Ewald sum for hydrodynamic interactions with periodicity in two dimensions

J Bleibel

Max-Planck-Institut für Intelligente Systeme, Heisenbergstr. 3, 70569 Stuttgart, Germany
Institut für Theoretische und Angewandte Physik, Universität Stuttgart, Pfaffenwaldring 57, 70569 Stuttgart, Germany

E-mail: bleibel@mf.mpg.de

Received 21 February 2012, in final form 26 April 2012
Published 17 May 2012
Online at stacks.iop.org/JPhysA/45/225002

Abstract

We carry out the Ewald summation for the Rotne–Prager–Yamakawa mobility tensor, the Oseen mobility tensor and further variations of both, relevant for the hydrodynamic interactions in colloidal suspensions, where all interacting particles are within a single plane, i.e. adsorbed at a fluid interface or other quasi-two-dimensional systems. We use the Poisson summation formula for systems periodic in two dimensions and finite in the third dimension in order to obtain simple formulae for applications, such as molecular dynamics or Brownian dynamics simulations. We show that for such systems as soon as noise is taken into account, a commonly used approximate three-dimensional Ewald summation leads to a spurious system size dependence, which may considerably affect the interpretation of simulation results and will be cured within our approach. Additionally, the resulting formulae are found to be computationally much less expensive than the approximate three-dimensional Ewald summation.

PACS numbers: 82.70.Dd, 47.11.Mn, 05.40.Jc

(Some figures may appear in colour only in the online journal)

1. Introduction

The influence of hydrodynamic interactions (HI) on the dynamics of colloidal systems (i.e. colloidal suspensions or colloids trapped at an interface) is subject to ongoing research [1–7], both from theoretical and experimental points of view. In many circumstances, these many-body systems can be investigated only with the help of simulations. For colloids floating in a bulk solvent at low Reynolds number, a reasonable treatment of HI can be achieved within Stokesian dynamics [8]. In particular, the Oseen or Rotne–Prager–Yamakawa far-field approximation [9, 10], treating the HI as pairwise additive interactions, allows an implementation of HI within Brownian dynamics computer simulations [8, 11]. Since the HI
in the bulk decay $\propto 1/r$, where $r$ is the distance between particles (a similar component is also present in the vicinity of or at fluid–fluid interfaces [12, 13]), they are considered to be long-ranged and demand special treatment within simulations. A suitable tool is provided by the Ewald summation of the Rotne–Prager–Yamakawa mobility tensor [1, 2, 4, 14].

If the colloidal particles in a solvent are not distributed in the full three-dimensional (3D) space, but rather form a thin (mono) layer, the system of interacting colloids may be considered quasi-two dimensional (2D). This may be realized by either trapping the particles at an interface [12, 15, 16] or by placing them in the vicinity of a free or hard boundary [1, 4, 13, 17], or by looking at thin fluid films only [5]. Then the question arises how to treat the HI in these quasi-2D systems. In the latter case, for colloids in a thin fluid film, an experimental study revealed that the 2D form of the Oseen hydrodynamic tensor provides a suitable description of the HI of this system [5]. However, for colloids trapped at fluid interfaces [15, 16], or in the vicinity of interfaces as in the experiment described in [1, 4], the situation is more involved. Owing to the flow fields extending over half of the 3D space, the system cannot be described with a 2D Oseen tensor with its peculiar long-ranged interactions decaying logarithmically with the interparticle distance. However, also the use of the full 3D Rotne–Prager–Yamakawa or Oseen tensors seems somewhat ill-founded, since both do not resemble solutions of the Stokes equation with respect to the underlying boundary conditions [13]. However, in the case of spherical objects these tensors still might serve as a rough approximation. As was advocated in [12], the presence of a free interface separating two fluid phases has only a little effect on the diffusion, and thus the mobility, of spherical particles half-immersed in both phases. The Green function in Stokes flow (Stokeslet) for the velocity field for a single particle in the presence of a boundary (free interface or rigid wall) consists of the Oseen tensor, the free (bulk) solution, plus a mirror term [12, 13, 18] (method of images). Therefore, neglecting the latter term while constructing a solution can be considered as a leading order approximation, where the particles are assumed to be far from any confining boundary. This has been successfully applied to simulate the diffusion of particles close to an interface in experiments [1, 4]. Alternatively, the quasi-2D version of the mobility tensor given in [13] for particles in the vicinity of a free interface may be used.

For computer simulations using the Ewald summation of a 3D mobility tensor, the question arises how to treat the long-ranged part in the third ($z$-)direction. Within the standard approach as developed by Beenakker [14], and used in the simulation studies presented in [1, 4], one has to assume periodic images also in the $z$-direction, i.e. the construction of a layered system of many interfaces. Although easily implemented, such a procedure is computationally expensive and not necessary from a physical point of view. In view of the usually implemented 3D Ewald algorithm, it is, however, unavoidable. Since we are interested in a quasi-2D system which is now extended into the third dimension, the question arises to what extent the influence of the artificial periodic images in the $z$-direction disturbs the result of the summation. Indeed, one could move the periodic images in the $z$-direction to large distances in order to study their influence within the main layer of particles as function of their distance, as was done for e.g. the Coulomb interaction (see [19] and references therein). Although the effective velocities for the particles within this procedure converge to the 2D result, we will show in the following that as soon as noise is considered, this approach generates a spurious system size dependence. Additionally, if one places the periodic images far away from the layer under consideration, this is even more expensive, since the sums within the usual Ewald formalism have to be cut off at larger $K$-values in reciprocal space. Thus for quasi-2D systems, 3D Ewald summation should be avoided. However, 2D Ewald summation formulae have been given so far only for the Oseen tensor in an implicit form [20]. With regard to broader applications, note that the Oseen tensor suffers from not being positive definite. This renders its usage problematic as
soon as a noise term requiring Cholesky decomposition of the tensor is taken into account. Therefore, 2D Ewald summation is needed for more suitable mobility tensors. Our method, as outlined in the following, naturally applies for the Rotne–Prager–Yamakawa tensor and also in the case of the quasi-2D mobility tensor of [13].

The paper is organized as follows. In section 2, we will first discuss the shortcomings of the 3D Ewald summation for quasi-2D systems. Then we will formally derive the 2D summation formula for the Rotne–Prager–Yamakawa tensor. As argued above, we consider this summation of the mobility tensor to be the more appropriate approximation than summing up the full 3D tensor since the result does not suffer from a spurious system-size dependence due to unphysical images across many additional interfaces and provides a computationally much cheaper way to incorporate HI within quasi-2D systems. We follow the procedures described in [21, 22], where a lower dimensional Ewald summation has been developed for electrostatic and dipole interactions, and derive summation formulae for the aforementioned mobility tensors with periodicity assumed in two of three dimensions. In section 3, we demonstrate the 2D Ewald summation procedure by carrying out simulations of a quasi-2D system. In a first step, we will show that conventional 3D Ewald summation leads to a divergent long-time diffusion constant, as the system size increases. We then apply our new summation formulæ, and show that the results are now independent of the system size. Finally, we compare our findings to experimental data, and show that for the particular system under consideration, a reasonable agreement with the data cannot be achieved using the Rotne–Prager–Yamakawa mobility tensor. Only upon summing the quasi-2D mobility tensor given by Cichocki and collaborators, simulations are found to approximate the experimental data. In view of the latter finding, and since the quasi-2D Ewald summation procedure outlined in the following could in principle be applied to any other geometrical setup or approximation (provided the system is quasi-2D and terms \( \propto \frac{1}{r^k} \) (\( k \geq 1 \)) are present), we derive and provide explicit formulæ ready to be used in quasi-2D simulations for the 2D Ewald sum of

(i) the Rotne–Prager–Yamakawa tensor (equations (39)–(41)),
(ii) the Oseen tensor (appendix C, equations (C.5)–(C.7)),
(iii) the quasi-2D mobility tensor of Cichocki et al (appendix C, equations (C.5)–(C.7)) and
(iv) the binary Rotne–Prager tensor (appendix D, equations (D.2)–(D.4)).

Finally, our conclusions are summarized and discussed in section 4.

2. Ewald summation for quasi-two-dimensional systems

Consider a tetragonal lattice with unit cells of volume \( L^2 \times L' \). The lattice is periodic in two dimensions (i.e. x and y) and finite in the third dimension (z). Each cell contains \( N \) spherical particles, arranged to form a single layer (monolayer) parallel to the x–y plane. The force acting on an individual particle \( i \) will be denoted by \( \mathbf{F}_i \). We assume that no external forces are present, thus the total force on the particles in the unit cell cancels to zero [14]:

\[
\sum_{i=1}^{N} \mathbf{F}_i = 0.
\] (1)

If the particles are surrounded by a solvent, one expects additional HI between the particles. For solvents with low Reynolds number, the motion of the colloidal particles is overdamped and inertia of the particles may be neglected [11]. Concerning the implementation of HI within computer simulations, this leads to the use of an position-dependent mobility tensor for the calculation of the viscous drag of the particles [8, 11]. One particular version of this mobility
tensor is the Rotne–Prager–Yamakawa tensor. For the integration of the equations of motion of the colloids, an effective velocity of each particle has to be calculated via

$$v_{\text{eff}} = \sum_{j=1}^{N} M_{ij} F_j$$

(2)

with the Rotne–Prager–Yamakawa mobility tensor

$$M_{ij} = (6\pi \eta a)^{-1} \left\{ \begin{array}{l} \frac{3}{4} a r_{ij}^{-1} \left( \frac{3}{2} \hat{r}_{ij} \hat{r}_{ij} \right) + \frac{1}{2} a^3 r_{ij}^{-3} \left( 1 - 3 \hat{r}_{ij} \hat{r}_{ij} \right) \end{array} \right\}, \quad (i \neq j, \ r \geq 2a)$$

(3a)

$$M_{ii} = (6\pi \eta a)^{-1}, \quad (i = j)$$

(3c)

and \( r_{ij} = |\mathbf{r}_i - \mathbf{r}_j| \). The product \( \hat{r}_{ij} \hat{r}_{ij} \) is the outer product of the normalized vectors \( \hat{r}_{ij} = \mathbf{r}_{ij}/r_{ij} \) and \( 1 \) denotes the unity matrix. Since the HI are long-ranged \( \propto r^{-1} \), Ewald summation has been suggested to treat the interactions of the periodic images [14]. This leads to a lattice sum

$$v_{\text{eff}} = \sum_{j=1}^{N} \sum_{n} \delta_{ij} M_{ij}(\mathbf{r}_{ij}, \mathbf{n}) F_j,$$

(4)

where the second sum runs over 2D lattice vectors \( \mathbf{n} = (n_x L, n_y L) \) with \( \mathbf{n} \neq 0 \) for \( \mathbf{r}_{ij} = 0 \) (indicated by the prime on the sum) and

$$M_{ij}(\mathbf{r}_{ij}, \mathbf{n}) = (6\pi \eta a)^{-1} \left\{ \begin{array}{l} \frac{3}{4} a r_{ij}^{-1} \left( \frac{1}{|\mathbf{r}_{ij} + \mathbf{n}|} + \frac{1}{2} a^3 \frac{1}{|\mathbf{r}_{ij} + \mathbf{n}|^3} \right) \hat{r}_{ij} \hat{r}_{ij} + \frac{3}{4} a^3 \frac{1}{|\mathbf{r}_{ij} + \mathbf{n}|^3} (\mathbf{r}_{ij} + \mathbf{n})(\mathbf{r}_{ij} + \mathbf{n}) - \frac{3}{2} a^3 \frac{1}{|\mathbf{r}_{ij} + \mathbf{n}|^3} (\mathbf{r}_{ij} + \mathbf{n})(\mathbf{r}_{ij} + \mathbf{n}) \end{array} \right\}.$$  

(5)

Note that the definition of \( M_{ij} \) for distances \( r < 2a \) (equation (3b)), introduced to guarantee the positive definiteness of \( M_{ij} \) [9], is not relevant for the following. It only contributes to the lattice sum for \( \mathbf{n} = 0 \) and can be added separately.

2.1. System size dependence of conventional 3D Ewald summation for monolayers

Concerning the above lattice sum, we will first consider its 3D analogue and the resulting Ewald summation derived by Beenakker [14]. In order to use it, one has to assume periodicity in the third dimension, thus the layer of particles is reproduced also in the \( z \)-direction at distances \( n_z L' \) [1, 2]. The 3D lattice sum is split into two sums, one in real and one in reciprocal space, respectively. We consider the sum in \( k \)-space (\( \mathbf{k} \) denotes the 3D wavevector),

$$S_k = \frac{1}{L'^2 L_z} \sum_{k \neq 0} \sum_{j=1}^{N} M^{(2)}(\mathbf{k}) F_j \cos(\mathbf{k} \cdot \mathbf{r}_{ij}),$$

(6)

where the matrix \( M^{(2)}(\mathbf{k}) \) contains the \( k \)-space part of the summed mobility tensor (see equations (4) and (6) in [14]), and concentrate on the wavevectors \( \mathbf{k} \) with \( k_x = k_y = 0 \). Since the particles are arranged within a single layer, say at \( z = 0 \), the product \( \mathbf{k} \cdot \mathbf{r}_{ij} \) vanishes for \( k_x = k_y = 0 \) and all pairs of particles. Thus the cosine above equals 1. If we move the layer in
the z-direction to large distances, the fundamental mode \( k_z^{\text{min}} = 2\pi/L_z \) approaches zero. For the matrix \( M^{(2)}(k) = M^{(2)}(k_z) \) then follows

\[
\lim_{L_z \to \infty} M^{(2)}(k_z) = \lim_{L_z \to \infty} \left( 1 - \frac{kk_z}{k_z^2} \right) \left( a - \frac{1}{3} a^3 k_z^3 \right) \left( 1 + \frac{k_z^2}{4a^2} + \frac{k_z^4}{8a^4} \right) \ldots
\]

\[
\approx \lim_{k_z \to 0} \left( \frac{6\pi a}{k_z^2} + 6\pi a^2 \right) - \frac{kk_z}{k_z^2} \left( \frac{6\pi a}{k_z^2} + 6\pi a^2 \right) = \frac{kk_z}{k_z^2} \left( \frac{6\pi a}{k_z^2} + 6\pi a^2 \right) \ldots
\]

(7)

Since the matrix \( kk_z/k_z^2 \) contains only one nonzero element \((kk_z/k_z^2)^{\infty} = 1\), the diagonal elements \( M^{(2)}_{ii} \) and \( M^{(2)}_{yy} \) of the matrix diverge as \( L_z \) increases. The value of these matrix elements does not depend on any dynamical variable; it is constant and is determined by the choice of the system size \( L_z \) and the particle radius \( a \). Therefore, after carrying out the 3D Ewald summation, the summed mobility matrix \( M_{ij}^*(r_{ij}, n) \) consists of a dynamical part and a constant part depending only on system parameters and diverging as \( L_z \) increases \((M^{(1)}(r_{ij}, n) \) denotes the spatial lattice sum of the mobility matrix, cf equation (6) in [14]):

\[
M_{ij}^*(r_{ij}) = \sum_n M_{ij}(r_{ij}, n)
\]

\[
= \left( 6\pi \eta a \right)^{-1} \left( 1 - \delta_{ij} \right) \left\{ \sum_n M^{(1)}(r_{ij}, n) \right\} + \ldots
\]

\[
+ \frac{1}{L^2 L_z} \sum_{k_z, k_x, k_y} M^{(2)}(k_z) \cos(k_z r_{ij}) + \frac{1}{L^2 L_z} \sum_{k_z, k_x, k_y} M^{(2)}(k_z) \right\}
\]

(8)

\( M_{ij}^*(r_{ij}) \) is in fact a \( 3N \times 3N \) matrix, consisting of a set of \( 3 \times 3 \) submatrices \( D_{ij}(r_{ij}) \) for each pair of particles. After the 3D Ewald summation it contains a diverging constant in the \( xx \)- and \( yy \)-diagonal elements of all off-diagonal submatrices \( D_{ij}(r_{ij}) \). Upon summing over the forces on all particles, this constant part does not contribute, since we assumed a zero net force (see equations (1) and (4)). If the latter requirement is relaxed, i.e. in order to study sedimentation, for 3D suspensions, it is necessary to include the backflow of the solvent as a pressure gradient in order to achieve a cancellation of the singular terms arising from \( k = 0 \) [8]. However, this would not cure the system size dependence in this special case, since the divergence with \( L_z \) does not require a vanishing wavevector. It is rather an artefact of the application of a summation technique appropriate for 3D periodic systems only. For the sum in equation (6) the divergence with \( L_z \) is therefore relevant for non-zero net forces only. However, the summed mobility matrix also enters the calculation of the correlated noise [11],

\[
\langle r_{ij}(t), r_{ij}(t + \Delta t) \rangle = 2M_{ij}^*(r_{ij})\Delta t,
\]

(9)

thus implying a diverging width of the correlator. Within the usual Ermak algorithm, one has to calculate the Cholesky decomposition of \( M_{ij}^*(r_{ij}) \) in order to compute \( 3N \) correlated random numbers for the random displacement of the particles. Therefore, if \( M_{ij}^*(r_{ij}) \) contains a diverging part, its ‘square root’ matrix \( \sigma_{ij}^*(r_{ij}) \) with \( \sigma_{ij}^* \sigma_{ij}^{**} = M_{ij}^* \) will also diverge and the random displacement of the particles may become arbitrarily large. A possible way out could be to simply subtract the divergent part, or to cut off the sum at a certain value; however, the latter would introduce an additional parameter, whereas in the first case, we are not aware of a consistent argument as to why the distances of the additional layers introduced in the z-direction should not matter. Therefore we consider it more appropriate to avoid this scenario by considering the system to be genuinely 2D from the beginning.

It is interesting to compare the setup of a monolayer of particles to another relevant case: particles confined between two parallel walls, rendering a 3D distribution of particles with
finite extent in one dimension (see e.g. [23–26]). A monolayer of colloids may be considered a limiting case of the more general configuration of a confined suspension of particles. However, there are also important differences. Within the confined geometry, particles are not only restricted to the slit by the walls, the different boundary conditions of the walls compared to the unbound fluid also alter the hydrodynamics. The distribution of particles is finite in one dimension; however, depending on the width of the slit, particles are able to move in the third direction also. The HI then also depend on the third spatial coordinate, and the 3D Ewald summation would therefore not diverge as in the case of a monolayer. The slit geometry has been dealt with in detail in [23], where the HI were included on the basis of a 2D Fourier series for the Green function of the Stokes equation with the corresponding no-slip boundary conditions. The method has been generalized to arbitrary domains in [24] as a new method for the computation of the hydrodynamical interactions for confined geometries, the so-called general geometry Ewald-like method. The latter method relies on the separation of forces into local and long-ranged parts [24, 25] just as in conventional Ewald-summation in electrostatics. Instead of solving the Stokes equation in order to obtain the Green function, our approach is rather to use an existing 3D solution in terms of well-known bulk mobility tensors and apply them to a 2D monolayer of particles immersed in an infinite 3D medium. In the following, we first keep the dependence on the non-periodic, third spatial coordinate throughout the derivation. Only for the final formulas do we then consider the limit of vanishing distances between the particles in the third direction, i.e. the spatial configuration of a monolayer. However, upon dropping this requirement and after some straightforward calculations, summation formulae could also be obtained for a 3D configuration of particles, with finite extent in one dimension.

2.2. Two-dimensional Ewald summation

In the following, we will now derive the result for the Ewald summation with periodicity in only two of three dimensions, which will cure the above spurious dependence on the system size. According to the procedure described in [21], we introduce

\[ \Phi(r_{ij}) = \sum_n \frac{1}{|r_{ij} + n|}, \quad (r_{ij} \neq 0); \]  
\[ \Psi(r_{ij}) = \sum_n \frac{1}{|r_{ij} + n|^3}, \quad (r_{ij} \neq 0); \]  
\[ \Theta(r_{ij}, \xi) = \sum_n \exp(-i\xi(n)) \frac{1}{|r_{ij} + n|^3}, \quad (r_{ij} \neq 0); \]  
\[ \chi(r_{ij}, \xi) = \sum_n \exp(-i\xi(n)) \frac{1}{|r_{ij} + n|}, \quad (r_{ij} \neq 0); \]

and denote the sums for \( r_{ij} = 0 \) and \( n \neq 0 \) by \( \Phi_0, \Psi_0, \Theta_0 \) and \( \chi_0 \), respectively:

\[ \Phi_0 = \sum_n \frac{1}{|n|}, \quad (n \neq 0); \]  
\[ \Psi_0 = \sum_n \frac{1}{|n|^3}, \quad (n \neq 0); \]  
\[ \Theta_0(\xi) = \sum_n \frac{\exp(-i\xi(n))}{|n|^3}, \quad (n \neq 0); \]  
\[ \chi_0(\xi) = \sum_n \frac{\exp(-i\xi(n))}{|n|}, \quad (n \neq 0). \]
Using these definitions, equation (4) can be written as

\[
(6\pi a^2) \nu_{\text{eff}} = G_l + \sum_{j=1}^{N} \left\{ \left( \frac{3}{4} a \Theta_{\mathbf{r}_j} + \frac{3}{4} a \Phi_{\mathbf{r}_j} + \frac{1}{2} a^2 \Psi_{\mathbf{r}_j} + \frac{1}{2} a^3 \Theta_{0} \right) \right. \\
- \left. \frac{3}{4} a \nabla_{\mathbf{r}_j} \Theta_{\mathbf{r}_j, \mathbf{\xi}} \right|_{|\mathbf{\xi}|=0} - \frac{3}{4} a \nabla_{\mathbf{r}_j} \Theta_{\mathbf{\xi} \mathbf{\xi}} \right|_{|\mathbf{\xi}|=0} \\
+ \frac{3}{2} a^3 \nabla_{\mathbf{r}_j} \nabla_{\mathbf{\xi}} \chi_{\mathbf{r}_j, \mathbf{\xi}} \right|_{|\mathbf{\xi}|=0} + \frac{3}{2} a^3 \nabla_{\mathbf{\xi}} \chi_{\mathbf{\xi} \mathbf{\xi}} \right|_{|\mathbf{\xi}|=0} \right\} \mathbf{F}_j,
\]

where \( \nabla_{\mathbf{\xi}} \) denotes the gradient with respect to \( \mathbf{\xi} \) and is evaluated for \( |\mathbf{\xi}| = 0 \) [21]. Note that the gradients appear with the opposite sign to compensate for the \( \mathbf{\xi}^2 \) due to differentiation of equations (12) and (13).

The sums appearing in equations (10)–(13) will now each be transformed into two rapid converging sums in real and reciprocal space. We repeat the formalism described in [21], in detail for the first component \( \Phi_{\mathbf{r}_j} \) and give the results for the other parts. For this purpose we use the definition of the Gamma function

\[
\frac{1}{t^{s(2)}} = \frac{1}{\Gamma(s)} \int_0^{\infty} t^{s-1} e^{-t^2} \, dt
\]

and Poisson’s summation formula in two dimensions for a sum of Gaussians

\[
\sum_n \exp(-t^2) = \frac{\pi}{L^2} \sum_K \exp(i\mathbf{K}\rho) \exp \left( -\frac{K^2}{4t} \right)
\]

where \( \rho = (x, y) \) and \( \mathbf{K} = (k_x, k_y) \) are 2D vectors in real and reciprocal space, respectively. Note that the 3D version of this summation formula differs only by an additional factor of \( \sqrt{\pi}/(L^{1/2}) \) on the right-hand side, stemming from the underlying Fourier transformation of the sum of Gaussians. Using the 3D summation formula instead, e.g. in order to derive the original result of Beenakker for the 3D Rotne–Prager tensor [14], would therefore not change the general outline of the calculation, but lead to different integrals in the following.

Inserting equation (19) for \( s = 1/2 \) into equation (10) leads to [21]

\[
\Phi = \sum_n \frac{1}{\Gamma(1/2)} \int_0^{\infty} t^{-1/2} \exp(-|\mathbf{r}_j + \mathbf{n}|^2 t) \, dt.
\]

The integral may be split up, by introducing a convergence factor \( \alpha \) [21] (still to be determined)

\[
\Phi = \sum_n \frac{1}{\sqrt{\pi}} \int_0^{\infty} t^{-1/2} \exp(-|\mathbf{r}_j + \mathbf{n}|^2 t) \, dt + \sum_n \frac{1}{\sqrt{\pi}} \int_0^{\infty} t^{-1/2} \exp(-|\mathbf{r}_j + \mathbf{n}|^2 t) \, dt.
\]

The first integral can be evaluated, and for the second we apply the summation formula (equation (20))

\[
\Phi = \sum_n \text{erfc}(\alpha|\mathbf{r}_j + \mathbf{n}|) + \sqrt{\pi} \frac{1}{L^2} \int_0^{\infty} \sum_{\mathbf{K}} \exp(i\mathbf{K}\rho_{\mathbf{r}_j}) \exp \left( -\frac{K^2}{4t} - z_{ij}^2 t \right) \, dt,
\]

where \( \text{erfc}(x) = 1 - \text{erf}(x) \) is the complementary error function and \( \mathbf{r}_{ij} = (\rho_{ij}, z_{ij}) \). The integral will contain a singularity for \( K = 0 \). This term deserves a special treatment; therefore, we separate the term with \( K = 0 \) from the sum to isolate the singularity [21]. As will be seen later on, we can use the absence of external forces (equation (1)) to get rid of all singular and
constant terms [14, 21].

\[
\Phi = \sum_n \frac{\text{erfc}(\alpha |r_{ij} + n|)}{|r_{ij} + n|} + \frac{\sqrt{\pi}}{L^2} \int_0^\infty t^{-3/2} \exp(-z_i^2 t) \, dt \\
+ \frac{\sqrt{\pi}}{L^2} \sum_{K \neq 0} \exp(iK\rho_{ij}) \int_0^\infty t^{-3/2} \exp\left(-\frac{K^2}{4t} - z_i^2 t\right) \, dt.
\]

(24)

The first integral evaluates to

\[
\int_0^\infty t^{-3/2} \exp(-z_i^2 t) \, dt = -2\sqrt{\pi}z_i \text{erf}(\alpha z_i) - \frac{2}{\alpha} e^{-\alpha^2 z_i^2} + \lim_{t \to 0^+} \frac{2e^{-z_i^2 t}}{\sqrt{t}},
\]

while the second integral can be performed using the substitution \(u^2 = 1/t\) [21]:

\[
\int_0^\infty t^{-3/2} \exp\left(-\frac{K^2}{4t} - z_i^2 t\right) \, dt = \frac{\sqrt{\pi}}{K} \left[ e^{-\alpha z_i} \text{erfc}\left(\frac{K}{2\alpha} - \alpha z_i\right) + e^{\alpha z_i} \text{erfc}\left(\frac{K}{2\alpha} + \alpha z_i\right) \right].
\]

(26)

Putting all the pieces together, one ends up with

\[
\Phi(r_{ij}) = \sum_n \frac{\text{erfc}(\alpha |r_{ij} + n|)}{|r_{ij} + n|} + \frac{\pi}{L^2} \sum_{K \neq 0} \exp(iK\rho_{ij}) \left[ e^{-\alpha z_i} \text{erfc}\left(\frac{K}{2\alpha} - \alpha z_i\right) \\
+ e^{\alpha z_i} \text{erfc}\left(\frac{K}{2\alpha} + \alpha z_i\right) \right] + \frac{2\sqrt{\pi}}{L^2} \left[ \sqrt{\pi} z_i \text{erfc}(\alpha z_i) + \frac{1}{\alpha} e^{-\alpha^2 z_i^2} - \lim_{t \to 0^+} \frac{e^{-z_i^2 t}}{\sqrt{t}} \right].
\]

(27)

Accordingly, for \(\Phi_0\) (equation (14)) one finds

\[
\Phi_0 = \sum_n \frac{\text{erfc}(\alpha |n|)}{|n|} + \frac{\pi}{L^2} \sum_{K \neq 0} \frac{2}{K} \text{erfc}\left(\frac{K}{2\alpha}\right) - 2\sqrt{\pi} \frac{L^2}{\alpha^2} + 2\sqrt{\pi} \lim_{t \to 0^+} \frac{1}{\sqrt{t}} = \frac{2\alpha}{\sqrt{\pi}},
\]

(28)

where the term for \(n = 0\) had to be inserted in the sum of the integral on \([0, \alpha^2]\) and subtracted separately in order to apply the Poisson summation formula [21].

For the terms \(\Psi(r_{ij})\) and \(\Psi_0\), the procedure is similar, so we just give the results. Note, that no singular terms will appear while performing the required integrations. Using equation (19) with \(s = 3/2\) leads to

\[
\Psi(r_{ij}) = \frac{2}{\sqrt{\pi}} \sum_n \int_0^\infty t^{1/2} \exp(-|r_{ij} + n|^2 t) \, dt \\
+ \frac{2\sqrt{\pi}}{L^2} \sum_K \exp(iK\rho_{ij}) \int_0^\infty t^{-1/2} \exp\left(-\frac{K^2}{4t} - z_i^2 t\right) \, dt
\]

(29)

and

\[
\Psi_0 = \frac{2}{\sqrt{\pi}} \sum_n \int_0^\infty t^{1/2} \exp(-|n|^2 t) \, dt + \frac{2\sqrt{\pi}}{L^2} \sum_K \int_0^\infty t^{-1/2} \exp\left(-\frac{K^2}{4t}\right) \, dt = \frac{4\alpha^3}{3\sqrt{\pi}}.
\]

(30)

The \(\Theta\) and \(\chi\) terms require a different form of the Poisson summation formula [21]:

\[
\sum_n \exp(-|\rho + n|^2 t - i\xi(\rho + n)) = \frac{\pi}{L^2 t} \sum_K \exp(iK\rho) \exp\left(-\frac{|K + \xi|^2}{4t}\right)
\]

(31)

which takes the additional \(\xi\)-dependence into account. Using equation (19) with \(s = 3/2\), equation (12) can be written in the following form:

\[
\Theta(r_{ij}, \xi) = \frac{2}{\sqrt{\pi}} \sum_n \exp(-i\xi(r_{ij} + n)) \int_0^\infty t^{1/2} \exp(-|r_{ij} + n|^2 t) \, dt.
\]

(32)
Again, one splits up the integral and applies equation (31) in the second integral. Thus

\[
\Theta(r_{ij}, \xi) = \frac{2}{V} \sum_n \exp(-i\xi(r_{ij} + n)) \int_0^\infty t^{1/2} \exp(-|r_{ij} + n|^2 t) \, dt \\
+ \frac{2\sqrt{\pi}}{L^2} \sum_k \exp(iK\rho_{ij} - i\xi z_{ij}) \int_0^\infty t^{-1/2} \exp\left(-\frac{|K + \xi|^2}{4t} - z_{ij}^2 t\right) \, dt,
\]

where \(\xi_\rho\) denotes the 2D x- and y-part of \(\xi\), and accordingly for \(\Theta_0(\xi)\).

\[
\Theta_0(\xi) = \frac{2}{V} \sum_n \exp(-i\xi(n)) \int_0^\infty t^{1/2} \exp(-|n|^2 t) \, dt \\
+ \frac{2\sqrt{\pi}}{L^2} \sum_k \int_0^\infty t^{-1/2} \exp\left(-\frac{|K + \xi|^2}{4t} \right) \, dt = \frac{4\alpha^3}{3\sqrt{\pi}}.
\]

The calculation for \(\chi(r_{ij}, \xi)\) is carried out with \(s = 5/2\) and after some short manipulations one finds

\[
\chi(r_{ij}, \xi) = \frac{4}{3\sqrt{\pi}} \sum_n \exp(-i\xi(r_{ij} + n)) \int_0^\infty t^{3/2} \exp(-|r_{ij} + n|^2 t) \, dt \\
+ \frac{4\sqrt{\pi}}{3L^2} \sum_k \exp(iK\rho_{ij} - i\xi z_{ij}) \int_0^\infty t^{1/2} \exp\left(-\frac{|K + \xi|^2}{4t} - z_{ij}^2 t\right) \, dt
\]

\[
\chi_0(\xi) = \frac{4}{3\sqrt{\pi}} \sum_n \exp(-i\xi(n)) \int_0^\infty t^{3/2} \exp(-|n|^2 t) \, dt \\
+ \frac{4\sqrt{\pi}}{3L^2} \sum_k \int_0^\infty t^{1/2} \exp\left(-\frac{|K + \xi|^2}{4t} \right) \, dt = \frac{8\alpha^3}{15\sqrt{\pi}}.
\]

The calculation of the components for \(r_{ij} = 0\), i.e. \(\Phi_0, \Psi_0, \Theta_0\) and \(\chi_0\) (see equations (14)–(17)) is only needed to extract the constant term arising from the constraint \(K \neq 0\). This term only contributes for \(i = j\), since it corresponds to the additional part summed up for \(r_{ij} = 0\) and \(n = 0\). Therefore it is placed outside the sum over \(j\). The remaining part can be absorbed into the main formulae by allowing \(r_{ij} = 0\). Note that for \(\Phi\) and \(\chi\) this constant term does not contribute, since it does not depend on \(\xi\) and only the matrix elements with gradients are used.

A similar singularity as in the integration (equation (25)) will appear when integrating \(\nabla_\xi \hat{V}_I\) (see also equation (A.1) in appendix A).

We are now left with expressions for \(\Phi, \Psi, \Theta\) and \(\chi\) as sums in real and reciprocal space. It remains to evaluate the gradients with respect to \(\xi\), perform the remaining integrals and collect all parts from equations (27)–(30) and equations (33)–(36) in order to insert them into equation (18). We list all components and integrals in appendix A. To obtain the final formula, we take the limit \(z_{ij} \to 0\) for all pairs of particles, i.e. all particles remain in a plane parallel to the \((x-y)\) plane. We introduce the following definitions:

\[
R_{jn} = |r_{ij} + n|, \quad \hat{R}_{jn} = \frac{(r_{ij} + n)}{|r_{ij} + n|}, \quad \hat{K} = \frac{K}{K}.
\]

Since we are only interested in the limit \(z_{ij} = 0\), we can assume without loss of generality that \(z = 0\) for all particles, i.e. \(r_{ij} = \rho_{ij}\), and thus consider \(\hat{R}_{jn}, \nabla_v, \hat{F}_I\) to be 2D only from now on. Before writing down the sum of all components we note that several terms cancel and, additionally, all divergent and constant parts stemming from equations (27)–(30) and (33)–(36) do not contribute to the final result. In the case of a net zero force on the particles in the system, (see equation (1)), any product with a (diverging) constant also vanishes. As
discussed in [8], the cancellation of the singular terms stemming from $K = 0$ can be achieved, even in the case when the average force on the particles is not zero. To this end, one has to introduce a pressure gradient representing the backflow of the fluid. The relevant physical quantities are then the velocities relative to the backflow of the fluid [8]. Taking all similar parts together, one then arrives at a rather compact result:

$$\begin{align*}
(6\pi \eta a)\nu_{\text{eff}} &= F_i = \frac{aa}{\sqrt{\pi}} \left( \frac{3}{2} + \frac{2}{3}a^2 \alpha^2 \right) F_i + \sum_{j=1}^{N} \left[ \sum_{n} \left( \frac{3}{4}a \frac{\text{erfc}(\alpha R_{jn})}{R_{jn}} \right) [1 + \hat{R}_{jn} \hat{R}_{jn}] 
\right.
\left. + \frac{1}{2}a \frac{\alpha^2 e^{-a^2 R_{jn}^2}}{R_{jn}^2} \left( \frac{3}{2} - 2a^2 \alpha^2 \right) \hat{R}_{jn} \hat{R}_{jn} \right] F_i
\right.
\left. + \frac{aa}{\sqrt{\pi}} e^{-a^2 R_{jn}^2} \sum_{k \neq n} \text{cos}(K r_{ij}) \left( \frac{3a}{2L^2} \text{erfc} \left( \frac{K}{2a} \right) \right) \right] F_i.
\end{align*}$$

If we cast this result into a similar form as in [14], the final formula reads

$$\begin{align*}
(6\pi \eta a)\nu_{\text{eff}} &= \sum_{j=1}^{N} \sum_{n} M^{(1)}(r_{jn}) F_j + \sum_{j=1}^{N} \sum_{k \neq n} M^{(2)}(K) \text{cos}(Kr_{ij}) F_j
\right.
\left. + 1 \left[ 1 - 3 \frac{\pi^{-1/2} a a}{\pi^{1/2} a a} - \frac{2}{3} \pi^{-1/2} a a \right] F_i.
\end{align*}$$

The prime in the first sum indicates that for $r_{ij} = 0$ the terms with $n = 0$ are omitted. We used the definitions:

$$\begin{align*}
M^{(1)}(r) &= \left\{ \left( \frac{3}{4}a^{-1} + \frac{1}{4}a^3 r^{-3} \right) \text{erfc}(ar) + a^3 a^{-2} \pi^{-1/2} \exp(-a^2 r^2) \right\}
\right.
\left. + \tilde{f} \left\{ \left( \frac{3}{4}a^{-1} - \frac{1}{4}a^3 r^{-3} \right) \text{erfc}(ar) 
\right.
\left. + \left( -3a^3 a^{-2} + \frac{1}{2}a^2 a^3 \right) \pi^{-1/2} \exp(-a^2 r^2) \right\}
\end{align*}$$

for the real part of the tensor and

$$\begin{align*}
M^{(2)}(K) &= 1 \left\{ \frac{2a}{\sqrt{2\pi}} \text{erfc} \left( \frac{k}{2a} \right) \right\} \left\{ \frac{3\pi}{2L^2 k} - \frac{1}{2} \cdot \frac{3a}{\sqrt{2\pi}} \text{erfc} \left( \frac{k}{2a} \right) 
\right.
\left. + \frac{aa^{-1}a^{-1} \pi^{-1/2}}{\sqrt{4a^2}} \exp \left( -\frac{k^2}{4a^2} \right) - \frac{3\pi}{2L^2 k} \right\}
\end{align*}$$

for the summation in Fourier space. This completes the derivation of the quasi-2D Ewald sum of the Rotne–Prager–Yamakawa mobility tensor.

The main advantage of the Rotne–Prager–Yamakawa tensor compared to the Oseen tensor is its positive definiteness [9], needed for the Cholesky decomposition as used in Brownian dynamics simulations [11]. Therefore, one also has to consider the part of the Rotne–Prager–Yamakawa tensor for $r_{ij} < 2a$ (equation (23) in [9]). However, this term does not contribute to the long-ranged part of the HI and thus can be simply taken out from the real space lattice sum for $n = 0$ in equation (38) or equation (39) and added separately. For the sum over the periodic images, this term will not appear since $|r_{ij} + n| > 2a$ is always fulfilled.

3. Results from simulations

In order to test the Ewald summation in a quasi-2D system using the procedure outlined above, we performed Brownian dynamics simulations with a single layer of $N$ colloids (radius $a$)
Figure 1. Comparison of the scaled mean-squared displacement for Brownian dynamics simulations of \( N = 100 \) colloidal particles (radius \( a = 2.35 \mu m \), 2D number density \( \varrho = 3.24 \times 10^{-3} \mu m^{-2} \)) with a repulsion strength of \( \Gamma = 8.2 \) without hydrodynamical interactions (open circles) and with HI included via 3D Ewald summation for a varying longitudinal system size \( L_z \). Experimental data and additional BD data (line) were taken from [1]. Error bars are of the order of the corresponding symbol size and have been omitted for clarity. A simulation with \( L_z \approx 7L \) (not shown) would accidentally match the data from the experiment. Such a fortuitous choice could well be the underlying reason for the good agreement between simulations with HI and the experimental data, as reported in [1].

within a fluid phase in a box with side lengths \( L_x = L_y = L_z \). This system resembles a model for the setup used in the experiments of [1, 17] where paramagnetic colloidal particles were placed atop a flat and stabilized air–water interface of a suspended droplet. The colloids are fully immersed in the fluid phase and, due to gravity pulling them downwards, just stay in the vicinity of the interface [27] without perturbing the latter considerably. Thus the colloidal particles constitute a quasi-2D system. More details can be found in [1, 17]. In the model, the layer of particles is placed parallel to the \((x–y)\) plane at \( z = L_z/2 \), with only in-plane motion allowed. The colloids interact through a repulsive potential \( v_{ij}/k_B T = \Gamma/d^3 \), where \( d \) is the distance between each two particles scaled by the mean interparticle separation: \( d = r/\sqrt{\varrho} \) (\( \varrho \) denotes the 2D number density of the colloids). In the experimental setup, this dipole repulsion is generated and controlled by an external magnetic field applied perpendicular to the layer of colloids.

First, we validate our simulation by attempting to reproduce the Brownian dynamics data published by Rinn et al [1]. We therefore set \( \Gamma = 8.2 \) and \( a = 2.35 \mu m \) for a system of \( N = 100 \) colloids at a number density of \( \varrho = 3.24 \cdot 10^{-3} \), which corresponds to an area fraction of \( \eta = 0.056 \). The time is measured in units of \( \tau = 1/(\varrho D_0) \), where \( D_0 \) is the single particle short-time diffusion constant, extracted from the simulations by extrapolating the mean-squared displacement towards \( t \to 0 \) [1]. In our simulations, we used the bulk value for the diffusion constant \( (D = (6\pi\eta a)^{-1}) \). If the scaling of the mean-squared displacement with \( D \) holds, then the extrapolated short-time diffusion constant \( D_0 \) should agree with this value. Indeed, for Brownian dynamics simulations we find \( D_0 = D \) and the data agree with [1] (see figure 1). Furthermore, we compare in figure 1 the mean-squared displacement (scaled by \( 4tD_0 \)) with the data agree with [1] (see figure 1). Furthermore, we compare in figure 1 the mean-squared displacement (scaled by \( 4tD_0 \)) for simulations with varying system size \( L_z \). First it should be noted that the extracted short-time diffusion constant \( D_0 \) differs from the corresponding bulk value \( D \), if 3D Ewald summation is applied. The extracted values increase with increasing system size in the \( z \)-direction. Additionally, as can be seen from the simulation
data presented in figure 1, the mean-squared displacement, scaled by the corresponding short-
time diffusion constant $D_0$, also increases with increasing system size. Thus the simulations
confirm the finding of a diverging mobility matrix for the 3D Ewald summation of this system.

Neglecting noise, the 3D summation method remains valid. If we then compare the
effective velocities of single particles, we find that for our current setup, the distance of the periodic images in the $z$-direction had to be scaled by a factor of 2 relative to the original size $L_z = L_x$, in order to reduce the impact on the single layer and to converge to the result from 2D Ewald summation according to equation (38). Note, that for large systems such a stretching of the third dimension could be unnecessary, since the size of a cubed box could be already sufficient. However, this has to be checked for each setup individually.

As the second test, we perform simulations with hydrodynamical interactions included,
but without applying any Ewald summation procedure. We extract the long-time self-diffusion constant $D_L$ by running the simulation for much longer times $t = 1.2\tau$ and fitting the scaled mean-squared displacement for times $t > 0.95\tau$ to a constant. Figure 2 depicts the long-time diffusion constant (scaled by $D_0$) for simulations with the same parameter setup as before and for various system sizes and constant number density. As the number of particles increases, for the Brownian dynamic simulation with HI, $D_L$ is found to decrease and approach the limit set by using 2D Ewald summation. The system size dependence of $D_L$ without Ewald summation becomes clearly visible. In order to roughly characterize the convergence, we extrapolated this decrease using a polynomial fit for $0 < N < 700$. It turned out that a system containing $N \gtrsim 25000$ particles would yield a similar result for $D_L/D_0$ as obtained in simulations with 2D Ewald summation. For the latter ones, we do not observe any significant dependence of $D_L$ on the system size.

Of course, it is now tempting to compare the 2D Ewald summation results to the experimental data from [1]. However, one has to keep in mind that the situation in the experiment is different. The particles are adsorbed to a free interface, whereas there are none in the simulations. The authors suggested to increase the hydrodynamical radius in the simulations, that is, the value of the particle radius within the calculation of the mobility tensor was increased by a factor of 2. Figure 3 depicts the scaled mean-squared...

Figure 2. Long-time self-diffusion constant (scaled by $D_0$) extracted from simulations with
hydrodynamical interactions, with and without 2D Ewald summation. As the number of particles
in the system increases, $D_L$ approaches the value as obtained from simulations with 2D Ewald
summation. Lines are drawn to guide the eye, and the dashed line corresponds to a polynomial fit.
Error bars correspond to the statistical error obtained from averaging over many simulation runs.
Figure 3. Comparison of the simulated scaled mean-squared displacement for the same setup as in figure 1 to experimental data as obtained by the experiment of Rinn et al [1]. Only representative error bars are shown for simulations. The error bars for the Brownian dynamics simulation without hydrodynamical interactions (stars) are smaller than the symbol size. The simulation data stemming from BD simulations with hydrodynamical interactions and 2D Ewald summation of the quasi-2D mobility tensor of Cichocki et al [13] (triangles) have been scaled by their different short-time diffusion constant $D_0 = 1.38 D$.

displacement for the 2D Ewald summation method and the experimental data of [1]. The data cannot be reproduced with this actual implementation. We refrained from trying out various hydrodynamical radii, since this would introduce a free parameter to the system. If we use the mobility tensor by Cichocki et al [13], derived from the two-sphere mobility tensor for particles close to a free interface as an asymptotic series in powers of $1/R$ (cf appendix C, equations (C.1a) and (C.1b)), the simulation data overestimate the experimental values. However, the short-time diffusion constant $D_0$ extracted from the simulation is also increased, compared to the simulations based on the Rotne–Prager mobility tensor. If we normalize the quasi-2D simulation data with respect to the extracted value for $D_0$, we find a rather good agreement. For longer times, however, the simulation data start to deviate slightly. Note that the deviation of the extrapolated short-time diffusion constant $D_0$ compared to $D = (6\pi \eta a)^{-1}$ may be anticipated by the inspection of equations (C.1a) and (C.1b) in the appendix C, since the matrix $Q_1$ for the self-diffusion already deviates by a factor of $\approx 1.38$ from the unity matrix in the Rotne–Prager case. Therefore, this deviation is only present in the quasi-2D case, for all other simulations the value of $D_0$ agrees with the diffusion constant $D$ initially plugged in.

Concerning the computational cost of the simulations, we find that using the 2D Ewald summation method leads to a reduction of at least an order of magnitude, compared to its 3D version, and depends on the particular choice of the convergence parameter $\alpha$. For a typical choice of $\alpha = 2/L$ the gain in speed was around a factor $\sim 15$. Note that the computational cost of the Ewald summation may vary, depending on the choice of $\alpha$.

4. Summary and conclusions

In summary, we have provided Ewald summation formulae for quasi-2D systems for the Rotne–Prager–Yamakawa mobility tensor and variants. We demonstrated, that for quasi-2D systems, 3D Ewald summation leads to a spurious system size dependence, stemming from the summation in the direction perpendicular to the 2D layer of particles. This problem was
solved by summing in two dimensions only, and, additionally, using the resulting formulae to calculate HI in computer simulations of quasi-2D systems was found to be much more efficient, due to the avoidance of summation in the third direction. We further found that the asymptotic value of the long-time diffusion constant for large systems could already be obtained for rather small systems using the 2D Ewald summation procedure. We demonstrated that the 2D Ewald sum of the quasi-2D analogue of the Rotne–Prager mobility tensor given by Cichocki and collaborators may be used to reproduce experimental data quite well. Together with recent advances for an approximate and efficient treatment of HI in computer simulations [28], inclusion of HI and the proper treatment of their long-ranged characteristic becomes feasible even for large systems in quasi-2D simulations of colloidal suspensions.

Acknowledgments

JB thanks M Oettel for fruitful discussions and the German Research Foundation (DFG) for the financial support through the Collaborative Research Center (SFB-TR6) ‘Colloids in External Fields’ Project N01.

Appendix A. Gradient terms and integrals

Here, we list the evaluation of the gradients $\nabla_{\mathbf{F}} \nabla_{\mathbf{f}}$ for the integrals appearing in the equations for $\Theta$ and $\chi$ (equations (33)–(36)), as well as the resulting integrals that need to be performed in order to obtain equation (38):

$$\nabla_{\mathbf{F}} \nabla_{\mathbf{f}} \int_0^{a^2} t^{-1/2} \exp \left(-\frac{|\mathbf{K}|^2}{4t} - z^2 t\right) dt \bigg|_{t=0} = -\frac{1}{2} \int_0^{a^2} t^{-3/2} \exp \left(-\frac{K^2}{4t} - z^2 t\right) dt$$

$$+ \frac{K}{4} \int_0^{a^2} t^{-5/2} \exp \left(-\frac{K^2}{4t} - z^2 t\right) dt$$

(A.1)

$$\nabla_{\mathbf{F}} \nabla_{\mathbf{f}} \int_0^{a^2} t^{1/2} \exp \left(-\frac{|\mathbf{K}|^2}{4t} - z^2 t\right) dt \bigg|_{t=0} = -\frac{1}{2} \int_0^{a^2} t^{-1/2} \exp \left(-\frac{K^2}{4t} - z^2 t\right) dt$$

$$+ \frac{K}{4} \int_0^{a^2} t^{-3/2} \exp \left(-\frac{K^2}{4t} - z^2 t\right) dt.$$  

(A.2)

The integral containing $t^{-3/2}$ has already been calculated (see equation (26)), and the remaining integrals can also be performed:

$$\int_0^{a^2} t^{-1/2} \exp \left(-\frac{K^2}{4t} - z^2 t\right) dt = \frac{\sqrt{\pi}}{2z} \left[e^{-Kz} \text{erfc} \left(\frac{K}{2\alpha} - \alpha z\right) + e^{Kz} \text{erfc} \left(\frac{K}{2\alpha} + \alpha z\right)\right]$$

(A.3)

$$\int_0^{a^2} t^{-5/2} \exp \left(-\frac{K^2}{4t} - z^2 t\right) dt = \frac{\sqrt{2\pi}}{K^3} \left(1 + Kz\right) e^{-Kz} \text{erfc} \left(\frac{K}{2\alpha} - \alpha z\right)$$

$$+ (1 - Kz)e^{Kz} \text{erfc} \left(\frac{K}{2\alpha} + \alpha z\right) + \frac{4}{\alpha K^2} \exp \left(-\frac{K^2}{4\alpha^2} - z^2 \alpha^2\right).$$

(A.4)

Taking the limit $z \to 0$ is straightforward for all terms except for the integral in equation (A.3). This evaluates to

$$\lim_{z \to 0} \int_0^{a^2} t^{-1/2} \exp \left(-\frac{K^2}{4t} - z^2 t\right) dt = 2\alpha \exp \left(-\frac{K^2}{4\alpha^2}\right) - \sqrt{\pi} K \text{erfc} \left(\frac{K}{2\alpha}\right).$$

(A.5)
Appendix B. Ewald sum of the Oseen tensor

For the Ewald sum according to equation (4) of the Oseen tensor
\[ O_{ij} = (8\pi \eta)^{-1} r_{ij}^{-1} (\mathbb{1} + \hat{r}_i \hat{r}_j), \quad (i \neq j) \] (B.1a)
\[ O_{ii} = (6\pi \eta a)^{-1} 1, \quad (i = j) \] (B.1b)

instead of \( M_{ij} \), we neglect all terms in equation (38) involving \( a^3 \), since these are the additional terms of the Rotne–Prager–Yamakawa tensor compared to the Oseen tensor. This leads to
\[(6\pi \eta a) v_{\text{eff}} = F_i - \frac{3\alpha a}{2\sqrt{\pi}} F_i + \sum_{j=1}^{N} \left\{ \sum_{n} \frac{3}{4} \alpha \text{erfc}(\alpha R_{jn}) \right\} + \frac{3}{2} \frac{a^2 \sqrt{\pi} e^{-a^2 r_{ij}^2}}{L^2} \hat{K} \hat{K} F_j \] (B.2)

which also may be cast in a similar form as equations (39)–(41):
\[(6\pi \eta a) v_{\text{eff}} = \sum_{j=1}^{N} \sum_{n} M^{(1)}_{O}(r_{jn}) F_j + \sum_{j=1}^{N} \sum_{K \neq 0} M^{(2)}_{O}(K) \cos(K r_{ij}) F_j + 1 \left( 1 - \frac{3}{2} \pi^{-1/2} a \alpha \right) F_i \] (B.3)

with
\[ M^{(1)}_{O}(r) = \mathbb{1} \left\{ \frac{3}{2} \pi^{-1} \text{erfc}(\alpha r) \right\} + \hat{R}_n \left\{ \frac{3}{2} \pi^{-1} \text{erfc}(\alpha r) + \frac{3}{2} a \alpha \pi^{-1/2} \exp(-a^2 r^2) \right\} \] (B.4)

for the real part of the tensor and
\[ M^{(2)}_{O}(K) = 1 \left\{ 2a \text{erfc} \left( \frac{k}{2\alpha} \right) \right\} - \frac{3\pi}{2L^2} \hat{K} \hat{K} \left\{ a \text{erfc} \left( \frac{k}{2\alpha} \right) + a \alpha^{-1} \pi^{-1/2} \exp \left( -\frac{k^2}{4\alpha^2} \right) \right\} \] (B.5)

for the Fourier space.

Appendix C. Ewald sum for the quasi-2D mobility tensor of Cichocki et al

The mobility tensor reported in [13] for a quasi-2D system of spherical particles close to a fluid interface differs from the 3D Rotne–Prager–Yamakawa tensor by a factor of 2 for the terms proportional to \( 1/r_{ij} \) and by a factor of \( q = 5.59027 \) for terms \( \propto 1/\alpha r_{ij}^3 \). Additionally, it uses different matrices for the self-mobility and for terms \( \propto 1/\alpha r_{ij}^3 \) instead of the unity matrix:
\[ M_{ij} = (6\pi \eta a)^{-1} \left\{ \frac{3\alpha a}{2\sqrt{\pi}} (1 + \hat{r}_i \hat{r}_j) + \frac{3}{2} a \alpha \pi^{-1/2} \exp(-a^2 r_{ij}^2) q(\mathbb{1} - 3\hat{r}_i \hat{r}_j) \right\}, \quad (i \neq j) \] (C.1a)
\[ M_{ii} = (6\pi \eta a)^{-1} Q_1, \quad (i = j) \] (C.1b)

with \( q = 5.59027 \) and matrices \( Q_1 \) and \( Q_3 \) [13],
\[ Q_1 = \begin{pmatrix} 1.3799554 & 0 \\ 0 & 1.3799554 \end{pmatrix} \] (C.2)
\[ Q_3 = \begin{pmatrix} -0.319658 & 0 \\ 0 & -0.319658 \end{pmatrix} \] (C.3)

reflecting the presence of a free interface (the additional scaling factor \( q \) and the matrix \( Q_1 \) may be derived from equation (8) in [13] by casting it into the Rotne–Prager form (equation (3a))).
As a consequence of that, the resulting summation formula involves more terms, since the different matrices avert cancellations. The corresponding analogue to equation (38) then reads

\[(6\pi \eta a)v_{i,\text{eff}} = Q_1 F_i - \frac{ax}{\sqrt{\pi}} \left( 3 + \frac{2}{3} qa^2 \alpha^2 Q_1 \right) F_i + \sum_{j=1}^{N} \left\{ \sum_{n} \left( 3 \frac{a}{2} \frac{\text{erfc}(aR_{jn})}{R_{jn}} [1 + \hat{R}_{jn} \hat{R}_{jn}] + \frac{qa^3 \alpha^2 e^{-a^2 R_{jn}^2}}{\sqrt{\pi}} \right) [Q_1 - 3 \hat{R}_{jn} \hat{R}_{jn}] \right\} + \sum_{j=1}^{N} \sum_{k \neq 0} \left\{ \cos(Kr_{ij}) \left( \frac{6a \pi}{L^2 K} \text{erfc} \left( \frac{K}{2a} \right) \right) \right\} F_j \]

and accordingly

\[(6\pi \eta a)v_{i,\text{eff}} = \sum_{j=1}^{N} \sum_{n} M^{(1)}_{\eta 2D}(R_{jn}) F_j + \sum_{j=1}^{N} \sum_{k \neq 0} M^{(2)}_{\eta 2D}(K) \cos(Kr_{ij}) F_j \]

\[= \left( Q_1 - 3 \pi^{-1/2} a x - \frac{2}{3} \pi^{-1/2} qa^3 \alpha^2 Q_1 \right) F_i \]

with the definitions:

\[M^{(1)}_{\eta 2D}(r) = 1 \left\{ \left( \frac{1}{2} \pi^{-1} + \frac{1}{2} qa^3 r^{-3} Q_1 \right) \text{erfc}(ar) + qa^3 \alpha r^{-2} \pi^{-1/2} \exp(-\alpha^2 r^2) Q_1 \right\} \]

\[= \hat{r} \left\{ \left( \frac{2}{12} \pi^{-1} - \frac{1}{3} qa^3 r^{-3} \right) \text{erfc}(ar) + (-3qa^3 \alpha r^{-2} + 3\alpha r - 2qa^3 \alpha^3) \pi^{-1/2} \exp(-\alpha^2 r^2) \right\} \]

for the real part of the tensor and

\[M^{(2)}_{\eta 2D}(k) = 1 \left\{ \left( 2a + \frac{1}{3} qa^3 k^2 (1 - Q_3) \right) \text{erfc} \left( \frac{k}{2a} \right) - \frac{2}{3} qa^3 \alpha k \pi^{-1/2} \exp \left( -\frac{k^2}{4a^2} \right) (\| - Q_3) \right\} \frac{3\pi}{L^2 k} \]

\[= \hat{k} \left\{ \left( a - \frac{1}{3} qa^3 k^2 \right) \text{erfc} \left( \frac{k}{2a} \right) + a \alpha^{-1} k \pi^{-1/2} \exp \left( -\frac{k^2}{4a^2} \right) \right\} \frac{3\pi}{L^2 k} \]

in Fourier space.

Appendix D. Ewald sum for the binary Rotne–Prager tensor

For binary mixtures of particles with radii \(a_i \in \{a_0, a_1\}\), we replace the particles radius \(a\) by \(a_i\), and within the sum over particles with radius \(a_i\), each factor \(a^3\) is replaced by \(\frac{4}{3} (a_i^2 + a_j^2)\) [29]:

\[M_{ij} = (6\pi \eta a_i)^{-1} \left\{ \frac{3}{4} a_i r_{ij}^{-1} (\| + \hat{r}_{ij} \hat{r}_{ij}) + \frac{a_i}{4} (a_i^2 + a_j^2) r_{ij}^{-1} (1 - 3\hat{r}_{ij} \hat{r}_{ij}) \right\}, \quad (i \neq j) \]

\[M_{ii} = (6\pi \eta a_i)^{-1}, \quad (i = j). \]
Carrying out this procedure for the previous results (equations (39)–(41)) leads to

$$(6\pi a_i) v_{\text{eff}} = \sum_{j=1}^{N} \sum_{n} M_b^{(1)}(R_{jn}) F_j + \sum_{j=1}^{N} \sum_{K \neq 0} M_b^{(2)}(K) \cos(Kr_{ij}) F_j$$

$$+ 1 \left( 1 - \frac{3}{2} \pi^{-1/2} a_i \alpha \right) \left( \frac{2}{3} \pi^{-1/2} a_i^3 \right) F_i,$$

\[ (D.2) \]

With the corresponding definitions

$$M_b^{(1)}(r) = 1 \left\{ \frac{3}{4} a_i r^{-1} + \frac{1}{4} a_i (a_i^2 + a_j^2) r^3 \right\} \text{erfc}(a r) + \frac{\alpha}{2} a_i (a_i^2 + a_j^2) r^{-2} \pi^{-1/2} \exp(-\alpha r^2)$$

$$+ \hat{F} \left\{ \frac{3}{4} a_i r^{-1} - \frac{3}{4} a_i (a_i^2 + a_j^2) r^3 \right\} \text{erfc}(a r)$$

$$+ \left( -\frac{3}{2} a_i (a_i^2 + a_j^2) r^{-2} + \frac{3}{2} a_i a_j - a_i (a_i^2 + a_j^2) a_j \right) \pi^{-1/2} \exp(-\alpha r^2)$$

\[ (D.3) \]

for the real part of the tensor and

$$M_b^{(2)}(k) = 1 \frac{2 a_i \text{erfc} \left( \frac{k}{2 a} \right)}{2 \ell^2 k} - \frac{\hat{k} \hat{k}}{\left( a_i - 1 a_i (a_i^2 + a_j^2) k^2 \right)} \text{erfc} \left( \frac{k}{2 a} \right)$$

$$+ a_i \alpha^{-2} k \pi^{-1/2} \exp \left( -\frac{k^2}{4 \alpha^2} \right) \frac{3 \pi}{2 \ell^2 k}$$

\[ (D.4) \]

for the summation in Fourier space.

References

[1] Rinn B, Zahn K, Maass P and Maret G 1999 Europhys. Lett. 46 537
[2] Pesché R, Kollmann M and Nägele G 2001 J. Chem. Phys. 114 8701
[3] Tanaka H and Araki T 2000 J. Chem. Phys. 112 7981
[4] Kollmann M, Hund R, Rinn B, Nägele G, Zahn K, König H, Maret G, Klein R and Dhont J K G 2002 Europhys. Lett. 58 919
[5] Leonardo R, Dietrich S, Domínguez A and Oettel M 2011 J. CHEM. PHYS. 135 104102
[6] Winter U and Geyer T 2009 J. CHEM. PHYS. 130 114905
[7] Zahn K, Mendez-Alcaraz J M and Maret G 1997 Phys. Rev. Lett. 79 175
[8] Swan J W and Brady J F 2007 Phys. Fluids 19 113306
[9] Yeh I C and Berkowitz M L 1999 J. CHEM. PHYS. 111 3155
[10] Pozrikidis C 1996 J. Eng. Math. 30 79
[11] Grzybowski A, Gwiazda E and Bródka A 2000 Phys. Rev. B 61 6706
[12] Porte M 2000 J. CHEM. PHYS. 112 164906
[13] Hernández-Ortiz J P, de Pablo J J and Graham M 2006 J. CHEM. PHYS. 125 164906
[14] Hernández-Ortiz J P, Pablo J J de and Graham M 2007 Phys. Rev. Lett. 98 140602
[15] Swan J W and Brady J F 2011 J. FLUID MECH. 687 254
[16] Zhai Y, de Pablo J J and Graham M 2012 J. CHEM. PHYS. 136 014901
[17] Oettel M and Dietrich S 2008 Langmuir 24 1425
[18] Winter U and Geyer T 2009 J. CHEM. PHYS. 130 114905
[19] García de la Torre J and Bloomfield V A 1977 Biopolymers 16 1747