denotes the integration variable, $\boldsymbol{n}'$ is the unit normal vector to the sphere at point $\boldsymbol{r}'$ and $\int_{S_j}$ denotes an integral over the surface of the sphere situated at $\boldsymbol{R}_j$. The curl of a tensor is defined in the following way:

$$(\nabla \times \boldsymbol{T})_{\alpha\beta} = \epsilon_{\alpha\gamma\zeta}\partial_\gamma \boldsymbol{T}_{\zeta\beta} \tag{3.4}$$

where the Greek letters denote the Cartesian components.

The Faxen laws (Kim & Karrila 1991) allow the velocity $\boldsymbol{U}_i$ and angular velocity $\boldsymbol{\Omega}_i$ of a sphere i immersed in an external flow $\boldsymbol{v}_0$, placed at $\boldsymbol{R}_i$ to be expressed as

$$\boldsymbol{U}_i = \frac{1}{4\pi a^2}\int_{S_i} \boldsymbol{v}_0\left(\boldsymbol{r}'\right)\,\mathrm{d}\sigma' = \left(\boldsymbol{1} + \frac{a^2}{6}\nabla^2\right)\boldsymbol{v}_0\Big|_{\boldsymbol{r}=\boldsymbol{R}_i}, \tag{3.5}$$

$$\boldsymbol{\Omega}_i = \frac{3}{8\pi a^3}\int_{S_i} \boldsymbol{n}' \times \boldsymbol{v}_0\left(\boldsymbol{r}'\right)\,\mathrm{d}\sigma' = \frac{1}{2}\nabla \times \boldsymbol{v}_0\Big|_{\boldsymbol{r}=\boldsymbol{R}_i}, \tag{3.6}$$

where the integration is performed over the sphere surface S_i. Thus substituting (3.2) and (3.3) into (3.6) we obtain the contribution to velocity $\boldsymbol{U}'_i$ and angular velocity $\boldsymbol{\Omega}'_i$ of a sphere i due to the force/torque acting on a sphere j

$$\begin{aligned}\boldsymbol{U}'_i &= \frac{1}{4\pi a^2}\int_{S_i} \boldsymbol{v}_0^t\left(\boldsymbol{r}'\right)\,\mathrm{d}\sigma' + \frac{1}{4\pi a^2}\int_{S_i} \boldsymbol{v}_0^r\left(\boldsymbol{r}'\right)\,\mathrm{d}\sigma' \\ &= \frac{1}{4\pi a^2}\int_{S_i}\mathrm{d}\sigma'\int_{S_j}\mathrm{d}\sigma''\boldsymbol{T}_0\left(\boldsymbol{r}'-\boldsymbol{r}''\right)\cdot\left[\frac{\boldsymbol{\mathcal{F}}}{4\pi a^2} + \frac{3}{8\pi a^3}\boldsymbol{\mathcal{T}}\times\boldsymbol{n}''\right],\end{aligned} \tag{3.7}$$

$$\begin{aligned}\boldsymbol{\Omega}'_i &= \frac{3}{8\pi a^3}\int_{S_i} \boldsymbol{n}'\times\boldsymbol{v}_0^t\left(\boldsymbol{r}'\right)\,\mathrm{d}\sigma' + \frac{3}{8\pi a^3}\int_{S_i} \boldsymbol{n}'\times\boldsymbol{v}_0^r\left(\boldsymbol{r}'\right)\,\mathrm{d}\sigma' \\ &= \frac{3}{8\pi a^3}\int_{S_i}\mathrm{d}\sigma'\int_{S_j}\mathrm{d}\sigma''\boldsymbol{n}'\times\boldsymbol{T}_0\left(\boldsymbol{r}'-\boldsymbol{r}''\right)\cdot\left[\frac{\boldsymbol{\mathcal{F}}}{4\pi a^2} + \frac{3}{8\pi a^3}\boldsymbol{\mathcal{T}}\times\boldsymbol{n}''\right].\end{aligned} \tag{3.8}$$

At this stage let us introduce the tensors

$$\boldsymbol{w}_i^t(\boldsymbol{r}) = \frac{1}{4\pi a^2}\boldsymbol{1}\delta\left(\rho_j - a\right), \quad \boldsymbol{w}_i^r(\boldsymbol{r}) = \frac{3}{8\pi a^3}\boldsymbol{\epsilon}\cdot\hat{\boldsymbol{\rho}}_j\delta\left(\rho_j - a\right), \tag{3.9}$$

where $(\boldsymbol{\epsilon}\cdot\hat{\boldsymbol{\rho}}_j)_{\alpha\beta} = \epsilon_{\alpha\beta\gamma}\hat{\rho}_{j\gamma}$. Above tensors multiplied by force $\boldsymbol{w}^t\cdot\boldsymbol{\mathcal{F}}$ and torque $\boldsymbol{w}^r\cdot\boldsymbol{\mathcal{T}}$ have the interpretation of the force densities on the surface of the sphere due to the force and torque acting on the sphere. We can now write down the following general formulae for the mobility matrix:

$$\boldsymbol{\mu}_{ij}^{tt} = \langle\boldsymbol{w}_i^t|\boldsymbol{T}_0|\boldsymbol{w}_j^t\rangle, \quad \boldsymbol{\mu}_{ij}^{rr} = \langle\boldsymbol{w}_i^r|\boldsymbol{T}_0|\boldsymbol{w}_j^r\rangle, \quad \boldsymbol{\mu}_{ij}^{rt} = \langle\boldsymbol{w}_i^r|\boldsymbol{T}_0|\boldsymbol{w}_j^t\rangle, \quad \boldsymbol{\mu}_{ij}^{tr} = \langle\boldsymbol{w}_i^t|\boldsymbol{T}_0|\boldsymbol{w}_j^r\rangle, \tag{3.10}$$

where we use the bra-ket notation defined in the following way:

$$\boldsymbol{\mu}_{ij}^{pq} = \left\langle\boldsymbol{w}_i^p|\boldsymbol{T}_0|\boldsymbol{w}_j^q\right\rangle = \int \mathrm{d}\boldsymbol{r}'\int \mathrm{d}\boldsymbol{r}''\left[\boldsymbol{w}_i^p\left(\boldsymbol{r}'\right)\right]^{\mathrm{T}}\cdot\boldsymbol{T}_0\left(\boldsymbol{r}'-\boldsymbol{r}''\right)\cdot\boldsymbol{w}_j^q\left(\boldsymbol{r}''\right), \tag{3.11}$$

with $p, q = r, t$ and T denoting tensor transposition. The method of calculation of the integrals in (3.7)–(3.8) is presented in the supplementary material available at http://dx.doi.org/10.1017/jfm.2013.402. Here, we simply quote the final results denoting $\boldsymbol{R}_{ij} = \boldsymbol{R}_i - \boldsymbol{R}_j$. Fortunately there is no need to integrate explicitly for non-overlapping spheres. For the translational–translational mobility, we get

$$\boldsymbol{\mu}_{ij}^{tt} = \begin{cases} \left(\mathbf{1} + \dfrac{a^2}{3}\nabla^2\right)\boldsymbol{T}_0\left(\boldsymbol{R}_{ij}\right) = \dfrac{1}{8\pi\eta R_{ij}}\left[\left(1 + \dfrac{2a^2}{3R_{ij}^2}\right)\mathbf{1} + \left(1 - \dfrac{2a^2}{R_{ij}^2}\right)\hat{\boldsymbol{R}}_{ij}\hat{\boldsymbol{R}}_{ij}\right], & R_{ij} > 2a, \\ \dfrac{1}{\zeta^{tt}}\left[\left(1 - \dfrac{9R_{ij}}{32a}\right)\mathbf{1} + \dfrac{3R_{ij}}{32a}\hat{\boldsymbol{R}}_{ij}\hat{\boldsymbol{R}}_{ij}\right], & R_{ij} \leqslant 2a, \end{cases} \tag{3.12}$$

which, in the limit of $R_{ij} \to 0$, yields the self-mobility

$$\boldsymbol{\mu}_{ii}^{tt} = \boldsymbol{\mu}_{jj}^{tt} = \lim_{R_{ij}\to 0} \boldsymbol{\mu}_{ij}^{tt} = \frac{1}{\zeta^{tt}}\mathbf{1}. \tag{3.13}$$

Next, for the rotational degrees of freedom

$$\boldsymbol{\mu}_{ij}^{rr} = \begin{cases} -\dfrac{1}{4}\nabla^2\boldsymbol{T}_0\left(\boldsymbol{R}_{ij}\right) = -\dfrac{1}{16\pi\eta R_{ij}^3}\left(\mathbf{1} - 3\hat{\boldsymbol{R}}_{ij}\hat{\boldsymbol{R}}_{ij}\right), & R_{ij} > 2a, \\ \dfrac{1}{\zeta^{rr}}\left[\left(1 - \dfrac{27}{32}\dfrac{R_{ij}}{a} + \dfrac{5}{64}\dfrac{R_{ij}^3}{a^3}\right)\mathbf{1} + \left(\dfrac{9}{32}\dfrac{R_{ij}}{a} - \dfrac{3}{64}\dfrac{R_{ij}^3}{a^3}\right)\hat{\boldsymbol{R}}_{ij}\hat{\boldsymbol{R}}_{ij}\right], & R_{ij} \leqslant 2a, \end{cases} \tag{3.14}$$

with the self-mobility given by

$$\boldsymbol{\mu}_{ii}^{rr} = \boldsymbol{\mu}_{jj}^{rr} = \lim_{R_{ij}\to 0} \boldsymbol{\mu}_{ij}^{rr} = \frac{1}{\zeta^{rr}}\mathbf{1}. \tag{3.15}$$

Finally, the translational–rotational mobility is described by the following tensor:

$$\boldsymbol{\mu}_{ij}^{rt} = \left[\boldsymbol{\mu}_{ij}^{tr}\right]^{\mathrm{T}} = \begin{cases} \dfrac{1}{2}\nabla\times\left(\mathbf{1} + \dfrac{a^2}{6}\nabla^2\right)\boldsymbol{T}_0\left(\boldsymbol{R}_{ij}\right) = \dfrac{1}{2}\nabla\times\boldsymbol{T}_0\left(\boldsymbol{R}_{ij}\right) = \dfrac{1}{8\pi\eta R_{ij}^2}\boldsymbol{\epsilon}\cdot\hat{\boldsymbol{R}}_{ij}, & R_{ij} > 2a, \\ \dfrac{1}{16\pi\eta a^2}\left(\dfrac{R_{ij}}{a} - \dfrac{3}{8}\dfrac{R_{ij}^2}{a^2}\right)\boldsymbol{\epsilon}\cdot\hat{\boldsymbol{R}}_{ij}, & R_{ij} \leqslant 2a, \end{cases} \tag{3.16}$$

with

$$\boldsymbol{\mu}_{ii}^{tr} = \boldsymbol{\mu}_{jj}^{tr} = \boldsymbol{\mu}_{ii}^{rt} = \boldsymbol{\mu}_{jj}^{rt} = \lim_{R_{ij}\to 0} \boldsymbol{\mu}_{ij}^{tr} = \lim_{R_{ij}\to 0} \boldsymbol{\mu}_{ij}^{rt} = \mathbf{0}. \tag{3.17}$$

Note that the formulae (3.12) for the translational mobility matrix, both for $R_{ij} > 2a$ and for $R_{ij} < 2a$ were derived earlier by Rotne & Prager (1969) and Yamakawa (1970) and are known as Rotne–Prager–Yamakawa mobility approximation. The expressions for the other components of the mobility matrix $\boldsymbol{\mu}_{ij}^{rr}$ and $\boldsymbol{\mu}_{ij}^{tr}$ are also known (Kim & Karrila 1991; Dhont 1996; Reichert 2006; de la Torre *et al.* 2007) but only for $R_{ij} > 2a$. However, to our knowledge, the regularizing corrections for $\boldsymbol{\mu}_{ij}^{rr}$ and $\boldsymbol{\mu}_{ij}^{tr}$ for the overlapping particles ($R_{ij} < 2a$) have not been derived so far. Importantly, as we will demonstrate in § 3.3, only with the use of these corrections does the mobility matrix $\boldsymbol{\mu}$ remain positive definite for all configurations of the particles.

Contrastingly, in the point-force (Stokeslet) model which is sometimes used for modelling the dynamics of colloidal suspensions (Pear & McCammon 1981), the mobility matrix, defined as

$$\boldsymbol{\mu}_{ij}^{tt} = \frac{1}{8\pi\eta R_{ij}}\left(\mathbf{1} + \hat{\boldsymbol{R}}_{ij}\hat{\boldsymbol{R}}_{ij}\right), \quad i \neq j, \quad \boldsymbol{\mu}_{ii}^{tt} = \frac{1}{\zeta^{tt}}\mathbf{1}, \tag{3.18}$$

is not positive definite even for non-overlapping spheres and does not possess the property (3.13).

3.2. *The shear disturbance tensor* $\boldsymbol{C}$

The formula for the third-rank convection tensor $\boldsymbol{C}$ can be obtained in the following way. Kim & Karrila (1991) provide a solution for the excess flow $\boldsymbol{v}_0^c(\boldsymbol{r})$, produced by a free sphere situated at $\boldsymbol{R}_j$ in the ambient shear flow $\boldsymbol{K}_\infty \cdot \boldsymbol{r}$, which is the difference between the total flow $\boldsymbol{v}(\boldsymbol{r})$ and the ambient flow

$$\boldsymbol{v}_0^c(\boldsymbol{r}) = \boldsymbol{v}(\boldsymbol{r}) - \boldsymbol{K}_\infty \cdot \boldsymbol{r} = \frac{20}{3}\pi\eta a^3 \left\{ \left[\left(1 + \frac{a^2}{10}\nabla^2 \right) \boldsymbol{T}_0\left(\boldsymbol{\rho}_j\right) \right] \overleftarrow{\nabla} \right\} : \boldsymbol{E}_\infty, \tag{3.19}$$

where $[\boldsymbol{T}(\boldsymbol{r})\overleftarrow{\nabla}]_{\alpha\beta\gamma} = \partial_\gamma T_{\alpha\beta}(\boldsymbol{r})$. The contribution to the surface force density due to the straining fluid motion is $3\eta\delta\left(\rho_j - a\right)\boldsymbol{E}_\infty \cdot \hat{\boldsymbol{\rho}}_j$; thus introducing tensor $\boldsymbol{w}^c(\boldsymbol{r})$

$$\boldsymbol{w}^c(\boldsymbol{r}) : \boldsymbol{E}_\infty = 3\eta\delta\left(\rho_j - a\right)\boldsymbol{E}_\infty \cdot \hat{\boldsymbol{\rho}}_j, \tag{3.20}$$

and using the Green's formula we may express the excess flow over the shear flow $\boldsymbol{K}_\infty \cdot \boldsymbol{r}$ in the following way:

$$\begin{aligned} \boldsymbol{v}_0^c(\boldsymbol{r}) &= 3\eta \int_{S_j} \boldsymbol{T}_0\left(\boldsymbol{r} - \boldsymbol{r}'\right) \cdot \boldsymbol{E}_\infty \cdot \boldsymbol{n}' \,\mathrm{d}\sigma' \\ &= \begin{cases} \dfrac{20}{3}\pi\eta a^3 \left\{ \left[\left(1 + \dfrac{a^2}{10}\nabla^2 \right) \boldsymbol{T}_0\left(\boldsymbol{\rho}_j\right) \right] \overleftarrow{\nabla} \right\} : \boldsymbol{E}_\infty, & \rho_j > a, \\ -\boldsymbol{E}_\infty \cdot \boldsymbol{\rho}_j, & \rho_j \leqslant a. \end{cases} \end{aligned} \tag{3.21}$$

Now, by the Faxen laws (3.6) the contribution to the velocity and angular velocity of another sphere (say number i) immersed in such flow is

$$\boldsymbol{U}_i' = \left\langle \boldsymbol{w}_i^t | \boldsymbol{T}_0 | \boldsymbol{w}_j^c \right\rangle : \boldsymbol{E}_\infty = \frac{1}{4\pi a^2} \int_{S_i} \boldsymbol{v}_0^c\left(\boldsymbol{r}'\right) \mathrm{d}\sigma' = \boldsymbol{\mu}_{ij}^{td} : \boldsymbol{E}_\infty, \tag{3.22}$$

$$\boldsymbol{\Omega}_i' = \left\langle \boldsymbol{w}_i^r | \boldsymbol{T}_0 | \boldsymbol{w}_j^c \right\rangle : \boldsymbol{E}_\infty = \frac{3}{8\pi a^3} \int_{S_i} \boldsymbol{n}' \times \boldsymbol{v}_0^c\left(\boldsymbol{r}'\right) \mathrm{d}\sigma' = \boldsymbol{\mu}_{ij}^{rd} : \boldsymbol{E}_\infty. \tag{3.23}$$

For the case of $R_{ij} > 2a$, the form of $\boldsymbol{\mu}_{ij}^{td}$, $\boldsymbol{\mu}_{ij}^{rd}$ is expressed using (3.6) and (3.21) in terms of differential operators

$$\boldsymbol{\mu}_{ij}^{td} : \boldsymbol{E}_\infty = \frac{20}{3}\pi\eta a^3 \left\{ \left[\left(1 + \frac{4a^2}{15}\nabla^2 \right) \boldsymbol{T}_0\left(\boldsymbol{R}_{ij}\right) \right] \overleftarrow{\nabla} \right\} : \boldsymbol{E}_\infty, \tag{3.24}$$

$$\boldsymbol{\mu}_{ij}^{rd} : \boldsymbol{E}_\infty = \frac{10}{3}\pi\eta a^3 \left\{ \left[\nabla \times \boldsymbol{T}_0\left(\boldsymbol{R}_{ij}\right) \right] \overleftarrow{\nabla} \right\} : \boldsymbol{E}_\infty, \tag{3.25}$$

where ∇ denotes derivation with respect to R_{ij}^α and $[\boldsymbol{T}(\boldsymbol{R}_{ij})\overleftarrow{\nabla}]_{\alpha\beta\gamma} = \partial_\gamma T_{\alpha\beta}(\boldsymbol{R}_{ij})$. This allows the final results to be written in the following form:

$$\left[\mu_{ij}^{td}\right]_{\alpha\beta\gamma} = \begin{cases} \dfrac{5}{6}a \left[-\dfrac{16}{5}\dfrac{a^4}{R_{ij}^4}\hat{R}_{ij}^\gamma \delta_{\alpha\beta} + \left(-3\dfrac{a^2}{R_{ij}^2} + 8\dfrac{a^4}{R_{ij}^4} \right) \hat{R}_{ij}^\alpha \hat{R}_{ij}^\beta \hat{R}_{ij}^\gamma \right], & R_{ij} > 2a, \\ \dfrac{5}{6}a \left[\left(-\dfrac{3}{5}\dfrac{R_{ij}}{a} + \dfrac{1}{4}\dfrac{R_{ij}^2}{a^2} \right) \hat{R}_{ij}^\gamma \delta_{\alpha\beta} - \dfrac{1}{16}\dfrac{R_{ij}^2}{a^2} \hat{R}_{ij}^\alpha \hat{R}_{ij}^\beta \hat{R}_{ij}^\gamma \right], & R_{ij} \leqslant 2a, \end{cases} \tag{3.26}$$

with the respective limit in the self-mobility case

$$\boldsymbol{\mu}_{ii}^{td} = \boldsymbol{\mu}_{jj}^{td} = \lim_{R_{ij}\to 0} \boldsymbol{\mu}_{ij}^{td} = \mathbf{0}, \tag{3.27}$$

and

$$\left[\mu_{ij}^{rd}\right]_{\alpha\beta\gamma} = \begin{cases} -\dfrac{5}{2}\left(\dfrac{a}{R_{ij}}\right)^3 \epsilon_{\alpha\beta\zeta}\hat{R}_{ij}^{\zeta}\hat{R}_{ij}^{\gamma}, & R_{ij} > 2a, \\ -\dfrac{5}{2}\left(\dfrac{3}{16}\dfrac{R_{ij}}{a} - \dfrac{1}{32}\dfrac{R_{ij}^3}{a^3}\right)\epsilon_{\alpha\beta\zeta}\hat{R}_{ij}^{\zeta}\hat{R}_{ij}^{\gamma}, & R_{ij} \leqslant 2a, \end{cases} \tag{3.28}$$

and in the self-mobility case as limit

$$\boldsymbol{\mu}_{ii}^{rd} = \boldsymbol{\mu}_{jj}^{rd} = \lim_{R_{ij}\to 0} \boldsymbol{\mu}_{ij}^{rd} = \mathbf{0}. \tag{3.29}$$

The expressions for $R_{ij} < 2a$ in (3.26) and (3.28) vanish for $\boldsymbol{R}_{ij} = 0$ and match with the $R_{ij} > 2a$ expressions at $R_{ij} = 2a$.

Note that (3.24) and (3.25) do not determine $\boldsymbol{\mu}^{td}$ and $\boldsymbol{\mu}^{rd}$ uniquely, since they define only the symmetric and traceless parts of the mobility matrix. Given this freedom, in (3.26)–(3.29) we take the matrices in the simplest algebraic form.

This completes our derivation making all the terms in mobility equation (2.4) directly computable under the Rotne–Prager–Yamakawa approximation.

3.3. *Positive definiteness*

It is now a straightforward task to demonstrate the positive definiteness of the mobility matrix given by (3.11). Cichocki *et al.* (2000) provide a simple proof of positive definiteness of a quadratic form such as in (3.11), which we will now summarize. Consider the following quadratic form:

$$\langle \boldsymbol{g}|\boldsymbol{T}_0|\boldsymbol{g}\rangle = \int \mathrm{d}\boldsymbol{r} \int \mathrm{d}\bar{\boldsymbol{r}} \boldsymbol{g}(\boldsymbol{r})^* \boldsymbol{\cdot} \boldsymbol{T}_0 (\boldsymbol{r} - \bar{\boldsymbol{r}}) \boldsymbol{\cdot} \boldsymbol{g}(\bar{\boldsymbol{r}}), \tag{3.30}$$

where $\boldsymbol{g}(\boldsymbol{r})$ is a complex-valued function and the asterisk denotes complex conjugation. We will show that from the positive definiteness of $\boldsymbol{T}_0$ it follows that $\boldsymbol{\mu}^{pq}$ is positive definite. Let

$$\boldsymbol{d}(\boldsymbol{r}) = \sum_{i,p} \boldsymbol{w}_i^p(\boldsymbol{r}) \boldsymbol{\cdot} \boldsymbol{d}_i^p, \tag{3.31}$$

where $\boldsymbol{d}_i^p$ denotes an arbitrary vector. Now we write

$$0 \leqslant \langle \boldsymbol{d}|\boldsymbol{T}_0|\boldsymbol{d}\rangle = \sum_{i,p}\sum_{j,q} \boldsymbol{d}_i^{p*} \boldsymbol{\cdot} \boldsymbol{\mu}_{ij}^{pq} \boldsymbol{\cdot} \boldsymbol{d}_j^q, \tag{3.32}$$

which ends the proof.

Note that the above proof of positivity does not hold for the point-force model (3.18). In this case the off-diagonal ($i \neq j$) terms of the mobility matrix can be cast in the form (3.11) using $\boldsymbol{w}_i^t(\boldsymbol{r}) = \mathbf{1}\delta(\boldsymbol{r} - \boldsymbol{R}_i)$. The diagonal terms, however, would then become infinite due to the singularity at $R_{ij} = 0$. This problem is circumvented in the formulation (3.18) by using single-particle mobilities $1/\zeta^{tt}$ for the diagonal terms. However, the resulting point-force mobility matrix is not positive definite for arbitrary configuration, thus cannot be used in Brownian dynamics simulations.

4. Generalization of the Rotne–Prager–Yamakawa mobility for arbitrary propagator

In this section we consider a general case of particles interacting hydrodynamically e.g. in confined geometry, periodic boundary conditions or in the presence of interfaces. We assume that for a given geometry a positive-definite Green's function, $\boldsymbol{T}(\boldsymbol{r}, \boldsymbol{r}')$, can be derived. Such solutions have indeed been constructed, e.g. for systems bounded by a cylinder and a sphere (Lorentz 1896; Oseen 1927; Liron & Shahar 1978), for periodic system (Hasimoto 1959) as well as for the system bounded by one (Blake 1971) and two walls (Bhattacharya, Bławzdziewicz & Wajnryb 2005).

We define the Rotne–Prager–Yamakawa approximation for the positive-definite mobility matrix in analogous way to (3.11):

$$\boldsymbol{\mu}_{ij}^{pq} = \langle \boldsymbol{w}_i^p | \boldsymbol{T} | \boldsymbol{w}_j^q \rangle = \iint \mathrm{d}\boldsymbol{r}' \, \mathrm{d}\boldsymbol{r}'' [\boldsymbol{w}_i^p(\boldsymbol{r}')]^{\mathrm{T}} \cdot \boldsymbol{T}(\boldsymbol{r}', \boldsymbol{r}'') \cdot \boldsymbol{w}_j^q(\boldsymbol{r}''). \tag{4.1}$$

To clarify notation we introduce differential operators

$$\overrightarrow{\boldsymbol{D}}^t(\boldsymbol{R}) = \boldsymbol{1}\left(1 + \frac{a^2}{6}\nabla_{\boldsymbol{R}}^2\right), \quad \overleftarrow{\boldsymbol{D}}^t(\boldsymbol{R}) = \boldsymbol{1}\left(1 + \frac{a^2}{6}\overleftarrow{\nabla}_{\boldsymbol{R}}^2\right), \tag{4.2}$$

$$\left[\overrightarrow{\boldsymbol{D}}^r(\boldsymbol{R})\right]_{\alpha\beta} = -\frac{1}{2}\epsilon_{\alpha\beta\gamma}\frac{\partial}{\partial R_\gamma}, \quad \left[\overleftarrow{\boldsymbol{D}}^r(\boldsymbol{R})\right]_{\alpha\beta} = \frac{1}{2}\epsilon_{\alpha\beta\gamma}\frac{\overleftarrow{\partial}}{\partial R_\gamma}, \tag{4.3}$$

where the arrow points in the direction of action of the differentiation operator. We rewrite (3.2) and (3.3) using these operators:

$$\boldsymbol{v}_0^t(\boldsymbol{r}) = \boldsymbol{T}_0(\boldsymbol{r} - \boldsymbol{R}_j) \cdot \overleftarrow{\boldsymbol{D}}^t(\boldsymbol{R}_j) \cdot \boldsymbol{\mathcal{F}}, \quad \boldsymbol{v}_0^r(\boldsymbol{r}) = \boldsymbol{T}_0(\boldsymbol{r} - \boldsymbol{R}_j) \cdot \overleftarrow{\boldsymbol{D}}^r(\boldsymbol{R}_j) \cdot \boldsymbol{\mathcal{T}}, \quad |\boldsymbol{r} - \boldsymbol{R}_j| > a. \tag{4.4}$$

For the external flow $\boldsymbol{v}_0(\boldsymbol{r})$ which is regular (has no sources within sphere i), by the use of the definition of $\boldsymbol{w}_i^p$ (3.9), the Faxen laws may be written in analogy to (3.6):

$$\left.\begin{aligned} \boldsymbol{U}_i &= \int_{S_i} \left[\boldsymbol{w}_i^t(\boldsymbol{r}')\right]^{\mathrm{T}} \cdot \boldsymbol{v}_0(\boldsymbol{r}') \, \mathrm{d}\sigma' = \overrightarrow{\boldsymbol{D}}^t(\boldsymbol{R}_i) \cdot \boldsymbol{v}_0(\boldsymbol{R}_i), \\ \boldsymbol{\Omega}_i &= \int_{S_i} \left[\boldsymbol{w}_i^r(\boldsymbol{r}')\right]^{\mathrm{T}} \cdot \boldsymbol{v}_0(\boldsymbol{r}') \, \mathrm{d}\sigma' = \overrightarrow{\boldsymbol{D}}^r(\boldsymbol{R}_i) \cdot \boldsymbol{v}_0(\boldsymbol{R}_i). \end{aligned}\right\} \tag{4.5}$$

We can now write down the Rotne–Prager–Yamakawa mobilities for the unbounded space (for Oseen propagator $\boldsymbol{T}_0$) for $R_{ij} > 2a$ using the differential operators

$$\boldsymbol{\mu}_{ij}^{pq} = \overrightarrow{\boldsymbol{D}}^p(\boldsymbol{R}_i) \cdot \boldsymbol{T}_0(\boldsymbol{R}_i - \boldsymbol{R}_j) \cdot \overleftarrow{\boldsymbol{D}}^q(\boldsymbol{R}_j). \tag{4.6}$$

Now we decompose the arbitrary propagator $\boldsymbol{T}(\boldsymbol{r}', \boldsymbol{r}'')$ as follows:

$$\boldsymbol{T}(\boldsymbol{r}', \boldsymbol{r}'') = \left[\boldsymbol{T}(\boldsymbol{r}', \boldsymbol{r}'') - \boldsymbol{T}_0(\boldsymbol{r}' - \boldsymbol{r}'')\right] + \boldsymbol{T}_0(\boldsymbol{r}' - \boldsymbol{r}'') = \boldsymbol{T}'(\boldsymbol{r}', \boldsymbol{r}'') + \boldsymbol{T}_0(\boldsymbol{r}' - \boldsymbol{r}''). \tag{4.7}$$

The operator $\boldsymbol{T}' = \boldsymbol{T} - \boldsymbol{T}_0$ has no singularities at $\boldsymbol{r}' = \boldsymbol{r}''$, thus, see (4.5), it has the property

$$\langle \boldsymbol{w}_i^p(\boldsymbol{r}') | \boldsymbol{T}'(\boldsymbol{r}', \boldsymbol{r}'') | \boldsymbol{w}_j^q(\boldsymbol{r}'') \rangle = \overrightarrow{\boldsymbol{D}}^p(\boldsymbol{R}_i) \cdot \boldsymbol{T}'(\boldsymbol{R}_i, \boldsymbol{R}_j) \cdot \overleftarrow{\boldsymbol{D}}^q(\boldsymbol{R}_j). \tag{4.8}$$

Using (4.7) and (4.8) we can cast the mobility $\boldsymbol{\mu}_{ij}^{pq}$ in the following form:

$$\begin{aligned} \boldsymbol{\mu}_{ij}^{pq} &= \overrightarrow{\boldsymbol{D}}^p(\boldsymbol{R}_i) \cdot \boldsymbol{T}(\boldsymbol{R}_i, \boldsymbol{R}_j) \cdot \overleftarrow{\boldsymbol{D}}^q(\boldsymbol{R}_j) + \left[\left\langle \boldsymbol{w}_i^p | \boldsymbol{T}_0 | \boldsymbol{w}_j^q \right\rangle - \overrightarrow{\boldsymbol{D}}^p(\boldsymbol{R}_i) \cdot \boldsymbol{T}_0(\boldsymbol{R}_i - \boldsymbol{R}_j) \cdot \overleftarrow{\boldsymbol{D}}^q(\boldsymbol{R}_j)\right] \\ &= \overrightarrow{\boldsymbol{D}}^p(\boldsymbol{R}_i) \cdot \boldsymbol{T}(\boldsymbol{R}_i, \boldsymbol{R}_j) \cdot \overleftarrow{\boldsymbol{D}}^q(\boldsymbol{R}_j) + \boldsymbol{Y}^{pq}(\boldsymbol{R}_{ij}). \end{aligned} \tag{4.9}$$

The correction

$$\boldsymbol{Y}^{pq}(\boldsymbol{R}_{ij}) = \left\langle \boldsymbol{w}_i^p | \boldsymbol{T}_0 | \boldsymbol{w}_j^q \right\rangle - \overrightarrow{\boldsymbol{D}^p}(\boldsymbol{R}_i) \cdot \boldsymbol{T}_0(\boldsymbol{R}_i - \boldsymbol{R}_j) \cdot \overleftarrow{\boldsymbol{D}^q}(\boldsymbol{R}_j), \tag{4.10}$$

is non-zero only for $|\boldsymbol{R}_{ij}| < 2a$ and is independent of the propagator $\boldsymbol{T}(\boldsymbol{r}_i, \boldsymbol{r}_j)$. We write down explicitly the corrections for all components of the mobility matrix (3.12) and (3.14),(3.16):

$$\begin{aligned} \boldsymbol{Y}^{tt}(\boldsymbol{R}_{ij}) = \Theta(2a - R_{ij}) \left\{ \frac{1}{\zeta^{tt}} \left[\left(1 - \frac{9R_{ij}}{32a} \right) \boldsymbol{1} + \frac{3R_{ij}}{32a} \hat{\boldsymbol{R}}_{ij} \hat{\boldsymbol{R}}_{ij} \right] \right. \\ \left. - \frac{1}{8\pi\eta R_{ij}} \left[\left(1 + \frac{2a^2}{3R_{ij}^2} \right) \boldsymbol{1} + \left(1 - \frac{2a^2}{R_{ij}^2} \right) \hat{\boldsymbol{R}}_{ij} \hat{\boldsymbol{R}}_{ij} \right] \right\}, \end{aligned} \tag{4.11}$$

$$\begin{aligned} \boldsymbol{Y}^{rr}(\boldsymbol{R}_{ij}) = \Theta(2a - R_{ij}) \left\{ \frac{1}{\zeta^{rr}} \left[\left(1 - \frac{27}{32} \frac{R_{ij}}{a} + \frac{5}{64} \frac{R_{ij}^3}{a^3} \right) \boldsymbol{1} + \left(\frac{9}{32} \frac{R_{ij}}{a} - \frac{3}{64} \frac{R_{ij}^3}{a^3} \right) \hat{\boldsymbol{R}}_{ij} \hat{\boldsymbol{R}}_{ij} \right] \right. \\ \left. + \frac{1}{16\pi\eta R_{ij}^3} \left(\boldsymbol{1} - 3\hat{\boldsymbol{R}}_{ij} \hat{\boldsymbol{R}}_{ij} \right) \right\}, \end{aligned} \tag{4.12}$$

$$\boldsymbol{Y}^{rt}(\boldsymbol{R}_{ij}) = \boldsymbol{Y}^{tr}(\boldsymbol{R}_{ij}) = \Theta(2a - R_{ij}) \left\{ \frac{1}{16\pi\eta a^2} \left(\frac{R_{ij}}{a} - \frac{3}{8} \frac{R_{ij}^2}{a^2} \right) \boldsymbol{\epsilon} \cdot \hat{\boldsymbol{R}}_{ij} - \frac{1}{8\pi\eta R_{ij}^2} \boldsymbol{\epsilon} \cdot \hat{\boldsymbol{R}}_{ij} \right\}. \tag{4.13}$$

For the self case, $i = j$, the mobility $\boldsymbol{\mu}_{ii}^{pq}$ is obtained from (4.9) (upper line) in the limit $\boldsymbol{R}_j \to \boldsymbol{R}_i$:

$$\begin{aligned} \boldsymbol{\mu}_{ii}^{pq} &= \lim_{\boldsymbol{R}_j \to \boldsymbol{R}_i} \left\langle \boldsymbol{w}_i^p(\boldsymbol{r}') | \boldsymbol{T}'(\boldsymbol{r}', \boldsymbol{r}'') | \boldsymbol{w}_j^q(\boldsymbol{r}'') \right\rangle + \frac{1}{\zeta_0^{pq}} \\ &= \lim_{\boldsymbol{R}_j \to \boldsymbol{R}_i} \left[\overrightarrow{\boldsymbol{D}^p}(\boldsymbol{R}_j) \cdot \boldsymbol{T}(\boldsymbol{R}_j, \boldsymbol{R}_i) \cdot \overleftarrow{\boldsymbol{D}^q}(\boldsymbol{R}_i) - \overrightarrow{\boldsymbol{D}^p}(\boldsymbol{R}_j) \cdot \boldsymbol{T}_0(\boldsymbol{R}_j - \boldsymbol{R}_i) \cdot \overleftarrow{\boldsymbol{D}^q}(\boldsymbol{R}_i) \right] + \frac{1}{\zeta_0^{pq}}. \end{aligned} \tag{4.14}$$

To sum up, we have shown how to evaluate the Rotne–Prager–Yamakawa approximation for an arbitrary propagator $\boldsymbol{T}(\boldsymbol{r}_i, \boldsymbol{r}_j)$, applying to $\boldsymbol{T}(\boldsymbol{r}_i, \boldsymbol{r}_j)$ differential operators in order to avoid the explicit and often infeasible surface integration. This allows one to construct the positive-definite hydrodynamic tensors in systems with non-trivial geometry (e.g. in the presence of a wall, in a channel or in periodic systems). For example, taking in (4.9) the Green's function for a Stokeslet in the presence of a wall (Blake 1971) leads (for non-overlapping spheres) to a Rotne–Prager–Blake tensor derived before by Bossis, Meunier & Sherwood (1991), see also Kim & Netz (2006), Gauger, Downton & Stark (2009) and Sing *et al.* (2010). However, these authors did not derive the regularizing correction for this tensor, which also prevented them from obtaining the self-term in a manner analogous to our (4.14). On the contrary, the self-term in the aforementioned works was inserted by hand, based on intuition, in which case great care must be taken since the resulting mobility matrix is, in general, not necessarily positive definite.

On a final note, let us stress that the regularizing correction (4.10) has the same simple analytical form in all cases, independently of the particular Green's function $\boldsymbol{T}(\boldsymbol{r}_i, \boldsymbol{r}_j)$.

5. Concluding remarks

In this paper, we have re-visited the problem of constructing the Rotne–Prager–Yamakawa approximation for mobility and shear disturbance matrices. A systematic method was presented which allows one to derive the RPY approximation in a systematic way, for translational, rotational and dipolar components of the generalized mobility matrix, both for non-overlapping and overlapping particles. The regularization corrections for translational–rotational and rotational–rotational mobility tensors have not been previously derived. These regularizations are crucial in obtaining positive-definite hydrodynamic matrices, which is essential for the Brownian dynamics simulations. The positive definiteness also allows the evaluation of the diffusion tensor and mobility for the bead models (including overlapping beads) of complicated molecules. Additionally, we have shown how our approach can be generalized to other boundary conditions and corresponding propagators.

Acknowledgements

E.W. and K.M. acknowledge the support of the Polish National Science Centre (Grant No 2012/05/B/ST8/03010). P.J.Z. acknowledges support of the Foundation for Polish Science (FNP) through TEAM/2010-6/2 project co-financed by the EU European Regional Development Fund. P.S. acknowledges the support of the Polish Ministry of Science and Higher Education (Grant No N N202 055440).

Supplementary data

Supplementary data is available at http://dx.doi.org/10.1017/jfm.2013.402.

References

ADAMCZYK, Z., CICHOCKI, B., EKIEL-JEŻEWSKA, M. L., SŁOWICKA, A., WAJNRYB, E. & WASILEWSKA, M. 2012 Fibrinogen conformations and charge in electrolyte solutions derived from DLS and dynamic viscosity measurements. *J. Colloid Interface Sci.* **385**, 244–257.

ANDO, T. & SKOLNICK, J. 2013 On the importance of hydrodynamic interactions in lipid membrane formation. *Biophys. J.* **104** (1), 96–105.

BHATTACHARYA, S., BŁAWZDZIEWICZ, J. & WAJNRYB, E. 2005 Hydrodynamic interactions of spherical particles in suspensions confined between two planar walls. *J. Fluid Mech.* **541**, 263–292.

BLAKE, J. A. 1971 A note on the image system for a Stokeslet in a no-slip boundary. *Proc. Camb. Phil. Soc.* **70**, 303.

BOSSIS, G., MEUNIER, A. & SHERWOOD, J. D. 1991 Stokesian dynamics simulations of particle trajectories near a plane. *Phys. Fluids* A **3**, 1853–1858.

BRADY, J. F. & BOSSIS, G. 1988 Stokesian dynamics. *Annu. Rev. Fluid Mech.* **20**, 111–157.

CICHOCKI, B., FELDERHOF, B. U., HINSEN, K., WAJNRYB, E. & BŁAWZDZIEWICZ, J. 1994 Friction and mobility of many spheres in Stokes flow. *J. Chem. Phys.* **100**, 3780–3790.

CICHOCKI, B., JONES, R. B., KUTTEH, R. & WAJNRYB, E. 2000 Friction and mobility for colloidal spheres in Stokes flow near a boundary: the multipole method and applications. *J. Chem. Phys.* **112**, 2548–2561.

DHONT, J. K. G. 1996 *An Introduction to Dynamics of Colloids*. Studies in Interface Science, vol. 2, Elsevier.

FELDERHOF, B. U. 1988 Many-body hydrodynamic interactions in suspensions. *Physica* A **151**, 1–16.

FREMBGEN-KESNER, T. & ELCOCK, A. H. 2009 Striking effects of hydrodynamic interactions on the simulated diffusion and folding of proteins. *J. Chem. Theory Comput.* **5**, 242–256.

GAUGER, E. M., DOWNTON, M. T. & STARK, H. 2009 Fluid transport at low Reynolds number with magnetically actuated artificial cilia. *Eur. Phys. J.* E **28** (2), 231–242.

HASIMOTO, H. 1959 On the periodic fundamental solutions of the Stokes equations and their application to viscous flow past a cubic array of spheres. *J. Fluid. Mech.* **5**, 317–328.

KIM, S. & KARRILA, S. J. 1991 *Microhydrodynamics: Principles and Selected Applications*. Butterworth-Heinemann.

KIM, Y. W. & NETZ, R. R. 2006 Electro-osmosis at inhomogeneous charged surfaces: hydrodynamic versus electric friction. *J. Chem. Phys.* **124**, 114709.

LARSON, R. G. & MAGDA, J. J. 1989 Coil-stretch transitions in mixed shear and extensional flows of dilute polymer solutions. *Macromolecules* **22**, 3004–3010.

LIRON, N. & SHAHAR, R. 1978 Stokes flow due to a Stokeslet in a pipe. *J. Fluid. Mech.* **86**, 727–744.

LORENTZ, H. A. 1896 A general theorem concerning the motion of a viscous fluid and a few consequences derived from it. *Versl. Konigl. Akad. Wetensch. Amst.* **5**, 168–175.

MAZUR, P. & VAN SAARLOOS, W. 1982 Many-sphere hydrodynamic interactions and mobilities in a suspension. *Physica* A **115**, 21–57.

NÄGELE, G. 2006 Brownian dynamics simulations. In *Computational Condensed Matter Physics* (ed. S. Blügel, G. Gompper, E. Koch, H. Müller-Krumbhaar, R. Spatschek & R. G. Winkler), Matter and Materials, vol. 32, Forschungszentrum Jülich.

OSEEN, C. W. 1927 *Neuere Methoden und Ergebnisse in der Hydrodynamik*. Akademische Verlagsgesellschaft.

PEAR, M. R. & MCCAMMON, J. A. 1981 Hydrodynamic interaction effects on local motions of chain molecules. *J. Chem. Phys.* **74**, 6922–6925.

POZRIKIDIS, C. 1992 *Boundary Integral and Singularity Methods for Linearized Viscous Flow*. Cambridge University Press.

REICHERT, M. 2006 Hydrodynamic interactions in colloidal and biological systems. PhD thesis, University Konstanz.

ROTNE, J. & PRAGER, S. 1969 Variational treatment of hydrodynamic interaction in polymers. *J. Chem. Phys.* **50**, 4831–4837.

SHAQFEH, E. S. G. 2005 The dynamics of single-molecule DNA in flow. *J. Non-Newtonian Fluid Mech.* **130**, 1–28.

SING, C. E., SCHMID, L., SCHNEIDER, M. F., FRANKE, T. & ALEXANDER-KATZ, A. 2010 Controlled surface-induced flows from the motion of self-assembled colloidal walkers. *Proc. Natl Acad. Sci.* **107** (2), 535–540.

SZYMCZAK, P. & CIEPLAK, M. 2011 Hydrodynamic effects in proteins. *J. Phys.: Condens. Matter* **23**, 033102.

TANAKA, H. 2001 Interplay between wetting and phase separation in binary fluid mixtures: roles of hydrodynamics. *J. Phys.: Condens. Matt.* **13**, 4637–4674.

DE LA TORRE, G. J., DEL RIO ECHENIQUE, G. & ORTEGA, A. 2007 Improved calculation of rotational diffusion and intrinsic viscosity of bead models for macromolecules and nanoparticles. *J. Phys. Chem.* B **111**, 955–961.

USTA, O. B., BUTLER, J. E. & LADD, A. J. C. 2007 Transverse migration of a confined polymer driven by an external force. *Phys. Rev. Lett.* **98**, 098301.

WOJTASZCZYK, P. & AVALOS, J. B. 1998 Influence of hydrodynamic interactions on the kinetics of colloidal particles' adsorption. *Phys. Rev. Lett.* **80**, 754–757.

YAMAKAWA, H. 1970 Transport properties of polymer chains in dilute solution: hydrodynamic interaction. *J. Chem. Phys.* **53**, 436–443.