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Stokesian Dynamics—The BBGKY Hierarchy for Correlation Functions

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Abstract A statistical description of a system of hard particles immersed in a viscous fluid is introduced and analyzed. Neglecting inertial effects and Brownian motion—i.e. assuming Stokesian dynamics for the particles, hierarchies of equations governing the time evolution of the reduced distribution functions, and for the correlation functions respectively, are derived. It is shown that all non-integrable expressions contained in these equations, stemming from long-range hydrodynamic interactions arising between particles, can be resummed to form physically meaningful fields. Applications of this theoretical scheme are sketched.

Keywords Stokesian dynamics · BBGKY hierarchy · Reduced distribution functions · Correlation functions · Suspensions · Hydrodynamic interactions

1 Introduction

In recent years, due to a growing interest in micro-fluidics, bio-physics as well as technological applications, the properties of suspensions, i.e. systems consisting of particles immersed in a viscous fluid, have attracted significant experimental [5, 16, 25, 26] and numerical research [10, 21, 22]. The theory has nevertheless stayed somewhat behind, and many important problems have been left unresolved. The aim of this work is to elucidate some of them.

Motion of a suspension of hard particles in an external force field and ambient fluid velocity field will be considered in this work. Particles and their velocities will be assumed to be small enough to validate the zero Reynold's number approximation to the Navier-Stokes equation describing fluid motion. On the other hand they will be expected to be large enough

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to neglect the influence of Brownian motion. These assumptions (physical particle size of the order of 10–100 μm) will justify the description of the system in terms of Stokesian particle dynamics. This implies that the relation between forces acting on particles or the ambient flow, and the velocities of the particles is linear. It is given by the so called mobility matrix when considering forces acting on particles, and the convection operator when the influence of the ambient flow is of interest. Both these operators depend on the configuration of all particles and describe the influence particles have on each other through fluid velocity fields they create when moving. These interactions are called hydrodynamic. Inertial effects are left out. Despite its apparent simplicity, an explicit theoretical analysis of the considered systems is surprisingly complicated due to properties of the hydrodynamic interactions. These are non-integrable, decaying as $1/r$ with the interparticle distance r, many-particle, and highly non-linear leading to very peculiar problems. The representative example is the long-standing dispute over system-size dependence of velocity fluctuations [6, 20, 26]. As noticed by Caflish and Luke [6], in a suspension, where particle positions are random, the velocity fluctuations scale with the size of the system. These are therefore formally infinite in the thermodynamic limit. A possible theoretical solution to this problem was found by Koch and Shaqfeh [20]. It was shown that once the pair-distribution function satisfies a strict criterion, the velocity fluctuations are indeed bounded. The mechanism by which this is accomplished may be called hydrodynamic screening, in analogy to the Debye-Hückel screening in electrolytes. Unfortunately the precise form of the pair-distribution function, describing the microstructure of a suspension in stationary state was not found.

A second, somewhat forgotten issue is the derivation of the pair correlation function describing the stationary microstructure of a polydisperse (i.e. consisting of particles of different size and density) and in particular a monodisperse (i.e. consisting of identical particles), sedimenting suspension. A solution valid for low concentrations of polydisperse particles was obtained by Batchelor and Wen [2, 3] in 1982. By considering only two-particle dynamics, they where able to derive and numerically solve an equation for the pair distribution function. Their analysis presents nevertheless several problem. The basic one is the lack of a solution for the case of a monodisperse suspension—in fact, for identical particles the equation considered by Batchelor vanishes identically. Even more striking is the non-uniqueness of the form of the distribution function and the value of the sedimentation coefficient in the limit of identical particles. As a polydisperse suspension is described by two parameters—the ratio of particle densities and particle radii—an arbitrary number of ways of calculating this limit exist. A proper solution of the problem would render the same result for all these ways, which is not true in the case of Batchelors derivation.

Our aim is to construct a statistical description of a homogeneous suspension, which will be free of the limitations present in Batchelors construction and will simultaneously be coherent with Koch and Shaqfeh's criterion for finite velocity fluctuations. Evolving away from an initial configuration, a suspension reaches a non-equilibrium stationary state. The problem of interest is then to quantify correlations arising between particle positions. We will here adopt a solution method based on the analysis of the BBGKY (Bogolubov-Born-Green-Kirkwood-Yvon) [1] hierarchy for those functions. Complications encountered are due to the extremely long-range of hydrodynamic interactions leading to non-locality of the equations. This is in fact a situation, which is very similar to the one confronted in systems of interacting charges. The known, in that case, solution along the lines of the Debye-Hückel theory might also be derived through an analysis of the equilibrium BBGKY hierarchy for that system. This has been described by A.E. Glauberman and I.P. Yukhnovskii [13–15] and summarized in the monograph by H. Falkenhagen [11]. A similar line of reasoning can be used to find the stationary, non-equilibrium distribution function for a system of particles described by Stokesian dynamics. In this work we will focus on the derivation of the BBGKY hierarchies for reduced distribution functions and correlation functions respectively. An analysis of these will lead to a resummation procedure, which will explicitly distinguish all non-integrable expressions and allow for a physical interpretation of correlation fields (between the suspension velocity fluctuations and particle concentration fluctuations) which then emerge. The feasibility of such a description is a crucial step toward the solution of the problem of finding the stationary state microstructure of a hydrodynamically interacting suspension of particles.

The structure of this paper is the following. In Sect. 2 a general statistical formalism is introduced. Next, in Sect. 3 hydrodynamic interactions arising between particles are described. The hierarchies of equations describing the time evolution of the reduced distribution functions and the correlation functions are derived in Sects. 4 and 5, respectively. Then in Sect. 6 all non-integrable expressions found in these equations are explicitly isolated and resummed to form physical fields. Concluding remarks have been placed in Sect. 7.

Supplementary results have been put in the Appendixes. Appendix A contains information and formulas necessary in order to generalize the results obtained in the main sections—valid for identical particles—to a polydisperse suspension. Appendix B presents briefly a diagrammatic technique, useful in the analysis of the considered BBGKY hierarchies.

2 Statistical Description of a Particle System with Stokesian Dynamics

In this paper we consider a system of N spherical hard particles of equal radii a, immersed in a viscous fluid of viscosity η and volume V. The Reynold's number is assumed to be small. Particles are nevertheless large enough to neglect the influence of Brownian motion and the timescale, at which the system is observed, justifies the description of the fluid by means of the stationary Stokes equations. In this regime the state of the whole system, at a given moment of time, is fully described once the configuration $\boldsymbol{X} = \{\boldsymbol{R}_1, \ldots, \boldsymbol{R}_N\}$ of all N particles is known. Such a system will be referred to as a monodisperse, non-Brownian suspension and the dynamics—Stokesian.

The evolution of the system is described once given the velocities of the particles $\boldsymbol{U}_i$, $i = 1, 2, \ldots$, as in Stokesian dynamics, inertial effects are absent. These quantities depend upon two elements

$$\boldsymbol{U}_i = \boldsymbol{U}_i^{\text{force}} + \boldsymbol{U}_i^{\text{flow}}. \tag{1}$$

One is the contribution from external forces acting on particles. It is of the form

$$\boldsymbol{U}_i^{\text{force}} = \sum_j \boldsymbol{\mu}_{ij}^{tt}(\boldsymbol{X})\boldsymbol{F}_j, \quad i = 1, 2, \ldots, \tag{2}$$

where $\boldsymbol{F}_j$ is the total external force acting on particle j, and $\boldsymbol{\mu}^{tt}$ is the translational part of the mobility matrix [8]. This matrix is defined in general as

$$\boldsymbol{\mu} = \begin{pmatrix} \boldsymbol{\mu}^{tt} & \boldsymbol{\mu}^{tr} \\ \boldsymbol{\mu}^{rt} & \boldsymbol{\mu}^{rr} \end{pmatrix}, \tag{3}$$

and binds the translational (t) and rotational (r) velocities of the particles with the forces and torques, according to the linear relation

$$\begin{pmatrix} \boldsymbol{U}_i \\ \boldsymbol{\Omega}_i \end{pmatrix} = \sum_{j=1}^{N} \begin{pmatrix} \boldsymbol{\mu}_{ij}^{tt} & \boldsymbol{\mu}_{ij}^{tr} \\ \boldsymbol{\mu}_{ij}^{rt} & \boldsymbol{\mu}_{ij}^{rr} \end{pmatrix} \begin{pmatrix} \boldsymbol{F}_j \\ \boldsymbol{\mathcal{T}}_j \end{pmatrix}. \tag{4}$$

Here $\mathcal{T}_j$ is the torque acting of particle j and $\boldsymbol{\Omega}_i$ the rotational velocity of particle i. In this work torques acting on the particles will be assumed to be zero and only the translational motion is of our interest. This restriction can nevertheless easily be relaxed and does not limit the generality of the considerations which follow.

In a homogeneous system of identical particles, the forces $\boldsymbol{F}_j$ are all equal. But in general, as the force field may depend on the position considered, so does the total external forces acting on particles. Then $\boldsymbol{F}_j \equiv \boldsymbol{F}(\boldsymbol{R}_j)$. The most universal case of a polydisperse suspension of different particles, when the external forces acting on particles depend on their radii and densities will be discussed in Appendix A, as apart from some notational problems, it does not introduce new qualitative challenges into this theory.

The particles may also be immersed in an ambient flow $\boldsymbol{v}_0$ which induces their motion through convection. This mechanism is described by the second element of (1), given by

$$\boldsymbol{U}_i^{\text{flow}} = \int d\boldsymbol{r}' \boldsymbol{\mathcal{C}}^t(\boldsymbol{R}_i, \boldsymbol{r}'|\boldsymbol{X})\boldsymbol{v}_0(\boldsymbol{r}') \equiv \boldsymbol{\mathcal{C}}_i^t(\boldsymbol{X})\boldsymbol{v}_0, \tag{5}$$

where $\boldsymbol{\mathcal{C}}_i^t(\boldsymbol{X})$ [12] is the translational part of the convection operator which provides the value of the translational velocity of the chosen i-th particle in the suspension, due to motion in an ambient flow $\boldsymbol{v}_0$. The derivation and properties of the introduced operators will be discussed in the next section.

If the suspension is enclosed in a rigid container, as is the case in a typical experimental setup, the ambient flow $\boldsymbol{v}_0$ is equal to zero. Due to the structure, and for the sake of generality of applications of the equations which follow, it is nevertheless preferable to keep the term with $\boldsymbol{v}_0$ in the evolution equations.

Summarizing, the evolution of the system state is in the situation considered, described by a set of coupled, first-order differential equations of the form

$$\frac{d\boldsymbol{R}_i}{dt} = \boldsymbol{U}_i \equiv \sum_{j=1}^{N} \boldsymbol{\mu}_{ij}^{tt}(\boldsymbol{X})\boldsymbol{F}_j + \boldsymbol{\mathcal{C}}_i^t(\boldsymbol{X})\boldsymbol{v}_0, \quad i = 1, 2, \ldots. \tag{6}$$

Such particle dynamics will be called Stokesian. These equations (6) provide a detailed description of the particle trajectories as they evolve in time. But due to the chaotic nature of this movement [18], a proper characterization of the future system state is essentially impossible. Therefore, a statistical description of a suspension is necessary. It is introduced through the particle distribution function $\rho(\boldsymbol{X}; t)$, defined as $\rho(\boldsymbol{X}; t)d\boldsymbol{X}$ being the probability of finding the particles in a configuration $d\boldsymbol{X}$ around $\boldsymbol{X}$ at time t. An equation governing the time evolution of this function is a particle number conservation equation in the configuration space of N particles. It follows from (6) and is given by

$$\frac{\partial \rho(\boldsymbol{X}; t)}{\partial t} + \sum_i \nabla_i \cdot \{\boldsymbol{U}_i \rho(\boldsymbol{X}; t)\} = 0. \tag{7}$$

This is the Liouville equation for the considered system.

The particle probability function $\rho(\boldsymbol{X}; t)$ contains all available information about the system. Equation (7) defining its time evolution is in fact equivalent to the set of (6) describing in detail the precise trajectories of all the particles. As usually done in non-equilibrium statistical mechanics [1], a description using the reduced distribution functions $n(\boldsymbol{r}_1, \ldots, \boldsymbol{r}_s; t), s = 1, 2, \ldots$ is extracted from the elaborate many-particle dynamics. In a homogeneous system $n(\boldsymbol{r}_1) = n$ is the concentration of particles. In general these functions should be interpreted as $n(\boldsymbol{r}_1, \ldots, \boldsymbol{r}_s; t) d\boldsymbol{r}_1 \cdots d\boldsymbol{r}_s$ being the average number of ensembles of s particles in which at time t, one of the particles is in position $d\boldsymbol{r}_1$ around $\boldsymbol{r}_1$, another in $d\boldsymbol{r}_2$ around $\boldsymbol{r}_2$ and so on. It should be stressed, that it usually suffices to know the two-particle distribution function in order to calculate the basic parameters of the system, such as its static or dynamic response coefficients.

In the sections which follow our aim will be to find and discuss in detail the equations which govern the time evolution of the reduced distribution functions. These will be derived from the Liouville equation (7). Their structure is dictated by the properties of the fluid-mediated interactions expressed by the hydrodynamic operators which describe the influence of external forces and ambient flows on the particle velocities. A brief summary of the derivation of these operators is given in the next section.

3 Hydrodynamic Operators

In this section we will derive and analyze the properties of the hydrodynamic operators $\boldsymbol{\mu}$ (3) and $\boldsymbol{C}$ (5) which describe the dynamics of the system.

It will be assumed, that the fluid velocity field $\boldsymbol{v}(\boldsymbol{r})$ is governed by the Stokes equations [19]

$$\begin{aligned} \eta \nabla^2 \boldsymbol{v} - \nabla p &= 0, \\ \nabla \cdot \boldsymbol{v} &= 0, \end{aligned} \tag{8}$$

where $p(\boldsymbol{r})$ is the pressure field.

For the system considered these must be solved together with appropriate boundary conditions which, in this case are

$$\boldsymbol{v}(\boldsymbol{r}) = \boldsymbol{u}_i(\boldsymbol{r}) \equiv \boldsymbol{U}_i + \boldsymbol{\Omega}_i \times (\boldsymbol{r} - \boldsymbol{R}_i) \quad \boldsymbol{r} \in S_i,\ i = 1, 2 \ldots, \tag{9}$$

for $\boldsymbol{r}$ pointing to a point on the surface S_i of the i-th particle—the fluid follows the motion of the particle at its surface. We will refer to this requirement as the stick-boundary condition. Further, conditions are specified at the boundaries of the whole system.

The Stokes equations as given by (8) describe fluid motion outside of the particles. The key problem is therefore, how to extract from those equations, information about particle movement, and in particular, the relation between the ambient flow, external forces acting on the particles and their velocities. A possible solution postulates an extension of the validity of the Stokes equations to the interior of the spheres, by introducing induced force densities $\boldsymbol{f}_i(\boldsymbol{r})$ located on their surfaces [23]. These force densities are chosen such, that the solution of

$$\eta \nabla^2 \boldsymbol{v} - \nabla p = -\sum_{i=1}^{N} \boldsymbol{f}_i, \tag{10}$$

$$\nabla \cdot \boldsymbol{v} = 0, \tag{11}$$

inside the particles describes a fictitious fluid motion which follows exactly their rigid body motion $\boldsymbol{u}_i(\boldsymbol{r})$ given by (9), simultaneously coinciding with the solution of the homogeneous Stokes equations (8) outside of the particles. Such a choice of force densities has a further advantage. The solution $\boldsymbol{v}(\boldsymbol{r}|\boldsymbol{X})$ of (10), when averaged over particle configurations $\boldsymbol{X}$, has the interpretation of the macroscopic suspension velocity.

Due to the linear character of the Stokes equation, the relation between the fluid velocity field and the force densities introduced is linear. It is thus written, using an integral operator with the kernel $\boldsymbol{T}$, in the following form:

$$\boldsymbol{v}(\boldsymbol{r}|\boldsymbol{X}) = \boldsymbol{v}_0(\boldsymbol{r}) + \int \boldsymbol{T}(\boldsymbol{r}, \boldsymbol{r}')\boldsymbol{f}(\boldsymbol{r}'|\boldsymbol{X})d\boldsymbol{r}, \tag{12}$$

where $\boldsymbol{f}$ stands for the complete force density which is the sum of all $\boldsymbol{f}_i$ for $i = 1, \dots, N$ (i.e. the term standing on the right side of (10)). In the operator notation, this equation has the simple form

$$\boldsymbol{v} = \boldsymbol{v}_0 + \boldsymbol{T}\boldsymbol{f}. \tag{13}$$

If considering an unbounded system, as will be done in this work, the operator kernel $\boldsymbol{T}(\boldsymbol{r}, \boldsymbol{r}') = \boldsymbol{T}_0(\boldsymbol{r} - \boldsymbol{r}')$ is the Oseen tensor

$$\boldsymbol{T}_0(\boldsymbol{r}) = \frac{1}{8\pi\eta r}(\boldsymbol{I} + \hat{\boldsymbol{r}}\hat{\boldsymbol{r}}), \tag{14}$$

where $\hat{\boldsymbol{r}} = \boldsymbol{r}/|\boldsymbol{r}|$.

The introduction of induced force densities and the formulation of the formal solution (12) can be used to find the relation between external forces acting on particles, their velocities and the ambient flow. The induced force densities are set by the boundary conditions (9) and the translational and rotational velocities of the particles. Taking into account the integral representation (12), these may be written as

$$[\boldsymbol{u}_i(\boldsymbol{r}) - \boldsymbol{v}_0(\boldsymbol{r})]_{\boldsymbol{r}\in S_i} = \sum_j \int d\boldsymbol{r}'\boldsymbol{T}_0(\boldsymbol{r} - \boldsymbol{r}')\boldsymbol{f}_j(\boldsymbol{r}'). \tag{15}$$

This now is an integral equation, where the known quantities are the velocities $\boldsymbol{U}_i$ and $\boldsymbol{\Omega}_i$, or equivalently $\boldsymbol{u}_i$ (see (9)), and the ambient flow $\boldsymbol{v}_0$, whereas the force densities $\boldsymbol{f}_i$ are to be found.

One of the solution methods of the integral equation (15), i.e. the calculation of induced force densities $\boldsymbol{f}_i$, is based on a multipole expansion. This scheme is described in detail for example in [9]. Here we present just the basic ideas and necessary formulas. To start with, all quantities present in (15) are projected onto a complete set of functions $\boldsymbol{v}^{\pm}_{lm\sigma}(\boldsymbol{r} - \boldsymbol{R}_i)$, which are solutions of the homogeneous Stokes equations (8). Indexes l, m, σ take values $l = 1, \dots, m = -l, \dots, l$ and $\sigma = 0, 1, 2$. The solutions $\boldsymbol{v}^{+}_{lm\sigma}(\boldsymbol{r} - \boldsymbol{R}_i)$ are regular at $\boldsymbol{r} = \boldsymbol{R}_i$ and grow to infinity, while $\boldsymbol{v}^{-}_{lm\sigma}(\boldsymbol{r} - \boldsymbol{R}_i)$ are singular at $\boldsymbol{r} = \boldsymbol{R}_i$ and vanish at infinity. The fluid velocity around each particle can thus be written as an infinite series

$$\boldsymbol{u}_i(\boldsymbol{r}) - \boldsymbol{v}_0(\boldsymbol{r}) = \sum_{lm\sigma} c(ilm\sigma)\boldsymbol{v}^{+}_{lm\sigma}(\boldsymbol{r} - \boldsymbol{R}_i), \tag{16}$$

where coefficients $c(ilm\sigma)$ are the multipole elements of this field. Respectively, the multipole elements of the force density, according to [9], are defined as

$$f(jlm\sigma) = \int d\boldsymbol{r}\boldsymbol{v}^{+}_{lm\sigma}(\boldsymbol{r} - \boldsymbol{R}_j) \cdot \boldsymbol{f}_j(\boldsymbol{r}). \tag{17}$$

In the multipole language, the integral equation (15) becomes a linear algebraic equation connecting various multipoles $c(i)$, $i = 1, 2, \dots$ of the fluid velocity on particles' surfaces and the multipoles $f(j),\ j = 1, 2, \ldots$ of the induced force densities. Because the influence of an induced force density located on a given particle, on that particles velocity is strictly different from the influence of force densities established on other particles, this equation is split into two parts. One is described by a single-particle operator $\boldsymbol{Z}_0$ defined by

$$\left[\boldsymbol{Z}_0^{-1}(i)\boldsymbol{f}_i\right](\boldsymbol{r}) \equiv \int \boldsymbol{T}_0(\boldsymbol{r}-\boldsymbol{r}')\boldsymbol{f}_i(\boldsymbol{r}')d\boldsymbol{r}', \quad \boldsymbol{r} \in S_i, \tag{18}$$

whereas the second part, describing the velocity field around particle i, coming from a force density present on another sphere j is written using the Green operator $\boldsymbol{G}(ij)$ defined by

$$\left[\boldsymbol{G}(ij)\boldsymbol{f}_j\right](\boldsymbol{r}) \equiv \int \boldsymbol{T}_0(\boldsymbol{r}-\boldsymbol{r}')\boldsymbol{f}_j(\boldsymbol{r}')d\boldsymbol{r}', \quad \boldsymbol{r} \in S_i, i \neq j. \tag{19}$$

In the multipole language these integral operators become infinite matrices. The Green operators matrix elements, given explicitly in [9], are characterized by two groups of four indexes $G(ilm\sigma, jl'm'\sigma')$, where i and j are the particle indexes, and $lm\sigma$, $l'm'\sigma'$—multipole indexes. It is here worth noting that the elements of the multipole expansion of the Green operator have a well defined dependence on the interparticle distance

$$G(ilm\sigma, jl'm'\sigma') \propto \frac{1}{R_{ij}^{l+l'+\sigma+\sigma'+1}}, \tag{20}$$

where $R_{ij} = |\boldsymbol{R}_i - \boldsymbol{R}_j|$. Further, the one-particle operator $\boldsymbol{Z}_0^{-1}$ has only diagonal elements in the particle indexes i and j. Its multipole matrix elements have therefore the form $Z_0^{-1}(ilm\sigma, il'm'\sigma')$ and are also derived explicitly in [9].

Summarizing briefly, in order to find the induced force densities, an expansion of (15) in a basis of functions $\boldsymbol{v}_{lm\sigma}^{\pm}$ is carried out. This then leads to an algebraic equation for the expansion coefficients, which is of the form

$$\boldsymbol{c} = (\boldsymbol{Z}_0^{-1} + \boldsymbol{G})\boldsymbol{f}. \tag{21}$$

Here, the infinite dimensional vectors $\boldsymbol{c}$ and $\boldsymbol{f}$ with components $c(ilm\sigma)$ and $f(jlm\sigma)$ have been introduced. Now the derivation of the induced force density multipoles boils down to finding the inverse of the infinite matrix $\boldsymbol{Z}_0^{-1} + \boldsymbol{G}$. Practically this is done numerically once a suitable truncation order has been chosen.

The first multipole moments of the force densities and the fluid velocity field around each particle correspond to the spherical components of the total force ($l = 1,\ \sigma = 0$), torque ($l = 1,\ \sigma = 1$) and translational ($l = 1,\ \sigma = 0$) and rotational ($l = 0,\ \sigma = 1$) velocities of the particles. The solution of (21) allows one to calculate all force multipoles, knowing the translational and rotational velocities of the particles and the multipoles of the ambient flow. But in the problems considered in this work, the known quantities are in fact forces and torques acting on the particles (i.e. the first multipoles of the induced force density) and the ambient flow, whereas the velocities of the particles are unknown. To find these, the equation for $\boldsymbol{f}$, derived by inverting the matrix $\boldsymbol{Z}_0^{-1} + \boldsymbol{G}$, must be inversed once again, but only in the space of the first two multipoles ($l = 1$ and $\sigma = 0, 1$). In this way the mobility matrix $\boldsymbol{\mu}$ and the convection operator $\boldsymbol{C}$ are derived. For a precise description of this method, the reference [9] should be addressed. The formulas which are obtained are of the form of scattering sequences

$$\boldsymbol{\mu} = \boldsymbol{\mu}_0 + \boldsymbol{\mu}_0\boldsymbol{Z}_0\frac{1}{1+\boldsymbol{G}\hat{\boldsymbol{Z}}_0}\boldsymbol{G}\boldsymbol{Z}_0\boldsymbol{\mu}_0 = \boldsymbol{\mu}_0 + \boldsymbol{\mu}_0\boldsymbol{Z}_0\sum_{k=0}^{\infty}(-\boldsymbol{G}\hat{\boldsymbol{Z}}_0)^k\boldsymbol{G}\boldsymbol{Z}_0\boldsymbol{\mu}_0, \tag{22}$$

$$\mathcal{C} = \boldsymbol{\mu}_0 \boldsymbol{Z}_0 - \boldsymbol{\mu}_0 \boldsymbol{Z}_0 \boldsymbol{G} \hat{\boldsymbol{Z}} = \boldsymbol{\mu}_0 \boldsymbol{Z}_0 \frac{1}{1 + \boldsymbol{G}\hat{\boldsymbol{Z}}_0} = \sum_{k=0}^{\infty} \boldsymbol{\mu}_0 \boldsymbol{Z}_0 (-\boldsymbol{G}\hat{\boldsymbol{Z}}_0)^k, \tag{23}$$

where the superposition of operators should be understood in the multipole language as a summation with respect to particle and multipole indexes. Here $\boldsymbol{\mu}_0$ is the translational and rotational mobility of a single sphere. Further

$$\hat{\boldsymbol{Z}}_0 = \boldsymbol{Z}_0 - \boldsymbol{Z}_0 \boldsymbol{\mu}_0 \boldsymbol{Z}_0, \tag{24}$$

is the convection-friction single-particle operator. When limited to the space of the first two multipoles ($l = 1,\ \sigma = 0, 1$), the one-particle mobility is the inverse of the one-particle friction operator. Therefore $\hat{\boldsymbol{Z}}_0$ as defined by (24) is zero in the space spanned by these multipoles.

The force densities induced on particle surfaces once subject to external forces and ambient flow, may also be formulated in terms of a scattering sequence. It has the following form [27]

$$\boldsymbol{f} = \frac{1}{1 + \hat{\boldsymbol{Z}}_0 \boldsymbol{G}} \boldsymbol{Z}_0 \boldsymbol{\mu}_0 \boldsymbol{\mathcal{F}} - \frac{1}{1 + \hat{\boldsymbol{Z}}_0 \boldsymbol{G}} \hat{\boldsymbol{Z}}_0 \boldsymbol{v}_0, \tag{25}$$

where

$$\boldsymbol{\mathcal{F}}_i = \begin{pmatrix} \boldsymbol{F}_i \\ \boldsymbol{\mathcal{T}}_i \end{pmatrix}, \tag{26}$$

is a vector of external forces and torques. This expression for the induced force densities will be necessary in further parts of this work.

Here, as we concentrate only on the translational movement of particles only that part of the mobility matrix (see (22)), convection operator (see (23)) and $\boldsymbol{\mathcal{F}}$ (refer to (26)) will be taken into account. Then only the single particle translational mobility

$$\boldsymbol{\mu}_0^{tt} = \frac{1}{6\pi\eta a} \boldsymbol{I}, \tag{27}$$

where $\boldsymbol{I}$ is the unit tensor, is used. For the sake of simplicity of notation, we will omit the superscript (t).

Indicating the particle indexes explicitly the mobility operator, as given in terms of a scattering sequence of the form as in (22), has the structure

$$\boldsymbol{\mu}_{11}(\boldsymbol{X}) = \boldsymbol{\mu}_0 + \sum_{i \neq 1} \boldsymbol{\mu}_0(1) \boldsymbol{Z}_0(1) \boldsymbol{G}(1i) \hat{\boldsymbol{Z}}_0(i) \boldsymbol{G}(i1) \boldsymbol{Z}_0(1) \boldsymbol{\mu}(1) + \cdots, \tag{28}$$

$$\begin{aligned} \boldsymbol{\mu}_{12}(\boldsymbol{X}) = {} & \boldsymbol{\mu}_0(1) \boldsymbol{Z}_0(1) \boldsymbol{G}(12) \boldsymbol{Z}_0(2) \boldsymbol{\mu}_0(2) \\ & - \sum_{i \neq 1,2} \boldsymbol{\mu}_0(1) \boldsymbol{Z}_0(1) \boldsymbol{G}(1i) \hat{\boldsymbol{Z}}_0(i) \boldsymbol{G}(i2) \boldsymbol{Z}_0(2) \boldsymbol{\mu}_0(2) + \cdots. \end{aligned} \tag{29}$$

In general, each scattering expression for the mobility operator $\boldsymbol{\mu}$, described by a specific term of (22) is constructed as follows [7, 24]: it begins by establishing the force density on a given particle j under the influence of an external force. This is given by the operator $\boldsymbol{Z}_0(j)\boldsymbol{\mu}_0(j)$. The force density localized on that particle induces a flow field in the viscous fluid, which propagates in the system. This propagation is described by the Green operator $\boldsymbol{G}(kj)$. Once the disturbance reaches another particle, it creates an additional force density given by the action of the operator $-\hat{\boldsymbol{Z}}_0(k)$. This force density is such that the total force and torques on that particle vanish, as these quantities are set already at the beginning of the scattering sequence. The mechanism described, can be replicated an arbitrary number of times. Finally, the velocity of a particle is determined by the operator $\boldsymbol{\mu}_0(i)\boldsymbol{Z}_0(i)$ which closes the scattering sequence. A strictly analogous interpretation is true in the case of the convection operator $\boldsymbol{\mathcal{C}}$ given by the sequence (23). The only difference takes place at the beginning of each term, where the initial force density is set by the influence of the ambient flow ant the operator $-\hat{\boldsymbol{Z}}_0(k)\boldsymbol{v}_0$.

The asymptotic distance behavior of the terms of the scattering sequence is set by the formula (20). In particular

$$\boldsymbol{Z}_0(i)\boldsymbol{G}(ij)\boldsymbol{Z}_0(j) \sim \frac{1}{R_{ij}}, \tag{30}$$

$$\hat{\boldsymbol{Z}}_0(i)\boldsymbol{G}(ij)\boldsymbol{Z}_0(j) \sim \boldsymbol{Z}_0(i)\boldsymbol{G}(ij)\hat{\boldsymbol{Z}}_0(j) \sim \frac{1}{R_{ij}^2}, \tag{31}$$

$$\hat{\boldsymbol{Z}}_0(i)\boldsymbol{G}(ij)\hat{\boldsymbol{Z}}_0(j) \sim \frac{1}{R_{ij}^3}, \tag{32}$$

for large interparticle distances R_{ij}. These formulas are due to the fact, that the lowest non-zero multipole element of the operator $\boldsymbol{Z}_0$ is of rank $l = 1, \sigma = 0$, but for the convection-friction operator $\hat{\boldsymbol{Z}}_0$ it is $l = 2, \sigma = 0$. Nevertheless all single hydrodynamic connections via the Green operator lead to very long-range interactions. Only when two Green operators connect chosen clusters of particles, a $1/r^4$ (doubled $1/r^2$, where r is the distance between clusters of particles) distance dependence emerges, as each of the Green propagators must in such a case be at least of the type as in (31).

4 The BBGKY Hierarchy for Reduced Distribution Functions

In order to obtain equations governing the time evolution of these functions, cluster expansions of the hydrodynamic operators $\boldsymbol{\mu}$ and $\boldsymbol{\mathcal{C}}$ must be derived. This is done on the following way. The hydrodynamic operators $\boldsymbol{\mu}$ and $\boldsymbol{\mathcal{C}}$ may be expressed in terms of scattering sequences of the form given by (22). Next, structures which contain the same set of particle position variables are grouped together. For the mobility matrix (or convection operator) describing self interaction of particles the following sequence is then obtained

$$\boldsymbol{\mu}_{11}(X) = \boldsymbol{\mu}_{11}^{(1)}(1) + \sum_{i=2}^{N} \boldsymbol{\mu}_{11}^{(2)}(1i) + \frac{1}{2!}\sum_{i,j=2}^{N} \boldsymbol{\mu}_{11}^{(3)}(1ij) + \cdots, \tag{33}$$

where $\boldsymbol{\mu}_{11}^{(s)}(i_1, \ldots i_s)$ is a cluster part which is the sum of all terms of the scattering expansion of the full mobility matrix (22), which contain exactly the positions of particles $i_1, \ldots, i_s$. Here, and in all following formulas the abbreviated notation $i \equiv \boldsymbol{R}_i$ will be used. The expansion introduced is unique and all its elements are well defined. The explicit expressions for the first terms of this expansion are given for example in [7].

The interaction between different particles, as described by the mobility matrix or convection operator may also by written in a cluster expansion of the form

$$\boldsymbol{\mu}_{12}(X) = \boldsymbol{\mu}_{12}^{(2)}(12) + \sum_{i=3}^{N} \boldsymbol{\mu}_{12}^{(3)}(12i) + \cdots. \tag{34}$$

Here, as above, $\boldsymbol{\mu}_{12}^{(s)}(i_1, \dots, i_s)$ denotes the sum of all terms of the scattering expansion of the mobility matrix, which begin (looking from the right) at particle 2, end at particle 1 and contain exactly $i_1, \dots, i_s$ particle positions. An identical expansion exists for the convection operator.

As already mentioned, in order to formulate a proper statistical description of the considered system, the set of reduced distribution functions is introduced instead of the function $\rho(\boldsymbol{X}; t)$. These are defined using the microscopic density

$$\hat{n}(\boldsymbol{r}_1, \dots, \boldsymbol{r}_s | \boldsymbol{X}) \equiv \sum_{k_1, \dots, k_s}{}^{'} \delta(\boldsymbol{r}_1 - \boldsymbol{R}_{k_1}) \cdots \delta(\boldsymbol{r}_s - \boldsymbol{R}_{k_s}), \tag{35}$$

as

$$n(\boldsymbol{r}_1, \dots, \boldsymbol{r}_s; t) \equiv \left\langle \hat{n}(\boldsymbol{r}_1, \dots, \boldsymbol{r}_s | \boldsymbol{X}) \right\rangle_t == \int d\boldsymbol{X} \hat{n}(\boldsymbol{r}_1, \dots, \boldsymbol{r}_s | \boldsymbol{X}) \rho(\boldsymbol{X}; t). \tag{36}$$

The symbol $\sum'$ denotes a summation over non-repeating index values and $\langle \cdot \rangle_t$ denotes the averaging over the particle distribution $\rho(\boldsymbol{X}; t)$.

In case of a system of identical particles this definition boils down to the formula

$$n(1, 2, \dots, s; t) = \frac{N!}{(N-s)!} \int d(s+1) \cdots dN \rho(\boldsymbol{X}; t), \tag{37}$$

where the particle notation introduced in (33) has been used, and the integration with respect to particle indexes should be understood as

$$\int di \equiv \int d\boldsymbol{R}_i. \tag{38}$$

The evolution equations for the reduced distribution functions are derived by calculating the time derivative of (37) and using the Liouville equation (7). The following equations are then obtained:

$$\left(\frac{\partial}{\partial t} + \mathfrak{L}(\boldsymbol{s}) \right) n(\boldsymbol{s}; t) = -\sum_{l=1}^{\infty} \int d(s + \boldsymbol{l}) \mathfrak{L}(\boldsymbol{s} | s + \boldsymbol{l}) n(\boldsymbol{s} + \boldsymbol{l}; t), \tag{39}$$

where

- $\boldsymbol{s}$ denotes the set of particles $\{1, \dots, s\}$,
- $s + \boldsymbol{l}$ denotes the l-particle set $\{s+1, \dots, s+l\}$,

and

$$\int d(s + \boldsymbol{l}) \equiv \frac{1}{l!} \int d(s+1) \cdots d(s+l). \tag{40}$$

Further

$$\mathfrak{L}(\boldsymbol{s}) n(\boldsymbol{s}; t) = \sum_{i=1}^{s} \nabla_i \cdot \left\{ [\boldsymbol{\mu} \boldsymbol{F} + \boldsymbol{\mathcal{C}} \boldsymbol{v}_0]_i \, (\boldsymbol{s}) \, n(\boldsymbol{s}; t) \right\}, \tag{41}$$

is the s-particle Liouville operator, where the abbreviated notation should be expanded as

$$[\boldsymbol{\mu} \boldsymbol{F} + \boldsymbol{\mathcal{C}} \boldsymbol{v}_0]_i (\boldsymbol{s}) = \sum_{j=1}^{s} \boldsymbol{\mu}_{ij}(\boldsymbol{s}) \boldsymbol{F}_j + \boldsymbol{\mathcal{C}}_i(\boldsymbol{s}) \boldsymbol{v}_0, \tag{42}$$

where $\boldsymbol{\mu}(\boldsymbol{s})$ and $\boldsymbol{\mathcal{C}}(\boldsymbol{s})$ are s-particle mobility and convection operators respectively. Likewise

$$\mathfrak{L}(\boldsymbol{s}|s+\boldsymbol{l})n(\boldsymbol{s}+\boldsymbol{l};t)=\sum_{i=1}^{s}\nabla_i\cdot\left\{[\boldsymbol{\mu}\boldsymbol{F}+\boldsymbol{\mathcal{C}}\boldsymbol{v}_0]_i\,(\boldsymbol{s}|s+\boldsymbol{l})\;n(\boldsymbol{s}+\boldsymbol{l};t)\right\}, \tag{43}$$

is the $s+l$-particle, extended Liouville operator. Here

$$[\boldsymbol{\mu}\boldsymbol{F}+\boldsymbol{\mathcal{C}}\boldsymbol{v}_0]_i(\boldsymbol{s}|s+\boldsymbol{l})=\sum_{j=1}^{s+l}(\boldsymbol{\mu}_{ij}(\boldsymbol{s}|s+\boldsymbol{l})\boldsymbol{F}_j+\boldsymbol{\mathcal{C}}_i(\boldsymbol{s}|s+\boldsymbol{l})\boldsymbol{v}_0) \tag{44}$$

and the following extended hydrodynamic operators have been introduced

$$\begin{aligned}
\boldsymbol{\mu}_{11}(1,\ldots,s|s+1,\ldots,s+l) &= \boldsymbol{\mu}_{11}^{(1+l)}(1,s+1,\ldots,s+l)\\
&\quad+\sum_{i=2}^{s}\boldsymbol{\mu}_{11}^{(2+l)}(1i,s+1,\ldots,s+l)+\cdots,\\
\boldsymbol{\mu}_{12}(1,\ldots,s|s+1,\ldots,s+l) &= \boldsymbol{\mu}_{12}^{(2+l)}(12,s+1,\ldots,s+l)\\
&\quad+\sum_{i=3}^{s}\boldsymbol{\mu}_{12}^{(3+l)}(12i,s+1,\ldots,s+l)+\cdots,\\
\boldsymbol{\mu}_{1s+1}(1,\ldots,s|s+1,\ldots,s+l) &= \boldsymbol{\mu}_{1s+1}^{(1+l)}(1,s+1,\ldots,s+l)\\
&\quad+\sum_{i=2}^{s}\boldsymbol{\mu}_{1s+1}^{(2+l)}(1i,s+1,\ldots,s+l)+\cdots.
\end{aligned} \tag{45}$$

These expansions are generalizations of the cluster structures (33) and (34) and may be interpreted as such expansions when limited to the set of particles $1,\ldots,s$. Here all terms must contain all the particles numbered $s+1,\ldots,s+l$ and at least one particle from the set $\{1,\ldots,s\}$, as all scattering structures considered here end (at the left) at one of those particles.

The set obtained by taking different values of $s=1,2,\ldots,$ in (39) forms a hierarchy of equations. This is the analogue of the BBGKY hierarchy introduced in the kinetic theory of gases. The summation in (39) formally extends to infinity. In a finite system, this means that $n(1,\ldots,s)$ is equal to zero for $s>N$, where N is the number of particles in the system. Usually the BBKGY equations are considered in the thermodynamic limit understood as $N\to\infty$, $V\to\infty$ while keeping $N/V=const$. Then formally all integrals in those equations extend to infinity, this set itself is infinite, and the equations contain an infinite number of terms, contrary to what is usually the case when considering two-body potential interactions. In order to find an s-particle distribution function it is therefore necessary to know all the higher-order functions, which is essentially impossible. A solution must therefore introduce a truncation at some point, which is further physically justifiable.

5 The BBGKY Hierarchy for Correlation Functions

When analyzing a system from the statistical perspective one is usually interested in the correlations which arise from particle interactions. These can be derived once the reduced distribution functions are known, and the following cluster expansion [17] is taken into account

$$
\begin{aligned}
n(1) &= h(1),\\
n(12) &= h(1)h(2) + h(12),\\
n(123) &= h(1)h(2)h(3) + h(12)h(3) + h(13)h(2) + h(23)h(1) + h(123),\\
\vdots\ &\ \ \vdots\\
n(\boldsymbol{s}) &= \sum_{\bigsqcup_i \boldsymbol{m}_i = \boldsymbol{s}} \prod_i h(\boldsymbol{m}_i),
\end{aligned}
\tag{46}
$$

where the functions $h(1,\dots,s)$ are s-particle correlation functions which vanish whenever any two particles from the set $1,\dots,s$ are an infinite distance from each other. Here, the explicit dependence on time has been left out.

These expansions, when inserted into (39) lead to an analogous hierarchy of equations for the correlation functions. As an equation for $n(1,\dots,s)$ has the same cluster structure as the function itself, finding the equation for an s-particle correlation function boils down to selecting terms where all s particles form a disjoint cluster. In other words this means that all these particles are contained in some correlation or hydrodynamic sequence. This property leads naturally to the notion of connectivity. To define it properly, some additional elements must be introduced. This will now be performed.

The Liouville operator $\mathfrak{L}(\boldsymbol{s}|s+\boldsymbol{l})$ is basically a sum of cluster structures of the hydrodynamic operators $\boldsymbol{\mu}$ and $\boldsymbol{C}$. All these are sums of certain terms of the appropriate scattering sequences. The Liouville operator has therefore itself a scattering structure and may be formulated in terms of one- and two-particle operators $\boldsymbol{Z}_0$ and $\boldsymbol{G}$. The consecutive terms of this series must end (at the left) with one of the particles indexed $1,\dots,s$ upon which acts the divergence derivative operator, and start with any of the particles $1,\dots,s+l$ indicated. The sequence is then acted upon from the right by the force exerted on the leading particle or the external flow. Every term of the expansion of $\mathfrak{L}(\boldsymbol{s}|s+\boldsymbol{l})$ is characterized by the first and last particle appearing in the scattering structure and the k-element subset $\mathcal{S}_k \in \boldsymbol{s}$ of particles which actually compose the hydrodynamic structure. This structure will be denoted by $\Lambda_{ij}^{(k+l)}(\mathcal{S}_k|s+\boldsymbol{l})$, where j and i represent the leading and closing particles [27].

The action of the Liouville operator on a reduced distribution function has the form of a product of two sequences. It is therefore a sequence itself, the terms of which have the structure

$$
\Phi[\Lambda, c](\boldsymbol{s}|s+\boldsymbol{l}) = \nabla_i \cdot \Lambda_{ij}^{(k+l)}(\mathcal{S}_k|s+\boldsymbol{l})c(\boldsymbol{s}+\boldsymbol{l}),
\tag{47}
$$

where

- $\mathcal{S}_k$ is a k-element subset of the set of particles $\boldsymbol{s}$, such that $i \in \mathcal{S}_k$,
- $\Lambda_{ij}^{(k+l)}(\mathcal{S}_k|s+\boldsymbol{l})$ is a scattering sequence which begins (looking from the right) at particle $j \in \mathcal{S}_k \cup (s+\boldsymbol{l})$, and ends at particle $i \in \mathcal{S}_k$,
- $c(\boldsymbol{s}+\boldsymbol{l})$ is a correlation structure—one of the terms of the expansion of the $s+l$-particle reduced distribution function in a sequence of products of correlation functions.

These structures are the main building blocks of the hierarchy equations and can graphically be represented by diagrams of the form introduced in Appendix B.

According to the topological features of $\Phi[\Lambda, c](\boldsymbol{s}|s+\boldsymbol{l})$, these structures may be classified as connected or non-connected. A non-connected term is one which may be factorized into functions depending on excluding sets of variables. A connected term is one, where all particles are linked through a sequence of Green propagators or additional correlation functions. The identification is best performed using the diagrammatic technique summarized in Appendix B. Connected terms then correspond to connected graphs, and the task of finding the connected part of the BBKGY hierarchy equation boils down to finding all such graphs with a given, predetermined structure. The distinction of these graphs may also be done by introducing for each hydrodynamic scattering expression a correlation function which ensures the connectivity of the underlying term $\Phi[\Lambda, c](\boldsymbol{s}|s+\boldsymbol{l})$. These functions are called connecting correlation function and are defined as follows:

Let $\mathcal{S}_k \subset \mathbf{s}+\boldsymbol{l}$ *be a* k*-element set and* $b(\boldsymbol{s}+\boldsymbol{l}|\mathcal{S}_k)$ *denote a correlation function, which ensures that each of the particles from this set is correlated with any particle from the complementary set* $\mathbf{s}+\boldsymbol{l}\setminus\mathcal{S}_k$.

For example

$$\begin{aligned}
b(12|1) &= n(12) - h(1)h(2) = h(12),\\
b(123|1) &= n(123) - n(23)h(1) = h(123) + h(12)h(3) + h(13)h(2),\\
b(123|12) &= n(123) - h(12)h(3) - h(23)h(1) - h(13)h(2) - h(1)h(2)h(3) = h(123),\\
&\vdots \quad \ddots
\end{aligned} \tag{48}$$

The connecting correlation function enables one to write the connected part of the discussed expressions. If

$$\{\mathfrak{L}(\boldsymbol{s}|s+\boldsymbol{l})n(\boldsymbol{s}+\boldsymbol{l})\}^c = \sum_{i=1}^{s} \nabla_i \cdot \{[\boldsymbol{\mu F} + \mathcal{C}\boldsymbol{v}_0]_i(\boldsymbol{s}|s+\boldsymbol{l})\, n(\boldsymbol{s}+\boldsymbol{l})\}^c, \tag{49}$$

where

$$[\boldsymbol{\mu F}]_i(\boldsymbol{s}|s+\boldsymbol{l}) = \sum_{k=1}^{s} \sum_{\mathcal{S}_k \in \boldsymbol{s}} [\boldsymbol{\mu F}]_i^{(k+l)}(\mathcal{S}_k|s+\boldsymbol{l}), \tag{50}$$

and

$$[\mathcal{C}\boldsymbol{v}_0]_i(\boldsymbol{s}|s+\boldsymbol{l}) = \sum_{k=1}^{s} \sum_{\mathcal{S}_k \in \boldsymbol{s}} [\mathcal{C}\boldsymbol{v}_0]_i^{(k+l)}(\mathcal{S}_k|s+\boldsymbol{l}), \tag{51}$$

$i \in \mathcal{S}_k$, then

$$\{[\boldsymbol{\mu F}]_i(\boldsymbol{s}|s+\boldsymbol{l})\, n(\boldsymbol{s}+\boldsymbol{l})\}^c = \sum_{k=1}^{s} \sum_{\mathcal{S}_k \subset \boldsymbol{s}} [\boldsymbol{\mu F}]_i(\mathcal{S}_k|s+\boldsymbol{l}) b(\boldsymbol{s}+\boldsymbol{l}|\boldsymbol{s}\setminus\mathcal{S}_k), \tag{52}$$

where the superscript c denotes the connected part of the expression.

The structure of the correlation functions is connected and such is the structure of the equation governing its time evolution. In fact the equation for an s-particle correlation function is exactly the connected part of the equation for the s-particle reduced distribution function. These equations have therefore the following form

$$\frac{\partial h(\boldsymbol{s};t)}{\partial t} = -\{\mathfrak{L}(\boldsymbol{s})n(\boldsymbol{s};t)\}^c - \sum_{l=1}^{\infty} \int d(s+\boldsymbol{l})\{\mathfrak{L}(\boldsymbol{s}|s+\boldsymbol{l})n(\boldsymbol{s}+\boldsymbol{l};t)\}^c, \tag{53}$$

where the superscript c denotes the connected parts of the expressions (49).

For example the equation for the two-particle correlation function has the form

$$\frac{\partial h(12;t)}{\partial t} = -\sum_{i=1,2} \nabla_i \cdot \{\boldsymbol{\mu}_{ii}^{(1)}(i)\boldsymbol{F}_i h(12;t)\} \\ - \sum_{i,j=1,2} \nabla_i \cdot \{\boldsymbol{\mu}_{ij}^{(2)}(12)\boldsymbol{F}_j(h(12;t)+h(1;t)h(2;t))\} + \cdots, \qquad (54)$$

assuming that $\boldsymbol{v}_0 = 0$.

Note that the term

$$\sum_{i=1,2} \nabla_i \cdot \{\boldsymbol{\mu}_{ii}^{(1)}(i)\boldsymbol{F}_i h(1;t)h(2;t)\}, \qquad (55)$$

which is not connected is not present in the above equation. On the other hand, this equations contains the structure

$$\nabla_1 \cdot \{\boldsymbol{\mu}_{12}^{(2)}(12)\boldsymbol{F}_2 h(1;t)h(2;t)\}, \qquad (56)$$

where (according to (34) and (28))

$$\boldsymbol{\mu}_{12}^{(2)}(12) = \boldsymbol{\mu}_0(1)\boldsymbol{Z}_0(1)\boldsymbol{G}(12)\boldsymbol{Z}_0(2)\boldsymbol{\mu}_0(2) \\ + \boldsymbol{\mu}_0(1)\boldsymbol{Z}_0(1)\boldsymbol{G}(12)\hat{\boldsymbol{Z}}_0(2)\boldsymbol{G}(21)\hat{\boldsymbol{Z}}_0(1)\boldsymbol{G}(12)\boldsymbol{Z}_0(2)\boldsymbol{\mu}_0(2) + \cdots. \qquad (57)$$

But as pointed out in (30), the first term of this expansion behaves asymptotically as $1/R_{12}$ for large interparticle distances R_{12}. Such terms may lead to non-integrability of the correlation function $h(12;t)$. Further, as similar expressions are present in the integrals of the right-hand side of the BBGKY hierarchy equations, these might not be convergent in the thermodynamic limit.

The next section will be devoted to the analysis of the derived hierarchy of equations. Special attention will be concentrated on expressions which lead to non-integrable expressions in these equations. A procedure to deal with such terms will be introduced.

6 Resummation of Non-integrable Expressions

In the previous section we obtained equations which govern the time evolution of the reduced distribution functions and correlation functions in a suspension of hydrodynamically interacting particles. We have also pointed out, that the right-hand sides of those equations contain expressions, which asymptotically decay proportionally to $1/r^\alpha$, where $\alpha = 1, 2, 3$. These are non-integrable. A parallel problem is well known in the Debye-Hückel theory of interacting charged particles, when the long-ranged electrostatic interactions lead to similar complications [11].

In this part we will concentrate on the selection of all expressions which might lead to non-integrability. These will later be treated in a special way and summed together to form physically meaningful fields.

We begin by invoking the formula for the local suspension velocity field. It is given by (12). This field is valid throughout the suspension. At points in the fluid, it describes the fluid velocity field, whereas at points inside the particles it follows their rigid-body motion.

Next the average (here and in subsequent equations we omit the explicit time dependence, whenever it does not lead to confusions)

$$\big\langle \boldsymbol{v}(\boldsymbol{r}|\boldsymbol{X})\hat{n}(\boldsymbol{r}_1,\dots,\boldsymbol{r}_s|\boldsymbol{X})\big\rangle, \tag{58}$$

is introduced. As all many-particle expressions it may be written in terms of a cluster expansion according to

$$\langle \boldsymbol{v}(\boldsymbol{r})\rangle = \boldsymbol{c}(\boldsymbol{r}), \tag{59}$$

$$\begin{aligned}
\big\langle \boldsymbol{v}(\boldsymbol{r})\hat{n}(\boldsymbol{r}_1)\big\rangle &= \boldsymbol{c}(\boldsymbol{r})n(\boldsymbol{r}_1) + \boldsymbol{c}(\boldsymbol{r}|\boldsymbol{r}_1),\\
\big\langle \boldsymbol{v}(\boldsymbol{r})\hat{n}(\boldsymbol{r}_1,\boldsymbol{r}_2)\big\rangle &= \boldsymbol{c}(\boldsymbol{r})n(\boldsymbol{r}_1,\boldsymbol{r}_2) + \boldsymbol{c}(\boldsymbol{r}|\boldsymbol{r}_1)n(\boldsymbol{r}_2)\\
&\quad + \boldsymbol{c}(\boldsymbol{r}|\boldsymbol{r}_2)n(\boldsymbol{r}_1) + \boldsymbol{c}(\boldsymbol{r}|\boldsymbol{r}_1,\boldsymbol{r}_2),\\
\vdots\quad & \quad\vdots,
\end{aligned} \tag{60}$$

where a vector field $\boldsymbol{c}(\boldsymbol{r}|\boldsymbol{r}_1,\dots,\boldsymbol{r}_s)$ describes correlations between the suspension velocity fluctuations at point $\boldsymbol{r}$ and fluctuations of the particle concentration at points $\boldsymbol{r}_1,\dots,\boldsymbol{r}_s$.

In order to proceed, the notion of reducibility has to be introduced. This will be done along the lines of [27], starting by the definition of the concept of connection lines.

A two-particle Green operator $\boldsymbol{G}$ is called a *connection line* when a term $\Phi[\Lambda, c]$ can be written in the form

$$\begin{aligned}
\Phi[\Lambda,c](\boldsymbol{s}|s+\boldsymbol{l}) &= \nabla_i\cdot[\Lambda^{(k+l)}_{ij}(\mathcal{S}_k; s+\boldsymbol{l})c(\boldsymbol{s}+\boldsymbol{l})]\\
&= \nabla_i\cdot\{\Lambda^{(n+m)}_{ip}(\sigma_n;\sigma_m)c(\Sigma)\}\boldsymbol{G}(pq)\cdot\Lambda^{(n'+m')}_{qj}(\bar{\sigma}_{n'};\bar{\sigma}_{m'})c(\bar{\Sigma}),
\end{aligned} \tag{61}$$

where

1. σ_n and $\bar{\sigma}_{n'}$ are n and n'-element $(n+n'=k)$ disjoint sets such that $\mathcal{S}_k = \sigma_n \cup \bar{\sigma}_{n'}$, $k = 1,2,\dots,s$, $n=1,\dots,k$. Further $i\in\sigma_n$, so this set is by definition not empty.
2. The sets σ_m and $\bar{\sigma}_{m'}$ are m and m'-element $(m+m'=l)$ disjoint sets such that $s+\boldsymbol{l} = \sigma_m\cup\bar{\sigma}_{m'}$, $m=0,\dots,l$.
3. Σ i $\bar{\Sigma}$ are disjoint sets such that $\boldsymbol{s}+\boldsymbol{l} = \Sigma\cup\bar{\Sigma}$ and

$$\Sigma \supseteq \sigma_n\cup\sigma_m, \tag{62}$$

$$\bar{\Sigma} \supseteq \bar{\sigma}_{n'}\cup\bar{\sigma}_{m'}. \tag{63}$$

4. $\boldsymbol{G}(pq)$ is a two-particle Green operator connecting the two disjoint structures which appear before and after it.

Connection lines appearing in the hierarchy mark those Green operators, which once removed, lead to non-connectivity of the discussed structures. The distinction of these operators is due to the extremely long range of interaction related by single Green operators (as described by (30)–(32)). When a structure does not contain connection lines, it is said to be *irreducible*. Each Green's function is then doubled by another hydrodynamic connection, or a sufficiently short-ranged correlation function. All terms then decay at least proportionally to $1/r^4$. This is illustrated in diagrammatic form in Appendix B.

In the first step toward a reformulation of the hierarchy equations, all irreducible structures are collected. These are written in terms of two operators defined by the formulas:

$$\mathbb{L}^{(s+l)}[n](\boldsymbol{s}) = \sum_{i=1}^{s} \int d(s+\boldsymbol{l})\nabla_i \cdot \left[(\boldsymbol{\mu F})_i(\boldsymbol{s}|s+\boldsymbol{l})n(\boldsymbol{s}+\boldsymbol{l};t)\right]^{irr}, \tag{64}$$

$$\mathbb{C}^{(s+l)}[n](\boldsymbol{s}) = \sum_{i=1}^{s} \int d(s+\boldsymbol{l})\nabla_i \cdot \left[\boldsymbol{\mathcal{C}}_i(\boldsymbol{s}|s+\boldsymbol{l})n(\boldsymbol{s}+\boldsymbol{l};t)\right]^{irr}. \tag{65}$$

Next all remaining terms are reduced according to the procedure described in detail in [27]. This is basically done in the following way: in all reducible terms having the structure (61) the first connection line (looking from the left in a scattering structure) is chosen. Then, the expression, which is on the left of it is by definition irreducible (as it does not contain connection lines). Its scattering structure is the same as that of a particular term of the expansion of the convection operator $\mathcal{C}$ given by (23). The expression which stands on the right of the connection line may be reducible or not. Its scattering structure is exactly the one of the expansion (25) of $\boldsymbol{f}$. Further, a connection line splits the set of $\boldsymbol{s}$ particles, which variables are not integrated in the equations, into two clusters. One, $\sigma_k \in \boldsymbol{s}$ enters the expressions on the left of the connection line, while the complimentary set $\bar{\sigma}_{k'} \in \boldsymbol{s}$, $k+k'=s$ takes part in the scattering and correlation structure on the right of the connection line. All other particles are distributed in either of the groups.

In the thermodynamic limit, when the number of all particles is infinite, a summation of all such reducible structures leads to the operator $\mathbb{C}^{(s+l)}[n](\sigma_k)$ (refer to (65)) standing on the left of the connection line, while on the right of the connection line, the exact hydrodynamic expression (25) of the induced force densities is established. Combined with the correlation structure, it boils down to the averaged field

$$\left\langle \boldsymbol{f}(\boldsymbol{r}|\boldsymbol{X})\hat{n}(\bar{\sigma}_{s-k}|\boldsymbol{X})\right\rangle. \tag{66}$$

The support of this field is limited to the particle surfaces, outside of which it is equal to zero. The convection operators (see (65)) support is also confined to these surfaces. Taking this into account, the definition (13) of the suspension velocity field, together with the definition (19) of the Green propagator connecting two particles, may be used to combine the connection line and the averaged induced force density into the field

$$\langle \boldsymbol{v}\ \hat{n}(\bar{\sigma}_{s-k})\rangle = \boldsymbol{v}_0 n(\sigma_{s-k}) + \boldsymbol{G}\left\langle \boldsymbol{f}(\boldsymbol{X})\hat{n}(\sigma_{s-k}|\boldsymbol{X})\right\rangle. \tag{67}$$

The BBGKY equations may now be written as

$$\frac{\partial n(\boldsymbol{s};t)}{\partial t} = \sum_{l=0}^{\infty}\left\{\mathbb{L}^{(s+l)}[n](\boldsymbol{s}) + \sum_{\sigma_k\cup\bar{\sigma}_{s-k}=\boldsymbol{s}} \mathbb{C}^{(s+l)}[n](\sigma_k)\langle \boldsymbol{v}\ \hat{n}(\bar{\sigma}_{s-k})\rangle\right\}, \tag{68}$$

where the operator $\mathbb{L}^{(s+l)}$ is defined according to (64), and $\mathbb{C}^{(s+l)}[n](\sigma_k)$ is given by the irreducible expression (65), where the set of $\boldsymbol{s}$ particles has been substituted by its k-element subset denoted by σ_k.

An analogous reformulation of the hierarchy equations is possible when considering the evolution of the correlation functions. Then in a natural way the correlation fields (60) and connected operators appear. The BBGKY hierarchy equations then have the following form

$$\frac{\partial h(\boldsymbol{s};t)}{\partial t} = \sum_{l=0}^{\infty}\left\{\mathbb{L}_c^{(s+l)}[n](\boldsymbol{s}) + \sum_{\sigma_k\cup\bar{\sigma}_{s-k}=\boldsymbol{s}} \mathbb{C}_c^{(s+l)}[n](\sigma_k)\boldsymbol{c}(|\sigma_{s-k})\right\}, \tag{69}$$

where

$$\mathbb{L}_c^{(s+l)}[n](\boldsymbol{s}) = \sum_{i=1}^{s} \int d(s+\boldsymbol{l}) \nabla_i \cdot \left[\{ (\boldsymbol{\mu F})_i (\boldsymbol{s} | s+\boldsymbol{l}) n(\boldsymbol{s}+\boldsymbol{l}; t) \}^c \right]^{irr} , \tag{70}$$

$$\mathbb{C}_c^{(s+l)}[n](\sigma_k) = \sum_{i \in \sigma_k} \int d(s+\boldsymbol{l}) \nabla_i \left[\{ \mathcal{C}_i (\sigma_k | s+\boldsymbol{l}) n(\sigma_k \cup s+\boldsymbol{l}; t) \}^c \right]^{irr} , \tag{71}$$

and $\boldsymbol{c}(\boldsymbol{r}|\sigma_{s-k})$ is defined by (60).

The operators $\mathbb{L}_c^{(s+l)}[n](\boldsymbol{s})$ and $\mathbb{C}_c^{(s+l)}[n](\sigma_k)$ introduced above are irreducible. By definition they do not contain connection lines. According to the reformulated equations, the evolution of an s-particle reduced distribution function or correlation function can therefore be expressed through irreducible, and what follows—local—operators and additional fields. These fields (67), or equivalently $\boldsymbol{c}(\boldsymbol{r}|\sigma)$, given by (60) contain all non-integrable expressions. This means that whenever a variable $\boldsymbol{R}_i$ is included in the set σ, this field contains (all present in the equations) non-integrable expressions which behave like $1/|\boldsymbol{r}-\boldsymbol{R}_i|^{\alpha}$, for $\alpha = 1, 2, 3$.

7 Conclusions

In this paper the statistical description of a system of hard particles immersed in a viscous fluid has been introduced. Assuming Stokesian particle dynamics, hierarchies of equations governing the time evolution of the reduced distribution functions and the correlation functions were derived. It was shown that all non-integrable expressions contained in these equations can be resummed to form physically meaningful fields, namely fields describing correlations between suspension velocity fluctuations and particle concentration fluctuations. This reformulation of the hierarchy equations is the basis on which a further analysis starts, the main aim of which will be to resolve the two coupled problems mentioned in the introduction—i.e. the derivation of the stationary particle distribution function and system-size dependence of velocity fluctuations. This will be done along the lines of [11], where the derivation of the Debye-Hückel distribution function was performed through an analysis of the equilibrium BBGKY hierarchy and a resummation of its non-integrable expressions. In fact a preliminary, although very promising result of this theoretical scheme, concerning monodisperse suspensions, has already been published [4].

Appendix A: Polydisperse Suspension

The derivation and analysis of the BBGKY hierarchy, as presented in this work is limited to the case of identical particles. It is nevertheless a straightforward task to generalize these results to the case when particles differ in size and density (i.e. the total external force). The only obstacle is a problematic and elaborate notation scheme, which must be introduced. Let us assume that all particles form K groups, such that within each group all particles are identical and characterized by a radius a_i, density ρ_i and number N_i, $i = 1, \ldots, K$. The difference in density of particles is important as in a gravitational field (the problem of sedimentation), it leads to a difference in forces acting on particles according to the formula

$$\boldsymbol{F}_i = \frac{4\pi a_i^3 (\rho_i - \rho) \boldsymbol{g}}{3} . \tag{72}$$

Now the definition of the reduced distribution function, as given by (35) and (36) must be modified to take into account that particles are distinguishable. In each group $i = 1, \dots, K$ of particles the s_i*-particle* microscopic density ($s_i = 1, \dots, N_i$) is defined by

$$\hat{n}^{(i)}(\boldsymbol{r}_1, \dots, \boldsymbol{r}_{s_i}) = \sum_{k_1,\dots,k_{s_i}}{}' \delta(\boldsymbol{r}_1 - \boldsymbol{R}^{(i)}_{k_1}) \cdots \delta(\boldsymbol{r}_{s_i} - \boldsymbol{R}^{(i)}_{k_{s_i}}), \tag{73}$$

where $k_1, \dots, k_{s_i} = 1, \dots, N_i$ and $\boldsymbol{R}^{(i)}_1, \dots, \boldsymbol{R}^{(i)}_{N_i}$ are the positions of particles of the i-th group. Then the generalized reduced distribution function can be defined as

$$n(\boldsymbol{r}^{(1)}_1, \dots, \boldsymbol{r}^{(1)}_{s_1}, \dots, \boldsymbol{r}^{(1)}_1, \dots, \boldsymbol{r}^{(K)}_{s_K}; t) = \langle \hat{n}^{(1)}(\boldsymbol{r}^{(1)}_1, \dots, \boldsymbol{r}^{(1)}_{s_1}) \dots \hat{n}^{(K)}(\boldsymbol{r}^{(K)}_1, \dots, \boldsymbol{r}^{(K)}_{s_K})\rangle. \tag{74}$$

These functions have the following interpretation:
$n(\boldsymbol{s}_1, \boldsymbol{s}_2, \dots, \boldsymbol{s}_K; t) d\boldsymbol{s}_1 \cdots d\boldsymbol{s}_k$ is the average number of ensembles of s_1 particles of kind 1, s_2 particles of kind 2, etc in a configuration $d\boldsymbol{s}_1$ around $\boldsymbol{s}_1$, $d\boldsymbol{s}_2$ around $\boldsymbol{s}_2$, etc. Here the abbreviated notation $\boldsymbol{s}_i \equiv \{\boldsymbol{r}^{(i)}_1, \dots, \boldsymbol{r}^{(i)}_{s_i}\}$ has been used.

The time evolution equation of the functions (74) may be obtained directly from the hierarchy of (39) once on the right-hand side, in expressions where additional particles are considered, the operation

$$\sum_{l=1}^{\infty} \frac{1}{l!} \int d(s + \boldsymbol{l}), \tag{75}$$

is substituted by

$$\sum_{l_1=1}^{\infty} \sum_{l_2=1}^{\infty} \dots \sum_{l_K=1}^{\infty} \frac{1}{l_1!} \frac{1}{l_2!} \cdots \frac{1}{l_K!} \int d(s_1 + \boldsymbol{l}_1) \cdots d(s_K + \boldsymbol{l}_K). \tag{76}$$

Then all further properties, such as the connectivity and reducibility follow.

Appendix B: Diagrammatic Technique

B.1 Definitions

In many cases it is convenient to represent the scattering sequences and correlation structures in terms of diagrams. We will introduce this technique by means of examples based on terms existing in the hierarchy equations. Notation used in a recent work [28] will be adopted:

1. A horizontal line - - - - corresponds to a particle (particle line) and is numbered on the left.
2. A term of the scattering sequence of a hydrodynamic operator begins (looking from the right) with the superposition $\boldsymbol{Z}_0 \boldsymbol{\mu}_0 \boldsymbol{F}$ or $-\hat{\boldsymbol{Z}}_0 \boldsymbol{v}_0$. These will be represented by diagram elements represented by Fig. 1a and Fig. 1b respectively.
3. A circle represents one-particle friction operators. Depending on its position in the scattering sequence, it may denote the superposition $\boldsymbol{\mu}_0 \boldsymbol{Z}_0$ or the operator $-\hat{\boldsymbol{Z}}_0$. In general, all diagrams used here end with $\boldsymbol{\mu}_0 \boldsymbol{Z}_0$ as it is always the velocity, which is of interest.
4. Each Green operator is represented by a single vertical line |. The hydrodynamic structure is formed by a sequence of these operators, where at each circle representing the operator $-\hat{\boldsymbol{Z}}_0$, one line enters tangent from the right and one leaves at the left. A hydrodynamic sequence is always connected and forms an ordered structure.

5. The divergence over a particles position variable is denoted by a rotated nabla symbol at the beginning of a particle line.
6. A filled symbol at the beginning of a given particle line denotes the operation of integration over that particles position variable.
7. Particle lines of correlated particles are connected by a double vertical lines ||. A node of the correlation function is depicted by a small circle between the double vertical lines and at a particles line.

For example the correlation-scattering structure

$$\int d3\nabla_2 \cdot \boldsymbol{\mu}_0(2)\boldsymbol{Z}_0(2)\boldsymbol{G}(21)\hat{\boldsymbol{Z}}_0(1)\boldsymbol{G}(13)\boldsymbol{Z}_0(3)\boldsymbol{\mu}_0(3)\boldsymbol{F}_3\, h(12), \tag{77}$$

is represented by the diagram (2), whereas the structure

$$\int d3\nabla_2 \cdot \boldsymbol{\mu}_0(2)\boldsymbol{Z}_0(2)\boldsymbol{G}(21)\hat{\boldsymbol{Z}}_0(1)\boldsymbol{G}(13)\hat{\boldsymbol{Z}}_0(3)\boldsymbol{G}(32)\hat{\boldsymbol{Z}}_0(2)\boldsymbol{v}_0\, h(23), \tag{78}$$

being part of the scattering sequence of $\boldsymbol{\mathcal{C}}_2\boldsymbol{v}_0$ is represented by the diagram found in Fig. 3.

B.2 Diagrams Representing BBGKY Hierarchy Equation

The diagrammatic representation of the right-hand side of the BBGKY hierarchy equation for the time evolution of an s-particle reduced distribution function is a sum of all graphs of the following class:

- containing s numbered particle-lines without integration, one of which begins with a divergence symbol.
- containing an arbitrary ($l \geq 0$) number of particle lines which begin with a black symbol.
- particle lines are connected by a sequence of Green propagators (hydrodynamic structure) represented by vertical lines.

Fig. 1

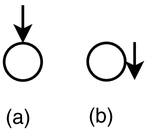


Fig. 2 Diagram representing structure (77)

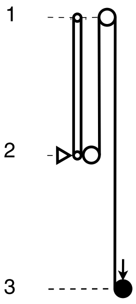
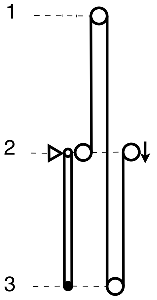


Fig. 3 Diagram representing structure (78)



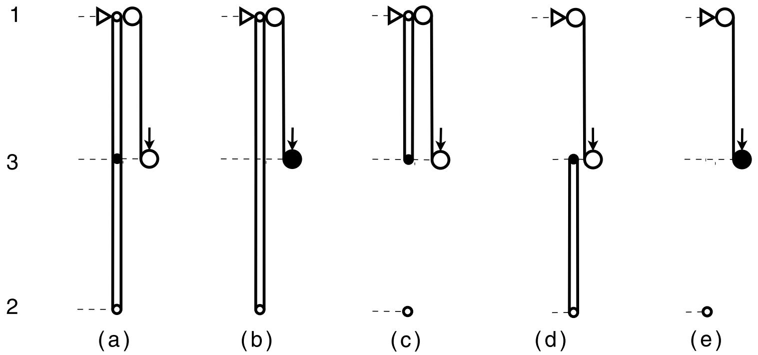


Fig. 4 Diagram representing structure (79)

- the correlation structure, represented by an arbitrary configuration of double vertical lines might introduce additional particle lines without integration not contained in the hydrodynamic structure.
- all diagrams begin (looking form the right) with one of the symbols depicted at Fig. 1.

The BBGKY hierarchy consists of all such, topologically non-equivalent diagrams. Whenever a suitable numbering scheme for particle lines beginning with a filled symbol is chosen, the combinatoric weight $1/l!$ falls out of the equations, as only one representative, out of $l!$ possible permutations is chosen (topologically non-equivalent). A inherent numbering procedure is provided by the structure of the hydrodynamic sequence itself. As no disjoint clusters may form it, it uniquely orders all the particle lines according to their first occurrence in that sequence.

B.3 Connected Expressions

Consider as an example the following correlation-scattering structure:

$$\int d3\nabla_1 \cdot \boldsymbol{\mu}_0(1)\boldsymbol{Z}_0(1)\boldsymbol{G}(13)\boldsymbol{Z}_0(3)\boldsymbol{\mu}_0(3)\boldsymbol{F}_3 n(123). \tag{79}$$

According to the expansion of $n(123)$ in terms of correlation functions (46) this structure is equal to the sum of diagrams found on Fig. 4a through Fig. 4e.

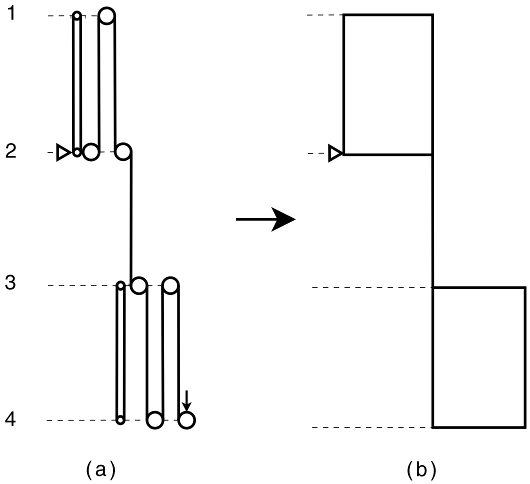


Fig. 5 Diagram representing structure (81)

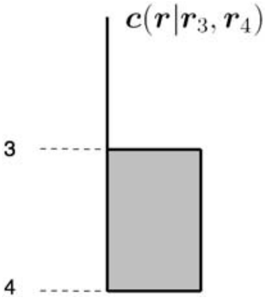


Fig. 6 Diagram representing the structure of the correlation field $\boldsymbol{c}(\boldsymbol{r}|\boldsymbol{r}_3, \boldsymbol{r}_4)$

Connected diagrams are Fig. 4a–d. Indeed

$$\begin{aligned}&\{\boldsymbol{\mu}_0(1)\boldsymbol{Z}_0(1)\boldsymbol{G}(13)\boldsymbol{Z}_0(3)\boldsymbol{\mu}_0(3)\boldsymbol{F}_3 n(123)\}^{(c)}\\&\quad=\boldsymbol{\mu}_0(1)\boldsymbol{Z}_0(1)\boldsymbol{G}(13)\boldsymbol{Z}_0(3)\boldsymbol{\mu}_0(3)\boldsymbol{F}_3(h(123)+h(13)h(2)+h(23)h(1)).\end{aligned}\tag{80}$$

In general, a non-connected expression always consists of a connected part, which holds the hydrodynamic structure, and the rest, which contains only correlation functions, and which itself might by connected or non-connected.

The BBGKY hierarchy equations for the correlation functions consist of a subclass of diagrams (Sect. B.2) which are connected.

B.4 Connection Lines and Reducibility

In the structure

$$\begin{aligned}&\nabla_2\cdot\boldsymbol{\mu}_0(2)\boldsymbol{Z}_0(2)\boldsymbol{G}(21)\hat{\boldsymbol{Z}}_0(1)\boldsymbol{G}(12)\hat{\boldsymbol{Z}}_0(2)\ \boldsymbol{G}(23)\\&\quad\times\hat{\boldsymbol{Z}}_0(3)\boldsymbol{G}(34)\hat{\boldsymbol{Z}}_0(4)\boldsymbol{G}(43)\hat{\boldsymbol{Z}}_0(3)\boldsymbol{G}(34)\\&\quad\times\boldsymbol{Z}_0(4)\boldsymbol{\mu}_0(4)\boldsymbol{F}_4\ h(12)h(34),\end{aligned}\tag{81}$$

presented in graphical form in Fig. 5a, the operator $\boldsymbol{G}(23)$ is a connection line.

As this expression contains a connection line it is said to be reducible. It can be depicted in a block-diagram form, as done on Fig. 5b. Notice that the structure formed by neglecting the expression on the left of the connection line is part of the correlation field $\boldsymbol{c}(\boldsymbol{r}|\boldsymbol{r}_3, \boldsymbol{r}_4)$. Indeed, this field is constructed as a sum of all diagrams of the form depicted in Fig. 6. Here the filled block represents an arbitrary number of particles, which are integrated over.

The distance dependence of connection lines is due to properties of the single-particle operators which stand at the beginning and at the end of it. Whenever a Green operator connects two $\boldsymbol{Z}_0$ operators, it behaves asymptotically as $1/r$ with the interparticle distance r. When at one end of a connection line the operator $\boldsymbol{Z}_0$ is present, whereas at the other, the convection-friction operator $\hat{\boldsymbol{Z}}_0$, the Green operator decays asymptotically as $1/r^2$. Finally when a connection line connects two $\hat{\boldsymbol{Z}}_0$ operators, as is the case in the structure (5), is behaves as $1/r^3$ because the multipole elements with $l = 1, \sigma = 0, 1$ are then equal to zero. Due to this property, whenever two Green operators connect clusters of particles, the interaction between those clusters decays at least as fast as $1/r^4$. Therefore only single Green operators lead to non-integrable interactions.

References

1. Balescu, R.: Equilibrium and Non-equilibrium Statistical Mechanics. Wiley, New York (1975)
2. Batchelor, G.K.: Sedimentation in a dilute polydisperse system of interacting spheres. Part 1. General theory. J. Fluid Mech. **119**, 379 (1982)
3. Batchelor, G.K., Wen, C.: Sedimentation in a dilute polydisperse system of interacting spheres. Part 2. Numerical results. J. Fluid Mech. **124**, 495 (1982)
4. Cichocki, B., Sadlej, K.: Steady-state particle distribution of a dilute sedimenting suspension. EuroPhys. Lett. **72**, 936 (2005)
5. Brenner, M.P., Mucha, P.J.: That sinking feeling. Nature **409**, 569 (2001)
6. Caflisch, R.E., Luke, J.H.C.: Variance in the sedimentation speed of a suspension. Phys. Fluids **28**, 759 (1985)
7. Cichocki, B., Ekiel-Jeżewska, M.L., Szymczak, P., Wajnryb, E.: Three-particle contribution to sedimentation and collective diffusion in hard-sphere suspensions. J. Chem. Phys. **117**, 1231 (2002)
8. Cichocki, B., Felderhof, B.U., Hinsen, K., Wajnryb, E., Awzdziewicz, J.B.: Friction and mobility of many spheres in stokes flow. J. Chem. Phys. **100**, 3780 (1994)
9. Cichocki, B., Jones, R., Kutteh, R., Wajnryb, E.: Friction and mobility for colloidal spheres in Stokes flow near a boundary: The multipole method and applications. J. Chem. Phys. **112**, 2548 (2000)
10. Cunha, F.R., Abade, G.C., Sousa, A.J., Hinch, E.J.: Modeling and direct simulation of velocity fluctuations and particle-velocity correlations in sedimentation. J. Fluids Engn. **124**, 957 (2002)
11. Falkenhagen, H.: Theorie der Elektrotyle. S. Hirzel, Leipzig (1971)
12. Felderhof, B.U., Jones, R.B.: Convective motion and transfer of force by many-body hydrodynamic interactions. Physica A **146**, 404 (1987)
13. Glauberman, A.E.: K teorii sistem elektricheski zaryashennykh chastits. Doklady Akad. Nauk S.S.S.R. **78**, 883 (1951)
14. Glauberman, A.E., Yukhnovskii, I.P.: K statisticheskoi teorii kontsentrirovannykh rastvorov sil'nykh elektolitov. i, ii. Zhur. Eksptl. Teoret. Fiz. **22**, 562 (1952)
15. Glauberman, A.E., Yukhnovskii, I.P.: Binarnaya funktsiya raspredeleniya dlya sistem vzaimodeistvuyshikh zaryashennikh chastits. i. Zhur. Eksptl. Teoret. Fiz. **27**, 690 (1954)
16. Guazzelli, E.: Evolution of particle-velocity correlations in sedimentation. Phys. Fluids **13**, 1537 (2001)
17. Huang, K.: Statistical Mechanics. Wiley, New York (1987)
18. Janosi, I.M., Tel, T., Wolf, D.E., Gallas, J.A.C.: Chaotic particle dynamics in viscous flows: The three-particle stokeslet problem. Phys. Rev. E **56**, 2858 (1997)
19. Kim, S., Karilla, S.J.: Microhydrodynamics: Principles and Selected Applications. Dover Publications, Inc., Mineola (2005)
20. Koch, D.L., Shaqfeh, E.S.G.: Screening in sedimenting suspensions. J. Fluid Mech. **224**, 275 (1991)
21. Ladd, A.J.C., Nguyen, N.Q.: Microstructure in a settling suspension of hard spheres. Phys. Rev. E **69**, 050,401 (2004)
22. Ladd, A.J.C., Nguyen, N.Q.: Sedimentation of hard-sphere suspensions at low reynolds number. J. Fluid Mech. **525**, 73 (2005)

23. Mazur, P., Bedeaux, D.: Generaization of faxen's theorem to nosteady motion of a sphere through an incompressible fluid in arbitrary flow. Physica **76**, 235 (1974)
24. Mazur, P., van Saarlos, W.: Many-sphere hydrodynamic interactions and mobilities in a suspension. Physica A **115**, 21 (1982)
25. Mucha, P.J., Brenner, M.P.: Diffusivities and front propagation in sedimentation. Phys. Fluids **15**, 1305 (2003)
26. Segré, P.N., Herbolzheimer, E., Chaikin, P.M.: Long-range correlations in sedimentation. Phys. Rev. Lett. **79**, 2574 (1997)
27. Szymczak, P., Cichocki, B.: Memory effects in collective dynamics of brownian suspensions. J. Chem. Phys. **121**, 3329 (2004)
28. Szymczak, P., Cichocki, B.: Diagrammatic approach to response problems in composite systems. J. Stat. Mech. (2008). P01025