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Three particle hydrodynamic interactions in suspensions

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We present an outline of a method to determine the components of the grand mobility matrix of a system of three hydrodynamically interacting spherical particles with arbitrary particle configuration. We present the results of a study of some components of the grand mobility matrix in the case of two special configurations. The results derived for one configuration, with the three particles on a line, are compared with some results from the literature. In the other configuration, we have put the particles on the corners of an equilateral triangle. Besides the study of the mobility matrix, we have derived virial expansions for the translational and rotational self-diffusion coefficients up to quadratic order in \( \varphi \), the volume fraction. Three particle contributions are incorporated in these expressions. We also present virial expansions, where three particle hydrodynamic interactions are included, for the translational and rotational sedimentation velocities. There are some results available in the literature in the case of the translational self-diffusion coefficient and the rotational sedimentation velocity, which are different from our results. We discuss these differences. As far as we know, the results of the three particle contribution to the virial expansions of the rotational self-diffusion coefficient and the rotational sedimentation velocity are new. All the virial expansions mentioned above are compared with experimental results and simulation data from the literature.

I. INTRODUCTION

In this article, we present the results of a study of three particle hydrodynamic interactions and the effects of these interactions on diffusion and sedimentation. The three-particle hydrodynamic interactions are studied by using the method to determine the grand mobility matrix of a system of \( N \) hydrodynamically interacting spherical particles, immersed in an unbounded fluid, presented by the authors in an earlier article, in the case \( N = 3 \) (this article is further abbreviated by I).\(^1\) In that paper we have presented virial expansions of the translational and rotational self-diffusion coefficients and the sedimentation velocities, where only two particle hydrodynamic interactions are included. Some of these results were already known and others were new such as, e.g., the virial expansion of the rotational counterpart of sedimentation \( \Omega_r/\Omega_0 \) [Eq. (1.6.29)]. We are now able to determine the corrections of these virial expansions caused by three particle hydrodynamic interactions. The results presented in this article can be compared with theoretical results on the one hand and with numerical and experimental data on the other hand. Before we present the way of obtaining our results, we give here a brief historical sketch of the area of interest, which is not claimed to be complete.

Recent developments in both numerical and theoretical research on the behavior of suspensions made it possible to improve the understanding of the transport and bulk properties of suspensions. In the area of numerical research, progress has been made by the development of methods to simulate systems of Brownian particles, called Stokesian dynamics. With this method, developed by Brady and Bossis,\(^2\) it became possible to study transport coefficients such as, e.g., translational and rotational self-diffusion coefficients, and bulk properties such as, e.g., sedimentation and effective viscosity, numerically. Some interesting results of Stokesian dynamics simulation, which we shall use to compare our results with, are presented in an article of Phillips, Brady, and Bossis.\(^3\) Many other authors have also presented results of simulation experiments on systems of Brownian particles to calculate transport coefficients of random hard sphere suspensions. Apart from this technique, one can calculate these transport coefficients numerically and results of Ladd will be used in this case to compare our theoretical results with.\(^4\) Apart from this numerical research, great efforts have also been made with theoretical studies of systems of hard spheres in an ambient fluid under low Reynolds number conditions. Besides the hydrodynamic pair interactions between the spheres, one has considered three and more particle hydrodynamic interactions. In the 1950s, Kynch used the reflection method to study hydrodynamic interactions between the particles of clusters of three and four spheres in a viscous fluid.\(^5\) Some decades later, in the early 1980s, Mazur and van Saarloos presented a general scheme to determine the components of the grand mobility matrix of a system of \( N \) spherical particles immersed in an unbounded fluid.\(^6\) The expressions determined by those authors are power expansions in \( a/R \), with \( a \) the particle radius and \( R \) typical interparticle distances. They derived explicit expressions for the components of the mobility matrix up to order \( R^{-7} \) and up to this order hydrodynamic interactions between two, three, and four particles only contribute to these expressions. Beenakker used the method of Mazur and van Saarloos to determine virial expansions of the short time self-diffusion coefficient, sedimentation, and high frequency
effective viscosity.\textsuperscript{7-9} In the same articles, he presented numerical results for the transport coefficients described above using an expansion in density fluctuation correlation functions. These results, taking into account many-particle hydrodynamic interactions, correspond with experimental and simulation data very well. A disadvantage of these results is the fact that they are of numerical type instead of an expression in terms of, e.g., volume fraction $\varphi$. In this way, it is not possible to estimate the relative importance of two, three, and more particle interactions, respectively. For a concise summary, we refer to a short review article by Mazur.\textsuperscript{10} Felderhof\textsuperscript{11} has given an outline how to proceed, using a method to study two particle hydrodynamic interactions,\textsuperscript{12,13} to study the more general $N$ particle problem. In another approach, Cichocki and Felderhof have presented a cluster expansion method to study the $N$ particle problem, but as far as we know, they used this method, in the case of hydrodynamic interactions in suspensions, in the two-body approximation only.\textsuperscript{14} Finally, we want to emphasize the work of Muthukumar et al. In articles of Muthukumar and Freed, a theory to study many-particle hydrodynamic interactions is expounded.\textsuperscript{15,16} Using this theory, Jones, Muthukumar, and Cohen were able to determine virial expansions of the cooperative and self-friction coefficients.\textsuperscript{17} We shall compare these expressions with some of our results.

In this article, we present in Sec. II the set of linear equations necessary to obtain the components of the grand mobility matrix. In Sec. III, we reformulate this set of equations for numerical purpose. We give the results of a study concerning the behavior of some components of the grand mobility matrix, in the case of two special configurations of the three particle cluster, in Sec. IV. A theoretical account of obtaining the virial expansions of some transport coefficients will be given in Sec. V as well as the final results. We end this article with some concluding remarks.

II. THE SET OF LINEAR EQUATIONS

Consider a system of $N$ spherical particles, all with the same radius $a$, immersed in an incompressible, unbounded fluid. We suppose that the fluid is at rest at infinity. The particles move with velocities $U_i$ and rotate with angular velocities $\Omega_i$ ($i \in \{1, \ldots, N\}$). They have position vectors $R_i$ with respect to the origin $O$. We can neglect the nonlinear term in the Navier–Stokes equation because we assume that the Reynolds number of the fluid motion induced by the particles is small $Re \ll 1$. The equations of motion of the fluid, the linearized, incompressible Navier–Stokes equations (later abbreviated by N.S. equations), have the following form:

\begin{align}
\eta_0 \nabla^2 v(r) - \nabla p(r) &= 0, \quad (2.1) \\
\nabla v(r) &= 0. \quad (2.2)
\end{align}

Here $v(r)$ is the fluid velocity, $p(r)$ is the pressure, and $\eta_0$ is the shear viscosity. We use stick boundary conditions. This means for $v(r)$,

\[ v(r) = U_i + \Omega_i \times (r - R_i), \quad \text{with } \forall S_i, \quad i \in \{1, \ldots, N\}, \quad (2.3) \]

where $S_i$ is the surface of particle $i$. To preserve linearity, we treat the position vector $R_i$ as independent of the time $t$. This linearization is equivalent to omitting a term of order $(U_i \cdot \nabla)v(r)$ in the Navier–Stokes equations, which is justified because of the smallness of the Reynolds number. The smallness of $Re$ implies that the characteristic time of particle displacement is much larger than the viscous time $a^2/\nu$, with $\nu$ the kinematic viscosity of the fluid. There is some literature on this point, e.g., in an article of Hauge and Martin-Lof.\textsuperscript{18} After calculation of $v(r)$ and $p(r)$, we are able to determine the pressure tensor $\Pi(r) = \rho(r) I - 2\eta_0 [\nabla v(r)]^2$. The pressure tensor can be used to determine the force $F_i$ and the torque $T_i$ exerted by the fluid on particle $i$. These quantities can be defined, in the case of spherical particles, in the following way:

\[ F_i = -a^2 \int_{|r| = a} \Pi_i(r) d\Omega_i, \]

\[ T_i = -a^3 \int_{|r| = a} [\hat{e} \times \Pi_i(r)] d\Omega_i, \quad (2.4) \]

with $d\Omega_i$ the element of solid angle with respect to the center of particle $i$ and $\hat{e}$, the radial unit vector in a spherical coordinate system. In these expressions, we have used

\[ \Pi_i(r)(r) \equiv \Pi(r) \hat{e}_i - \eta_0 \left( \frac{\partial}{\partial r} - \frac{1}{r} \right) v(r) \]

\[- \frac{\eta_0}{r} \nabla [r v(r)]. \quad (2.5) \]

We introduce furthermore the grand mobility matrix, which is defined by

\[ \begin{pmatrix} U \\ \Omega \end{pmatrix} = - \begin{pmatrix} \mu^n U \\ \mu^n \Omega \end{pmatrix} (F), \quad (2.6) \]

where we have used the shorthand notation $U = (U_1, \ldots, U_N)$ etc. $\mu^n$ etc. are matrices with, e.g.,

\[ \mu^n = \begin{pmatrix} \mu^n_{11} & \cdots & \mu^n_{1N} \\ \vdots & \ddots & \vdots \\ \mu^n_{N1} & \cdots & \mu^n_{NN} \end{pmatrix}, \quad (2.7) \]

where $\mu^n_{ij}$ is a mobility tensor. The upper indices $t$ and $r$ refer to “translational” and “rotational,” respectively. The dot in Eq. (2.6) stands for a contraction of tensors. The components of the grand mobility matrix satisfy the following symmetry relations:\textsuperscript{13}

\[ \mu^n_{ij,ab} = \mu^n_{ji,ba}, \quad \mu^n_{ij,ab} = \mu^n_{ji,ba}, \quad \mu^n_{ij,ab} = \mu^n_{ji,ba}. \quad (2.8) \]

We now restrict ourselves to three particle hydrodynamic interactions. The fluid velocity $v(r)$, created if three spherical particles move in a fluid, can be seen as a sum of three contributions because of the linearity of the N.S. equations. The contributions are scattered velocity fields defined with respect to the origins of the particles. These velocity fields can be expanded in terms of basic solutions. We have expounded the set of basic solutions in article I, where we have used the basic functions introduced by Schmitz and Felderhof.\textsuperscript{12} For a review, we refer the reader to article I. With these basic functions, we can write for the velocity field $v(r)$,
\[ \mathbf{v}(\mathbf{r}) = \sum_{\vec{m}, \vec{n}}^3 \left[ \alpha'_{\vec{m}, \vec{n}} \mathbf{v}(\mathbf{r}_i) + \beta'_{\vec{m}, \vec{n}} \mathbf{v}(\mathbf{r}_j + \mathbf{R}_i) \right] \\
+ \sum_{\vec{j} \neq \vec{i}}^3 \left[ \alpha'_{\vec{m}, \vec{n}} \mathbf{v}(\mathbf{r}_i + \mathbf{R}_j) \right] \\
+ \beta'_{\vec{m}, \vec{n}} \mathbf{v}(\mathbf{r}_j + \mathbf{R}_i) \right] + \gamma'_{\vec{m}, \vec{n}} \mathbf{v}(\mathbf{r}_j + \mathbf{R}_i), \tag{2.9} \]

where \( \mathbf{r}_i = \mathbf{r} - \mathbf{R}_i \), and \( \mathbf{R}_j = \mathbf{R}_j - \mathbf{R}_i \), the position vector of the center of particle \( i \) with respect to the center of particle \( j \).

One can obtain in an analogous way an expression for the pressure field \( p(\mathbf{r}) \). The way to determine the set of linear algebraic equations is expounded in I and the result is:

\[ \int_{[r, l, \alpha]} \mathbf{v}(\mathbf{r}) \cdot \mathbf{A}_{\vec{p}}(\theta, \phi) d\Omega_i = p(p+1) \mathbf{a} - \frac{m_{pq}}{(p+q)!} \mathbf{a} - \frac{1}{2} p(p+1)(2p+1) \mathbf{a} - \frac{m_{pq}}{(p+q)!} \mathbf{a} - \frac{p}{2} - \frac{1}{2} p(p+1) \sum_{\vec{j} \neq \vec{i}}^3 \left[ \frac{1}{l(l+1)} \beta_{\vec{m}, \vec{n}} \mathbf{M}_{\vec{m}, \vec{n}}^{l \vec{n}, \vec{m}} \right] \\
+ \frac{1}{2} i(p+1) \sum_{\vec{j} \neq \vec{i}}^3 \left[ \frac{1}{l(l+1)} \mathbf{M}_{\vec{m}, \vec{n}}^{l \vec{n}, \vec{m}} \right] \tag{2.10} \]

\[ \int_{[r, l, \alpha]} \mathbf{v}(\mathbf{r}) \cdot \mathbf{B}_{\vec{p}}(\theta, \phi) d\Omega_i = p(p+1) \mathbf{a} - \frac{m_{pq}}{(p+q)!} \mathbf{a} - \frac{1}{2} p(p+1)(2p+1) \mathbf{a} - \frac{m_{pq}}{(p+q)!} \mathbf{a} - \frac{p}{2} - \frac{1}{2} p(p+1) \sum_{\vec{j} \neq \vec{i}}^3 \left[ \frac{1}{l(l+1)} \gamma_{\vec{m}, \vec{n}} \mathbf{M}_{\vec{m}, \vec{n}}^{l \vec{n}, \vec{m}} \right] \\
+ \frac{1}{2} i(p+1) \sum_{\vec{j} \neq \vec{i}}^3 \left[ \frac{1}{l(l+1)} \mathbf{M}_{\vec{m}, \vec{n}}^{l \vec{n}, \vec{m}} \right] \tag{2.11} \]

\[ \int_{[r, l, \alpha]} \mathbf{v}(\mathbf{r}) \cdot \mathbf{C}_{\vec{p}}(\theta, \phi) d\Omega_i = a - \frac{1}{2} p(p+1)(2p+1) \mathbf{a} - \frac{m_{pq}}{(p+q)!} \mathbf{a} - \frac{1}{2} p(p+1) \sum_{\vec{j} \neq \vec{i}}^3 \left[ \frac{1}{l(l+1)} \gamma_{\vec{m}, \vec{n}} \mathbf{M}_{\vec{m}, \vec{n}}^{l \vec{n}, \vec{m}} \right] \\
+ \frac{1}{2} i(p+1) \sum_{\vec{j} \neq \vec{i}}^3 \left[ \frac{1}{l(l+1)} \mathbf{M}_{\vec{m}, \vec{n}}^{l \vec{n}, \vec{m}} \right] \tag{2.12} \]

where \( i \) is the imaginary unit. In these expressions, we have used the following shorthand notations:

\[ m_{pq} = \left[ \frac{4\pi}{(2p+1)(2q+1)} \right]^{1/2}, \tag{2.13} \]

and

\[ M_{\vec{m}, \vec{n}}^{l \vec{n}, \vec{m}} = (-1)^{p+q} \frac{(l+p-m+q)!}{n_{\vec{m}}(l-m)!} \times \mathbf{Y}_{l+p-m-q}^{m-p} \left( \xi_y, \eta_y \right) \frac{R_y^{l+p+1}}{R_y^{l+p+1}}, \tag{2.14} \]

with \( \xi_y \) and \( \eta_y \) the polar angles of the vector \( \mathbf{R}_y \). This expression is obtained from the general form of the Hobson formula (1.5.5). With this formula, one can express a solid spherical harmonic, defined with respect to an origin \( O \), in terms of solid spherical harmonics defined with respect to another origin.

The left-hand sides of Eqs. (2.10)–(2.12) are also equal with those presented in I [see Eqs. (1.5.8)–(1.5.10)].

The constants \( n_{10} \) and \( n_{11} \) are special cases of the expression \( m_{pq} \) [Eq. (2.13)]. Combining Eqs. (2.10), (2.11), and (2.12) with Eqs. (2.15), (2.16), and (2.17), respectively, we obtain an infinite set of linear equations of the coefficients \( \{\alpha'_{\vec{m}, \vec{n}}, \beta'_{\vec{m}, \vec{n}}, \gamma_{\vec{m}, \vec{n}}\} \) with \( ie \{1,2,3\} \). In I, we have explained the way one can determine the coefficients which one is interested in. Furthermore, we have expressed the force \( \mathbf{F}_i \) and...
torque $T_i$, in terms of some coefficients. These expressions are

$$
\begin{align*}
F_i &= -\frac{3}{2} \eta \left[ n_{11} (\gamma_i,_{-1} - \gamma_i,_{1}) \hat{e}_x \\
&\quad - in_{11} (\gamma_i,_{-1} + \gamma_i,_{1}) \hat{e}_y + 2n_{10} \gamma_i,_{0} \hat{e}_z \right], \\
T_i &= -\frac{3}{2} \eta \left[ n_{11} (\beta_i,_{-1} - \beta_i,_{1}) \hat{e}_x \\
&\quad - in_{11} (\beta_i,_{-1} + \beta_i,_{1}) \hat{e}_y + 2n_{10} \beta_i,_{0} \hat{e}_z \right].
\end{align*}
$$

(2.18) (2.19)

III. REFORMULATION OF THE SET OF LINEAR EQUATIONS FOR NUMERICAL CALCULATIONS

In Sec. II, we have determined a set of linear equations (2.10)-(2.12) appropriate to describe the problem of three hydrodynamically interacting spherical particles in a viscous fluid. We have obtained this result starting with an arbitrary configuration of the three particles, but simplify it considerably by introducing a special coordinate system without affecting the generality of the set of linear equations. We number the particles and put particle 1 in the origin of a coordinate system, particle 2 on the negative z axis, and particle 3 in the $xz$ plane. Consequently, we have $\xi_{12} = 0$, $\eta_{13} = \pi$, and $\eta_{23} = \pi$. We furthermore use the relations $\xi_{ij} = \pi - \xi_{ij}$ and $\eta_{ij} = \eta_{ij} + \pi$. This procedure is in principle equivalent to the one used in I, where we have put both particles on the z axis to describe two particle hydrodynamic interactions. In that special case, we were able to decouple the set of linear equations with respect to the azimuthal indices. This simplification is of course not possible in the three particle case. An exception should be made if the third particle is also on the z axis. Introduce furthermore the new coefficients $\{A_{m,n}^{\pm \ell}, B_{m,n}^{\pm \ell}, C_{m,n}^{\pm \ell}\}$,

$$
\begin{align*}
A_{m,n}^{\pm \ell} &= \frac{(-1)^{m+\ell}}{n_{m,n}^{l+2}} \left[ A_{m,n}^{\ell} \pm (-1)^{m} \alpha_{m,n}^{l} \right], \\
B_{m,n}^{\pm \ell} &= \frac{(-1)^{m+\ell}}{n_{m,n}^{l+1}} \left[ B_{m,n}^{\ell} \pm (-1)^{m} \beta_{m,n}^{l} \right], \\
C_{m,n}^{\pm \ell} &= \frac{(-1)^{m+\ell}}{n_{m,n}^{l}} \left[ C_{m,n}^{\ell} \pm (-1)^{m} \gamma_{m,n}^{l} \right].
\end{align*}
$$

(3.1a) (3.1b) (3.1c)

We see from these relations that it is sufficient to use the plus coefficients $A_{m,n}^{\ell}$ etc., with $m>0$ and the minus coefficients $A_{m,n}^{-\ell}$ etc. with $m>0$ only. With this new set of coefficients, we are able to derive two simplified sets of linear equations, which are independent of each other. With one set, we can study the components of the grand mobility matrix relating translations of the particles in the $z$ and $x$ direction and rotations of the particles in the $y$ direction, on the one hand, with the forces exerted by the fluid on the particles in the $z$ and $x$ directions and the torques exerted on the particles in the $y$ direction, on the other hand. In the other set, the role of translations and rotations is interchanged as is the case with the forces and torques. We do not present the total derivation of the rewritten set of linear equations, but only the final result which comes to

\begin{align*}
\int_{r_{12}} \nu(r) \cdot [A_{pq}^{\ell}(\theta,_{i},\varphi,_{i}) + (-1)^{n}A_{pq}^{\ell-1}(\theta,_{i},\varphi,_{i})] d\Omega_{i} \\
&= -\frac{3}{2} \delta_{n,1} (\delta_{t,1} U_{tx} + 2\delta_{t,0} U_{ta}) = C_{pq}^{\ell} + \sum_{j=1}^{3} \frac{1}{(p+1)(l+1)(2l+1)} \left[ \frac{1}{n_{m,n}^{l+1}} \sum_{m=0}^{l} \left( P_{m,n}^{+\ell} x_{ij}^{l+1} + r_{m,n}^{+\ell} C_{m,n}^{+\ell} \right) \right. \\
&\quad \times \sum_{m=0}^{l} l(l+1) Q_{m,n}^{+\ell} x_{ij}^{l+1} + r_{m,n}^{+\ell} B_{m,n}^{+\ell} + \frac{1}{2(p+1)} \sum_{m=0}^{l} l(2l-1) P_{m,n}^{+\ell} x_{ij}^{l+1} + r_{m,n}^{+\ell} C_{m,n}^{+\ell} \\
&\quad - \frac{(2p+1)}{2(p+1)} \sum_{m=0}^{l} l(2l-1) P_{m,n}^{+\ell} x_{ij}^{l+1} + r_{m,n}^{+\ell} C_{m,n}^{+\ell} \left. \right] \\
&= \frac{1}{2} (p+1) (2p-1) C_{pq}^{\ell} - \frac{3}{2p+3} \sum_{j=1}^{3} l(l+1) P_{m,n}^{+\ell} x_{ij}^{l+1} + r_{m,n}^{+\ell} C_{m,n}^{+\ell},
\end{align*}

(3.2) (3.3)

\begin{align*}
\int_{r_{ij}} \nu(r) \cdot [C_{pq}^{\ell}(\theta,_{i},\varphi,_{i}) + (-1)^{n}C_{pq}^{\ell-1}(\theta,_{i},\varphi,_{i})] d\Omega_{i} \\
&= 2\alpha \delta_{n,1} \delta_{t,1} \Omega_{ij} = B_{pq}^{-\ell} - \frac{1}{2p+3} \sum_{m=0}^{l} \left[ P_{m,n}^{-\ell} x_{ij}^{l+1} + r_{m,n}^{-\ell} B_{m,n}^{-\ell} + \frac{1}{2} (2l-1) Q_{m,n}^{-\ell} x_{ij}^{l+1} + r_{m,n}^{-\ell} C_{m,n}^{-\ell} \right].
\end{align*}

(3.4)


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In the derivation of these equations, which can be done straightforwardly, we have used Eqs. (2.15)-(2.17). Furthermore, we have introduced the following shorthand notations:

\[
P_{lm;pq}^{\pm} = \left( -\frac{1}{2} \delta_{m,0} \right) \frac{(-1)^{l+p+m+q}}{(p+q)!} n_{lm} R_{ij}^{l+p+1} [M_{lm;pq}^{\pm} \pm (-1)^{m}M_{lm;pq}^{\mp}],
\]

\[
Q_{lm;pq}^{\pm} = \left( -\frac{1}{2} \delta_{m,0} \right) \frac{(-1)^{l+p+m+q}}{(p+q)!} n_{lm} R_{ij}^{l+p}[ (lq+mp)M_{lm;pq}^{\pm -1,4} \pm (-1)^{m}(lq-mp)M_{lm;pq}^{\pm -1,4}],
\]

\[
R_{lm;pq}^{\pm} = \left( -\frac{1}{2} \delta_{m,0} \right) \frac{(-1)^{l+p+m+q}}{(p+q)!} n_{lm} R_{ij}^{l+p+1} [\{(lq+mp)[2(lq+mp)-(m+q)]} - (lp+mq)(l+p-1)]M_{lm;pq}^{\pm -2,q} \pm (-1)^{m}[(lq-mp)[2(lq+mp)+(m-q)]}
\]

\[
- (lp-mp)(l+p-1)]M_{lm;pq}^{\pm -2,q} \right].
\]

It is possible to solve the set of linear equations (3.2)-(3.4) partially if we express all coefficients \(A_{lm;pq}^{i,l}, B_{lm;i}, C_{lm;i}^{i,l}, \) with \(i > 1, \) for \(i \in \{1,2,3\}\) in terms of the coefficients \(B_{lm;i}, C_{lm;i}^{1,l}, \) and \(C_{lm;i}^{1,l}.\) This procedure resembles the one used in article I. We can express the remaining coefficients in terms of components of the force \(F_i\) and torque \(T_j\) by using Eqs. (2.18) and (2.19)

\[
B_{1l;i} = \frac{1}{4\pi \eta_0 a^2} T_{ij}, \quad C_{10;i} = \frac{1}{2\pi \eta_0 a} F_{ij}, \quad C_{11;i} = \frac{1}{4\pi \eta_0 a} F_{ij}.
\]

Finally, we obtain the following relation among \(U_x, U_y, \) and \(\Omega_x\) on the one hand and \(F_x, F_y, \) and \(T_y\) on the other hand, with \(U_x = (U_{1x}, U_{2x}, U_{3x}) \) etc.

\[
\begin{pmatrix}
U_x
\end{pmatrix} = \left( \begin{array}{ccc}
\mu_{x1} & \mu_{x2} & \mu_{x3} \\
\mu_{z1} & \mu_{z2} & \mu_{z3} \\
\mu_{y1} & \mu_{y2} & \mu_{y3}
\end{array} \right) \begin{pmatrix}
F_x \\
F_y \\
T_y
\end{pmatrix}.
\]

In an analogous way, using the shorthand notations defined above [Eqs. (3.5)-(3.7)], we obtain

\[
\frac{1}{p(p+1)n_{pq}} \int_{\Omega} v(r) \cdot [A_{pq}^{i,l}(\theta_i, \varphi_i) - (-1)^q A_{pq; -q}^{*}(\theta_i, \varphi_i)] d\Omega,
\]

\[
= \frac{1}{2} \delta_{\rho,1} \delta_{q,1} U_{ij} = C_{pq}^{i,l} \frac{2(2p+1)}{p(p+1)} \sum_{m>1} \frac{1}{l(l+1)} \frac{Q_{lm;pq}^{\pm} X_{ij}^{l+p+1} A_{lm;i}^{j} + (2p+1)}{p(p+1)}
\]

\[
\times \sum_{m>1} \frac{l(2l-1)}{(2l+2p-1)} R_{lm;pq}^{\pm} \frac{1}{2p+3} \sum_{m>1} \frac{2l+2p-1}{l(l+1)} \frac{2p+1}{2p+1} \frac{R_{lm;pq}^{\pm} X_{ij}^{l+p+1} C_{lm;i}^{j}}{2p+1},
\]

\[
A_{pq; -q}^{i,l} = \frac{1}{2} \frac{2p+1}{(2p+1)(2p-1)} \sum_{m>1} \frac{1}{l(l+1)} \frac{1}{2p+3} \sum_{m>1} \frac{2l+2p-1}{l(l+1)} \frac{1}{2p+1} \frac{R_{lm;pq}^{\pm} X_{ij}^{l+p+1} C_{lm;i}^{j}}{2p+1},
\]

\[
i \frac{(-1)^{p+q+1}}{n_{pq}} \int_{\Omega} v(r) \cdot [C_{pq; -q}^{i,l}(\theta_i, \varphi_i) + (-1)^q C_{pq; -q}^{*}(\theta_i, \varphi_i)] d\Omega
\]

\[
= 2ia \delta_{\rho,1} \delta_{q,1} \Omega_{1x} + 2\delta_{q,1} \Omega_{1z}
\]

\[
= B_{pq}^{i,l} \frac{1}{3} \sum_{j\neq 1} \sum_{m>1} \frac{1}{l(l+1)} \frac{1}{2p+3} \sum_{m>1} \frac{2l+2p-1}{l(l+1)} \frac{R_{lm;pq}^{\pm} X_{ij}^{l+p+1} B_{ij}^{i,l} + (2p-1)}{2p+1} \frac{R_{lm;pq}^{\pm} X_{ij}^{l+p+1} C_{lm;i}^{j}}{2p+1}.
\]
In the way described above, we are able to solve partially this set of linear equations. Using the relations

$$B_{1i}^+ = \frac{i}{2\pi \eta_0 a^3} \mathcal{T}_{ix}, \quad B_{i1}^+ = \frac{i}{4\pi \eta_0 a^3} \mathcal{T}_{ix},$$

$$C_{1i}^+ = \frac{i}{4\pi \eta_0 a} F_{iy},$$  \hspace{1cm} (3.13)

we can present the final result in the following form:

$$\begin{pmatrix} \Omega_x \\ \Omega_z \\ \bar{U}_p \end{pmatrix} = - \begin{pmatrix} \mu_{xx}^T \\ \mu_{zx}^T \\ \mu_{xx}^T \\ \mu_{zz}^T \\ \mu_{xy}^T \\ \mu_{yx}^T \end{pmatrix} \begin{pmatrix} T_x \\ T_y \\ T_z \end{pmatrix}. \hspace{1cm} (3.14)$$

We have made a computer program in FORTRAN-77 to calculate the components of the grand mobility matrix (represented by Eqs. (3.9) and (3.14)) for arbitrary particle configurations. This computer program is available from the authors upon request.

### IV. TWO SPECIAL CONFIGURATIONS

We shall now present the results of a study of the behavior of some components of the grand mobility matrix in the case of two special configurations. In the first configuration, we put the three particles on the z axis of a coordinate system with origin at the center of particle 1. Particle 2 is placed between the two other particles. This is a suitable configuration because there are some numerical results available to compare our results with. In the second configuration, we put the three particles on the corners of an equilateral triangle. This configuration has the property that the three particles can touch each other simultaneously. There are many other configurations possible which are suitable to study, e.g., configurations with a very pronounced three particle contribution to the components of the grand mobility matrix. However, we have to restrict ourselves and therefore we pay attention to the two symmetric configurations described above only.

We consider now the first configuration with

<table>
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### TABLE I. Dimensionless translational mobilities of three particle clusters (the particles on a line) as a function of interparticle distance and upper limit. L3—pure three particle contribution; $\infty$—limiting values obtained by our method; Ladd—results obtained by Ladd (Ref. 19); PE—contribution according to Eqs. (4.4).
Use of Eq. (4.1) in the set of linear equations results in a decoupling of this set concerning the azimuthal indices [see Eq. (2.14)]. This decoupling procedure is comparable to the one used in I. We use it to simplify the computer program, necessary to solve the set of linear equations, considerably. The set of linear equations solved allows us to determine the components of the grand mobility matrix. Formally, we have an infinite set of linear equations. We can avoid the problem of solving an infinite set of linear equations by introducing an upper limit for the allowed values of \( l \) and \( p \), i.e., \( l_{max} = p_{max} = L \). With this upper limit, it is possible to solve the set of linear equations. This restriction is nothing but the assumption that all the coefficients \( A^{l,m}_i, B^{l,m}_i, \) and \( C^{l,m}_i \) are zero for \( l > L \). With the upper limit \( L \), we have 9L linear equations with the same number of unknown coefficients and can calculate the so-called \( L \)-th order solution of the set of linear equations. In the present problem, we confine ourselves to the presentation of the components of the mobility matrices \( \mu_{l}^{zz} \) and \( \mu_{zz}^{zz} \). Using symmetry arguments, we see that \( \mu_{zz}^{zz} = 0, \mu_{zz}^{zz} = 0, \mu_{zz}^{zz} = 0, \) and \( \mu_{zz}^{zz} = 0 \) (more mobility matrices are zero in this special case, but they are of no significance to us). Using Eqs. (3.9) and (3.14), we obtain, respectively,

\[
U_z = - \mu_{zz}^{zz} F_z, \quad \Omega_z = - \mu_{zz}^{zz} T_z, \tag{4.2}
\]

where we used the shorthand notations \( U_z = (U_{1z}, U_{2z}, U_{3z}) \) etc. We present our results in Table I. The rows labeled with \( L = \infty \) represent limiting values of the components of the mobility matrix resulting from our method. We also give the results obtained by Ladd.\(^{19}\) We have used his limiting results only; he obtained them by a numerical implementation of the work of Mazur and van Saarloos.\(^{6}\) In this table, \( \mu_{zz}^{zz} \equiv 6 \pi \eta_0 a \mu_{zz}^{zz} \). We can conclude that there are differences between our results and those of Ladd in the case of touching spheres \( |R_{1z}| = 2a \). The origin of this difference is the fact that Ladd used a low order moment approximation to determine his results. In this case, it is necessary to evaluate the calculations with large \( L \) in order to obtain reasonable results (in our case, up to \( L = 100 \)). Furthermore, we can conclude that, if \( |R_{1z}| = 2a \), the components of \( \mu_{zz}^{zz} \) tend to the same value, which we denote by \( \alpha \) and

\[
U_z = - \alpha \sum_{j=1}^{3} F_{zj}, \quad \Omega_z = - \alpha \sum_{j=1}^{3} T_{zj}, \tag{4.3}
\]

where \( \alpha \approx 0.649 \). This approximate value is obtained by considering the numerical data up to \( L = 100 \) and by assuming that the components of \( \mu_{zz}^{zz} \) should be equal, which will be argued below. This leads to an upper and lower limit for \( \alpha \), 0.6485 \(< \alpha < 0.6495 \). In the two particle problem, we can see the same situation as in the case of \( \mu_{zz}^{zz} \), with \( \alpha \approx 0.7750 \) (see, e.g., Refs. 1 and 19). This can be explained as follows: if a force \( F \), in the \( z \) direction, is exerted on particle 1, that particle will translate with velocity \( U \), in the \( z \) direction too, but the second particle will also translate with this velocity \( U \). This behavior is easy to understand if the force on particle 1 is in the direction of particle 2 because the touching particles are supposed to be hard spheres. That is why we find the same behavior if the force is in the opposite direction is a direct consequence of the stick boundary conditions [Eq. (2.3)]. If the first particle tends to move away, a large fluid velocity difference arises in a small region of space between the surfaces of the two particles.\(^{20}\) This difference can be canceled if the second particle translates with the first. The three particle case described above can be explained in the same way, but it is also necessary to use the symmetry relations for the mobility matrix \( \mu_{zz}^{zz} \) to understand the equivalence of the three particles, although the outer particles see a different environment in comparison to the particle between them. The results in Table I in the columns labeled with P3 represent the contribution of pure three particle hydrodynamic interactions to \( \mu_{1,zz}^{zz} \) and \( \mu_{2,zz}^{zz} \). These results, we see that three particle interactions are important, especially for small interparticle distances, and that the convergence behavior is slow. Finally, in Table I, in the row denoted by PE, we have presented the pure three particle contributions obtained from the first term of the power expansion in inverse interparticle distances, which has the following form:\(^{6}\)

\[
6 \pi \eta_0 a \mu_{1,zz}^{zz} = - \frac{75}{8} x_{1z}^2 x_{2z} x_{3z} \xi_1 [1 - 3 \xi_2^2] [1 - 3 \xi_3^2] + 6 \xi_2^2 \xi_3^2 + 6 \xi_1 \xi_2 \xi_3] = - \frac{75}{8} x_{1z}^2, \tag{4.4a}
\]

\[
6 \pi \eta_0 a \mu_{2,zz}^{zz} = - \frac{75}{8} x_{1z}^2 x_{2z} x_{3z} \xi_2 [1 - 3 \xi_1^2] [1 - 3 \xi_3^2] + 6 \xi_1^2 \xi_3^2 + 6 \xi_1 \xi_2 \xi_3] = \frac{75}{16} x_{1z}^2. \tag{4.4b}
\]
### TABLE II. Dimensionless rotational mobilities of three particle clusters (the particles on a line) as a function of interparticle distance and upper limit L. P3—pure three particle contribution; ∞—limiting values obtained by our method.

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### TABLE III. Dimensionless translational mobilities of three particle clusters (equilateral triangle) as a function of interparticle distance and upper limit L. P3—pure three particle contribution; ∞—limiting values obtained by our method; PE—contribution according to the general form of Eqs. (4.4).

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<th>μ₂₂,xx</th>
<th>μ₃₃,xx</th>
<th>P3</th>
<th>μ₁₁,zz</th>
<th>μ₂₂,zz</th>
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We now consider a more complicated problem with the third particle in the $xz$ plane. The decoupling of the set of linear equations disappears and it becomes more difficult to solve the set of linear equations for large $L$. In the case described above, we had to solve $9L$ linear equations, but in general, one has to solve $9L(L + 2)$ linear equations. It will be obvious that the number of linear equations to be solved increases sharply with the increase of $L$. Consequently, we have a numerical upper limit for $L$ itself. Despite this limitation, we are able to study the general problem reasonably well. Suppose the following situation:

$$| \mathbf{R}_{12} | = | \mathbf{R}_{13} | = | \mathbf{R}_{23} | , \quad \xi_{12} = 0 , \quad \xi_{13} = \frac{1}{3} \pi , \quad \xi_{23} = \frac{3}{4} \pi .$$

In Tables III and IV, we present the results of $\mu^{\text{am}}_{11, \alpha \alpha}$ and $\mu^{\text{am}}_{11, \alpha \alpha}$, respectively, with $\alpha \in \{x, y, z\}$ for several values of $| \mathbf{R}_{12} |$. It is not necessary to study the components of the mobility matrices $\mu^{\text{am}}$ and $\mu^{\text{am}}$ for $i = 2$ or $3$ because they are related to those presented in both tables. The values in the columns labeled with $P3$ are again the pure three particle contributions, the rows labeled with $L = \infty$ represent limiting results obtained by our method, and the values in the rows denoted by $\text{PE}$ are determined with Eq. (4.4a). This expression gives, in our special configuration, results for higher. The conclusion here is that three particle hydrodynamic interactions cannot be accounted for by a single term. This becomes especially clear if we consider nearly touching spheres. Mazur and van Saarloos have presented some expressions for $\mu^{\text{am}}_{11, \alpha \alpha}$ and $\mu^{\text{am}}_{11, \alpha \alpha}$. These expressions have the following inverse interparticle distance dependence, with $R$ representing the interparticle distances, all equal in the present case:

$$\mu^{\text{am}}_{11,xx} = O(R^{-9}) , \quad \mu^{\text{am}}_{11,yy} = O(R^{-9}) , \quad \mu^{\text{am}}_{11,zz} = O(R^{-7}) , \quad \mu^{\text{am}}_{11,\alpha \alpha} = O(R^{-8}) , \quad \alpha \in \{x, y, z\} .$$

This behavior is not present in Tables III and IV, as we may expect, keeping in mind the same kind of problems in the two particle case. So it is obvious that we have to take into account many more terms of the power expansion in inverse interparticle distances. It should be noted that our method gives results which are not expressed in terms of single power expansions. See in this context the discussion in article I. It is remarkable that for nearly touching spheres, the rotational mobilities are more influenced by the pure three particle hydrodynamic interactions than the translational mobilities.

We want to point out that it could be interesting to work out these three particle problems by using algebraic computer programs. The final expressions of the components of the grand mobility matrix are then expressed as a fraction with both the numerator and the denominator described in terms of spherical harmonics and inverse typical interparticle distances.

Finally, we want to discuss some results for sedimentation of this system of particles with the special configuration introduced above. Durlofsky, Brady, and Bossis have discussed this problem, also using some unpublished results of Kim (1985). We consider sedimentation of this three particle cluster in the $y$ direction, being the configuration appro-

### Table IV. Dimensionless rotational mobilities of three particle clusters (equilateral triangle) as a function of interparticle distance and upper limit $L$. $P3$—pure three particle contribution; $\infty$—limiting values obtained by our method.

<table>
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<th>$L$</th>
<th>$\mu^{\text{am}}_{11,xx}$</th>
<th>$\mu^{\text{am}}_{11,yy}$</th>
<th>$\mu^{\text{am}}_{11,zz}$</th>
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<td>0.767 50</td>
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<td>0.716 39</td>
<td>0.608 53</td>
<td>0.787 31</td>
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<td>15</td>
<td>0.682 13</td>
<td>0.561 13</td>
<td>0.758 35</td>
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<td>6</td>
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<tr>
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<td>10</td>
<td>0.911 12</td>
<td>0.885 57</td>
<td>0.940 32</td>
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<tr>
<td></td>
<td>$\infty$</td>
<td>0.911 04</td>
<td>0.885 43</td>
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<td>- 0.000 02</td>
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<td>0.965 39</td>
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<td>$\infty$</td>
<td>0.998 60</td>
<td>- 0.000 01</td>
<td>0.997 93</td>
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</table>

appropriate to compare our results with those of Durlofsky et al. We are interested in the drag coefficient $\lambda$ which is defined as follows:

$$\lambda = (\mu_{1,1}^{xx} + \mu_{1,2}^{xx} + \mu_{1,3}^{xx})^{-1}. \quad (4.8)$$

Kim was able to study the coefficient $\lambda$ up to $x = a/R = 0.4$. At smaller interparticle spacings, the convergence behavior of his power expansion is very slow. In a paper concerning this sedimentation problem, Kim used extrapolated values for smaller interparticle spacings. We were able to study $\lambda$ up to $x = 0.5$ and the results resemble those of Durlofsky et al., but it seems that at very small interparticle spacings, our results are slightly lower than those of Durlofsky et al. (see Fig. 3, Ref. 21). Unfortunately, we cannot compare our results in further detail because they have not presented tabulated results. We present some results for nearly touching spheres in Table V. In the case of touching spheres $x = 0.5$, we have calculated a limiting value $\lambda = 0.556$. The maximum value of the upper limit is $L = 15$. Durlofsky et al. discussed the existence of a minimum of $\lambda$ near $x = 0.495$. This minimum of $\lambda$ means that the sedimentation velocity of the cluster is maximal. One of their arguments is the existence of such a minimum in the two particle analog, as shown by Batchelor. Up to this order, we were not able to pass around this minimum, although we too believe that this minimum of $\lambda$ exists. Using the method presented in article I, we can round this minimum in the two particle case, but for values $L > 20$ only. For the maximum value of $L$ used in our calculations ($L = 150$) we have obtained correct results up to $x = 0.4995$. The result for $x = 0.4995$ is $0.713(6)$ with uncertainty in the decimal between brackets. Jeffrey and Onishi have presented expressions derived from lubrication theory. For the two particle case, as described above, lubrication theory gives $0.713(1)$. We expect that also in the three particle problem, it must be possible to round the minimum, but we have to use higher values of the upper limit $L$ then. Concerning the results presented in Table V, we want to point out that we are able to determine $\lambda$ in the case of small interparticle spacings in contrast with the results of Kim.

He gave two possible arguments to explain the divergence behavior of his results for $\lambda$. In the first place, he pointed out that it might be inherently impossible to obtain results for $\lambda$ in terms of a single expansion in $a/R$ and second he has not ruled out an error in his analysis. Our expression for $\lambda$ can be expressed as a fraction of expansions in $a/R$, so we cannot decide if one of the reasons of Kim is correct to explain the divergence behavior.

Furthermore, it is noteworthy that the pure three particle contribution to the components of the grand mobility matrix, although relatively small for large interparticle distances, become very important in calculating three particle configuration averages. This has consequences when we study the virial expansions of, e.g., the translational and rotational short time self-diffusion coefficients $D_i^r$ and $D_i^l$. In Sec. V, we present the results of the virial expansions of the self-diffusion coefficients and the virial expansions of the sedimentation velocity $U_i/U_0$ and its rotational counterpart $\Omega_i/\Omega_0$.

V. RESULTS FOR DIFFUSION AND SEDIMENTATION

The study of the transport coefficients of suspensions is often aimed at the determination of the virial expansions of these coefficients. In article I, we have explained how virial expansions can be derived by using pair hydrodynamic interactions between the spherical particles. We have used the pair distribution function up to order $q$, the volume fraction of dispersed particles in the suspension. Some of the results are new and other results had already been obtained by others. We can think, e.g., of the first-order virial coefficients of the short time translational self-diffusion tensor and of the sedimentation velocity, obtained by Batchelor, and the first-order virial coefficient of the short time rotational self-diffusion tensor, obtained by Cichocki and Felderhof. The results for the translational and rotational self-diffusion tensors are

$$D_i^r = D_0^r I(1 - 1.83q - 1.13q^2), \quad (5.1)$$

$$D_i^l = D_0^l I(1 - 0.63q - 1.02q^2), \quad (5.2)$$

where $D_0^r = k_B T/(6\pi \eta_0 a)$, $D_0^l = k_B T/(8\pi \eta_0 a^2)$, $k_B$ is the Boltzmann constant, and $T$ the absolute temperature. The virial expansions of the translational and rotational sedimentation velocities are

$$U_i/U_0 = (1 - 6.55q + 12.51q^2), \quad (5.3)$$

$$\Omega_i/\Omega_0 = (1 - 1.52q - 0.79q^2), \quad (5.4)$$

with $U_0$ and $\Omega_0$ the translational and rotational sedimentation velocity of one particle in an unbounded fluid, respectively. The second-order virial coefficient of $U_i/U_0$ is not given in article I, but it is obvious that this coefficient can be obtained in the same way as other second-order virial coefficients. Our aim in this section is to correct the second-order virial coefficients by including three particle hydrodynamic interactions.

The short time self-diffusion tensors $D_i^r$ and $D_i^l$ can be defined by means of the grand mobility matrix. These diffusion tensors can be used to describe the translational and rotational diffusion of a single test particle on a time scale in which the particle configuration remains nearly constant.

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</tr>
<tr>
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<td>15</td>
<td>0.5562</td>
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and are defined in the following way:

\[
D'_i = \frac{k_B T}{N} \left( \sum_{n=1}^{N} \mu_{ii}^n \right) = k_B T \langle \mu_{ii}^n \rangle, \tag{5.5}
\]

\[
D''_i = \frac{k_B T}{N} \left( \sum_{n=1}^{N} \mu_{ii}^n \right) = k_B T \langle \mu_{ii}^n \rangle, \tag{5.6}
\]

with \( N \) the number of particles in the suspension and \( \langle \cdots \rangle_c \) denotes an average over all configurations of the \( N \) particles inside a volume \( V \). The configuration average of the mobility tensors can be rewritten in the following form, where we restrict ourselves to the moment to the translational case:

\[
\langle \mu_{ii}^n \rangle_c = \mu_i^0 I + \sum_{n=2}^{N} \langle \left[ \mu_{ii}^n \right]_e \rangle_c, \tag{5.7}
\]

where \( \mu_i^0 = 1/(6 \eta_0 a) \) and \( \langle \left[ \mu_{ii}^n \right]_e \rangle_c \) denotes a configuration average of the \( n \) particle cluster. The average \( \langle \left[ \mu_{ii}^n \right]_e \rangle_c \) is, up to order \( \varphi^2 \), already known [see Eq. (5.1)]

\[
\langle \left[ \mu_{ii}^n \right]_e \rangle_c = \frac{1}{2} \frac{(N-1)(N-2)}{\nu^2} \int dR_{12} \int dR_{13} g(R_{12}, R_{13}, R_{23}) \left[ \mu_{ii}^n (R_{12}, R_{13}) \right]_3
\]

\[
= n_0^2 \frac{4\pi^3}{3} \int R_{12} dR_{12} \int R_{13} dR_{13} \int \sin \xi_{12} d\xi_{13} \left( g(R_{12}, R_{13}, R_{23}) \right) \left[ \mu_{ii}^n (R_{12}, R_{13}) \right]_3, \tag{5.9}
\]

where we have assumed that \( N \gg 1 \). Furthermore, we have used rotational symmetries to determine the integrations over the azimuthal angles \( \varphi_{12} \) and \( \varphi_{13} \), and over the angle \( \xi_{12} \). Beenakker has calculated the configuration average of the first term of the power expansion in inverse interparticle distances [Eq. (4.4a)]. The integral was evaluated numerically, where he used Monte Carlo techniques. This average leads finally to the following three particle contribution to the self-diffusion tensor:

\[
\left[ D'_i \right]_3 = 1.8 \varphi^3 D_b^0 I. \tag{5.10}
\]

This is slightly lower than the result of an analytical determination of the configuration average

\[
\left[ D''_i \right]_3 = 1.8 \varphi^3 D_b^0 I. \tag{5.11}
\]

In this special case, there is no essential difference between Eqs. (5.10) and (5.11) in contrast with the sedimentation problem, where it is difficult to evaluate some kind of integrals because of numerical problems. The remaining part of the configuration average of \( \left[ \mu_{ii}^n \right]_3 \) will be calculated numerically which should be done very carefully, because the three particle contribution of the components of the mobility matrix of particle 1 is influenced strongly by two nearly touching spheres at a great distance from the first particle. In that situation, there is also the problem that the components of the mobility matrix of particle 1 have a poor convergence behavior with increasing \( L \), the upper limit of our set of linear equations. Consequently, we have to take into account this area in the numerical integration with great care. This can be done by considering symmetry arguments, viz.

\[
\mu_{ii}^n (R_{12}, R_{13}) = \mu_{i2}^n (R_{12}, R_{23}). \tag{5.12}
\]

With this relation, we can rewrite the configuration integral and the result is and if we restrict ourselves to three particle hydrodynamic interactions only, we have to calculate \( \langle \left[ \mu_{i1}^n \right]_3 \rangle_c \). The configuration average of the three particle cluster can be evaluated with the help of the following simple form of the three particle distribution function:

\[
g(R_{12}, R_{13}, R_{23}) = g(R_{12}, R_{13}, R_{23}) = \begin{cases} 0, & R_{12} < 2a \text{ or } R_{13} < 2a \text{ or } R_{23} < 2a, \\ 1, & \text{elsewise} \end{cases} \tag{5.8}
\]

with \( R_g = |R_g| \). It should be noted that \( R_{23} \) is a function of the interparticle distances \( R_{12} \) and \( R_{13} \). Despite the simplicity of the three particle distribution function, the configuration integral itself is an integral over a very complicated domain. The configuration average of the three particle cluster is now

\[
\langle \left[ \mu_{i1}^n \right]_3 \rangle_c = - \frac{1}{2} n_0^2 \int dR_{12} \int dR_{13} H(R_{23} - R_{13}) \times g(R_{12}, R_{13}, R_{23}) \left( \mu_{i1}^n [R_{12}, R_{13}], \mu_{i2}^n [R_{12}, R_{23}] \right), \tag{5.13}
\]

The function \( H(x) \) is the Heaviside function with \( H(x) = 1 \) if \( x > 0 \) and \( H(x) = 0 \) if \( x < 0 \). The coefficients \( \epsilon_1 \) and \( \epsilon_2 \) are the virial coefficients obtained from both configuration integrals. The results of these coefficients are presented in Table VI, where we see that \( \epsilon_2 \) is relatively large and shows slow convergence behavior. Despite these numerical problems, we can make an estimate of the pure three particle contribution of the virial expansion of \( D'_i \).

### Table VI

<table>
<thead>
<tr>
<th>( L )</th>
<th>( \epsilon_1 )</th>
<th>( \epsilon_2 )</th>
<th>( \left[ D'_i \right]_3 )</th>
<th>( \epsilon_1 )</th>
<th>( \epsilon_2 )</th>
<th>( \left[ D'_i \right]_3 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>0.044</td>
<td>-0.604</td>
<td>1.25</td>
<td>0.218</td>
<td>-0.046</td>
<td>0.17</td>
</tr>
<tr>
<td>3</td>
<td>0.235</td>
<td>-0.880</td>
<td>1.17</td>
<td>0.320</td>
<td>-0.075</td>
<td>0.25</td>
</tr>
<tr>
<td>4</td>
<td>0.271</td>
<td>-1.015</td>
<td>1.07</td>
<td>0.353</td>
<td>-0.086</td>
<td>0.27</td>
</tr>
<tr>
<td>5</td>
<td>0.285</td>
<td>-1.085</td>
<td>1.01</td>
<td>0.366</td>
<td>-0.092</td>
<td>0.27</td>
</tr>
<tr>
<td>6</td>
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<td>-1.124</td>
<td>0.98</td>
<td>0.373</td>
<td>-0.097</td>
<td>0.28</td>
</tr>
<tr>
<td>7</td>
<td>0.297</td>
<td>-1.147</td>
<td>0.96</td>
<td>0.378</td>
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<tr>
<td>8</td>
<td>0.300</td>
<td>-1.162</td>
<td>0.95</td>
<td>0.381</td>
<td>-0.102</td>
<td>0.28</td>
</tr>
<tr>
<td>9</td>
<td>0.301</td>
<td>-1.171</td>
<td>0.94</td>
<td>0.384</td>
<td>-0.104</td>
<td>0.28</td>
</tr>
<tr>
<td>10</td>
<td>0.302</td>
<td>-1.178</td>
<td>0.93</td>
<td>0.386</td>
<td>-0.105</td>
<td>0.28</td>
</tr>
</tbody>
</table>
\[ [D^r_3] = 0.93 \varphi^2 D_0^r I, \]  
\( (5.14) \)

which is approximately 50\% lower than the value obtained by Beenakker (see in this context his remarks concerning the higher-order corrections\(^7\)). The final expression of the virial expansion of \( D^r_3 \) is now

\[ D^r_3 = D_0^r I (1 - 1.83 \varphi - 0.20 \varphi^2), \]  
\( (5.15) \)

where we should take into account that the value of the second-order virial coefficient might be some few hundredth parts lower. It is obvious that our value of the second-order virial coefficient is much lower than Beenakker's value 0.88. This difference can be partially explained by the fact that Beenakker has used a value \(-0.92\) instead of \(-1.13\) for the two particle contribution to the second-order virial coefficient. Recently, Jones, Muthukumar, and Cohen have obtained a value of the second-order virial coefficient which is even higher than Beenakker’s value, viz. 1.21.\(^17\) We cannot explain this difference. In Fig. 1, we have plotted our result, Beenakker's result, the result of Jones \textit{et al.}, and some experimental results of Pusey and van Megen,\(^{26}\) Ottewill and Williams,\(^{27}\) and van Megen and Underwood.\(^{28}\) In Fig. 2, we have plotted our result, the numerical results of Ladd,\(^4\) the simulation data of Phillips \textit{et al.},\(^3\) and the result of \( D^r_3 \) obtained by Beenakker and Mazur, where they used an expansion in powers of the fluctuation in the concentration of the suspended particles.\(^7\) We may conclude that our expression is reasonable up to \( \varphi \approx 0.4 \), especially if we compare our result with the most recent experiments of van Megen and Underwood, and we may hope that the contribution of four and more particle hydrodynamic interactions to the virial expansion will be an improvement if \( \varphi > 0.4 \). The result of Jones \textit{et al.}\(^17\) seems too high, as well as the virial expansion of Beenakker.\(^7\) Beenakker has already made this conclusion concerning his virial expansion and developed a new method to include many-particle hydrodynamic interactions (see Fig. 2). The numerical data of Ladd and the simulation results of Phillips \textit{et al.}\(^2\) confirm our remarks above.

In the same way, we can determine the virial expansion of \( D^r_2 \). This can be achieved completely by numerical means. The results of the numerical integrations are, for different values of \( L \), presented in Table VI. The virial expansion, including three particle hydrodynamic interactions, is

\[ D^r_2 = D_0^r I (1 - 0.63 \varphi - 0.74 \varphi^2). \]  
\( (5.16) \)

In article I, we pointed out that the three particle contribution to the rotational self-diffusion coefficient would be less important than in the translational case. This is confirmed by considering the two contributions 0.28\( \varphi^2 \) for \( [D^r_3] \) and 0.93\( \varphi^2 \) for \( [D^r_2] \). In Fig. 3, we have plotted our expression for the virial expansion of \( D^r_2 \) and compare this with computer simulation results of Phillips \textit{et al.} We can conclude that Eq. (5.16) corresponds reasonably well with the computer simulation data. Unfortunately, there are no experimental results available.

The derivation of an expression of the sedimentation velocity is more complicated. In article I, we have used an expression of Pusey and Toug\(^{29}\) of the short time effective diffusion coefficient to determine the virial expansion of the sedimentation velocity. This expression has the following form:

![Fig. 1](image1.png)

**FIG. 1.** The short time self-diffusion coefficient, normalized with \( D_0^r \), is plotted vs \( \varphi \), the volume fraction. The solid curve represents our result [Eq. (5.15)], the dashed curve represents the virial expansion obtained by Beenakker and Mazur, and the dashed–dotted curve represents the virial expansion obtained by Jones \textit{et al.}\(^1\) The symbols represent experimental results of van Megen and Underwood (circles), Ottewill and Williams (triangles), and Pusey and van Megen (squares).

![Fig. 2](image2.png)

**FIG. 2.** The short time self-diffusion coefficient, normalized with \( D_0^r \), is plotted vs \( \varphi \), the volume fraction. The solid curve represents our result [Eq. (5.15)], the dashed curve represents the virial expansion obtained by Beenakker and Mazur, and the dashed–dotted curve represents the virial expansion obtained by Jones \textit{et al.} The open symbols represent numerical data of Ladd (squares) and simulation results of Phillips \textit{et al.} (triangles). The plusses represent the results obtained by Beenakker and Mazur, which includes many-particle hydrodynamic interactions.
with \( \mathbf{k} \) a wave vector with direction \( \hat{\mathbf{k}} \) and \( S(k) \) the static structure factor. Using Eq. (5.9) and taking \( \hat{\mathbf{k}} = \hat{\mathbf{e}}_z \), we write the three particle contribution as

\[
D_{\text{eff}}(k) = \frac{k_b T}{NS(k)} \sum_{j=1}^{N} \langle \hat{k}_j \mu_{ij} \hat{k} \exp(i\mathbf{k} \cdot \mathbf{R}_{ij}) \rangle_c.
\]

(5.17)

The shorthand notation \( \langle \mu_{12}^{\text{II}}, \mathbf{k} \rangle_c \) is introduced for later use. The sedimentation velocity is now defined as

\[
U/U_0 - 1 \sim D_s(k) S(k)
\]

(5.19)

The calculation of the three particle contribution to \( U/U_0 \) will be achieved in two steps, an analytical and a numerical step. In the first step, we write the mobility tensor \( [\mu_{12}^{\text{II}}]_3 \) in the following way:

\[
[\mu_{12}^{\text{II}}]_3 = \left[ [\mu_{12}^{\text{II}}]_3 - \nu_{12} \right] + \nu_{12}.
\]

(5.20)

The configuration average of \( \nu_{12} \), defined as in Eq. (4.4), will be determined analytically and the same kind of configuration average of the tensor \( [ [\mu_{12}^{\text{II}}]_3 - \nu_{12} ] \) will be calculated numerically. The tensor \( \nu_{12} \) consists of four terms, which have the following form according to Mazur and van Saarloos:

\[

\begin{align*}
\langle v_{12}^{(1)} \rangle_1 &= \frac{2}{\mu_0} \mu_0 (\xi_{13} x_{12} x_{23} - 3 \xi_{12}^2 \hat{R}_{13} \hat{R}_{23}) \\
\langle v_{12}^{(2)} \rangle_2 &= -3 \mu_0 (\xi_{13} x_{12} x_{23} (1 - 3 \xi_{12}^2) \hat{R}_{13} \hat{R}_{23} + x_{12} x_{23} (2 \xi_{13} \hat{R}_{23} \hat{R}_{13} + (1 - 5 \xi_{13}^2) \hat{R}_{13} \hat{R}_{23} + \hat{R}_{13} \hat{R}_{23} )) \\
\langle v_{12}^{(3)} \rangle_3 &= \frac{2}{\mu_0} \mu_0 (\xi_{13} x_{12} x_{23} [49 - 117 \xi_{12}^2] \hat{R}_{13} \hat{R}_{23} + (93 - 315 \xi_{12}^2) \hat{R}_{13} \hat{R}_{23} + (783 - 1575 \xi_{12}^2) \hat{R}_{13} \hat{R}_{23} + (1 - 5 \xi_{12}^2) \hat{R}_{13} \hat{R}_{23} \hat{R}_{13} )) \\
\langle v_{12}^{(4)} \rangle_4 &= \frac{2}{\mu_0} \mu_0 (\xi_{13} x_{12} x_{23} (1 - 3 \xi_{12}^2) \hat{R}_{13} \hat{R}_{23} - x_{12} x_{23} (1 - 3 \xi_{12}^2) \hat{R}_{13} \hat{R}_{23} ) \\
\end{align*}
\]

(5.21a) (5.21b) (5.21c) (5.21d)

with \( \zeta_1, \zeta_2, \) and \( \zeta_3 \) defined below Eq. (4.4). We shall furthermore use the special coordinate system introduced in Sec. III. The configuration average of the sum of \( \langle v_{12} \rangle_i \) is, in the limit \( k \to 0 \),

\[
\lim_{k \to 0} \sum_{i=1}^{4} \langle \langle v_{12} \rangle_i \rangle_c = 8.0 \mu_0 \varphi^2.
\]

(5.22)

The numerical calculation of the remaining term in Eq. (5.20) gives the following contribution:

\[
\lim_{k \to 0} \langle \langle [ [\mu_{12}^{\text{II}}]_3 - \nu_{12} ] , \mathbf{k} \rangle_c \rangle_c = -0.6 \mu_0 \varphi^2.
\]

(5.23)

In Table VII, we present the numerical results of the virial coefficients \( \epsilon_1 \) and \( \epsilon_2 \) as a function of the upper limit \( L \).

<table>
<thead>
<tr>
<th>( L )</th>
<th>( \epsilon_1 )</th>
<th>( \epsilon_2 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>0.80</td>
<td>-0.042</td>
</tr>
<tr>
<td>3</td>
<td>-0.20</td>
<td>-0.040</td>
</tr>
<tr>
<td>4</td>
<td>-0.40</td>
<td>-0.009</td>
</tr>
<tr>
<td>5</td>
<td>-0.50</td>
<td>-0.005</td>
</tr>
<tr>
<td>6</td>
<td>-0.55</td>
<td>-0.012</td>
</tr>
<tr>
<td>7</td>
<td>-0.57</td>
<td>-0.015</td>
</tr>
<tr>
<td>8</td>
<td>-0.59</td>
<td>-0.016</td>
</tr>
<tr>
<td>9</td>
<td>-0.60</td>
<td>-0.017</td>
</tr>
<tr>
<td>10</td>
<td>-0.61</td>
<td>-0.017</td>
</tr>
</tbody>
</table>

FIG. 3. The short time rotational self-diffusion coefficient, normalized with \( D_s(\varphi) \), is plotted vs \( \varphi \), the volume fraction. The solid curve represents our result [Eq. (5.16)], and the squares, triangles, and plusses represent simulation results of Phillips et al.
coefficients, denoted by \( \epsilon_1 \), for different values of the upper limit \( L \). This gives us some understanding of the convergence behavior of the numerical coefficient in Eq. (5.23). The total three particle contribution to the virial expansion of the sedimentation velocity is now \( \langle [\mu_{12}^3]_3, k \rangle_\epsilon = 7.4\mu_0^3\varphi^2 \) and the virial expansion, including the three particle contribution to the self-diffusion coefficient, is

\[
U_1/U_0 = 1 - 6.55\varphi + 19.7\varphi^2.
\]  

(5.24)

We have compared this result with numerical results obtained by Ladd\(^4\) and computer simulation results of Phillips, Brady, and Bossis.\(^3\) We have plotted these results in Fig. 4. The data of Phillips et al. differ, for small \( \varphi \), with our result and the results obtained by Ladd. This difference is caused by the insertion of a degree of periodicity into the model of Phillips et al. (see also Ref. 3). In Fig. 4, we have also plotted the linear virial expansion (the Batchelor expression) and we can conclude that this expression describes sedimentation reasonably well up to \( \varphi \approx 0.05 \). Our expression [Eq. (5.24)] is reasonable up to \( \varphi \approx 0.12 - 0.13 \). Beyond this value of \( \varphi \), the virial expansion describes sedimentation very badly and it can be expected that the third-order virial coefficient should be large (comparable with the second virial coefficient) and negative to compensate the quadratic term. It should be emphasized that the numerical results of Beenakker and Mazur correspond well with the results of Ladd very well up to high volume fractions.\(^8\) They included many-particle hydrodynamic interactions. However, the relative importance of two and three particle interactions cannot be estimated with numerical results as those of Beenakker and Mazur. We have not plotted experimental results in Fig. 4. These results are of course available, but some recent experimental results from Buscall et al.\(^{30}\) and Bacri et al.\(^{31}\) resemble the numerical data of Ladd very well up to \( \varphi \approx 0.5 \). Recently, Jones et al.\(^{17}\) have presented results of the second-order virial coefficient and their virial expansion has the following form:

\[
U_1/U_0 = 1 - 6.57\varphi + 18.27\varphi^2.
\]  

(5.25)

Their virial expansion resembles ours, although the three particle contribution of this expression is nearly 20\% lower than in our result. The difference becomes larger if we split off in both results the three particle contribution belonging to the self-diffusion coefficient. The result of Jones et al.\(^{17}\) is then 4.6, which is nearly 40\% lower than our result, viz. 7.4. As is the case with the three particle contribution to the self-diffusion coefficient, we cannot explain this difference.

In an analogous way, we have studied the virial expansion of the rotational counterpart of sedimentation. The only difference is that \( \mu' \) should be replaced by \( \mu'' \) in the equations above. Here we have to calculate one term analytically, viz.\(^6\)

\[
\left[ [\mu_{12}^2]_3 \right] = ( [\mu_{12}^2]_3 - v_{12}) + v_{12},
\]  

(5.26)

with

\[
v_{12} = \frac{1}{2} \mu_0 \xi_{12}^2 \left( \langle \hat{R}_{13} \times \hat{R}_{23} \rangle \langle \hat{R}_{13} \times \hat{R}_{23} \rangle + \xi_{13} \hat{R}_{23} \hat{R}_{13} \right)
\]  

and \( \mu_0 = 1/(8\pi \eta_0 a^4) \). The configuration average of \( v_{12} \) is

\[
\lim_{k \to 0} (v_{12}, k)_\epsilon = 1.17\mu_0^2 \varphi^2.
\]  

(5.27)

The numerical calculation of the remaining part yields the contribution \(- 0.02\mu_0^2 \varphi^2 \). In Table VII, the numerical result is tabulated as a function of the upper limit \( L \). The virial expansion of \( \Omega_1/\Omega_0 \) is

\[
\Omega_1/\Omega_0 = 1 - 1.52\varphi + 0.64\varphi^2.
\]  

(5.29)

In Fig. 5, we have plotted this result as a function of volume fraction \( \varphi \) and compared the result with computer simulation results of Phillips et al.\(^3\) This virial expansion and the simulation results agree very well up to \( \varphi \approx 0.4 \), which leads to the conclusion that rotational sedimentation can be described reasonably well by using two and three particle hydrodynamic interactions among the hard spheres in a suspension.

VI. CONCLUSION

The set of linear equations derived in Sec. II has been used to study three particle hydrodynamic interactions. After studying some components of the grand mobility matrix, one important conclusion can be drawn, namely that it is necessary to include many terms of the power expansion in typical inverse interparticle distances to describe three particle hydrodynamic interactions sufficiently, even at intermediate interparticle distances. This is a tedious task and as far as we know there are no results available of arbitrary configurations of the three particle cluster. The method presented in this chapter seems attractive to perform these calculations, although the final result is not in the form of a single power expansion. Analytical expressions can be obtained by using algebraic computer programs. We have restricted ourselves to calculating these components numerically.
The study of the three particle contribution to the translational self-diffusion coefficient leads to the conclusion that the contribution calculated in Sec. V [Eq. (5.14)] is much smaller than predicted by several other authors, but the derived expression of $D_1$ [Eq. (5.15)] is reasonable, if compared with experimental data and simulation results, up to $\varphi \approx 0.4$ in contrast with expressions derived by Beenakker and Jones et al.$^{17}$ The rotational counterpart is described reasonably well using two and three particle hydrodynamic interactions only. This is also the case for the rotational sedimentation velocity. At this moment, we can compare these theoretical results with simulation data only because, to our knowledge, there are no experimental results available. The translational sedimentation velocity cannot be described very easily in terms of a virial expansion. With the expression derived in this paper [Eq. (5.24)], we can extend the range, where that expression is reasonable, to higher volume fractions, up to $\varphi \approx 0.13$. For higher volume fractions, we cannot use this virial expansion. Despite this disappointing result, which was expected, we can learn something different from it and also from the result of Jones et al.$^{17}$ Both results show that it is dangerous to derive virial coefficients from experimental results with fitting procedures. There are several results available of the second-order virial coefficient of the sedimentation velocity, which are obtained from experiment. Kops-Werkhoven and Fijnaut obtained a value $10 \pm 4,^{32}$ which is too small, and Cheng and Schachman obtained a value of roughly $20,^{33}$ which is reasonable. The same remark is also valid for other virial expansions such as the virial expansion of the translational self-diffusion coefficient.