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High-frequency effective viscosity of hard-sphere suspensions

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We present a method to determine a theoretical expression for the high-frequency effective viscosity of a suspension of hard spheres. In the explicit calculations we have used two-particle hydrodynamic interactions only. The expression of the effective viscosity has a Saito-like form with a pole near \( \varphi = 0.70 \), which is comparable to the maximum packing fraction of a hard-sphere system. The final result is in good agreement with experimental and numerical results up to higher volume fractions, as will be shown in comparisons with recent experiments and computer-simulation results. Three- and more-particle hydrodynamic interactions are expected to be important for dense suspensions, but with the presented method it appears that these higher-order interactions give more subtle contributions to the presented expression of the effective viscosity in comparison with the commonly used methods to derive a virial expansion of the effective viscosity itself. This advantageous point will be discussed briefly in the discussion.

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I. INTRODUCTION

In recent years several attempts have been made to calculate, on theoretical grounds, the high-frequency effective viscosity \( \eta_{\text{eff}} \) (in the following abbreviated to effective viscosity) of a suspension of undeformable spherical particles, in a fluid with shear viscosity \( \eta_0 \), as a function of the volume fraction \( \varphi \) of suspended particles. The effective viscosity of a suspension of hard spheres can be measured experimentally by studying the behavior of this suspension in an oscillating shear flow at high frequencies (see, e.g., the measurements of van der Werff, de Kruijff, Blom, and Mellema [1]). At high frequencies, the effective viscosity \( \eta_{\text{eff}} \) will be determined by hydrodynamic interactions between the spherical particles only, because the effects of Brownian motion can be neglected. At lower frequencies Brownian motion becomes important as shown experimentally by e.g., van der Werff et al. [1] and theoretically by the work of, e.g., Batchelor [2], who calculated \( \eta_{\text{eff}} \) the steady-shear limit of the effective viscosity. In the high-frequency limit we can furthermore assume that the pair distribution function \( g(\mathbf{R}) \) is isotropic, because within this limit it is not very likely that the oscillating shear flow disturbs the equilibrium pair distribution function. The behavior of \( \eta_{\text{eff}} \) at low volume fraction of suspended particles is well understood and the virial expansion of \( \eta_{\text{eff}} \), to second order in \( \varphi \), is

\[
\eta_{\text{eff}} = \eta_0 \left(1 + C_1 \varphi + C_2 \varphi^2\right). \tag{1.1}
\]

The first-order virial coefficient \( C_1 \) has already been calculated by Einstein in the beginning of this century [3]. The value is \( C_1 = \frac{1}{2} \). Experiments with suspensions of spherical particles show that it is necessary to incorporate higher-order corrections if \( \varphi > 0.05 \). For that reason many attempts were made in the past few decades to calculate the second-order virial coefficient \( C_2 \) and, related to this number, the Huggins coefficient \( k_h \) with \( k_h = \frac{5}{3} C_2 \). This coefficient was studied by Peterson and Fixman, who derived the value \( k_h = 0.69 \) [4]. Later on Batchelor and Green determined \( C_2 \) in an exact way [5]. Their result is \( C_2 = 5.2 \pm 0.3 \). They came to this coefficient by taking into account two-particle hydrodynamic interactions between suspended particles in a shear flow only. For this calculation they used the simplest form of the pair distribution function, i.e., exclusion of particle overlap (hard spheres). Recently Chichocki and Felderhof determined a more accurate value for the term quadratic in volume fraction of \( \eta_{\text{eff}} \), \( C_2 = 5.00 \) [6]. Their calculations are based upon a multipole expansion of the hydrodynamic interactions [7]. The difference between the result of Batchelor and Green on the one hand and Chichocki and Felderhof on the other can be explained by realizing that Chichocki and Felderhof used more accurate hydrodynamic functions to evaluate \( C_2 \). Russel and Gast extended the formalism of Batchelor and Green by including a volume-fraction-dependent equilibrium pair distribution function [8].

There exist other expressions in the literature describing the effective viscosity. One of the alternatives is the expression derived by Saito [9,10], which is

\[
\eta_{\text{eff}} = \eta_0 \left[1 + \frac{5}{2} \left(1 - \varphi \right)\right]. \tag{1.2}
\]

Chichocki, Felderhof, and Schmitz have derived a relation for \( \eta_{\text{eff}} \) with the help of a cluster expansion [11]. Their result takes a form similar to the Saito formula [Eq. (1.2)] and is obtained by including the two-body approximation in the cluster expansion only. The expression for the effective viscosity has a pole for \( \varphi = 0.364 \), a relatively low volume fraction. To bring into line the theoretical expression with the experimental results, they concluded that higher-order correction terms in the cluster expansion should be taken into account to shift the pole to a higher volume fraction.Beenakker obtained the effective
viscosity using an expansion in density fluctuation correlation functions [12]. His numerical result, taking into account many-particle hydrodynamics, is in good agreement with the experimental results of van der Werff et al. for \( \varphi \leq 0.45 \) [1]. Beenakker did not present results for higher volume fractions because he expected that those would be less accurate. The list of results for \( \eta_{\text{eff}} \) mentioned above is not complete and could be extended by work of other authors [13–17]. Several authors, like Mellema and Willemse [13] and Bedeaux [14], treated the suspension as a mixture of two fluids with different shear viscosities. To describe suspensions they assumed that the viscosity of one fluid, present with volume fraction \( \varphi \), should be infinitely large. Their result in this limit is

\[
\eta^*_{\text{eff}} = \eta_0 \left( 1 + \frac{\frac{1}{2} \varphi + 1.42 \varphi^2}{(1-\frac{1}{2} \varphi)} \right). \tag{1.3}
\]

We see in this equation that the effective viscosity has a pole for \( \varphi = 0.4 \). This seems contradictory to the experimental results. The same formula was earlier derived by Lundgren [15].

Finally we focus attention on some numerical results. Some years ago Brady and Bossis developed a method for simulating a system consisting of spherical particles in shear flow. They called it “Stokesian dynamics simulation” (for a summary of the general Stokesian dynamics method we refer to a paper of Brady and Bossis [18]). In a later article Phillips, Brady, and Bossis present computer-simulation results of the effective viscosity [19]. We also mention here the numerical results for \( \eta^*_{\text{eff}} \) of Ladd [20,21]. All these numerical results for the effective viscosity can be used to compare theoretical results with.

In this article we present a method to determine the effective viscosity theoretically. The final result is

\[
\eta^*_{\text{eff}} = \eta_0 \left( 1 + \frac{\frac{1}{2} \varphi + 1.42 \varphi^2}{(1-\frac{1}{2} \varphi)} \right) \tag{1.4}
\]

and can be obtained in a way similar to Saito's way of deriving his expression for \( \eta_{\text{eff}} \) [Eq. (1.2)]. The presented expression will be discussed in Sec. VI. Our theoretical expression is in good agreement with experimental results of van der Werff et al. [1] for volume fractions up to 0.6. This result is remarkable because we incorporated two-particle hydrodynamic interactions only. Equation (1.4) also agrees reasonably well with computer-simulation results. It is important to note that three- and more-particle hydrodynamic interactions are important for higher volume fractions (\( \varphi > 0.2 \)) but we expect that the contributions of these higher-order corrections are more subtle than is the case in the commonly used expressions to describe the effective viscosity [viz., Eq. (1.1)]. The advantage is that the final result tends faster to the experimental results and simulation data if many-particle hydrodynamic interactions are included in comparison with the quadratic virial expansion, Eq. (1.1). Future research should find out if the presented procedure can also be followed in the case of other problems. We can think, e.g., of the steady-shear limit of the effective viscosity \( \eta^*_{\text{eff}} \). Finally we want to point out that Eq. (1.4) has a pole for \( \varphi \approx 0.70 \), which is nearly equal to the maximum packing fraction of a hard-sphere system.

In this article we show in Secs. II and III the method to derive the set of linear equations needed to obtain the grand mobility matrix. In Sec. IV we present the derivation of the stresslet \( \vec{S} \), exerted by the fluid on a particle, in terms of some coefficients. Equation (1.4) is derived in Sec. V and we discuss this result in Sec. VI. We end this article with some concluding remarks.

## II. THE MOBILITY MATRIX

We start the study of the effective viscosity by considering a system of two spherical particles, both with radius \( a \), immersed in an unbounded fluid with an incoming fluid velocity field \( \vec{v}_0(r) \). For the moment we assume that \( \vec{v}_0(r) \) is arbitrary but satisfies the incompressible, linearized Navier-Stokes (NS) equations. Later on in this article we restrict ourselves to the relatively simple case of shear flow. The particles have velocities \( \vec{U}_i \) and rotational velocities \( \omega_i \) (\( i=1,2 \)). Their centers have position vectors \( \vec{R}_i \) with respect to the origin \( O \). The particles are of such small size that the Reynolds number of the fluid motion induced by the particles is small, \( Re << 1 \). With this assumption we can neglect the nonlinear term in the Navier-Stokes equation. The equations of motion for the fluid, the NS equations, are now

\[
\eta_0 \vec{v}(r) - \vec{v}_p(r) = 0, \tag{2.1}
\]

\[
\vec{v} \cdot \vec{v} = 0, \tag{2.2}
\]

with \( \vec{v}(r) \) the fluid velocity, \( p(r) \) the pressure, and \( \eta_0 \) the shear viscosity. We suppose stick boundary conditions at the surface \( S_i \) of particle \( i \),

\[
\vec{v}(r) = \vec{U}_i + \omega_i \times (r - \vec{R}_i) \quad \text{with} \quad r \in S_i \quad \text{and} \quad i=1,2. \tag{2.3}
\]

To preserve linearity we treat the position vector \( \vec{R}_i \) as independent of the time \( t \). This linearization is equivalent to omitting a term of order \( (\vec{U}_i \cdot \vec{v})(r) \) in the NS 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 [22] or the remarks in a review article of Herczynski and Piekowska [23]. After calculation of \( \vec{v}(r) \) and \( p(r) \) we are able to determine the pressure tensor

\[
\Pi(r) = p(r) - 2 \eta_0 \vec{v}(r). \tag{2.4}
\]

The pressure tensor can be used to determine the force \( \vec{F}_i \), the torque \( \vec{T}_i \), and the stresslet \( \vec{S}_i \), exerted by the fluid on particle \( i \). The stresslet \( \vec{S}_i \) is the symmetric and traceless part of the first moment of the force distribution integrated over the surface of particle \( i \). These quantities can be defined, in the case of spherical particles, in the following way:

\[
\vec{F}_i = -a^2 \int_{|\vec{r}|=a} \vec{\Pi}_i(r) \, dS_i, \tag{2.4}
\]

\[
\vec{T}_i = -a^3 \int_{|\vec{r}|=a} (\vec{\varepsilon}_r \times \vec{\Pi}_i(r)) \, dS_i, \tag{2.5}
\]

\[
\vec{S}_i = -a^3 \int_{|\vec{r}|=a} \langle \vec{\varepsilon}_r, \vec{\Pi}_i(r) \rangle \, dS_i, \tag{2.6}
\]
with \(d\Omega\), the element of solid angle with respect to the center of particle \(i\) and \(\hat{e}_r\), the radial unit vector in a spherical coordinate system. Furthermore

\[
\Pi_i(r) = \left( \hat{e}_r \right) \cdot \left( r \cdot \nabla v(r) \right),
\]

and \(\left( \hat{e}_r \Pi_i(r) \hat{e}_r \right)\) is the traceless and symmetric part of

\[
\left( \hat{e}_r \Pi_i(r) \hat{e}_r \right) = \frac{1}{2} \left[ \left( \hat{e}_r \Pi_i(r) + \Pi_i(r) \hat{e}_r \right) - \frac{1}{2} \left[ \Pi_i(r) \hat{e}_r \right] \right].
\]

We are especially interested in the grand mobility matrix, the analogue of the grand resistance matrix defined by Happel and Brenner [24]. This mobility matrix relates the velocity difference between the particle and the surface averaged incoming fluid velocity field with the forces, torques, and stresses, exerted by the fluid on the particles, etc. This relation is expressed, using the shorthand notations \(U = (U_1, \ldots, U_N)\), \(U^c = (U_{01}, \ldots, U_{0N})\), etc., by the following equation:

\[
\begin{bmatrix}
U - U_0 \\
\Omega - \Omega_0 \\
-\vec{G}_0
\end{bmatrix} = - \begin{bmatrix} \mu' & \mu'' & \mu''' \\ \mu'' & \mu'' & \mu''' \\ \mu''' & \mu'' & \mu'' \end{bmatrix} \begin{bmatrix} F \\ T \\ \vec{S} \end{bmatrix},
\]

where \(U_0, \Omega_0,\) and \(\vec{G}_0\) are the incoming fluid velocity, vorticity, and rate of strain, respectively, averaged over the surface of the respective particles. The \(\mu''\) etc. are matrices with

\[
\mu'' = \begin{bmatrix} \mu''_{11} & \cdots & \mu''_{1N} \\ \vdots & \ddots & \vdots \\ \mu''_{N1} & \cdots & \mu''_{NN} \end{bmatrix},
\]

where \(\mu''_{ij}\) is a mobility tensor. The upper indices \(i, r,\) and \(d\) refer to “translational,” “rotational,” and “dipole,” respectively. The dot in Eq. (2.9) stands for a contraction of tensors. It should be emphasized that in this notation for the mobility tensors no distinction has been made between tensors of different rank (an exception is made for vectors, of course). In Eq. (2.9), e.g., the matrix \(\mu''\) consists of second-rank tensors and the matrix \(\mu'''\) consists of fourth-rank tensors. For studying the effective viscosity we are especially interested in \(\mu''''\). Finally we want to point out the following symmetry relations for the components of the mobility matrix which have the same form as those presented by Schmitz and Felderhof, although they used a different definition of the grand mobility matrix [25]:

\[
\begin{aligned}
\mu''_{ij, ab} &= \mu''_{ji, ba}, & \mu''_{ij, ab} &= \mu''_{ji, ba}, \\
\mu'''_{ij, ab} &= \mu''_{ij, ab}, & \mu'''_{ij, ab} &= \mu''_{ij, ab}, \\
\mu''''_{ij, ab} &= \mu''''_{ji, ba}, & \mu''''_{ij, ab} &= \mu''''_{ji, ba}.
\end{aligned}
\]

Our aim is the determination of the grand mobility matrix [Eq. (2.9)] which can be achieved by the method presented by the authors in an earlier article [26] (hereafter referred to as I).

### III. THE SET OF LINEAR EQUATIONS

The fluid velocity field \(v(r)\), created if two spherical particles move in a fluid with an externally imposed flow, can be seen, because of the linearity of the NS equations, as a sum of three contributions. One contribution originates from the incoming flow field and the other two contributions are scattered velocity fields defined with respect to the origins of the particles, or

\[
v(r) - v_0(r) = \sum_{i=1}^{N} v_i(r), \quad r_i = r - R_i.
\]

The velocity fields \(v_i(r)\) 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 [27]. These basic solutions, that behave regularly for \(|r| \to \infty\), are

\[
\begin{align}
\psi_{lm\alpha}(r) &= \frac{1}{(l+1)(2l+1)} r^{-l} B_{lm}(\theta, \varphi), \\
\psi_{lm\beta}(r) &= \frac{1}{l(l+1)} r^{-l} C_{lm}(\theta, \varphi), \\
\psi_{lm\gamma}(r) &= \frac{1}{2(2l+1)} r^{-l} \left[ (l+1) A_{lm}(\theta, \varphi) - \frac{l(2l-1)}{2} B_{lm}(\theta, \varphi) \right].
\end{align}
\]

The indices \(l\) and \(m\) are restricted to \(l \geq 1\) and \(|m| \leq l\). The accompanying basic solutions for the pressure are

\[
\begin{align}
p_{lm\alpha}(r) &= 0, & p_{lm\beta}(r) &= 0, \\
p_{lm\gamma}(r) &= \eta d (2l-1) r^{-l} Y_{lm}(\theta, \varphi).
\end{align}
\]

These basic solutions all satisfy the NS equations (2.1) and (2.2). The vectors \(A_{lm}(\theta, \varphi), B_{lm}(\theta, \varphi),\) and \(C_{lm}(\theta, \varphi)\) are special vector spherical harmonics related to the vector spherical harmonics \(Y_{JM}(\theta, \varphi)\) defined by Edmonds [28]. In I we have paid some attention to these vector spherical harmonics [26]. The incoming fluid velocity field should be expanded in terms of an alternative set of basic solutions, which have been introduced by Schmitz and Felderhof also [27]. These basic solutions behave regularly for \(|r| \to 0\), and have the following form:

\[
\begin{align}
w_{lm\alpha}(r) &= r^{l-1} A_{lm}(\theta, \varphi), \\
w_{lm\beta}(r) &= r^l C_{lm}(\theta, \varphi), \\
w_{lm\gamma}(r) &= r^{l+1} \left[ (l+1)(2l+3) A_{lm}(\theta, \varphi) + B_{lm}(\theta, \varphi) \right].
\end{align}
\]

For the pressure we have

\[
\begin{align}
p_{lm\alpha}(r) &= 0, & p_{lm\beta}(r) &= 0, \\
p_{lm\gamma}(r) &= \eta \eta d (2l-1) r^{-l} Y_{lm}(\theta, \varphi).
\end{align}
\]
\begin{equation}
\begin{aligned}
& p_{lm}^{\text{inc}}(r) = 0, \quad p_{lm}^{\text{ext}}(r) = 0, \\
& p_{lm}^{\text{inc}}(r) = \frac{(l+1)(2l+1)2(l+3)}{l} r Y_{lm}^{\theta \varphi}.
\end{aligned}
\end{equation}

For these incoming basic functions the indices \( l \) and \( m \)
are again restricted to \( l \geq 1 \) and \( |m| \leq l \). It is evident that this set of basic solutions satisfies the NS equations for each \( l, m \) and \( \sigma \in [\alpha, \beta, \gamma] \) separately as is the case with the outgoing basic solutions. With these basic solutions we can write for the velocity field \( v'(r_i) = v(r_i + R_i) \), the fluid velocity defined with respect to the origin of particle \( i \) (with \( i = 1, 2 \)),

\begin{equation}
\begin{aligned}
& v'(r_i) - \sum_{l \geq 1, m} \left[ \alpha_{lm}^i \omega_{lm}(r_i + R_i) + \beta_{lm}^i \omega_{lm}(r_i + R_i) + \gamma_{lm}^i \omega_{lm}(r_i + R_i) \right] \\
& = \sum_{l \geq 1, m} \left[ \alpha_{lm}^i \nu_{lm}(r_i) + \beta_{lm}^i \nu_{lm}(r_i) + \gamma_{lm}^i \nu_{lm}(r_i) \right] + \sum_{l \geq 1, m} \left[ \alpha_{lm}^i \nu_{lm}(r_i + R_{ij}) + \beta_{lm}^i \nu_{lm}(r_i + R_{ij}) + \gamma_{lm}^i \nu_{lm}(r_i + R_{ij}) \right],
\end{aligned}
\end{equation}

where \( R_{ij} = R_i - R_j \) is the position vector of the center of particle \( i \) with respect to the center of particle \( j \). We furthermore assume that \( j \neq i \). In an analogous way we have for the pressure field \( p'(r_i) \)

\begin{equation}
\begin{aligned}
& p'(r_i) = \sum_{l \geq 1, m} \left[ \alpha_{lm}^{\text{inc}} \nu_{lm}(r_i + R_i) \right] = \sum_{l \geq 1, m} \left[ \alpha_{lm}^{\text{inc}} \nu_{lm}(r_i) \right] + \sum_{l \geq 1, m} \left[ \alpha_{lm}^{\text{inc}} \nu_{lm}(r_i + R_{ij}) \right] + \sum_{l \geq 1, m} \left[ \alpha_{lm}^{\text{inc}} \nu_{lm}(r_i + R_{ij}) \right].
\end{aligned}
\end{equation}

To determine the set of linear equations, which is needed to calculate the coefficients \( \{ \alpha_{lm}^i, \beta_{lm}^i, \gamma_{lm}^i \} \) for \( i = 1 \) and \( 2 \), it is necessary to calculate the following inner products:

\begin{equation}
\int_{|r_i|=a} [v'(r_i) - v_0(r)] \cdot A_{pq}^* (\theta_i, \varphi_i) d\Omega_i,
\end{equation}

keeping in mind that one can write \( r = r_i + R_i \). We have to determine similar inner products with \( A_{pq}^* (\theta_i, \varphi_i) \) replaced by \( B_{pq}^* (\theta_i, \varphi_i) \) or \( C_{pq}^* (\theta_i, \varphi_i) \). We do not go into the details of the calculation of these inner products. There is no difference with the inner products already calculated in article I [see Eqs. (1.5.8)-(1.5.13), Ref. [26]] except the part concerning the incoming fluid velocity \( v_0(r) \). We can calculate the inner products [Eq. (3.8)] by substituting the right-hand side of Eq. (3.6) for \([v'(r_i) - v_0(r)]\) in these inner products. This kind of calculation is already done in article I and the final result is

\begin{equation}
\begin{aligned}
& \int_{|r_i|=a} [v'(r_i) - v_0(r)] \cdot A_{pq}^* (\theta_i, \varphi_i) d\Omega_i = p(p + 1) a^{-p} y_{pq}^i + \frac{n_{pq}}{(p + q)!} a^{-1} \\
& \quad \times \left[ p (p + 1) \sum_{l \geq 1, m} \frac{l}{(l + 1)(2l + 1)} \alpha_{lm}^i M_{lm;p,q}^i \right. \\
& \quad - \frac{1}{2} (p + 1) \sum_{l \geq 1, m} \frac{l_q + mp}{l(l + 1)} \beta_{lm}^i M_{lm;p-1,q}^i + \frac{1}{2} p a^2 \sum_{l \geq 1, m} \frac{l(2l - 1)}{2l + 2p - 1} \gamma_{lm}^i M_{lm;p,q}^i \\
& \quad - \frac{1}{2} p (p + 1) \sum_{l \geq 1, m} \frac{l(2l - 1)}{2l + 2p - 1} R_{ij}^l M_{lm;p,q}^i \\
& \quad + \frac{(2p + 1)}{(2p - 1)} \sum_{l \geq 1, m} \frac{2l + 2p - l p - 1}{2l + 2p - 1} \left[ (l_q + mp)(2l_q + mp) - (m + q) \right] \\
& \quad \left. - (l p + mq)(l + p - 1) \right] \gamma_{lm}^i M_{lm;p-2,q}^i, \\
& \int_{|r_i|=a} [v'(r_i) - v_0(r)] \cdot B_{pq}^* (\theta_i, \varphi_i) d\Omega_i = p a^{-p} \beta_{pq}^i - \frac{1}{2} \rho (p + 1)(2p - 1) a^{-p} \gamma_{pq}^i \\
& + \frac{n_{pq}}{(p + q)!} \frac{p}{2p + 3} \sum_{l \geq 1, m} \frac{l(2l - 1)}{2l + 2p - 1} \gamma_{lm}^i M_{lm;p,q}^i.
\end{aligned}
\end{equation}
\[ \int_{|r|=a} [v'(r) - v_0(r)] \cdot C_{pq}^* (\theta, \phi) d\Omega = a^{-(p+1)} \beta_{pq} - \frac{n_{pq}}{(p+q)!} a^p \sum_{l \geq 1, m} \beta_{lm}^p M_{lm;pq}^{(p+1)} + \gamma_{lm}^p M_{lm;pq}^{(p+1)} \]  

where \( j \neq i \) and \( \gamma \) is the imaginary unit. In these expressions we have used the following shorthand notations: the constant \( n_{pq} \) with

\[ n_{pq} = \left( \frac{4\pi (p+q)!}{(2p+1)(p-q)!} \right)^{1/2} \tag{3.12} \]

and

\[ M_{lm;pq}^{(1)} = (-1)^{l+p+q} M_{lm;pq}^{(1)} R^{-(l+p+1)} S_{m,q} \tag{3.13} \]

where \( R = |R_{ij}| \). These expressions are obtained from the Hobson formula, Eqs. (I.6.1)–(I.6.3). With the Hobson formula a solid spherical harmonic, defined with respect to an origin \( O \), can be expressed in terms of solid spherical harmonics defined with respect to another origin. In the present problem we are not interested in the general form of the inner products but in the special case with

\[ v_0(r) = \vec{G}_0 \cdot r. \tag{3.14} \]

In this equation for the incident flow \( \vec{G}_0 \) represents the rate of strain tensor. If we define the incoming fluid velocity with respect to the origin of particle \( i \) we obtain

\[ v_0(r_i + R_j) = \vec{G}_0 \cdot r_i + \vec{G}_0 \cdot R_j = \vec{v}_0(r_i) + U_{0i}, \tag{3.15} \]

with \( U_{0i} \) the incoming flow at the center of particle \( i \) or equivalently the incoming fluid velocity averaged over the surface of particle \( i \) [see in this context Eq. (2.9)]. We can expand \( \vec{v}_0(r_i) \) in terms of basic solutions [Eqs. (3.4a)–(3.4c)]. There are few coefficients which are not zero. These are the coefficients related to the linear basic functions, i.e., \( \alpha_{2m}^0 \neq 0 \), with \(|m| \leq 2 \), and \( \beta_{lm}^1 \neq 0 \), \(|m| \leq 1 \). We can rewrite Eq. (3.15) in the following way:

\[ \vec{v}_0(r_i + R_j) = \sum_{m=-2}^1 \alpha_{2m}^0 \vec{w}_{2m}(r_i) \]

\[ \quad + \sum_{m=-1}^1 \beta_{lm}^1 \vec{w}_{1m}(r_j) + U_{0i}. \tag{3.16} \]

We can now determine the inner products [Eq. (3.8)], using Eqs. (I.5.8)–(I.5.10). We now use the boundary conditions explicitly by using Eq. (2.3), which is a result of the assumption of stick boundary conditions, and Eq. (3.16) which is determined using boundary conditions at infinity concerning the external flow. This results in the following expressions:

\[ \int_{|r|=a} [v'(r) - v_0(r)] \cdot A_{pq}^* (\theta, \phi) d\Omega_i = a^{(p+1)/2} \delta_{pq} \left[ n_{11}(\delta_{q,-1}-\delta_{q,1}) (U_{ix} - U_{Oix}) + i n_{11}(\delta_{q,-1}+\delta_{q,1}) (U_{iy} - U_{Oiy}) \right. \]

\[ + 2 n_{10} \delta_{q,0} (U_{ix} - U_{Oix}) \left. - 10 a \alpha_{2q}^0 \delta_{p,2} \right], \tag{3.17} \]

\[ \int_{|r|=a} [v'(r) - v_0(r)] \cdot B_{pq}^* (\theta, \phi) d\Omega_i = 0, \tag{3.18} \]

\[ \int_{|r|=a} [v'(r) - v_0(r)] \cdot C_{pq}^* (\theta, \phi) d\Omega_i = a \delta_{pq} \left[ n_{11}(\delta_{q,-1}-\delta_{q,1}) \hat{e}_x + i n_{11}(\delta_{q,-1}+\delta_{q,1}) \hat{e}_y + 2 n_{10} \delta_{q,0} \hat{e}_z \right] - 2 a \beta_{1q}^0 \delta_{p,1}. \tag{3.19} \]

The constants \( n_{10} \) and \( n_{11} \) are special cases of \( n_{pq} \) [Eq. (3.12)]. The last inner product can be rewritten by using the surface averaged vorticity,

\[ \Omega_{0i} = \frac{1}{4\pi} \int_{|r|=a} [\nabla \times v_0(r)] d\Omega_i = \frac{3}{8\pi} \left[ n_{11}(\beta_{l,-1}^0 - \beta_{l,1}^0) \hat{e}_x - i n_{11}(\beta_{l,-1}^0 + \beta_{l,1}^0) \hat{e}_y + 2 n_{10} \beta_{l,0}^0 \hat{e}_z \right] = \Omega_0, \tag{3.20} \]

with \( \hat{e}_x \), \( \hat{e}_y \), and \( \hat{e}_z \) the unit vectors in a Cartesian coordinate system. This expression is independent of the chosen origin. Combination of Eqs. (3.19) and (3.20) gives

\[ \int_{|r|=a} [v'(r) - v_0(r)] \cdot C_{pq}^* (\theta, \phi) d\Omega_i = a \delta_{pq} \left[ n_{11}(\delta_{q,-1}-\delta_{q,1}) \hat{e}_x + i n_{11}(\delta_{q,-1}+\delta_{q,1}) \hat{e}_y + 2 n_{10} \delta_{q,0} \hat{e}_z \right], \tag{3.21} \]

Combining (3.9), (3.10), and (3.11) and (3.17), (3.18), and (3.21), respectively, we obtain an infinite set of linear equations of the coefficients \( \{ \alpha_{lm}, \beta_{lm}, \gamma_{lm} \} \) with \( i \in \{1, 2\} \). In I we have expounded on the way the coefficients of interest can be determined.

The obtained set of equations [Eqs. (3.9)-3.11)] can be used to determine the velocities and angular velocities of a system of two particles moving in an arbitrary externally imposed flow field and we see that this set of equations resembles the set used to study the effects of hydro-
dynamic interactions between two particles moving in a fluid without external flow field. The main difference is found in the inner products determined just above [Eqs. (3.17), (3.18), and (3.21)].

IV. THE STRESSLET AS FUNCTION OF THE COEFFICIENTS

In I we have expressed the force \( F_i \) and torque \( T_i \) in terms of some coefficients. These expressions remain unchanged and are

\[
F_i = -\frac{1}{2} \eta_0 \pi n_1 (\gamma_1^i - 1) \hat{e}_x,
\]

\[
T_i = -\frac{1}{2} \eta_0 \pi n_1 (\beta_1^i - 1) \hat{e}_x,
\]

\[
\gamma_2^{i,0} = \frac{1}{\sqrt{15}} \tilde{\gamma}_{2,0}, \quad \gamma_1^{i,1} = -\frac{1}{2} \frac{1}{\sqrt{5}} \tilde{\gamma}_{2,1}.
\]

Furthermore we define the following linear operator [working on the fluid velocity field \( v(r) \)]:

\[
Q[v(r)] \equiv -\eta_0 \left( \frac{\partial}{\partial r} - \frac{1}{r} \right) v(r) - \frac{\eta_0}{r} \nabla \cdot [v(r)] \nabla v(r).
\]

Consequently we can write for \( \Pi_i(r) \) [see Eq. (2.7)]

\[
\Pi_i(r) = \rho(r) \hat{e}_r + Q[v(r)]
\]

and for the stresslet on particle \( i \)

\[
\tilde{S}_i = P[\Pi_i(r)] = P[\rho(r) \hat{e}_r] + P[Q[v(r)]].
\]

There are three sources contributing to \( \tilde{S}_i \), the incoming fluid velocity field \( v_i(r) \), which gives the stresslet \( \tilde{S}_i^2 \), and the two velocity fields \( v_i(r_1) \) and \( v_i(r_2) \) scattered from both particles, which give \( \tilde{S}_i^6 \) and \( \tilde{S}_i^7 \), respectively. We first calculate the contribution of the incoming flow field. We can write [see Eq. (3.16)]

\[
Q[v_0(r_1 + R_1)] = \sum_{m=-2}^{+2} \alpha_{2m}^0 Q[w_{2m}(r_1)]
\]

\[
+ \sum_{m=-1}^{+1} \beta_{2m}^0 Q[w_{1m}(r_1)] + Q(U_{01}).
\]

The last term of Eq. (4.10) equals zero. This is obvious because a constant velocity field is expanded in terms of the basic functions \( w_{1m}(r_1) \) and \( Q[w_{1m}(r_1)] = 0 \). The final result for Eq. (4.10) is

\[
Q[v_0(r_1 + R_1)] = -2\eta_0 \sum_{m=-2}^{+2} \alpha_{2m}^0 A_{2m}(\theta_1, \varphi_1).
\]

There is no contribution from the pressure \( p_0(r) \) because all \( \gamma_{1m}^0 = 0 \) [see Eq. (3.5)]. The contribution to the stresslet \( \tilde{S}_i \) is

\[
\tilde{S}_i^2 = -2\eta_0 \sum_{m=-2}^{+2} \alpha_{2m}^0 [A_{2m}] = \eta_0 a^3 \sum_{m=-2}^{+2} \alpha_{2m}^0 \tilde{\gamma}_{2m}.
\]

In an analogous way, using the expansion in basic solutions of \( p_1(r_1) \) and \( v_1(r_1) \), we obtain

\[
\tilde{S}_i^4 = \eta_0 \sum \gamma_{1m}^{(l+1)} \gamma_{2m}^{(l)} \left[ I(2l - 1)P[Y_{lm} \hat{e}_r] \right.
\]

\[
+ \frac{(l+1)}{2(l+1)} \left[ P[A_{lm}] \right] \right)
\]

\[
= -\frac{9}{10} \eta_0 \sum_{m=-2}^{+2} \gamma_{1m}^{(l+1)} \gamma_{2m}^{(l)}.
\]

In the first sum of this equation we have omitted terms which are zero [see Eq. (4.5)]. The determination of \( \tilde{S}_i^4 \) requires more extensive calculations. First we have to express the pressure \( p_2(r_2) \) and the fluid velocity \( v_2(r_2) \) in terms of \( r_1 \). This is possible with the Hobson formula (see I, Secs. 5 and 6) and the result is
\[ p_2(r_1) = \eta_0 \sum_{l \geq 1} \sum_{m=0}^{\infty} \frac{n_{pq}}{(p+q)!} \left[ \frac{l}{(l+1)(2l+1)} \right] \gamma_{lm}^2 M_{lm;pq}^{21} r_q^2 Y_{pq}(\theta_1, \phi_1) , \]

\[ v_2(r_2) = \sum_{l \geq 1} \sum_{m=0}^{\infty} \frac{n_{pq}}{(p+q)!} \left[ \frac{l}{(l+1)(2l+1)} \right] \alpha_{lm}^2 M_{lm;pq}^{21} r_q^{-1} A_{pq}(\theta_1, \phi_1) + \frac{1}{(l+1)^2} \beta_{lm}^2 M_{lm;pq}^{21} r_q^2 C_{pq}(\theta_1, \phi_1) - r_q^{-1} \left[ \mathbf{R}_{12} \times A_{pq}(\theta_1, \phi_1) \right] + (l+1) \gamma_{lm}^2 M_{lm;pq}^{21} \left[ r_q^2 (r_1 + \mathbf{R}_{12}) Y_{pq}(\theta_1, \phi_1) - \frac{(l-2)}{2(l+1)} r_q^{-1} (r_1 + \mathbf{R}_{12})^2 A_{pq}(\theta_1, \phi_1) \right] , \]

where \( M_{lm;pq}^{21} \) is defined in Eq. (3.13) and \( \mathbf{R}_{12} = \mathcal{R}_{\mathbf{e}} \). Using both expressions it is not difficult to work out \( P[p_2(r, \mathbf{e})] \) and \( P[Q(v_2(r_1))] \) although it requires much space. We restrict ourselves to presenting the final result of the stresslet \( S_1^2 \).

It has the following form:

\[ \mathbf{S}_1^2 = \eta_0 \mathbf{a}^2 \sum_{l=1}^{\infty} \sum_{m=0}^{\infty} \frac{n_{2q}}{(2+q)!} M_{lm;2q}^{21} \left[ \frac{l}{(l+1)(2l+1)} \right] \alpha_{lm}^2 + \frac{1}{2} \epsilon_{lm}^{21} \mathbf{r} \cdot \mathbf{R} \frac{1}{l(l+1)} \beta_{lm}^2 + \frac{2}{(l+1)^2} \gamma_{lm}^2 \right] \mathbf{r}_q \cdot \mathbf{r}_q , \]

\[ \mathbf{S}_1^1 \equiv \mathbf{S}_1^0 + \mathbf{S}_1^2 = -\frac{1}{2} \eta_0 \sum_{q=-2}^{+2} \gamma_{12}^l \mathbf{r}_q \cdot \mathbf{r}_q = \frac{1}{3} \mathbf{S}_1^1 . \]

The final form for the stresslet \( \mathbf{S}_1^1 \), exerted by the fluid on particle 1, is now

\[ \mathbf{S}_1 = \mathbf{S}_1^0 + \mathbf{S}_1^1 + \mathbf{S}_1^2 = -\frac{1}{2} \eta_0 \sum_{q=-2}^{+2} \gamma_{12}^l \mathbf{r}_q \cdot \mathbf{r}_q = \frac{1}{3} \mathbf{S}_1^1 . \]

In an analogous way one can determine \( \mathbf{S}_2^i \) and the result is Eq. (4.18) with \( \gamma_{2q}^l \) replaced by \( \gamma_{12}^l \). It may be noted that in the limit \( R \to \infty \) the stresslet \( \mathbf{S}_2^0 \) should be zero, so that Eqs. (4.17) and (4.18) lead to

\[ \lim_{R \to \infty} \mathbf{S}_1 = \frac{1}{3} \mathbf{S}_1^0 , \]

where the coefficient \( \frac{1}{3} \) corresponds to the Einstein coefficient \( C_1 \) of Eq. (1.1).

We end this section with a final remark. The expressions for the force \( \mathbf{F}_1 \) and torque \( \mathbf{T}_1 \), as functions of some coefficients [Eqs. (4.1) and (4.2)] are even valid in the case of the \( N \)-particle problem. Although it has not been calculated explicitly it can be expected that Eq. (4.3), which expresses the stresslet \( \mathbf{S}_1^i \) as a function of some expansion coefficients, is also valid for the \( N \)-particle problem.

V. THE DERIVATION
OF THE EFFECTIVE VISCOSITY

Our study of the effective viscosity is in line with the ideas presented by Saito [9] because we try to find a relation for the effective viscosity \( \eta_{\text{eff}} \) assuming that the bulk stress on the fluid remains constant independent of the number of suspended particles. We can achieve this by keeping in mind a special experimental setup with which the physical ideas become more clear. Imagine a viscosimeter, which is an apparatus of Couette, where the radii of the cylinders are considered infinite compared with the distance between the inner and outer cylinder. We can regard the surface of the two cylinders as two parallel planes. We assume that the distance between the two planes is large compared to the particle dimensions, so we can ignore wall effects. In this viscosimeter we put a fluid, which is the ambient fluid of the suspension, with viscosity \( \eta_0 \) and we exert an oscillating torque \( \mathbf{T}_0 \) on, say, the outer cylinder while the inner cylinder is kept fixed. The outer cylinder will rotate with an oscillating angular velocity \( \omega_0 \). Under these conditions we can conclude that, between the two planes, an oscillating shear flow exists, which can be described with a rate of strain tensor. This experimental setup is a special case because of the geometry used, but in general we can introduce the (oscillating) rate of strain tensor \( \mathbf{G}_0 \). In the next experiment we add some small undeformable spherical particles to the pure fluid creating a suspension with a volume fraction \( \varphi \) of dispersed particles. This suspension will have an effective viscosity \( \eta_{\text{eff}} \). Again we exert the same oscillating torque \( \mathbf{T}_0 \) on the outer cylinder but now the oscillating angular velocity of it, \( \omega_0 \), is smaller than \( \omega_0 \). This is a consequence of the fluid velocity perturbations caused by the suspended particles. The rate of strain ten-
or of this system, assuming a fixed particle configuration, is \( \tilde{G}_\text{eff} \) and is effectively a volume average of the rate of strain in the system,
\[
\tilde{G}_\text{eff} = \frac{1}{V} \int [\nabla \mathbf{v}(\mathbf{r})] dV \quad (5.1)
\]
The rate of strain depends also on the configuration of the suspended particles. However, if many particles are present in a macroscopic volume \( V \) then the configuration average and the volume average yield the same result. Consequently the calculation of the configuration average of \( \tilde{G}_\text{eff} \) is superfluous. The shear forces per unit surface on the outer cylinder of the apparatus of Couette of the experimental setup described above are equal in both experiments so we know the following relation between \( \tilde{G}_0 \) and \( \tilde{G}_\text{eff} \):
\[
\eta_0 \tilde{G}_0 = \eta_\text{eff} \tilde{G}_\text{eff} \quad (5.2)
\]
The calculations to determine \( \eta_\text{eff} \) consist of two main parts, but in both the same numerical problem has to be solved. In the first step we express the rate of strain tensor of a suspension, with a volume fraction \( \varphi \) of dispersed particles, in terms of the mobility matrix and the stresslets. If the system contains \( N \) force- and torque-free particles, then Eq. (2.9) implies
\[
\tilde{G}_{0,i} = \sum_{j=1}^{N} \tilde{\mu}^{dd}_{ij} \tilde{S}_{j} \quad (5.3)
\]
We assume that the stresslets are independent of particle configuration. Summing over \( i \), taking the configurational average, and using the assumption of constant stresslets, we have
\[
\langle \tilde{G} \rangle_c = \langle \tilde{\mu}^{dd}_{11} + \tilde{\mu}^{dd}_{12} \rangle_c \tilde{S} \quad (5.4)
\]
where
\[
\tilde{\mu}^{dd}_{ij, a\mu
u} = \langle \mu^{dd}_{ij, a\mu
u} \rangle_0 + \frac{3}{20\pi \eta_\text{eff} a^2} \left[ \frac{3}{2} A^{dd}_{ij}(R) \langle \hat{\mathbf{R}}_a \hat{\mathbf{R}}_b \rangle^{(ab)} \langle \hat{\mathbf{R}}_\mu \hat{\mathbf{R}}_\nu \rangle^{(\mu\nu)} \right. \\
+ 2B^{dd}_{ij}(R) \langle \delta^{(ab)} \delta^{(\mu\nu)} \rangle - \langle \hat{\mathbf{R}}_a \hat{\mathbf{R}}_b \rangle^{(ab)} \langle \hat{\mathbf{R}}_\mu \hat{\mathbf{R}}_\nu \rangle^{(\mu\nu)} \\
+ C^{dd}_{ij}(R) \langle \delta^{(ab)} \delta^{(\mu\nu)} \rangle - 2 \langle \delta^{(ab)} \delta^{(\mu\nu)} \rangle + \frac{1}{2} \langle \hat{\mathbf{R}}_a \hat{\mathbf{R}}_b \rangle^{(ab)} \langle \hat{\mathbf{R}}_\mu \hat{\mathbf{R}}_\nu \rangle^{(\mu\nu)} \right] , \quad (5.6)
\]
where \( \hat{\mathbf{R}}_a = R_a/R \) and \( i, j \in \{1, 2\} \). The symbol \( \langle \cdots \rangle^{(ab)} \) denotes a projection onto the symmetric and traceless part in the index pair \( (a, b) \). The projected tensors in Eq. (5.6) read explicitly, according to Cichocki, Felderhof, and Schmitz [7],
\[
\langle \hat{\mathbf{R}}_a \hat{\mathbf{R}}_b \rangle^{(ab)} = \hat{\mathbf{R}}_a \hat{\mathbf{R}}_b - \frac{1}{3} \delta^{ab} \langle \delta^{\mu \nu} \rangle^{(\mu \nu)} - \frac{1}{3} \delta^{ab} \delta^{\mu \nu} \\
\langle \delta^{(ab)} \delta^{(\mu \nu)} \rangle = \frac{1}{2} \delta^{\mu \nu} \langle \delta^{(ab)} \rangle^{(ab)} + \frac{1}{2} \delta^{\mu \nu} \langle \delta^{(ab)} \rangle^{(ab)} - \frac{1}{2} \delta^{ab} \delta^{\mu \nu} \quad (5.7)
\]
Furthermore,
\[
\langle \mu^{dd}_{ij, a\mu \nu} \rangle_0 = \frac{3}{40\pi \eta_\text{eff} a^2} \langle \delta^{\mu \nu} \rangle^{(ab)} - \frac{1}{3} \delta^{ab} \delta^{\mu \nu} \delta_{ij} \quad (5.8)
\]

\[\langle \mu^{dd}_{11} \rangle_c = \mu^{dd}_{0} + \rho_0 \int_{|\mathbf{R}| \geq 2a} \langle \mu^{dd}_{11} \rangle_c \mathbf{R} d\mathbf{R} \] 
\[\langle \mu^{dd}_{12} \rangle_c = \rho_0 \int_{|\mathbf{R}| \geq 2a} \mu^{dd}_{12} d\mathbf{R} \] 
\[\mu^{dd}_{0} \text{ is the pure one-particle mobility, } \rho_0 = N/V, \text{ and } \langle \cdots \rangle_c \text{ denotes a configuration average. The pair correlation is simply the no-overlap condition. One should be cautious with the calculation of } \langle \mu^{dd}_{12} \rangle_c \text{ because the integral is conditionally convergent. We shall discuss this point later on. Furthermore, } \tilde{G} \text{ and } \vec{S} \text{ now stand for an average over particles:} \]
\[
\tilde{G} = \frac{1}{N} \sum_{i=1}^{N} \tilde{G}_{0,i} \quad (5.5)
\]
We may identify the configuration average of \( \tilde{G} \) with \( \tilde{G}_\text{eff} \).

The double dot in Eqs. (5.3) and (5.4) stands for a double contraction between the fourth-rank dipole-dipole mobility tensor and the stresslet.

We do not present the details of the calculation of the components of the dipole-dipole mobility matrix \( \mu^{dd} \). This calculation is more or less a technical matter. The procedure is in principle the same as presented in 1. An important difference, however, is the fact that the particles are force- and torque-free. This means that \( \beta_{1m} = 0 \) and \( \gamma_{1m} = 0 \), with \( m \in \{-1, 0, 1\}, \) which is a simple consequence of Eqs. (4.1) and (4.2). Furthermore we express the coefficients \( \alpha^{ab}_{pq} \), with \( p \geq 1, \beta^{ab}_{pq} \), with \( p \geq 2, \) and \( \gamma^{ab}_{pq} \), with \( p \geq 3 \) (and the accompanying allowed values for the azimuthal indices \( q \) ), for \( i = 1, 2 \), in terms of the coefficients \( \gamma_{1m} \), \( |m| \leq 2 \) and \( j = 1, 2 \). For more details see article 1 [26].

The dipole-dipole mobility matrix is made up of tensors of rank four, \( \mu^{dd}_{ij} \), and these tensors have the following structure, according to Cichocki, Felderhof, and Schmitz [7], if hydrodynamic pair interactions are included (we use a slightly different notation):

\[
\langle \mu_{ij, a\mu \nu} \rangle_0 = \langle \mu^{dd}_{ij, a\mu \nu} \rangle_0 - \frac{3}{40\pi \eta_\text{eff} a^2} \left[ \frac{3}{2} A^{dd}_{ij}(R) \langle \hat{\mathbf{R}}_a \hat{\mathbf{R}}_b \rangle^{(ab)} \langle \hat{\mathbf{R}}_\mu \hat{\mathbf{R}}_\nu \rangle^{(\mu\nu)} \right. \\
+ 2B^{dd}_{ij}(R) \langle \delta^{(ab)} \delta^{(\mu \nu)} \rangle - \langle \hat{\mathbf{R}}_a \hat{\mathbf{R}}_b \rangle^{(ab)} \langle \hat{\mathbf{R}}_\mu \hat{\mathbf{R}}_\nu \rangle^{(\mu\nu)} \\
+ C^{dd}_{ij}(R) \langle \delta^{(ab)} \delta^{(\mu \nu)} \rangle - 2 \langle \delta^{(ab)} \delta^{(\mu \nu)} \rangle + \frac{1}{2} \langle \hat{\mathbf{R}}_a \hat{\mathbf{R}}_b \rangle^{(ab)} \langle \hat{\mathbf{R}}_\mu \hat{\mathbf{R}}_\nu \rangle^{(\mu\nu)} \right] , \quad (5.6)
\]
index \( m \) and the set of linear equations is decoupled considering these indices. The \( A^{ij}_{m}(R) \) are related to the set of linear equations for \( m = 0 \), the \( B^{ij}_{m}(R) \) to \( |m| = 1 \), and the \( C^{ij}_{m}(R) \) to \( |m| = 2 \). Consequently the set of linear equations with \( |m| > 2 \) is of no interest to us.

The lowest-order solutions (\( L = 2 \), see article I) of the hydrodynamic functions result in the following expressions:

\[
\begin{align*}
A^{ij}_{11}(x) &= O(x^8), \\
B^{ij}_{11}(x) &= O(x^8), \\
C^{ij}_{11}(x) &= O(x^8) \\
A^{ij}_{12}(x) &= -5x^3 + 12x^5 + O(x^8), \\
B^{ij}_{12}(x) &= -\frac{2}{3}x^3 - 8x^5 + O(x^8), \\
C^{ij}_{12}(x) &= 2x^5 + O(x^8),
\end{align*}
\]  
(5.9)

where \( x = a/R \). Our aim is now the calculation of the configuration average of the difference \([\mu^{dd}_{ij} - \mu^{dd}_{ij}(0)]\) using the simplest form of the pair distribution function, excluding particle overlap only, or \( g(R) = g(|R|) = 0 \) if \( |R| < 2a \) and \( g(|R|) = 1 \) if \( |R| \geq 2a \). The configuration average is now defined as

\[
\langle [\mu^{dd}_{ij} - \mu^{dd}_{ij}(0)] \rangle_c = n_0 \int_{|R| \geq 2a} [\mu^{dd}_{ij} - \mu^{dd}_{ij}(0)] dR,
\]
(5.10)

with \( n_0 = N/V \) the particle density. In the following we denote a configuration average with \( \langle \cdot \rangle_c \). This averaging procedure must be done very carefully because of the appearance of conditionally convergent integrals. This has to do with the terms proportional to \( x^5 \) in the expressions for \( A^{dd}_{12}(R) \) and \( B^{dd}_{12}(R) \) [see Eq. (5.9)]. If we consider the hydrodynamic functions with terms up to \( x^5 \) only we arrive at the situation already considered by Saito. Proper volume averaging gives the so-called Saito contribution, which gives rise to the virial coefficient \( c_S = -1 \) [9]. This coefficient comes out in work of other authors, e.g., Fedderhof reproduced it using a local field argument [29,30] or from a virtual overlap contribution to the two-body cluster integral [31]. See in this context also the work of Bedaux, Kapral, and Mazur [16]. With the theory of renormalized cluster expansions it is possible to avoid these conditionally convergent integrals [32,33,11]. This theory confirms the value of the Saito coefficient \( c_S \). These renormalized cluster expansions have also been used in the theory of sedimentation [34]. This kind of conditionally convergent integrals also appears in the work of Batchelor and Green although in another form [5]. For further calculations we split off the terms proportional to \( x^3 \) and \( x^5 \); the remaining parts of the hydrodynamic functions are denoted by \( \tilde{A}^{dd}_{ij}(R), \tilde{B}^{dd}_{ij}(R), \) and \( \tilde{C}^{dd}_{ij}(R) \). From Eq. (5.9) we see that \( A^{dd}_{12}(R) = \tilde{A}^{dd}_{12}(R), \) \( B^{dd}_{12}(R) = \tilde{B}^{dd}_{12}(R), \) and \( C^{dd}_{12}(R) = \tilde{C}^{dd}_{12}(R) \). To avoid misunderstanding we shall label the remaining part of the configurational averages with \( SR \), which stands for short-range part. The contribution of the terms just split off will be represented by the Saito coefficient \( c_S \).

It is not difficult to evaluate the angular average of the short-range part of Eq. (5.10) keeping \( R \), the interparticle distance, constant. The result is

\[
\langle [\mu^{dd}_{ij} - \mu^{dd}_{ij}(0)] \rangle_{c, SR} = \int [\mu^{dd}_{ij} - \mu^{dd}_{ij}(0)] d\Omega = \frac{3}{20\pi \eta a^2} \int_{ij} f^{dd}_{ij}(R) \left[ \delta_{\alpha \beta} \delta_{\mu \nu} + \delta_{\alpha \mu} \delta_{\beta \nu} - \frac{1}{3} \delta_{\alpha \beta} \delta_{\mu \nu} \right],
\]
(5.11)

with

\[
f^{dd}_{ij}(R) = \frac{1}{2} \pi [ \tilde{A}^{dd}_{ij}(R) + 2 \tilde{B}^{dd}_{ij}(R) + 2 \tilde{C}^{dd}_{ij}(R) ].
\]
(5.12)

We are now able to determine the double contraction between the angular-averaged mobility tensor and the stresslet \( \tilde{S} \),

\[
\langle [\mu^{dd}_{ij} - \mu^{dd}_{ij}(0)] \rangle_{c, SR} \tilde{S} = \frac{3}{10\pi \eta a^3} f^{dd}_{ij}(R) \tilde{S}.
\]
(5.13)

The total configuration average is now

\[
\langle [\mu^{dd}_{ij} - \mu^{dd}_{ij}(0)] \rangle_{c, SR} = \frac{3}{20\pi \eta a^3} f^{dd}_{ij}(R) \tilde{S}.
\]
(5.14)

where we have changed the variable of integration using \( x = a/R \). Substitution of this result for the configuration averageds in Eq. (5.4) gives

\[
\tilde{G}_{eff} = \frac{3}{20\pi \eta a^3} \tilde{S} + 1 + \frac{1}{3} \frac{\eta \phi}{20\pi \eta a^3} \left[ \frac{3}{2} \int_0^{1/2} \sum_{ij} \left[ \tilde{A}^{dd}_{ij}(x) + 2 \tilde{B}^{dd}_{ij}(x) + 2 \tilde{C}^{dd}_{ij}(x) \right] \frac{dx}{x^4} \right].
\]
(5.15)

The integral in Eq. (5.15) can be calculated numerically and the result is \(-0.694 \). The final result is

\[
\tilde{G}_{eff} = \frac{3}{20\pi \eta a^3} (1 - 1.42\phi) \tilde{S}.
\]
(5.16)
sor contractions $\vec{M}_{ij}^{dd} S$ are introduced as a shorthand notation to describe the rate of strain of the scattered velocity fields around the two particles in terms of the stresslet $S$ (in the case that pair interactions are included only),

$$
\eta \vec{G}_0 = \eta \vec{G}_{\text{eff}} + (\vec{M}_{11}^{dd} + \vec{M}_{12}^{dd}) \cdot \vec{S}.
$$

With this relation and Eq. (5.16) we can eliminate the stresslet $S$. Comparison of the final relation with Eq. (5.2) gives us $\eta_{\text{eff}}$. The determination of $\eta_{\text{eff}}(\vec{G}_0 - \vec{G}_{\text{eff}})$, as a function of volume fraction $\varphi$, is possible by studying the following quantity:

$$
[\nabla v_\beta(x)]' - [\nabla v(x)]' = - \sum_{i=1}^N [\nabla v_i(x)]' \equiv (\vec{\Delta}_1 + \vec{\Delta}_2)/\eta_0,
$$

(5.18)

with $v_i(x)$ a scattered velocity field defined with respect to the center of particle $i$. We have introduced $N$ scattered velocity fields because there are $N$ particles present in the suspension, but we study two-particle hydrodynamic interactions only. The tensors $\vec{\Delta}_1$ and $\vec{\Delta}_2$ will be defined below. In the first place we have to calculate the volume average of the left-hand side of Eq. (5.18). We know the rate of strain in the pure fluid case, so the result of the volume averaging of $[\nabla v_\beta(x)]'$ is

$$
\frac{1}{V} \int [\nabla v_\beta(x)]' dV = \vec{G}_0.
$$

(5.19)

We also know the effective rate of strain of a suspension with volume fraction $\varphi$ of dispersed spherical particles. We can study this rate of strain by measuring the angular frequency $\omega_{\text{eff}}$ of the outer cylinder of a Couette apparatus with the same torque on the outer cylinder as in the pure fluid case described above. The rate of strain is $\vec{G}_{\text{eff}}$ and is a volume average in the above sense [see Eq. (5.19)], with $v_\beta(x)$ replaced by $v(x)$,

$$
\frac{1}{V} \int [\nabla v(x)]' dV = \vec{G}_{\text{eff}}.
$$

(5.20)

We are also able to calculate the volume and configuration average of the right-hand side of Eq. (5.18). To obtain the final result we distinguish between a quasi-one-particle contribution, denoted by $\vec{\Delta}_1$, and an excluded volume contribution, $\vec{\Delta}_2$. It is noteworthy that we have to consider all scattered velocity fields, but the averages of the terms in the sum of Eq. (5.18), concerning the fluid velocity fields, yield the same result. For convenience we calculate the expressions below by using the velocity fields $v_i(x)$ and $v_2(x)$ only; this makes no difference in the calculation of the final result. The quasi-one-particle contribution can be determined very easily with the following relation:

$$
\langle \vec{\Delta}_{1,\text{eff}} \rangle_c = - \eta \varphi \int_{|x_2| \geq a} [\nabla v_2(x_2)]' dV = \frac{1}{\eta_0 S}.
$$

(5.21)

We call $\langle \vec{\Delta}_{1,\text{eff}} \rangle_c$ a quasi-one-particle contribution because the expansion coefficients of the velocity field $v_2(x)$ contain the hydrodynamic pair interactions. Our expression for the stresslet $S$ follows from (5.16),

$$
\vec{S} = \frac{\eta_0 \varphi}{\eta_0} \frac{1}{1 - 1.42 \varphi} \vec{G}_{\text{eff}},
$$

(5.22)

and we obtain for $\langle \vec{\Delta}_{1,\text{eff}} \rangle_c$

$$
\langle \vec{\Delta}_{1,\text{eff}} \rangle_c = \frac{\varphi}{\eta_0} \frac{1}{1 - 1.42 \varphi} \eta_0 \vec{G}_{\text{eff}}.
$$

(5.23)

It is obvious that we have not excluded the volume of the other particles to obtain Eq. (5.23). The final result can be obtained by addition of the extra amount is $\langle \vec{\Delta}_{2,\text{eff}} \rangle_c$. This extra amount is the negative value of the integral, defined in Eq. (5.21), within the volume occupied by the other particles. The extra amount $\langle \vec{\Delta}_{2,\text{eff}} \rangle_c$ is

$$
\langle \vec{\Delta}_{2,\text{eff}} \rangle_c = \eta_0 \varphi \int_{|x_2| \geq a} dV \int_{|x_1| \leq a} dV \int_{|x_1| \leq a} dV = \langle \vec{G}_{\text{eff}} \rangle_c + \langle \vec{G}_{\text{eff}} \rangle_c + \langle \vec{G}_{\text{eff}} \rangle_c,
$$

(5.24)

with $\vec{R}$ the interparticle distance. Although we did not write down the $\vec{R}$ dependence of the integrand in the equation above it should be remembered that this dependence is implicitly assumed in the expansion coefficients $\{a_{m,\beta}, b_{m,\gamma}, \gamma_{m, \beta} \}$. Evidently we have used the simplest form of the pair distribution function which excludes overlap of the two particles only. The integral over the volume of particle 1 can be determined easily from the following integral theorem for tensors (see, e.g., Ref. [35]):

$$
\int \vec{A}_i \cdot dV = \int \vec{A}_i \cdot \frac{\partial x_k}{\partial x_i} dA - \int \frac{\partial A_{ik}}{\partial x_k} x_j dV,
$$

(5.25)

with $V$ the enclosed volume, $A$ the surface enclosing $V$, and $n_k$ a component of an outward unit vector. In our case this unit vector is pointing into the fluid because $V$ is the volume occupied by particle 1. Define now the symmetric and traceless tensor $\vec{B}$ with components

$$
B_{ij} = - \Pi_{ij} + \frac{1}{2} \Pi_{ii} \Pi_{ij},
$$

(5.26)

where we used the summation convention for double indices. The tensor $\Pi$ is the pressure tensor,

$$
\Pi = - \vec{p} - 2 \eta_0 (\nabla v)^2.
$$

(5.27)

It is obvious that $\vec{B} = 2 \eta_0 (\nabla v)^2$. Combination of Eqs. (5.25)–(5.27) gives, with the identity $\nabla \cdot \vec{B} = 0$,

$$
2 \eta_0 \int_{|x_1| \leq a} dV \int_{|x_1| \leq a} dV \int_{|x_1| \leq a} dV = \int_{|x_1| \leq a} \vec{B}_{ij} x_j n_k dA = \langle \vec{S}_{1,ij} \rangle_c,
$$

(5.28)

the stresslet exerted by the fluid velocity field $v_2$ on particle 1. Substitution of this result for the integrand in Eq. (5.24), use of Eq. (4.16), and using the set of linear equations (3.9)–(3.11) to express the coefficients $\alpha_{pq}^2$ with $p \geq 1$, $\beta_{pq}^2$ with $p \geq 2$, and $\gamma_{pq}^2$ and $p \geq 3$ (and the accompanying allowed values for the azimuthal indices $q$) in terms of the coefficients $\gamma_{m, \beta} |m| \leq 2$, we obtain, after calculation of the configuration average,
or, by using Eq. (5.16),
\[
\langle \hat{\Delta}_{2,\text{eff}} \rangle_c = \frac{1.42q^2}{(1 - 1.42q^2)} \eta_0 G_{\text{eff}}.
\] (5.30)

Consequently, using the constraint \( \eta_0 G_0 = \eta_{\text{eff}} G_{\text{eff}} \), we obtain the relation for the high-frequency effective viscosity already presented in Sec. I, viz.,
\[
\eta_{\text{eff}} = \eta_0 \left[ 1 + \frac{1}{1 + 1.42q^2} \right] \eta_0 G_{\text{eff}}.
\] (5.31)

Finally we express the stresslet \( \bar{S} \) as a function of \( \varphi \) and \( \bar{G}_0 \) only, using Eqs. (5.2) and (5.31),
\[
\bar{S} = \frac{1}{\eta_0 \pi \eta_0 \rho} \left( 1 + \frac{1}{1 + 1.08q^2 + 1.42q^2} \right) \bar{G}_0.
\] (5.32)

The stresslet \( \bar{S} \) is a function of volume fraction, although independent of particle configuration as assumed below Eq. (5.3). This relation is an extension of an expression presented by Saito [Ref. [9] Eq. (8) with \( k_0 \) replaced by \( G_0 \) and \( \frac{1}{\eta_0 \pi \rho a^3} \langle \kappa \rangle \) replaced by \( \bar{S} \). The numerical coefficient 1.08 arises from the subtraction \( \frac{1}{2} - 1.42 \) and in the Saito expression this coefficient is \( \frac{1}{2} + c_1 \). The term of quadratic order in \( q \) is an excluded volume effect and cannot be present in the Saito expression.

**VI. DISCUSSION**

In this section we shall compare our theoretical expression for the effective viscosity with experiment, other theories, and results from simulation experiments and also discuss the limitations of our expression of \( \eta_{\text{eff}} \). We start with a remark on the divergent behavior of our expression and the position of the pole. We see that our expression of the effective viscosity [Eq. (5.31)] has a pole near \( \varphi \approx 0.7 \) which is something higher than \( \varphi_{RC} \approx 0.64 \), the random-close-packing volume fraction. Furthermore it does not differ much from the maximum packing fraction. There are arguments that we should compare our pole with \( \varphi_{RC} \). One of these arguments is the fact that we used the isotropic pair distribution function while averaging over all possible particle configurations. In the case of maximum packed structures the isotropy has disappeared because there is some crystal-like structure. It is interesting to note that Krieger and Dougherty [36] introduced an empirical expression for \( \eta_{\text{eff}} \) later derived by Ball and Richmond using a mean field argument [see Eqs. (6.4)–(6.6) in Ref. [37]],
\[
\eta_{\text{eff}} = \eta_0 \left( 1 - k_f \varphi \right) \left( 1 + \frac{1}{1 + 1.42q^2} \right),
\] (6.1)

where \( k_f \) is the reciprocal of the maximum packing fraction or the reciprocal of the random-close-packing fraction (Ball and Richmond). This expression is often used to fit experimental data, with \( k_f \) as a fitting parameter (see, e.g., Ref. [38]).

In Fig. 1 we have plotted the effective viscosity \( \eta_{\text{eff}} \) as a function of volume fraction. In this figure we compare our results of \( \eta_{\text{eff}} \) with results of the effective viscosity of monodisperse hard-sphere systems, experimentally obtained by van der Werff et al. [1]. We have made no distinction among the measurements on monodisperse systems with different particle radii because the data of van der Werff et al. do not show any effect of particle size. This can be expected on theoretical grounds. The theoretical result corresponds with the experimental data up to \( \varphi \approx 0.6 \). This is remarkable because we used two-particle hydrodynamic interactions only to derive Eq. (5.31). At the moment it is not clear how three- and more-particle hydrodynamic interactions will change our result of \( \eta_{\text{eff}} \) but these interactions may shift the pole in Eq. (5.31) to a slightly lower value (see the simulation results and numerical data discussed below). For reasons of completeness we have included in Fig. 1 the theoretical results of Beenakker, derived by using many-particle hydrodynamic interactions [12], and the virial expansion of Batchelor and Green, derived by using two-particle hydrodynamic interactions only [5]. The analytical result of Batchelor and Green is
\[
\eta_{\text{eff}} = \eta_0 \left( 1 + \frac{1}{2} q + 5.2q^2 \right),
\] (6.2)

where they calculated the second-order virial coefficient with an accuracy of 6%. More recent calculations, using more accurate hydrodynamic functions, give for the second-order virial coefficient \( C_2 = 5.00 \) (see, e.g., Refs. [6] and [20]). We should obtain the same second-order virial coefficient by expanding our result for small \( \varphi \) and the result is \( C_2 = 4.97 \). This difference may be caused by small numerical errors. Recently Thomas and Muthukumar presented results for the effective viscosity which include three-particle hydrodynamic interactions [39]. Their expression is
\[
\eta_{\text{eff}} = \eta_0 \left( 1 + \frac{1}{2} q + 5.00q^2 + 6.40q^3 \right).
\] (6.3)

This expression is a poor improvement that leads to the conclusion that many higher-order terms should be in-
cluded to obtain an expression that describes the experimental results and simulation data reasonably well. Nevertheless we can use the third-order virial coefficient to make an estimate of the extra contribution to Eq. (5.31), which is a combination of a pure three-particle contribution and the two-particle contribution, configurationally averaged with the $\varphi$-dependent part of the pair distribution function. We assume that our expression should give the same cubic virial expansion in the low $\varphi$ regime, but it is noteworthy that this estimate gives an indication only. Supposing that $\varepsilon \varphi^2$ is the combined three-two-particle contribution, we obtain the following expression:

$$\eta_{\text{eff}} = \eta_0 \left( 1 + \frac{1}{1 + 0.42 \varphi + \varepsilon \varphi^2} \right),$$

(6.4)

Low $\varphi$ expansion and comparing with Eq. (6.3) results in $\varepsilon \approx 0.2$. There are some other estimates of the third-order virial coefficient e.g., by using the Krieger-Dougherty relation [Eq. (6.1)] or the Mooney equation [40], viz.,

$$\eta_{\text{eff}} = \eta_0 \exp \left( \frac{1}{1 - k_f \varphi} \right),$$

(6.5)

with $k_f$ a fitting parameter. The estimates made by Thomas and Muthukumar, based on the Krieger-Dougherty relation and the Mooney equation, give $C_1 \approx 8.5$ [39]. In this case $\varepsilon \approx -0.4$. In both cases the three-body coefficient $\varepsilon$ is much smaller than the coefficient of the term representing two-body effects, viz., 1.42. As such the use of a virial expansion in the numerator and in the denominator of Eq. (6.4) converges much faster than the commonly used virial expansions of the viscosity itself. Future research should clarify this matter. The above discussion does not intend to question the importance of many-particle hydrodynamic interactions, necessary to understand the behavior of transport coefficients in dense suspensions. It tries to separate the effects of many-particle hydrodynamic interactions from the divergence of the effective viscosity which is already found if one considers the lower-order contributions to the viscosity.

In Fig. 2 we have compared our theoretical result with simulation results of Phillips, Brady, and Bossis [19] and with numerical data of Ladd [21] and we see that our theoretical result of $\eta_{\text{eff}}$ is systematically somewhat lower than the numerical data for $\varphi \geq 0.4$. This difference may be explained by the fact that we have ignored many-particle hydrodynamic interactions while both numerical results of $\eta_{\text{eff}}$ include the effect of many-particle interactions. Finally we want to point out that Beenakker's remark about the range of validity of his theoretical results is also applicable to our results [12]. He derived a lower- and an upper-frequency limit and his results are valid between the two. The upper limit is determined by the fre-

Fig. 2. The high-frequency effective viscosity is plotted vs volume fraction $\varphi$. The solid curve represents our result, Eq. (5.31). The crosses are the simulation results of Phillips et al. and the open circles are the numerical results of Ladd.

VII. CONCLUSION

In this article we have derived an expression for the high-frequency effective viscosity in a way similar to Saito's in the early 1950s. The result is in good agreement with experimental data and the agreement with simulation results and numerical data is reasonable. It is known that many-particle hydrodynamic interactions are, in general, important so higher-order corrections to Eq. (5.31) are to be studied, but these hydrodynamic interactions are likely to give small corrections to the expression derived in this article because the divergence in our expression for the effective viscosity is found if one considers two-particle hydrodynamic interactions only. This is not the case if one considers the virial expansion (6.2) that results from the approach by Batchelor and Green [5]. This advantage should be exploited in future research. In a future article we plan to present some results for diffusion coefficients and sedimentation (translational and rotational) where the effect of three-particle interactions is included [41]. The procedure presented in that paper, combined with the ideas outlined in this article, might give us the answer to the question of how Eq. (5.31) should be corrected for three-particle hydrodynamic interactions.