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Statistical description of a non-Brownian suspension

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A suspension of non-Brownian particles in a constant, external force field is studied. Statistical description of the system, based on reduced distribution functions is found. A hierarchy of equations for the correlation functions, analogous to the BBGKY hierarchy known from the kinetic theory of gases, is derived and thoroughly analysed. As an example, the problem of deriving the two-particle correlation function for a sedimenting system is considered.

1. Introduction

Suspensions of non-Brownian particles, although seemingly simple systems, are surprisingly difficult to treat theoretically ([1–3]). Vivid research has been going on in the field for a few decades. Despite efforts, many puzzles still remain unsolved. The main problem which we address is the question of finding a statistical description of a suspension of particles.

Particles suspended in a viscous fluid interact in an indirect manner. The movement of a particle creates a fluid velocity field slowly decaying with distance. This field induces motion of other particles giving rise to nonlinear, so-called hydrodynamic interactions. Although these can, in the time-scales adopted, be treated as effectively instantaneous, even in the low-concentration limit the hydrodynamic interaction usually cannot be limited solely to two-particle encounters. It is important to keep in mind that two isolated particles sedimenting in an infinite, quiescent fluid do not change their relative position. If the dynamics is limited only to two-particle interactions, the equation for the pair distribution function vanishes. The initial particle distribution does not change with time. It is therefore essential to consider the many-body character of the hydrodynamic interactions in order to find the steady-state micro-structure of a monodisperse, non-Brownian suspension.

In this paper we summarise the derivation of a statistical description of a non-Brownian suspension. The methods used originate in the kinetic theory of gases. A hierarchy of equations governing the time evolution of the correlation and reduced distribution functions is found and analysed. Further, as an example, we show how this sequence of equations can be used to find the pair distribution function and the sedimentation coefficient for a low-concentration monodisperse suspension in steady state.

2. Statistical description of a system with hydrodynamic interactions

2.1. Liouville equation

The system under consideration is a suspension of N spheres of equal radii a immersed in an incompressible fluid of shear viscosity η. In the regime studied, the particle Reynolds number is assumed to be small enough to treat inertial effects as virtually absent and therefore negligible. Likewise, the Peclet number defined as the ratio $U_0 a/D$ (U_0 – Stokes settling velocity, D – diffusion constant) describing the relative influence of hydrodynamic effects and Brownian motion, is presumed to be large. On these assumptions and with the time scales imposed, the fluid motion is governed by the stationary Stokes equation:

$$\eta\nabla^2\mathbf{v}(\mathbf{r}) - \nabla p(\mathbf{r}) = \mathbf{0}, \qquad \nabla\cdot\mathbf{v}(\mathbf{r}) = 0. \tag{2.1}$$

Stick boundary conditions on the particles are adopted.

The linearity of the Stokes equation induces a linear relation between the velocities of the suspended particles and the forces acting on them. This relation establishes the equations of motion of the particles:

$$\frac{d\mathbf{R}_i}{dt} = \mathbf{U}_i = \sum_{j=1}^{N}\left\{\boldsymbol{\mu}_{ij}(\mathbf{X})\mathbf{F_j}(\mathbf{X})\right\}. \tag{2.2}$$

$\boldsymbol{\mu}$ is the translational part of the mobility matrix. This is a hydrodynamic matrix function of the configuration $\mathbf{X} = (\mathbf{R}_1, \ldots, \mathbf{R}_N)$ of all the particles, describing the indirect, fluid mediated interactions between particles. We will assume the forces acting on the particles to originate in an external field.

Let $\rho(\mathbf{X};t)$ be the probability distribution function for the discussed system. It describes the probability of finding at time t the system in the state $\mathbf{X}$. Due to (2.2) the probability conservation equation follows:

$$\frac{\partial\rho(\mathbf{X};t)}{\partial t} + \sum_{i,j}\nabla_i\cdot\{\boldsymbol{\mu}_{ij}(\mathbf{X})\mathbf{F}_j\rho(\mathbf{X};t)\} = 0. \tag{2.3}$$

This is the Liouville equation for the discussed system.

2.2. Hydrodynamic interactions

In order to analyse the Liouville Equation (2.3), first a formula for the mobility matrix has to be generated. This matrix function quantifies the hydrodynamic interactions in the regime of Stokes dynamics. It describes the relation between the forces acting on the particles and their velocities and is defined by

$$\mathbf{U} = \boldsymbol{\mu}\boldsymbol{\mathcal{F}}, \tag{2.4}$$

where $\boldsymbol{\mathcal{F}} = (\mathbf{F}_1, \ldots, \mathbf{F}_N)$ are the forces acting on the particles. Similarly, $\mathbf{U} = (\mathbf{U}_1, \ldots, \mathbf{U}_N)$ are the translational velocities. The matrix $\boldsymbol{\mu}$ is a square $3N \times 3N$ Cartesian tensor describing the translational components of the interactions. We assume here that there is no torque exerted on the particles and that the rotational degrees of freedom are therefore irrelevant. To find the mobility matrix $\boldsymbol{\mu}$, the Stokes equations (2.1) have to be solved

To solve the Stokes equations, given the stick boundary conditions on the particles' surfaces, a density $\mathbf{f}_i(\mathbf{r})$ of induced forces is introduced for each particle [4]. Inserting these into the equations, the rigid-body motion of the particles may be interpreted as a fictitious fluid flow which obeys the Stokes equations now valid in the entire space. Then the solution of the hydrodynamic equations can be written in terms of a Green function $\mathbf{G}(\mathbf{r}, \mathbf{r}')$

$$\mathbf{v}(\mathbf{r}) = \int \mathbf{G}(\mathbf{r}, \mathbf{r}')\mathbf{f}(\mathbf{r}')d\mathbf{r}. \tag{2.5}$$

In the case of an infinite fluid, the Green operator is the Oseen tensor [7].

Restricting the expression to the surface of a given particle, solution (2.5) can be written in terms of two distinct parts

$$\mathbf{v}(\mathbf{r}) = \mathbf{u}_i(\mathbf{r}) = \int \mathbf{G}(\mathbf{r}, \mathbf{r}')\mathbf{f}_i(\mathbf{r}')d\mathbf{r}' + \sum_{j \neq i} \int \mathbf{G}(\mathbf{r}, \mathbf{r}')\mathbf{f}_j(\mathbf{r}')d\mathbf{r}', \qquad \mathbf{r} \in S_i, \quad i = 1, \ldots, N. \tag{2.6}$$

The first part accounts for the contribution to the surface velocity $\mathbf{u}_i$ from the force density $\mathbf{f}_i(\mathbf{r})$ concentrated on the surface of the i-th particle, whereas the second term describes the influence of force densities present on particles with indexes other that i. These two distinct elements can be written using respectively one-particle and two-particle integral operators defined as [5]

$$\begin{aligned} \left[\mathbf{Z}_0^{-1}(i)\mathbf{f}_i\right](\mathbf{r}) &\equiv \int \mathbf{G}(\mathbf{r}, \mathbf{r}')\mathbf{f}_i(\mathbf{r}')d\mathbf{r}', \qquad \mathbf{r} \in S_i, \\ \left[\mathbf{G}(ij)\mathbf{f}_j\right](\mathbf{r}) &\equiv \int \mathbf{G}(\mathbf{r}, \mathbf{r}')\mathbf{f}_j(\mathbf{r}')d\mathbf{r}', \qquad \mathbf{r} \in S_i, \quad i \neq j. \end{aligned} \tag{2.7}$$

The equation (2.6) can be rewritten in the form

$$\mathbf{u}_i = \sum_j (\boldsymbol{\mathcal{Z}}_0^{-1} + \boldsymbol{\mathcal{G}})_{ij} \mathbf{f}_j, \tag{2.8}$$

where

$$\boldsymbol{\mathcal{Z}}_{0ij} = \mathbf{Z}_0(i)\delta_{ij}, \qquad \boldsymbol{\mathcal{G}}_{ij} = \mathbf{G}(ij)(1-\delta_{ij}). \tag{2.9}$$

The solution to the many-body mobility problem may be formulated in terms of a scattering sequence. The force densities, determined by fluid flow around each particle, are treated as a source of subsequent fluid velocity fields. These fields are propagated through the system and reflect off other particles, thereby causing interaction. An analysis of the above equations leads to an explicit expression for the mobility matrix formulated in terms of one and two-body operators [5]:

$$\boldsymbol{\mu} = \boldsymbol{\mu}_0 + \boldsymbol{\mu}_0 \boldsymbol{\mathcal{Z}}_0 \frac{1}{1+\boldsymbol{\mathcal{G}}\hat{\boldsymbol{\mathcal{Z}}}_0} \boldsymbol{\mathcal{G}} \boldsymbol{\mathcal{Z}}_0 \boldsymbol{\mu}_0 = \boldsymbol{\mu}_0 + \boldsymbol{\mu}_0 \boldsymbol{\mathcal{Z}}_0 \sum_{k=0}^{\infty} (-\hat{\boldsymbol{\mathcal{Z}}}_0 \boldsymbol{\mathcal{G}})^k \boldsymbol{\mathcal{G}} \boldsymbol{\mathcal{Z}}_0 \boldsymbol{\mu}_0, \tag{2.10}$$

where $\boldsymbol{\mu}_0$ is the mobility of a single particle and $\hat{\boldsymbol{\mathcal{Z}}}_0 = \boldsymbol{\mathcal{Z}}_0 - \boldsymbol{\mathcal{Z}}_0 \boldsymbol{\mu}_0 \boldsymbol{\mathcal{Z}}_0$. The consecutive elements of the series expansion (2.10) are the terms of a scattering sequence.

To further analyse the problem, the multipole expansion is introduced [7]. All the irreducible multipoles are characterised by three numbers l, m and σ. Then all the operators introduced become infinite matrices and the integral equations transform to an infinite system of linear algebraic equations for the individual matrix elements. The multipole expansion further provides an expansion of the Green operator and, what follows, the two-particle mobility matrix in terms of the inverse of the inter-particle distance. In general the matrix elements of the two-particle Green operator $\langle lm\sigma|\mathbf{G}(ij)|l'm'\sigma'\rangle$ ($l = 1, 2, \ldots$, $m = -l, \ldots, l$, $\sigma = 0, 1, 2$) behave as $1/R_{ij}^{l+l'+\sigma+\sigma'-1}$ with the scalar inter-particle distance R_{ij}. Hence the interactions between low multipoles are of infinite range as they contain terms which decay as $1/R_{ij}^{\gamma}$, $\gamma = 1, 2, 3$.

2.3. BBGKY hierarchy

Our aim is to find the equations governing the evolution of the reduced distribution functions defined as [6]

$$n(1,2,\ldots,s;t) = \frac{N!}{(N-s)!} \int d\mathbf{R}_{(s+1)} \cdots d\mathbf{R}_N \rho(\mathbf{X};t). \tag{2.11}$$

To translate from a system description based on the Liouville equation to a description using the introduced reduced distribution function, a further analysis of the mobility matrix is needed. Due to its structure of a scattering series (2.10), the mobility function may also be formulated in terms of a cluster expansion. For all the self-interaction terms:

$$\boldsymbol{\mu}_{11}(X) = \boldsymbol{\mu}_{11}^{(1)}(1) + \sum_{i=2}^{N} \boldsymbol{\mu}_{11}^{(2)}(1i) + \frac{1}{2!} \sum_{\substack{i,j=2 \\ i \neq j}}^{N} \boldsymbol{\mu}_{11}^{(3)}(1ij) + \ldots \tag{2.12}$$

Here $\boldsymbol{\mu}_{11}^{(1)}(1)$ is the single particle mobility matrix given by the Stokes formula, $\boldsymbol{\mu}_{11}^{(2)}(1i)$ is the sum of all terms of the scattering expansion (2.10) which include particles 1 and i, etc. The interaction of different particles is described by

$$\boldsymbol{\mu}_{12}(X) = \boldsymbol{\mu}_{12}^{(2)}(12) + \sum_{i=3}^{N} \boldsymbol{\mu}_{12}^{(3)}(12i) + \ldots, \tag{2.13}$$

where now $\boldsymbol{\mu}_{12}^{(2)}(12)$ stands for the sum of all terms of the scattering sequence (2.10) of $\boldsymbol{\mu}_{12}(X)$ which contain only particles 1 and 2, $\boldsymbol{\mu}_{12}^{(3)}(12i)$ includes all terms which contain three particles 1, 2 and i, etc.

We also introduce the extended cluster expansion according to the formula ($s > 1$):

$$\begin{aligned}
\boldsymbol{\mu}_{11}(1\ldots s|s+1\ldots s+l) &= \boldsymbol{\mu}_{11}^{(l+1)}(1s+1\ldots s+l) \\
&\quad + \sum_{i=2}^{s} \boldsymbol{\mu}_{11}^{(l+2)}(1is+1\ldots s+l) + \ldots, \\
\boldsymbol{\mu}_{12}(1\ldots s|s+1\ldots s+l) &= \boldsymbol{\mu}_{12}^{(l+2)}(12s+1\ldots s+l) \\
&\quad + \sum_{i=3}^{s} \boldsymbol{\mu}_{12}^{(l+3)}(12is+1\ldots s+l) + \ldots, \\
\boldsymbol{\mu}_{1s+1}(1\ldots s|s+1\ldots s+l) &= \boldsymbol{\mu}_{1s+1}^{(l+1)}(1s+1\ldots s+l) \\
&\quad + \sum_{i=2}^{s} \boldsymbol{\mu}_{1s+1}^{(l+2)}(1is+1\ldots s+l) + \ldots .
\end{aligned} \tag{2.14}$$

For the case of $s = 2$, $l = 1$:

$$\boldsymbol{\mu}_{11}(12|3) = \boldsymbol{\mu}_{11}^{(2)}(13) + \boldsymbol{\mu}_{11}^{(3)}(123), \tag{2.15}$$

$$\boldsymbol{\mu}_{12}(12|3) = \boldsymbol{\mu}_{12}^{(3)}(123), \tag{2.16}$$

$$\boldsymbol{\mu}_{13}(12|3) = \boldsymbol{\mu}_{13}^{(2)}(13) + \boldsymbol{\mu}_{13}^{(3)}(123). \tag{2.17}$$

The following notation will be used to simplify the derived equations:

- $\mathbf{s}$ denotes the set of particles $\{1,\ldots,s\}$,
- $s+\mathbf{l}$ denotes the l-particle set $\{s+1,\ldots,s+l\}$.

The integration with respect to the particle indexes is to be understood as

$$\int d(s+\mathbf{l}) \equiv \frac{1}{l!}\int d(s+1)\cdots d(s+l). \tag{2.18}$$

Consider the integration of the Liouville equation with respect to all but the first s particles. The boundary terms emerging due to integration of the divergence operator vanish. The remaining expressions may be simplified upon introducing the cluster expansion of the mobility matrix. The result is a sequence of multiple cluster integrals containing the reduced distribution function of order s and higher, and the hydrodynamic functions. The system of equations obtained by taking different values of $s=1,2,\ldots$ forms a hierarchy:

$$\left(\frac{\partial}{\partial t}+\mathfrak{L}(\mathbf{s})\right)n(\mathbf{s};t) = -\sum_{l=1}^{N-s}\int d(s+\mathbf{l})\mathfrak{L}(\mathbf{s}|s+\mathbf{l})n(\mathbf{s}+\mathbf{l};t), \tag{2.19}$$

where

$$\mathfrak{L}(\mathbf{s})n(\mathbf{s};t) = \sum_{i=1}^{s}\nabla_i\cdot\left\{\sum_{j=1}^{s}\boldsymbol{\mu}_{ij}(\mathbf{s})\mathbf{F}_j\; n(\mathbf{s};t)\right\} \tag{2.20}$$

is the s-particle Liouville operator and $\boldsymbol{\mu}(\mathbf{s})$ is the full s-particle mobility operator. Whereas

$$\mathfrak{L}(\mathbf{s}|s+\mathbf{l})n(\mathbf{s}+\mathbf{l};t) = \sum_{i=1}^{s}\nabla_i\cdot\left\{\sum_{j=1}^{s+l}\boldsymbol{\mu}_{ij}(\mathbf{s}|s+\mathbf{l})\mathbf{F}_j\; n(\mathbf{s}+\mathbf{l};t)\right\} \tag{2.21}$$

is the $(s+l)$-particle, extended Liouville operator for the evolution of a s-particle reduced distribution function. It is defined with the help of extended hydrodynamic functions (2.14) introduced above.

The system of equations (2.19) is the analogue of the BBGKY (Bogolubov–Born–Green–Kirkwood-Yvon) hierarchy [6] introduced in the kinetic theory of gases. In the case of a system of infinitely many particles this set is infinite, and the equations contain an infinite number of terms, contrary to what is usually the case when considering two-body potential interactions.

The reduced distribution functions (2.11) are usually assumed to have the following property. Whenever particles may be grouped inside two (or more) spheres $C_1, C_2\ldots$ such that the radii of these are much smaller than the distance between them, then the partial distribution function describing the particle densities factorizes. This leads to a cluster expansion in terms of the correlation functions

$$
\begin{aligned}
n(1) &= h(1),\\
n(12) &= h(1)h(2) + h(12),\\
n(123) &= h(1)h(2)h(3) + h(12)h(3) + h(13)h(2) + h(23)h(1) + h(123),\\
\vdots\ &\ \ \vdots\\
n(\mathbf{s}) &= \sum_{\bigsqcup_i \mathbf{m}_i = \mathbf{s}} \prod_i h(\mathbf{m}_i),
\end{aligned}
\tag{2.22}
$$

where $h(1\dots s)$ is a s-particle correlation function, which vanishes whenever any particle is pulled away from the rest.

This structure of the reduced distribution functions allows a further study of the hierarchy equations. These may be analysed qualitatively by noting two important properties. First, if a given term of the cluster expansion of $\mathfrak{L}(\mathbf{s}|s+\mathbf{l})n(\mathbf{s}+\mathbf{l};t)$ ($s = 1, \dots$ and $l = 0, \dots$) may be factorised into two distinct functions depending on excluding particle subsets, then this term is said to be non-connected. Contrary, if such a factorisation is impossible, the expression is called connected. Notice that the hydrodynamic structure of a given expression is by definition connected. The factorisation may then be a consequence only of the correlation bonds introduced on the particles. Therefore if a term is non-connected, it contains a disjoint part existing solely as a correlation function block. For example,

$$\boldsymbol{\mu}(1)\mathbf{Z}_0(1)\mathbf{G}(14)\mathbf{Z}_0(4)\boldsymbol{\mu}_0(4)\mathbf{F}_4\ h(14)h(23) \tag{2.23}$$

is non-connected, as particles 2 and 3, although linked by a correlation bond, are not linked to any of the other particles. In contrast, the structure

$$\boldsymbol{\mu}(1)\mathbf{Z}_0(1)\mathbf{G}(14)\mathbf{Z}_0(4)\boldsymbol{\mu}_0(4)\mathbf{F}_4\ h(12)h(34) \tag{2.24}$$

where only the correlation structure has been changed, is connected. Both expressions are parts of $\boldsymbol{\mu}_{14}(123|4)\mathbf{F}_4\ n(1234)$. The connected part of an expression will be denoted by adding a superscript c.

This property of the hierarchy equation may be used to formulate the equations governing the evolution of the correlation function. Indeed, instead of considering the set of reduced distribution functions, it is often more desired to consider the equivalent set of correlation functions. Since these are linked to the reduced distribution functions by the cluster expansion of the latter (2.22), a relevant hierarchy of equations governing the time evolution of the correlation functions may be derived. The right-hand side of these contain only connected terms. In other words, an equation governing the time evolution of an s-particle correlation function may be deduced from the analogous equation for the reduced distribution function by neglecting all non-connected terms.

$$\frac{\partial h(\mathbf{s};t)}{\partial t} = -\{\mathfrak{L}(\mathbf{s})n(\mathbf{s};t)\}^c - \sum_{l=1}^{\infty} \int d(s+\mathbf{l})\{\mathfrak{L}(\mathbf{s}|s+\mathbf{l})n(\mathbf{s}+\mathbf{l};t)\}^c. \tag{2.25}$$

For example, for the two-particle correlation function, the first terms of the equation have the following structure:

$$\begin{aligned}\frac{\partial h(12;t)}{\partial t} = &-\sum_{i=1,2} \nabla_i \cdot \{\boldsymbol{\mu}_{ii}^{(1)}(i)\mathbf{F}_i h(12;t)\} \\ &-\sum_{i,j=1,2} \nabla_i \cdot \{\boldsymbol{\mu}_{ij}^{(2)}(12)\mathbf{F}_j(h(12;t)+h(1;t)h(2;t))\} + \ldots .\end{aligned} \tag{2.26}$$

Notice that both in the equation for the reduced distribution and correlation functions, there exist terms $\mathfrak{L}(\mathbf{s}|s+\mathbf{l})n(\mathbf{s}+\mathbf{l};t)$ ($s = 1,\ldots$ and $l = 0,\ldots$) which factorise into two distinct expressions upon removing a single Green operator $\mathbf{G}$. Such terms are called reducible [5]. This concept takes on importance upon invoking the dependence of the Green operator on the inter-particle distance. Remember that these operators contain parts which decay very slowly with distance. Reducible terms therefore introduce long-range expressions into the hierarchy, and further could cause problems in the limit of an infinite system leading to non-existence of certain integrals. In particular, the appearance of such terms in the equations leads to the conclusion that the correlation functions must also decay slowly with inter-particle distance, leading to non-integrability of these function.

The hierarchy Equations (2.19) and (2.25) are not immediately useful, because they express one unknown correlation or reduced distribution function by all the higher-order function. At some stage, an approximation based on a physical analysis must be made that closes the hierarchy. One of the methods is an expansion of the correlation functions in terms of density. As indicated, even in the low-concentration limit, the reducibility of some terms of the r.h.s. of the hierarchy equations poses a problem. We will show in the next section that using a mechanism of hydrodynamic screening, a solution for the pair distribution function might nevertheless be obtained.

3. Stationary state pair distribution function for a sedimenting suspension

Consider the stationary sedimentation of a suspension of identical particles. The long range of the correlation functions, being a consequence of the structure of the hierarchy, contradicts the usual behaviour of these functions. We therefore explore the possibility to reconcile the existence of the long-range terms in the hierarchy equations with the finite range of the correlations. It is tempting here to invoke the concept of hydrodynamic screening. Consider such a particle distribution, that all long-range contributions are cut off and the correlations stay integrable. It can be shown that indeed such a particle configuration can be constructed [8]. This can be accomplished in stationary state by assuming the dilute limit when the reduced distribution function can in leading order be approximated by the dominating part which, for an s-particle function, is proportional to n^s. The hierarchy equations can then be truncated on a given order of density and in particular provide closed equations for the reduced distribution function or the correlation functions. As it has been already pointed out in the introduction, the two-particle equation cannot yield a condition for the two-particle correlation function due to the symmetry of the two-body hydrodynamic interactions. It is necessary to consider three-particle interaction to achieve non-trivial results in a monodisperse suspension.

Analysing the stationary equation for the three-particle correlation or reduced distribution function truncated to terms proportional to the density cubed

$$\mathfrak{L}(123)n(123) = 0, \tag{3.1}$$

one can extract all long-range contributions. These appear only when the configuration of particles is such, that a single particle (eg. 1) is situated far away from a group of two close particles (eg. 2 and 3). Then $n(123) \to n(23)n(1)$ and non-integrable terms appear only in the expression

$$\boldsymbol{\nabla}_2\cdot\big\{[\boldsymbol{\mu}_{21}(1\cdot 2)+\boldsymbol{\mu}_{21}(1\cdot[23])]n(23)\big\}+\boldsymbol{\nabla}_3\cdot\big\{[(\boldsymbol{\mu}_{31}(1\cdot 3)+\boldsymbol{\mu}_{31}(1\cdot[23])]n(23)\big\}, \tag{3.2}$$

where

$$\begin{aligned}
\boldsymbol{\mu}_{21}(1\cdot 2) &= \boldsymbol{\mu}_0(2)\mathbf{Z}_0(2)\mathbf{G}(21)\mathbf{Z}_0(1)\boldsymbol{\mu}_0(1), && (3.3)\\
\boldsymbol{\mu}_{31}(1\cdot 2) &= \boldsymbol{\mu}_0(3)\mathbf{Z}_0(3)\mathbf{G}(31)\mathbf{Z}_0(1)\boldsymbol{\mu}_0(1), && (3.4)\\
\boldsymbol{\mu}_{21}(1\cdot[23]) &= [\mathbf{C}_{22}(23)\mathbf{G}(21)+\mathbf{C}_{23}(23)\mathbf{G}(31)]\mathbf{Z}_0(1)\boldsymbol{\mu}_0(1), && (3.5)\\
\boldsymbol{\mu}_{31}(1\cdot[23]) &= [\mathbf{C}_{33}(23)\mathbf{G}(31)+\mathbf{C}_{32}(23)\mathbf{G}(21)]\mathbf{Z}_0(1)\boldsymbol{\mu}_0(1) && (3.6)
\end{aligned}$$

and

$$\mathbf{C}_{ij} = \left[\boldsymbol{\mu}_0\boldsymbol{\mathcal{Z}}_0\frac{1}{1+\boldsymbol{\mathcal{G}}\hat{\boldsymbol{\mathcal{Z}}}_0}\boldsymbol{\mathcal{G}}\hat{\boldsymbol{\mathcal{Z}}}_0\right]_{ij}(23) \tag{3.7}$$

which stands for all those scatterings in the formula (2.10) which contain only particles 2 and 3 and begin at particle $i = 2, 3$, and end respectively at particle $j = 2, 3$.

Upon analysing the distance dependence of the multipole elements of the long-range Green operators $\mathbf{G}(21)$ and $\mathbf{G}(31)$, all the non-integrable contributions of (3.2) can be singled out. These originate in the rotational-translational (rt, $l = 1$, $\sigma = 1$; $l' = 1, \sigma' = 0$) and dipole-translational (dt, $l = 2$, $\sigma = 0$; $l' = 1$, $\sigma' = 0$) multipoles of $\mathbf{G}$. Now suppose that the two-particle distribution function $n(23)$ is such that these long-range structures disappear. This assumption leads to two equations for the pair distribution function. One, associated with the (rt) multipole describes the dependence of the distribution function upon the direction of the vector $\mathbf{R} = \mathbf{R}_3 - \mathbf{R}_2$

$$\mathbf{R} \times \nabla n(\mathbf{R}) = 0, \tag{3.8}$$

giving the immediate result $n(\mathbf{R}) = n(R)$. The second equation, related to the (dt) multipole yields an equation for the now isotropic function $n(R) = n^2 g(R)$

$$\left\{(1 - A)\frac{1}{g}\frac{dg}{dR} - \frac{3(A - B)}{R} - \frac{dA}{dR}\right\} = 0. \tag{3.9}$$

A and B are functions of the scalar distance R given by the relations $A = 2(\alpha^{td}_{11} + \alpha^{td}_{12})/R$ and $B = 2(\beta^{td}_{11} + \beta^{td}_{12})/R$, where α^{td}_{ij} and β^{td}_{ij} are functions appearing in the explicit form of the generalized two-particle mobility matrix connecting the translational velocity of the particles to the stresslet [9].

$$C^{td}_{i\alpha;j\mu\nu}(23) = \mu^{td}_{i\alpha;j\mu\nu}(23) = \alpha^{td}_{ij}\hat{R}_\alpha\overline{\hat{R}_{\boldsymbol{\mu}}\hat{R}_\nu} + \beta^{td}_{ij}\overline{(\delta_{\alpha\mu} - \hat{R}_\alpha\hat{R}_\mu)\hat{R}_\nu}^{(\mu\nu)}, \tag{3.10}$$

where the overline denotes the irreducible part of the tensor.

Since $\lim_{R\to\infty} g(R) = 1$

$$g(R) = \frac{1}{1 - A}\exp\left[\int_R^\infty \frac{3(B - A)}{R(1 - A)}dR\right]. \tag{3.11}$$

The knowledge of the pair distribution function makes a calculation of the steady-state sedimentation coefficient possible. Indeed, if

$$\frac{U}{U_0} = 1 + \phi\lambda + \mathcal{O}(\phi^2), \tag{3.12}$$

where ϕ is the volume fraction, U is the mean sedimentation velocity and U_0 is the Stokes velocity, then the linear coefficient λ may be expressed in terms of the two-body mobility matrix and the pair distribution function. A numerical calculation leads to the value $\lambda = -3.87$. A fit to the experimental data ([10, 11]) leads to the sedimentation coefficient equal to -3.9, in excellent agreement with the calculated value [8].

4. Concluding remarks

A hierarchy of equations for the correlation and reduced distribution functions has been found and its structure analysed. It has been shown that the derived equations may be used to investigate the phenomenon of sedimentation at low-concentration, of a non-Brownian suspension. In particular, in spite of the long range of the hydrodynamic interactions, a short range two-particle correlation function was derived in a self-consistent manner.

References

1. G. K. Batchelor, *Sedimentation in a dilute dispersion of spheres*, J. Fluid Mech., **52**, 245, 1972.
2. G. K. Batchelor, *Sedimentation in a dilute polidisperse system of interacting spheres. Part 1. General theory*, J. Fluid Mech., **119**, 379, 1982.
3. G. K. Batchelor, C. S. Wen, *Sedimentation in a dilute disperse system of interacting spheres. Part 2. Numerical results*, J. Fluid Mech., **124**, 495, 1982.
4. P. Mazur, D. Bedeaux, *Generalization of Faxen's theorem to non-steady motion of a sphere through an incompressible fluid in arbitrary flow*, Physica, **76**, 2, 235–246, 1974.
5. B. Cichocki, P. Szymczak, *Memory effects in collective dynamics of Brownian suspensions*, J. Chem. Phys., **121**, 3329, 2004.
6. R. Balescu, *Equilibrium and nonequilibrium statistical mechanics*, Wiley, New York 1975.
7. B. Cichocki, R. B. Jones, R. Kutteh, E. Wajnryb, *Friction and mobility for colloidal spheres in a Stokes flow near a boundary: The multipole method and applications*, J. Chem. Phys., **112**, 2548, 2000.
8. B. Cichocki, K. Sadlej, *Steady-state particle distribution of a dilute sedimenting suspension*, Europhys. Lett., **72**, 6, 1–7, 2005.
9. B. Cichocki, B. U. Felderhof, R. Schitz, *Hydrodynamic interactions between two spherical particles*, Phys. Chem. Hydrodynamics, **10**, 383, 1988.
10. J. M. Ham, G. M. Homsy, *Hindered settling and hydrodynamic dispersion in quiescent sedimenting suspensions*, Int. J. Multiphase Flow, **14**, 533, 1988.
11. T.J. Hanratty, A. Bandukwala, *Fluidization and sedimentation of spherical particles*, AIChE J., **3**, 293–6, 1957.

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