Steady state particle distribution of a dilute sedimenting suspension

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Sedimentation of a non-Brownian suspension of hard particles is studied. It is shown that in the low concentration limit a two-particle distribution function ensuring finite particle correlation length can be found and explicitly calculated. The sedimentation coefficient is computed. Results are compared with experiment.

I. INTRODUCTION

In recent years the familiar problem of sedimentation of a non-Brownian suspension of particles has gained interest as new insight into the phenomenon is gathered. Despite vivid research in the field, some questions still remain intriguing puzzles. Even when considering observation time scales and dimension regimes, which sanction the neglect of Brownian motion, the dynamics show to be many-body and highly chaotic leading to astonishing consequences.

One of the main, still open problems, is the determination of the particle probability distribution function for a steadily sedimenting suspension. It was first challenged by Batchelor [1]. In order to derive an equation for the pair-distribution function he considered a polidisperse suspension. Taking then the limit of identical particles, a particle distribution function for monodisperse suspensions could be derived. But these results where shown to be ambiguous (depending on the way the limit is taken) and therefore doubtful [1], [2], [3]). The problem of particle distribution is often skinned over by the assumption, that the particles are randomly distributed in the fluid. Indeed this will be the case, if Brownian motions play a significant role in the dynamics of the system, creating a randomising mechanism. But whenever this type of stochastic motion is absent, there seems to be no justification for such assumptions. Further, it has been [2] pointed out, that a equilibrium (equal probability) distribution of particles results in characteristics of a non-Brownian suspension, which have not been detected in experiments [2]. Moreover it can actually be explicitly shown that such a distribution cannot be a solution of the Liouville equation incorporating full multi-body interactions, therefore falling completely out of consideration.

In this work we propose a way to tackle this issue. The derivation of the statistical properties of the system will be based on the conjecture that correlations in steady state must be of finite length. This assumption, originating in the basic ideas of statistical mechanics, has profound consequences. It, as will be shown, leads to an effective equation for the pair distribution function. Truncation of long range hydrodynamic interactions will be achieved through a screening mechanism similar in spirit to that introduced by Koch & Shaqfeh [3].

II. BBGKY HIERARCHY

The system under consideration is a suspension of $N$ spheres of equal radius $a$ immersed in an incompressible fluid of shear viscosity $\eta$. In the regime studied the particle Reynolds number is assumed to be small enough to treat inertial effects as virtually absent. Moreover the Peclet number describing the relative influence of hydrodynamic effects to Brownian motion, is presumed to be large. On those assumptions and on the laboratory time scales imposed the fluid motion is governed by the stationary Stokes equation. The dynamics of the particles are therefore given by the equations of motion

$$\frac{dR_i}{dt} = U_i = \sum_{j=1}^{N} \mu_{ij}(X) F_j$$

(1)

where $\mu$ is the translational part of the mobility matrix, principally a matrix function of the configuration $X = (R_1, \ldots, R_N)$ of all the particles (if not indicated otherwise, dimensionless - normalised to $2a$ - distances will be used). It describes the hydrodynamic interactions between particles and connects the forces acting on particles to the velocities they acquire in a given configuration. The forces $F_j$ will be assumed to be constant and equal $F$ for all particles.

The particle distribution function satisfies the Liouville equation, which for the system considered, has the following structure

$$\frac{\partial \rho(X,t)}{\partial t} + \sum_{i,j} \nabla_i \cdot [\mu_{ij}(X) F \rho(X,t)] = 0$$

(2)

The mobility matrix can be found following a formal procedure presented for example in ref. [3]. Summarising, in order to calculate the hydrodynamic interaction, the boundary conditions on the particles surfaces are substituted by induced force densities introduced for each particle. These force densities, induced by fluid flow, produce subsequent fluid velocity fields which are propagated and reflect off other particles and thereby cause interactions. In consequence, the mobility matrix can
be expressed as a scattering sequence equivalent to a superposition of all these reflections. This sequence contains one-particle scattering operators, which construct induced force densities on particles given velocity fields around them, and Green operators which propagate the influence of a force density concentrated on a given particle, resulting in fluid velocity fields around other particles. In an unbounded, infinite system, the Green operator is the Oseen tensor.

The full \(N\)-body mobility matrix can therefore be expanded in terms of the number of particles which enter each term of the scattering sequence. This so called cluster expansion has the structure

\[
\mu_{11}(X) = \mu_{11}^{(1)}(1) + \sum_{i=2}^{N} \mu_{11}^{(i)}(1) + \ldots
\]

\[
\mu_{12}(X) = \mu_{12}^{(2)}(12) + \sum_{i=3}^{N} \mu_{12}^{(i)}(12i) + \ldots
\]

where \(\mu_{ij}^{(s)}(1, \ldots, s)\) denotes all the terms of the scattering sequence of \(\mu\) which involve only, but all the particles \(\{1, \ldots, s\}\).

Next we introduce reduced distribution functions

\[
n(1, 2, \ldots, s) = \frac{N!}{(N-s)!} \int d(s+1) \cdots dN \rho(X)
\]

These functions represent the average densities of pairs, triplets, etc. of particles in given configurations.

An integration of the Liouville equation in all the variables except the first \(s\), together with the cluster expansion of the hydrodynamic interactions, leads to an infinite hierarchy of equations governing the time evolution of the reduced distribution functions. This is the analogue of the BBGKY (Bogolubov-Born-Green-Kirkwood-Yvon) hierarchy introduced in the kinetic theory of gases. It should be considered in the thermodynamic limit when the number of particles and the volume of the system go to infinity, while the density is kept constant. The equations for the two and three particle distribution functions have the structure

\[
\frac{\partial n(12)}{\partial t} = -\sum_{i,j=1,2} \nabla_i \cdot \mu_{ij}(12) F n(12)
\]

\[
- \int d^3 x \sum_{i,j=1,2} \nabla_i \cdot [\mu F]_{ij}(12; 3) n(123) + \ldots
\]

where for example

\[
[\mu F]_{11}(12; 3) = [\mu_{11}^{(2)}(13) + \mu_{13}^{(2)}(13) + \sum_{j=1}^{3} \mu_{1j}^{(3)}(123)] F
\]

and

\[
\frac{\partial n(123)}{\partial t} = -\sum_{i,j=1}^{3} \nabla_i \cdot \mu_{ij}(123) F n(123) + \ldots
\]

where in both equations, terms depending on higher-order distribution functions have been omitted.

The reduced distribution functions factorize for groups of particles that are far away from each other. Therefore these functions can be expanded in terms of correlation functions according to

\[
n(12) = h(1)h(2) + h(12)
\]

\[
n(123) = h(1)h(2)h(3) + h(12)h(3) + h(13)h(2) + h(23)h(1) + h(123)
\]

where \(h(1 \ldots s)\) is a \(s\)-particle correlation function which vanishes whenever any subset of particles is dragged away from the rest. If the correlations in a system are to be of finite length, which will be assumed, the correlation functions must decay faster then the inverse of the inter-particle distance cubed.

Using the expansion, the BBGKY hierarchy for the reduced distribution functions can be transcribed into a hierarchy of equations for the time evolution of the successive correlation functions. An analysis of these equations shows that if system configuration is such that particles form two uncorrelated clusters, some expressions on the r.h.s., formulated in terms of the scattering sequence, contain single Green operators (so called connection or articulation lines) joining these two groups of particles. Such terms result in long range contributions because a solitary Green operator includes slowly decaying parts proportional to \(1/r^\gamma\) where \(r\) is the relative distance between the groups and \(\gamma = 1, 2, 3\). The evolution of a correlation function is therefore given by an equation which contains long range terms and is therefore non-local in space. This structure of the hierarchy equations is inconsistent with the finite correlation length assumed for the correlation functions. As will be shown in the next section, considering the low concentration limit a particle distribution can be found, which cancels out all long range terms in the hierarchy, thereby saving finite correlation length.

### III. LOW CONCENTRATION LIMIT PAIR DISTRIBUTION FUNCTION

In the low concentration limit the dominating part of a \(s\)-particle reduced distribution function is proportional to \(n^s\). Consequently, the first terms of the hierarchy equations have a well established order in density if the concentration of particles is low.

Consider the equation giving the time evolution of the two-particle correlation function. It is derived through a substitution of \(\bar{\mu}\) into \(\mu\). In the lowest order of density it vanishes due to the relation

\[
\sum_{i,j=1,2} \nabla_i \cdot \mu_{ij}(12) = 0.
\]

which is a consequence of the isotropic nature of the hydrodynamic interactions. The two-particle equation
yields therefore in the dilute limit no condition for the two-particle correlation function. This peculiarity was also encountered by Batchelor [1]. His solution was based on the analysis of a polidisperse suspension where the two-particle equation yields a condition, but was shown to be ambiguous (1, 2, 3). We turn our attention to the next hierarchy equation, i.e. the third equation which emerges when $µ$ is addressed to $µ$, truncated to the terms proportional to the density cubed.

As pointed out, the r.h.s of this equation contains long range terms. If all three particles are far away from each other, both sides of the equation contain only shot range terms. But consider the case when one particle (e.g. the particle with index 1) is far away from a close pair of particles (consequently particles with indices 2 and 3). All the terms, which in such a configuration lead to long range contributions can be identified and singled out resulting in the expression

\[ n \nabla_2 \cdot \left[ (\mu_{23}(2|1) + \mu_{21}(23|1) + \mu_{21}(2|3)n(23)) + n \nabla_3 \cdot (\mu_{31}(3|1) + \mu_{31}(32|1) + \mu_{31}(3|2)1)n(23) \right] \]

where each vertical line stands for a single Green operator connecting uncorrelated particles. For example $\mu_{21}(23|1)$ stands for the sum of all terms of the scattering sequence of $\mu_{21}(123)$ (123) that contain only a single Green operator connecting particle 1 and the group (2,3).

In what follows, we will show that there exists a two-particle probability distribution function which cancels out these long range terms and therefore leads to finite correlation length.

In equation (10) the long range contributions appear in the Green operators binding particle 1 with the group consisting of particles 2 and 3. Their explicit form is retrieved through an analysis of the multipole matrix elements of the Green operators, which can be found e.g. in [4].

When taking into account the symmetry relations upon exchange of particles, and a change of variables where $r$ becomes the vector joining the distant particle and the center of the group of close particles, while $R$ the vector denoting the relative position of the two particles within the group, we reach the conclusion that most of the long range terms cancel out automatically - only the multipoles proportional to the inverse of the inter-particle distance survive.

A further expansion in terms of $R = |R| \, \langle R \ll r \rangle$ shows that only terms proportional to $1/r^2$ remain. The equation which emerges has an identical structure to the one derived by Batchelor&Green [11] and describing the relative motion of a pair of particles in a shear flow. The resulting particle distribution function $n(R) = n^2 g(R)$ is isotropic. It is given by the solution of the differential equation

\[ \left\{ (1-A) \frac{d}{dR} \left[ \frac{3(A-B)}{R} - \frac{dA}{dR} \right] \right\} = 0 \]

where $\alpha_{ij}^{(2d)}$ and $\beta_{ij}^{(2d)}$ are functions appearing in the explicit form of the generalised two-particle mobility matrix connecting the translational velocity of the particles to the stresslet [11]. Since $\lim_{R \to \infty} g(R) = 1$

\[ g(R) = \frac{1}{1-A} \exp \left[ \int_{R}^{\infty} \frac{3(B-A)}{R(1-A)} dR \right] \]

Figure 1: The two-particle reduced distribution function $g(R)$ satisfying the finite correlation length criterion.

Note that this solution is independent of the direction distinguished by the external force field, the effects of which are in the low concentration limit completely dispersed by the isotropic hydrodynamic interactions. Further, to verify the consistency of the reasoning imposed, it can be shown that the two-particle distribution function derived guarantees convergence of all terms up to order $n^3$ in equation (6) governing the time evolution of the two-particle reduced distribution function.

\[ A \cdot \text{Stationary particle distribution function} \]

The functions $A(R)$ and $B(R)$ can be expressed as series in inverse powers of inter-particle distance [11]. In numerical approximations of the integral these sequences are truncated at 1000 and 800 terms respectively. lubrication corrections and far limit asymptotic with $g(R) - 1 \approx 0.1953/R^6$ are taken into account. The computed function $g(R)$ is plotted at fig. 11. Further, the structure factor at $k = 0$ is calculated: $S(0) = 1 - 1.64\phi$.

\[ \text{B. Sedimentation coefficient} \]

In the low concentration limit the average sedimentation velocity $U$ of a particle in the suspension measured relative to the Stokes velocity $U_0$ may be expanded in a series in the powers of volume fraction $\phi = 4\pi a^3 n/3$.

\[ U = U_0 + \phi K + \mathcal{O}(\phi^2) \]
Figure 2: Dimensionless sedimentation velocity as a function of volume fraction - experimental results after Hanratty et al. [14]. Points represent experimental data and the solid curve is a fit. The solid line represents the sedimentation coefficient $K = -3.87$, whereas the dotted line is the coefficient for a suspension in equilibrium with $K = -6.55$.

The linear coefficient $K$ can be expressed in terms of a microscopic expression [13] involving two-body mobility matrices and the pair distribution function.

$$K = K_0 + \frac{2}{\pi \mu_0} \int_{R_{12} \geq 1} (g(12) - 1) \text{Tr} \left[ \sum_{i=1}^{2} \mu_i^{(2)}(12) \right] dR_{12}$$

where $K_0 = -6.546$ [13, 12] is the sedimentation coefficient for an equilibrium distribution of hard spheres and $\mu_0$ is the mobility of a single sphere. Dimensionless distance $R_{12}$ normalised to $2a$ is adopted. The integral can be interpreted as a correction emerging due to the change of the distribution away from equilibrium. A numerical calculation yields the result $K = -3.87$. It is compared with experimental data of Hanratty et al. on fig. 2. A fit to the experimental data of Ham et al. [15] leads to the sedimentation coefficient equal to $-3.9$, in excellent agreement with the calculated value.

V. CONCLUDING REMARKS

We have shown that a mono-disperse suspension of hard, non-Brownian particles can develop a micro-structure which insures finite correlation length. A scheme based on the formal schemes of non-equilibrium statistical mechanics and a detailed analysis of hydrodynamic interaction was used in the low-concentration regime to derive an equation for the two-particle distribution function. The solution shows to be isotropic due to the domination of the isotropic hydrodynamic interactions. The mechanism which insures the cutoff of long range correlations can be interpreted as hydrodynamic screening in the sense that the micro-structure of the suspension arranges itself in a way which truncates long range parts of the interactions. A close pair of particles feels the influence of a third distant particle through an effective shearing flow. Screening leads to a configuration of the close pair, which hinders the effect of the distant particle. The idea, that a suspension might develop a micro-structure which changes the characteristics of it, was introduced by Koch & Shaqfeh [6] upon the analysis of the problem of diverging velocity fluctuation.

Further, the sedimentation coefficient was calculated. The value $-3.87$ was found. It agrees very well with the experimental results cited.

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