Generalization of Clausius-Mossotti approximation in application to short-time transport properties of suspensions

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Abstract

In 1983 Felderhof, Ford and Cohen gave microscopic explanation of the famous Clausius-Mossotti formula for the dielectric constant of nonpolar dielectric. They based their considerations on the cluster expansion of the dielectric constant, which relates this macroscopic property with the microscopic characteristics of the system.

In this article, we analyze the cluster expansion of Felderhof, Ford and Cohen by performing its resummation (renormalization). Our analysis leads to the ring expansion for the macroscopic characteristic of the system, which is an expression alternative to the cluster expansion. Using similarity of structures of the cluster expansion and the ring expansion, we generalize (renormalize) the Clausius-Mossotti approximation. We apply our renormalized Clausius-Mossotti approximation to the case of the short-time transport properties of suspensions, calculating the effective viscosity and the hydrodynamic function with the translational self-diffusion and the collective diffusion coefficient. We perform calculations for monodisperse hard-sphere suspensions in equilibrium with volume fraction up to 45%. To assess the renormalized Clausius-Mossotti approximation, it is compared with numerical simulations and the Beenakker-Mazur method. The results of our renormalized Clausius-Mossotti approximation lead to comparable or much less error (with respect to the numerical simulations), than the Beenakker-Mazur method for the volume fractions below $\phi \approx 30\%$ (apart from a small range of wave vectors in hydrodynamic function). For volume fractions above $\phi \approx 30\%$, the Beenakker-Mazur method gives in most cases lower error, than the renormalized Clausius-Mossotti approximation.

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I. INTRODUCTION

Einstein was the first, who applied statistical physics to calculate the viscosity of suspension [1]. Having in mind nanometer size sugar molecules immersed in water, he considered a model of sufficiently big spherical particles immersed in viscous liquid. Experiments show, that in this case the observed viscosity increases [2]. In his work, Einstein related the observed (effective) viscosity $\eta_{\text{eff}}$ of suspension with its structure on the microscopic level. His result, $\frac{\eta_{\text{eff}}}{\eta} = 1 + \frac{5}{2}\phi$ - where $\eta$ denotes the viscosity of solvent and $\phi$ denotes the volume fraction of the system - is valid only for dilute suspensions. This limitation is caused by the assumption, that the particles immersed in fluid do not influence their mutual motion. The problem of the influence of the particles on their mutual motion is essential to go beyond the diluted regime and was already addressed by Smoluchowski.

His analysis for two sedimenting spheres leads to the following conclusions. The two spheres sediment faster than a single one. Moreover, the velocities of both spheres are deviated from the direction of the gravity field, as shown in the figure. This example clearly demonstrates, that two sedimenting particles in gravity field behave differently than a single particle, because a single particle would sediment vertically downward. Despite the fact, that there are no direct forces between the particles, they influence their motion. This "interaction" of the immersed particles is mediated by fluid and is called the "hydrodynamic interaction".

Apart from the considerations for the finite number of particles, Smoluchowski analyzed also an infinite set of particles. He concluded, that behavior of suspension strongly depends on the shape of the system. The shape matters, even if its boundaries are extended to infinity. In other words, Smoluchowski identified the problem of long-range hydrodynamic interactions. Another important feature of the hydrodynamic interactions is their many-body character. Motion of three particles cannot be described as a superposition of the two-particle characteristics. Similar holds for larger number of particles. In general, many-body characteristics are needed in the macroscopic considerations for suspensions. From the perspective of transport properties, even the two-body hydrodynamic interactions are problematic. Analysis of the two-body problem reveals, that two particles at a small distance in incompressible, viscous fluid, strongly "interact" hydrodynamically. In order to keep
constant velocity of the approaching particles, asymptotically an infinite force is needed [3].

Extension of Einstein’s analysis for more concentrated systems appeared to be difficult, because of the long-range hydrodynamic interactions. One of the first successful approaches was made by Saito [4], who obtained the following formula for the effective viscosity, \( \eta_{\text{eff}}/\eta = (1 + 3\phi/2)/(1 - \phi) \). Saito took the hydrodynamic interactions into consideration partially. He also discussed the long-range character of the hydrodynamic interactions and strongly emphasized difficulties unsolved at that time [5]. The first systematic extension of Einstein’s work for more concentrated suspensions was performed by Peterson and Fixman [6]. They obtained a virial expansion of the effective viscosity up to the second order, which includes the two-body hydrodynamic interactions. It was the first approach, in which the transport coefficient in the second order was given by absolutely convergent integrals. Despite of this success, they did not express the transport coefficients by absolutely convergent integrals for higher orders of virial expansion. Therefore, the problem with long-range hydrodynamic interactions was still not solved at that time. Solution came with the work of Felderhof, Ford and Cohen in 1982 [7]. The above authors considered a dielectric system, but their analysis can be directly carried over to the physics of suspensions. They proved, that the dielectric constant is a local quantity, which does not depend on the shape of the system. Their idea is related to the Brown’s approach, who obtained similar result limited to the lowest terms in the single-particle polarizability expansion of a dielectric constant [8]. Felderhof, Ford and Cohen also gave the microscopic explanation of the famous Clausius-Mossotti formula [9], which is an analog of the Saito formula [4] for the effective viscosity in the physics of suspensions. It is worth mentioning here the effective medium approaches [10, 11] and their extensions including the two-body hydrodynamic interactions in a more accurate way [12, 13].

Nowadays, the most prominent statistical physics approach to the short-time transport properties of suspensions is the Beenakker-Mazur method [14–16], which was developed and applied for different suspensions [17–22]. The method gives reasonable results for a wide range of volume fractions, but it does not take the two-body hydrodynamic interactions fully into account. It is known from virial expansion [23–25] and from numerical simulations, [26, 27] that the two-body hydrodynamic interactions of close particles are essential to grasp the dynamics of the system. Therefore, there is still an open problem in the physics of suspensions: formulation of a systematic method, which would take the two-body hydro-
dynamic interactions fully into consideration and which would give reasonable results for at least the intermediate volume fractions, say $\phi \approx 25\%$. Systematic consideration of the two-body hydrodynamic interactions in the Beenakker-Mazur method is difficult, because the method relies on the expansion of the transport properties in the series of the so-called renormalized fluctuations. This series expansion is then truncated in the second order in the fluctuations. To consider the full two-body hydrodynamic interactions in the Beenakker-Mazur expansion, one needs summation of all orders in the series, which is impossible in practice.

In this article, we develop the approach of Felderhof, Ford and Cohen. As mentioned above, they introduced the cluster expansion of the macroscopic characteristics of dispersive media such as e.g. the polarizability of dielectric and the effective viscosity of suspension [7]. Felderhof, Ford and Cohen also gave the microscopic explanation of the Clausius-Mossotti formula for dielectrics (related to the Saito formula in case of suspensions) [9]. Their cluster expansion is a starting point of this article. We perform a rigorous analysis of the cluster expansion leading to a formula, which we call ring expansion of the macroscopic characteristics. We also generalize the Clausius-Mossotti approximation, basing on a similarity between the Felderhof, Ford and Cohen’s cluster expansion and the ring expansion introduced in this article. Using the generalized Clausius-Mossotti approximation, we calculate the effective viscosity and the hydrodynamic function (with the translational short-time self-diffusion and the collective diffusion coefficient) for suspension of monodisperse hard-spheres in equilibrium.

The generalization (renormalization) of the Clausius-Mossotti approximation based on the ring expansion introduced in this article is motivated by the results of the virial expansion for the effective viscosity and the sedimentation coefficients [24, 25]. One of the dominant contributions to the virial expansion for the sedimentation coefficient on the three-body level comes from the terms with a virtual overlap of spheres. The idea of resummation of the above terms with a virtual overlap of spheres for more dense suspensions was presented to the author of this article by Prof. Bogdan Cichocki, to whom the author is very grateful.

The outline of this article is as follows. In second section, we describe the suspension on the microscopic level and discuss the macroscopic characteristics of suspensions. In third section, we repeat the analysis of Felderhof, Ford and Cohen leading to the cluster expansion of the macroscopic characteristics. In fourth section, we introduce the novel ring expansion
of the macroscopic characteristics, which is a rigorous result. The ring expansion is further used in fifth section of the article, to introduce a generalization of the Clausius-Mossotti approximation. Here, we also present the short-time transport properties calculated by this novel method. The generalized Clausius-Mossotti approximation is discussed and its results are compared with the results of the numerical simulations and with the Beenakker-Mazur method.

II. MACROSCOPIC PROPERTIES OF SUSPENSIONS

We consider suspension of hard spheres of radius $a$ in incompressible Newtonian fluid of kinematic viscosity $\eta$. We also assume sufficiently slow motion of the fluid and the condition of no slip on the surface of immersed particles. As a result, the fluid is described by the stationary Stokes equations with the stick boundary conditions [28]. The stationary Stokes equations for the problem of the suspension of $N$ spheres, centered at positions $X \equiv R_1, \ldots, R_N$, freely-moving in ambient flow $v_0(\mathbf{r})$, under action of external force density field $f_{\text{ext}}(\mathbf{r})$, can be represented in the following integral form [29, 30]

$$U_i(X, \mathbf{r}) = \int d^3 r' M_0 (R_i, \mathbf{r}, \mathbf{r}') f_{\text{ext}}(\mathbf{r}')$$

$$+ \int d^3 r' M_< (R_i, \mathbf{r}, \mathbf{r}') \left[ v_0(\mathbf{r}') + \sum_{j=1}^{N} \int d^3 r'' g_0 (\mathbf{r}', \mathbf{r}'') f_j(X; \mathbf{r}'') \right],$$

$$f_i(X, \mathbf{r}) = \int d^3 r' \hat{M} (i, \mathbf{r}, \mathbf{r}') \left[ v_0(\mathbf{r}') + \sum_{j=1}^{N} \int d^3 r'' g_0 (\mathbf{r}', \mathbf{r}'') f_j(X; \mathbf{r}'') \right]$$

$$+ \int d^3 r' M_> (i, \mathbf{r}, \mathbf{r}') f_{\text{ext}}(\mathbf{r}).$$

(1)

In the above equations, the particle velocity field $U_i(X; \mathbf{r})$ is defined inside the particle, i.e. for $|\mathbf{r} - R_i| \leq a$. For hard spheres it has always the following form

$$U_i(X; \mathbf{r}) = V_i(X) + \Omega_i(X) \times (\mathbf{r} - R_i), \quad \text{for} \quad |\mathbf{r} - R_i| \leq a,$$

(2)

with translational $V_i$ and rotational $\Omega_i$ velocity of the particles. Moreover, $f_i(X; \mathbf{r})$ describes the force density [31–33] acting on the fluid by the surface of the particle number $i$ and is defined by

$$f_i(X; \mathbf{r}) = -\sigma(\mathbf{r}) \cdot \hat{n}_i(\mathbf{r}) \delta(|\mathbf{r} - R_i| - a),$$

(3)
where \( \sigma \) represents the stress tensor in the fluid, \( \hat{n}_i(r) = (r - R_i) / |r - R_i| \) is a vector normal to the surface of the sphere \( i \), whereas \( \delta(x) \) stands for the one-dimensional Dirac delta function. \( G_0(r) \) in equations (1) is the Oseen tensor,

\[
G_0(r) = \left( 1 + \hat{r}\hat{r} \right) / (8\pi\eta|r|),
\]

with \( \hat{r} = r / |r| \). The Oseen tensor is a Green function of the Stokes equations [34], hence flow of the whole suspension \( v(r) \) is given by

\[
v(r) = v_0(r) + \sum_{i=1}^{N} \int d^3r' G_0(r - r') \cdot f_i(r').
\]

The equations (1) are linear both in the ambient flow \( v_0 \) and in the external force density \( f_{\text{ext}} \). Therefore, to describe the response operators \( M_0, \hat{M}, M_<, M_> \), it is sufficient and convenient to consider special cases of a single particle problem. \( M_0 \) in the equations (1), in the case of the single particle problem in the external force density field \( f_{\text{ext}}(r) \), and in absence of the ambient flow, \( v_0 = 0 \), yields the velocity field of the particle,

\[
U_1(R_1; r) = \int d^3r' M_0(R_1, r, r') f_{\text{ext}}(r').
\]

The single particle operator \( M_< \) gives the particle velocity field \( U_1 \), when the particle is placed in the ambient flow \( v_0 \),

\[
U_1(R_1; r) = \int d^3r' M_< (R_1, r, r') v_0(r').
\]

\( \hat{M}(R, r, r') \), called the single particle convective friction kernel, yields the force density \( f_1(R_1; r) \) on the surface of the single particle at the position \( R_1 \), when it is placed in the ambient flow \( v_0(r) \),

\[
f_1(R_1; r) = \int d^3r' \hat{M}(R_1, r, r') v_0(r').
\]

Finally, \( M_> \) describes the force density \( f_1(R_1; r) \) on the surface of the single particle at the position \( R_1 \), under the action of the external force \( f_{\text{ext}} \),

\[
f_1(R_1; r) = \int d^3r' M>(R_1, r, r') f_{\text{ext}}(r').
\]

In this article, we investigate the equations (1) mostly without referring to the specific form of the response operators \( M_0, M_<, M_>, \hat{M} \). For their detail form, we refer the reader to the references [30, 35].
To facilitate further analysis of the equations (1), we omit integral variables in those equations, writing them in the following form

\[
U_i(X) = M_0(i) f_{\text{ext}} + M_< (i) \left[ v_0 + \sum_{j=1 \atop j \neq i}^{N} G_0 f_j(X) \right],
\]

\[
f_i(X) = \tilde{M} (i) \left[ v_0 + \sum_{j=1 \atop j \neq i}^{N} G_0 f_j(X) \right] + M_>(i) f_{\text{ext}}.
\] (10)

For the position of the particle \(i\) in the single particle response operators \(M_0, M_<, \tilde{M}, M_>\), we also use the following abbreviation: \(i \equiv R_i\). Finally, we write the above equations as follows [30],

\[
\begin{bmatrix}
U_i(X) \\
f_i(X)
\end{bmatrix} = M (i) \left( \begin{bmatrix}
f_{\text{ext}} \\
v_0
\end{bmatrix} + \sum_{j=1 \atop j \neq i}^{N} G \begin{bmatrix}
U_j(X) \\
f_j(X)
\end{bmatrix} \right),
\] (11)

introducing \(6 \times 6\) dimensional matrices \(M\) and \(G\) defined by the below equations,

\[
M (R, r, r') = \begin{bmatrix}
M_0 (R, r, r') & M_< (R, r, r') \\
M_> (R, r, r') & \tilde{M} (R, r, r')
\end{bmatrix}
\] (12)

and

\[
G (r, r') = \begin{bmatrix}
0 & 0 \\
0 & G_0 (r, r')
\end{bmatrix}.
\] (13)

A. Scattering series

To solve the equations (11), several methods can be used. One of the possible approaches is the method of reflections [36]. It relies on taking successive iterations of the equation (11) which leads to the following formula:

\[
\begin{bmatrix}
U_i(X) \\
f_i(X)
\end{bmatrix} = M (i) \left( \begin{bmatrix}
f_{\text{ext}} \\
v_0
\end{bmatrix} + \sum_{j=1 \atop j \neq i}^{N} M (i) G M (j) \begin{bmatrix}
f_{\text{ext}} \\
v_0
\end{bmatrix} \right) + \sum_{j=1 \atop j \neq i}^{N} \sum_{k=1 \atop k \neq j}^{N} M (i) G M (j) G M (k) \begin{bmatrix}
f_{\text{ext}} \\
v_0
\end{bmatrix} + \ldots.
\] (14)
The above expression of the force densities, \( f_i(X) \), and the velocities of the particles, \( U_i(X) \), has a form of a multiple scattering series. It means, that \( f_i(X) \) and \( U_i(X) \) are given by the sum of the scattering sequences, for example:

\[
M(1)G \neq M(2)
\] (15)

and

\[
M(1)G \neq M(2)G \neq M(3)G \neq M(2).
\] (16)

As we interpret - each scattering sequence is a superposition of the single-particle scattering operators \( M(i) \), which "scatter" the flow and of Green functions \( G \), which "propagates" the flow.

It is convenient and useful to represent the scattering sequences graphically [29]. The above two sequences can be represented respectively by

\[
\begin{align*}
2 & \quad \circ \quad \cdots \\
1 & \quad \circ \quad \cdots
\end{align*}
\quad ,
\begin{align*}
3 & \quad \circ \quad \cdots \\
2 & \quad \circ \quad \cdots \\
1 & \quad \circ \quad \cdots
\end{align*}
\] (17)

In general, to represent a scattering sequence graphically, we draw horizontal dashed lines \(-\quad -\quad -\quad -\quad -\quad .\) Each line corresponds to a particle in the scattering sequence. Then, reading the sequence from left to right, we put successively: the circle \( \circ \) on the dashed line \( i \) for the operator \( M(i) \) and the vertical line \( \mid \) connecting the dashed lines \( i \) and \( j \) for the Oseen tensor \( G \), when it appears in the configuration \( M(i)G \neq M(j) \).

The scattering series plays a major role in our considerations. We denote the scattering series by \( T_{ij}(X) \):

\[
T_{ij}(X) = M(i)\delta_{ij} + M(i)G \neq M(j)(1 - \delta_{ij})
\]

\[
+ \sum_{k=1 \atop k \neq i,k \neq j}^{N} M(i)G \neq M(k)G \neq M(j) + \ldots .
\] (18)

Therefore, the velocity \( U_i(X) \) and the force density \( f_i(X) \) in the expression (14) are given by the formula

\[
\begin{bmatrix}
U_i(X) \\
f_i(X)
\end{bmatrix}
= \sum_{j=1}^{N} T_{ij}(X) \begin{bmatrix}
f_{ext} \\
v_0
\end{bmatrix}.
\] (19)
B. Macroscopic response

To describe properties of suspension on the macroscopic level, we consider an ensemble of configurations of particles $X \equiv R_1, \ldots, R_N$, which is described by a probability distribution function $p(X)$. We also introduce the average force density defined by the equation

$$\langle f(R, r) \rangle = \left\langle \sum_{i=1}^{N} \delta(R - i) f_i(X, r) \right\rangle$$

and the average particle velocity field

$$\langle U(R, r) \rangle = \left\langle \sum_{i=1}^{N} \delta(R - i) U_i(X, r) \right\rangle,$$

where the three-dimensional Dirac delta function $\delta(R - i) \equiv \delta(R - R_i)$ and the average over the probability distribution $\langle [...] \rangle = \int d^3 R_1 \ldots d^3 R_N \ p(X) \ [...]$ are used. Averages of the equations (19) - multiplied by proper Dirac delta functions - lead to the following expression for the average velocity and the average force density,

$$\left[ \langle U(R, r) \rangle \quad \langle f(R, r) \rangle \right] = \int d^3 R' d^3 r' T(R, r; R', r') \left[ f_{\text{ext}}(r') \quad v_0(r') \right],$$

where the averaged scattering series is denoted by $T(R, r; R', r')$ and defined with the formula

$$T(R, r; R', r') = \left\langle \sum_{i=1}^{N} \sum_{j=1}^{N} \delta(R - i) T_{ij}(X, r, r') \delta(R' - j) \right\rangle.$$

Notice, that in the above operator $T(R, r; R', r')$, the Dirac delta functions fix positions of the first ($i$) and the last ($j$) particle in the scattering series $T_{ij}$ at the positions $R$ and $R'$ respectively. The average flow of the suspension $\langle v(r) \rangle$ is a combination of the ambient flow $v_0(r)$, in which the particles are immersed and of flow generated by the presence of the particles

$$\langle v(r) \rangle = v_0(r) + \int d^3 R \int d^3 r' G_0(r, r') \langle f(R, r') \rangle,$$

which is obtained by averaging the formula (5). We eliminate the flow $v_0$ from the equations (24) and (22), which leads to the formula

$$\left[ \langle U \rangle \quad \langle f \rangle \right] = T \left[ \langle f_{\text{ext}} \rangle \quad \langle v \rangle \right] - TG \left[ \langle U \rangle \quad \langle f \rangle \right],$$

where the averaged scattering series $T(R, r; R', r')$ is defined with the formula
in which we also facilitate the notation by omitting the integral variables. Its subsequent iterations lead to a relation of the average particle velocity $\langle U \rangle$ and the force density $\langle f \rangle$ to the external force density $f_{\text{ext}}$ and the average flow of suspension $\langle v \rangle$,

$$\begin{bmatrix} \langle U(R, r) \rangle \\ \langle f(R, r) \rangle \end{bmatrix} = \int d^3R'd^3r'T^{\text{irr}}(R, r; R', r') \begin{bmatrix} f_{\text{ext}}(r') \\ \langle v(r') \rangle \end{bmatrix},$$  

(26)

which defines $T^{\text{irr}}$ operator given by

$$T^{\text{irr}} = T(1 + GT)^{-1}.$$  

(27)

The equation (26) is directly related to the macroscopic properties of the suspension. For example, the effective viscosity $\eta_{\text{eff}}$ can be inferred from the relation between the average force density $\langle f(R, r) \rangle$ and the average suspension flow $\langle v(r) \rangle$, when no external forces act on the particles, $f_{\text{ext}} = 0$. The relation between $\langle f(R, r) \rangle$ and $\langle v(r) \rangle$ in this situation results from the equation (26), after projection it into the lower half of the double vectors $[(U), \langle f \rangle]$ and $[f_{\text{ext}}, \langle v \rangle]$. In order to do that, we introduce a projector $P_L$ defined by

$$P_L \begin{bmatrix} \langle U \rangle \\ \langle f \rangle \end{bmatrix} = \langle f \rangle,$$

(28)

with its transposition $P_L^T$. After projection, the equation (26) reads

$$\langle f(R, r) \rangle = \int d^3R'd^3r'P_LT^{\text{irr}}(R, r; R', r')P_L^T \langle v(r') \rangle.$$  

(29)

If the $T^{\text{irr}}$ operator is known, by calculating the following four rank Cartesian tensor

$$X_{\alpha\beta\gamma}^{\alpha\beta\gamma}(R, R') = \int d^3r \int d^3r'(r - R)_\alpha [P_LT^{\text{irr}}(R, r; R', r') P_L^T]_{\beta\delta} (r'-R')_\gamma,$$

(30)

and by symmetrizing it over the first and the second pair of the Cartesian indexes

$$X_{\alpha\beta\gamma}^{\delta\delta\gamma}(R, R') = \frac{1}{4} (X_{\alpha\beta\gamma}^{\alpha\beta\gamma}(R, R') + X_{\beta\alpha\gamma}^{\beta\alpha\gamma}(R, R') + X_{\alpha\beta\gamma}^{\alpha\beta\gamma}(R, R') + X_{\beta\alpha\gamma}^{\beta\alpha\gamma}(R, R')),$$

(31)

we obtain the effective viscosity $\eta_{\text{eff}}$, using the formula [28, 29]

$$\eta_{\text{eff}} = \eta + \lim_{N \to \infty} \frac{1}{10N} \sum_{\alpha, \beta = 1}^3 \int d^3R \int d^3R'X_{\alpha\beta\beta\alpha}^{\delta\delta\delta\delta}(R, R').$$  

(32)

Thermodynamic limit $\lim_{N \to \infty}$ is performed in the above equation.
Apart from the short-time effective viscosity $\eta_{\text{eff}}$, we also consider the short-time wave dependent sedimentation coefficient $H(q)$. The sedimentation coefficient describes response of the suspension to the external force of the plane wave form,

$$F_{\text{ext}}(R) = F_0 \hat{q} \Re \exp(-i q R). \quad (33)$$

We show in the appendix A, that under the action of the above force, the average translational velocity of the particles defined by

$$\langle V(R) \rangle = \left\langle \sum_{i=1}^{N} \delta(R - R_i) V_i(X) \right\rangle, \quad (34)$$

in an isotropic and homogeneous suspension, has also a plane wave form,

$$\langle V(R) \rangle = V(q) \hat{q} \Re \exp(-i q R). \quad (35)$$

Linearity of the Stokes equations implies, that the coefficient $V(q)$ in the above formula is proportional to the force $F_0$,

$$V(q) = H(q) \mu_0 F_0.$$  

This formula defines the wave dependent sedimentation coefficient $H(q)$, which is also called the hydrodynamic function. The factor $\mu_0 = 1/(6\pi\eta a)$ denotes the Stokes coefficient. $H(q)$ is a dimensionless function with the property $H(q) \rightarrow 1$ in the limit of a diluted suspension, i.e. when the volume fraction $\phi \rightarrow 0$. As we also discuss in the appendix A, the microscopic expression for the hydrodynamic function $H(q)$ has the following form [29]

$$H(q) = \frac{1}{\mu_0} \frac{1}{3} \text{Tr} \left[ \int d^3 R \ e^{-iqR} Y(R) \right], \quad (36)$$

where $3 \times 3$ matrix $Y(R)$ is defined by the following equation

$$Y(R - R') = \frac{1}{\left(\frac{4}{3} \pi a^3\right)^2 \lim_{\infty}} \int d^3 r \int d^3 r' P_U T^{\text{irr}}(R, r; R', r') P_U^T. \quad (37)$$

The projector $P_U$ projects on the upper half of the double vectors $[(\mathbf{U}), (\mathbf{f})]$,

$$P_U \begin{bmatrix} \langle \mathbf{U} \rangle \\ \langle \mathbf{f} \rangle \end{bmatrix} = \langle \mathbf{U} \rangle. \quad (38)$$

$P_U^T$ denotes transposition of $P_U$. 

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It is worth noting, that the hydrodynamic function for the zero wave vector, $q = 0$, describes the sedimentation rate, $K$, of the suspension in a gravity field,

$$K = H (q = 0) \quad (39)$$

and is also related to the short-time collective diffusion coefficient $D_c$,

$$D_c = D_0 H (q = 0) , \quad (40)$$

whereas for infinite wave vector length $H (q \to \infty)$ is related to the short-time self-diffusion coefficient $D_s$,

$$D_s = D_0 H (q \to \infty) . \quad (41)$$

In both expressions $D_0 = k_B T / (6 \pi \eta a)$ is the diffusion coefficient of a single particle.

Both, the effective viscosity, and the hydrodynamic function can be inferred from the $T_{\text{irr}}$ operator. It is shown by the expressions (30-32) for the effective viscosity $\eta_{\text{eff}}$ and by the equations (36-37) for the wave dependent sedimentation coefficient $H (q)$. Therefore, $T_{\text{irr}}$ becomes the quantity of the main interest in this article.

III. FELDERHOF, FORD AND COHEN ANALYSIS OF $T_{\text{irr}}$

In the first stage of our analysis of $T_{\text{irr}}$ defined by the equation (26), we follow the idea of Felderhof, Ford and Cohen. They obtained the microscopic expression for $T_{\text{irr}}$ for the dielectric system in the form of a cluster expansion [7]. The application of their idea to the physics of suspensions is straightforward, because the governing equations are similar for suspensions and dielectrics [29, 37]. To perform the cluster expansion of the operator $T_{\text{irr}}$ on the basis of the expression (27), Felderhof, Ford and Cohen introduced the cluster expansion of the operator $T$.

A. Cluster expansion of $T$

In the expression (23), $T_{ij} (r, r'; X)$ includes infinitely many scattering sequences, as shown in the formula (18). There are scattering sequences with different number of particles: single particle scattering sequences, e.g.

$$1 - \bigcirc- , \quad 4 - \bigcirc- . \quad (42)$$
two-particle scattering sequences, e.g.

\[ \begin{array}{ccc}
  1 & \circ & \cdots \\
  2 & \circ & \cdots \\
\end{array}, \quad \begin{array}{ccc}
  2 & \circ & \cdots \\
  4 & \circ & \cdots \\
\end{array}, \quad (43) \]

and scattering sequences with higher number of particles, up to \( N \). The scattering sequences with the same number of particles may include different particles. It is noticeable in the examples above, where the first scattering sequence is between the particles from the group \( C = \{1, 2\} \). The second scattering sequence is between the particles from the group \( C = \{2, 4\} \). All the scattering sequences

\[ \sum_{i=1}^{N} \sum_{j=1}^{N} \delta (\mathbf{R} - i) \mathbf{T}_{ij} (\mathbf{r}, \mathbf{r}'; X) \delta (\mathbf{R}' - j) \]

can be divided, regarding which particles appear in a scattering sequence. To perform this division, from all scattering sequences \( \sum_{i=1}^{N} \sum_{j=1}^{N} \delta (\mathbf{R} - i) \mathbf{T}_{ij} (\mathbf{r}, \mathbf{r}'; X) \delta (\mathbf{R}' - j) \), we extract only the scattering sequences between the particles from the group \( C \):

\[ \mathbf{T}^{(s)} (\mathbf{R}, \mathbf{r}; \mathbf{R}', \mathbf{r}' || C) = \text{all } s - \text{particle scattering sequences from } \sum_{i=1}^{N} \sum_{j=1}^{N} \delta (\mathbf{R} - i) \mathbf{T}_{ij} (\mathbf{r}, \mathbf{r}'; X) \delta (\mathbf{R}' - j), \]

which include all particles from \( s - \text{particle group } C \). (44)

The above definition allows to represent the cluster expansion of the scattering series as follows

\[ \sum_{i=1}^{N} \sum_{j=1}^{N} \delta (\mathbf{R} - i) \mathbf{T}_{ij} (\mathbf{r}, \mathbf{r}'; X) \delta (\mathbf{R}' - j) = \sum_{s=1}^{N} \sum_{C \subset X; |C| = s} \mathbf{T}^{(s)} (\mathbf{R}, \mathbf{r}; \mathbf{R}', \mathbf{r}' || C). \] (45)

In the above expression, \( |C| \) stands for the number of particles in the group \( C \), whereas \( \sum_{C \subset X; |C| = s} \) denotes summation over the \( s \)-particle groups of particles among \( X = \{1, \ldots, N\} \). Number of such \( s \)-particle groups is given by the Newton symbol \( \binom{N}{s} \).

Average of the equation (45) over the probability distribution function, leads to the cluster expansion for the average scattering series \( \mathbf{T} \) given be the equation (23),

\[ \mathbf{T} (\mathbf{R}, \mathbf{r}; \mathbf{R}', \mathbf{r}') = \left\langle \sum_{s=1}^{N} \sum_{C \subset X; |C| = s} \mathbf{T}^{(s)} (\mathbf{R}, \mathbf{r}; \mathbf{R}', \mathbf{r}' || C) \right\rangle. \] (46)

Since all particles are identical, i.e. the probability distribution \( p \) is symmetric with respect to interchange of the positions \( \mathbf{R}_i \), all terms with the same number of particles \( s \) in the above expression give the same contribution. Therefore, we simplify the last expression, by
taking one $s$–particle group $C = \{1, \ldots, s\}$ and multiplying it by the factor $\left(\begin{array}{c} N \\ s \end{array}\right)$. It yields

$$T(R, r; R', r') = \left\langle \sum_{s=1}^{N} \left(\begin{array}{c} N \\ s \end{array}\right) T^{(s)}(R, r; R', r'|1 \ldots s) \right\rangle. \quad (47)$$

Introducing $s$-particle distribution functions defined by

$$n(1 \ldots s) = \frac{N!}{(N-s)!} \int d^3 R_{s+1} \ldots \int d^3 R_N p(1 \ldots N), \quad (48)$$

we obtain the cluster expansion of the response operator $T$ in the following form,

$$T(R, r; R', r') = \sum_{s=1}^{N} \frac{1}{s!} T^{(s)}(R, r; R', r'), \quad (49)$$

$$T^{(s)}(R, r; R', r') = \int d^3 R_1 \ldots \int d^3 R_s n(1 \ldots s) T^{(s)}(R, r; R', r'|1 \ldots s). \quad (50)$$

Its thermodynamic limit is achieved by extending of the summation up to $N = \infty$ and performing the thermodynamic limit of the $s$–particle distribution functions $n$. From now on, we will consider the suspension in the thermodynamic limit.

**B. Nodal line**

To perform the cluster expansion of $T^{\text{irr}}$ operator, Felderhof, Ford and Cohen used the relation (27), which may be represented in the following form

$$T^{\text{irr}} = T - TGT + TGTGT - \ldots. \quad (51)$$

Let us look at the second term, i.e. $TGT$. Representing the $T$ by the cluster expansion (49), produces many terms, each of the form

$$T^{(s_1)}G T^{(s_2)}. \quad (52)$$

In the expression $T^{(s_1)}G T^{(s_2)}$, the scattering sequences between $s_1$ particles appearing in $T^{(s_1)}$ are ”connected” by the Green function $G$ with the scattering sequences consisted of $s_2$ particles appearing in $T^{(s_2)}$. Altogether, $T^{(s_1)}G T^{(s_2)}$ forms $s_1 + s_2$-particle scattering sequences. The scattering sequences built from the $T^{(s_1)}G T^{(s_2)}$ are of a special type, i.e. there is a line $G$ connecting a particle from $T^{(s_1)}$ to a particle from $T^{(s_2)}$. This line $G$ is the only ”connection” between the particles from $T^{(s_1)}$ and $T^{(s_2)}$. It is critical to distinguish the lines $G$, which are the only connections between some groups of the particles in a scattering
sequence. Those $G$ are called the nodal lines [29]. It is described by the following examples. In the scattering sequence given by expression (15), there is one propagator $G$. It is a nodal line, because it is the only connection between the groups of particles $\{1\}$ and $\{2\}$. In the scattering sequence (16), there are three propagators $G$. The underlined propagator,

$$M(1)GM(2)GM(3)GM(2),$$

is a nodal line, because it is the only connection between the group $\{1\}$ and the group $\{23\}$. In diagrammatic language, the last scattering sequence reads

$$\begin{array}{c}
\text{nodal line} \\
1 \quad 2 \quad 3 \\
1 \quad \text{nodal line}
\end{array}$$

It is easy to identify a nodal line in diagrammatic language: if cutting a line of a propagator $G$, divides the diagram into two separate pieces, then the propagator $G$ is a nodal line.

**C. Cluster expansion of $T$ with nodal lines specified**

In the previous section, we indicated, that an important element of the analysis of $T^{\text{irr}}$ is the notion of the nodal line. Therefore, we perform further division of the scattering series $T$, by specifying the nodal lines in the scattering sequences.

In agreement with the definition (44), $T^{(s)}(R,r;R',r'|1\ldots s)$ represents infinitely many $s$–particle scattering sequences. We divide them into disjoint sets, by specifying the number of the nodal lines and by specifying the groups of particles separated by the nodal lines in the scattering sequence. Those sets are characterized by the number of groups $g$, the number of particles in each group $s_1 = |C_1|, \ldots , s_g = |C_g|$, and by saying which particles from $\{1, \ldots , s\}$ are in the group $C_i$. The number of the groups $g$ is larger by one than the number of the nodal lines. Since we consider the $s$-particle scattering sequences, we have the condition $s_1 + \ldots + s_g = s$. We extract from all $s$–particle scattering sequences $T^{(s)}(R,r;R'',r''|1\ldots s)$, the scattering sequences with specified groups of particles $C_1, \ldots , C_g$ separated by nodal
lines, defining

\[ \tilde{T}^{(s)}(R, r; R', r'|C_1|\ldots|C_g) \]

= all scattering sequences from \( T^{(s)}(R, r; R', r'|C_1\ldots C_g) \) with \( g - 1 \) nodal lines

separating particles from the groups \( C_1, \ldots, C_g \). \hspace{1cm} (55)

With the above definition, the \( s \)-particle scattering sequences can be divided as follows,

\[ T^{(s)}(R, r; R', r'|C) = \sum_{g=1}^{s} \sum_{s_1+\ldots+s_g=s} \sum_{C_1,\ldots,C_g\subset C, |C_1|+\ldots+|C_g|=s} \tilde{T}^{(s)}(R, r; R', r'|C_1|\ldots|C_g) \]. \hspace{1cm} (56)

Here, \( \sum_{C_1,\ldots,C_g\subset C, |C_1|=s_1,\ldots,|C_g|=s_g} \) denotes summation over all possible divisions of the set of \( s \) particles \( \{1, \ldots, s\} \) into \( g \) subsets, with \( s_1 \) particles in the first subset, \( s_2 \) particles in the second subset, etc. There are \( s!/(s_1!\ldots s_g!) \) possible divisions.

Let us now consider the lowest order term \( \tilde{T}^{(s)}(C) \) in the expression (56), i.e. the term with \( g = 1 \), which have no nodal lines. The definition (55) implies that

\[ \tilde{T}^{(s)}(R, r; R', r'|C) = \text{all scattering sequences from } T^{(s)}(R, r; R', r'|C) \text{ without nodal lines.} \]

The above scattering sequences without nodal lines play significant role. They are called the irreducible scattering sequences [29].

The second order term in the expression (56) is the term with \( g = 2 \),

\[ \sum_{s_1+s_2=s} \sum_{C_1, C_2 \subset C, |C_1|+|C_2|=s} \tilde{T}^{(s)}(R, r; R', r'|C_1|C_2) \]. \hspace{1cm} (57)

From the definition (55), it follows, that \( \tilde{T}^{(s)}(C_1|C_2) \) has one nodal line \( G \) separating the particles from the groups \( C_1 \) and \( C_2 \). Therefore, all scattering sequences in \( \tilde{T}^{(s)}(C_1|C_2) \) have the following structure: first - there are some reflections between the particles from the group \( C_1 \), then - there is exactly one reflection \( G \) to a particle in the group \( C_2 \) (nodal line), and then - there are reflections between the particles from the group \( C_2 \). The reflections between the particles from the group \( C_1 \) must be irreducible (without a nodal line). The same holds for the group \( C_2 \). It suggests, that \( \tilde{T}^{(s)}(C_1|C_2) \) has the following structure,

\[ \tilde{T}^{(s)}(C_1|C_2) = \tilde{T}^{(s_1)}(C_1) G \tilde{T}^{(s_2)}(C_2) \], \hspace{1cm} (58)
in which the irreducible scattering sequences $\bar{T}^{(s)}(C)$ appear and the nodal line is written explicitly. The above formula can be simply proved using the definition (55). Similar results hold for the higher terms of the expansion (56),

$$\bar{T}^{(s)}(C_1\ldots|C_g) = \bar{T}^{(s_1)}(C_1)G\ldots G\bar{T}^{(s_g)}(C_g),$$

(59)

for the groups $C_1,\ldots,C_g$, including $s_1,\ldots,s_g$ particles respectively. In the above formula, the nodal lines separate different irreducible sections $\bar{T}^{(s_i)}(C_i)$ of the scattering sequences $\bar{T}^{(s)}(C_1\ldots|C_g)$. Each irreducible section $\bar{T}^{(s_i)}(C_i)$ is referred to as "the block". Moreover, by "the block structure" we mean the way particles are distributed in the blocks. A block structure is specified as follows: $C_1\ldots|C_g$. The block structure of the scattering sequence given by the expression (16) is 1|23. It is convenient to introduce the following notation for the irreducible scattering sequences,

$$S_I(R,r;|R',r'||C) = \frac{1}{s!}\bar{T}^{(s)}(R,r;R',r'||C),$$

(60)

with the factor $s!$. Using the above two formulas, we rewrite the expansion (56) as follows

$$T^{(s)}(C) = \sum_{g=1}^{s} \sum_{s_1+\ldots+s_g=s} \sum_{C_1,\ldots,C_g\subseteq C, |C_1+\ldots+|C_g|=s} s_1!\ldots s_g!S_I(C_1)G\ldots GS_I(C_g).$$

(61)

The above representation used in the cluster expansion of $T$ operator represented by equations (49) and (50), after a simple algebra, leads to the expression

$$T = \sum_{g=1}^{\infty} \sum_{C_1,\ldots,C_g} \int dC_1\ldots dC_g n(C_1,\ldots,C_g)S_I(C_1)G\ldots GS_I(C_g).$$

(62)

Cancellation of the factors $s_i!$ results from the symmetry of the probability distribution density $p$ and the fact, that the particles are identical. The symbol $\sum_{C_1,\ldots,C_g} \int dC_1\ldots \int dC_g$ denotes summation over different numbers of particles in each of $g$ groups and integration over the positions of particles as follows

$$\sum_{C_1,\ldots,C_g} \int dC_1\ldots dC_g f(C_1,\ldots,C_g) = \sum_{n_1=1}^{\infty} \ldots \sum_{n_g=1}^{\infty} \int d^3 R_1^{n_1}\ldots d^3 R_1^{n_1} \ldots d^3 R_1^{n_g}\ldots d^3 R_1^{n_g} f(R_1^{n_1},\ldots,R_1^{n_1},\ldots,R_1^{n_g},\ldots,R_1^{n_g})$$

(63)
D. Cluster expansion of $T^{\text{irr}}$

We go back to the expression (51),

$$T^{\text{irr}} = T - TGT + TGTGT - \ldots,$$  \hspace{1cm} (64)

in order to derive the cluster expansion of the $T^{\text{irr}}$ operator. As we discussed before, the average scattering series $T$ includes all possible scattering sequences. $TGT$ in the equation (64) produces scattering sequences with at least one nodal line, $TGTGT$ with at least two nodal lines, etc. The analysis of the above equation relies on a consideration of the scattering sequences with given number of nodal lines. Therefore, we will consider terms with different block structures $C_1|\ldots|C_g$ on the right-hand side of the equation (64).

Let us focus first on the block structure $C_1$, i.e. on the scattering sequences without nodal lines. They appear only in the first term, $T$, of the equation (64), because the higher terms, $TGT$, $TGTGT$, $\ldots$, include at least one nodal line. In the expression (62) for $T$, the irreducible scattering sequences come only from the term $g = 1$. Therefore, all terms on the right hand-side of the equation (64) with the block structure $C_1$, are of the form

$$n(C_1)S_I(C_1).$$  \hspace{1cm} (65)

Next, we consider the terms on the right-hand side of the equation (64) with the block structure $C_1|C_2$, i.e. with one nodal line. Such scattering sequences, i.e. $S_I(C_1)GS_I(C_2)$, appear only in the first term, $T$, and in the second term, $TGT$. A contribution from the $T$ comes from the term with $g = 2$ of the equation (62) and is of the form $n(C_1C_2)S_I(C_1)GS_I(C_2)$. A contribution from $TGT$ has a form of $n(C_1)n(C_2)S_I(C_1)GS_I(C_2)$ and is produced by the terms with $g = 1$ in both $T$. Those two terms altogether yield

$$(n(C_1, C_2) - n(C_1)n(C_2))S_I(C_1)GS_I(C_2).$$  \hspace{1cm} (66)

For the block structure consisted of the three groups $C_1|C_2|C_3$, analysis of the right-hand side of the expression (64) leads to the contribution of the following form

$$(n(C_1C_2C_3) - n(C_1C_2)n(C_3) - n(C_1)n(C_2C_3) + n(C_1)n(C_2)n(C_3))S_I(C_1)GS_I(C_2)GS_I(C_3).$$  \hspace{1cm} (67)
In such manner, analysis of all block structures $C_1|...|C_g$ is possible. Functions appearing along with the block structures of the form $S_I(C_1)G...GS_I(C_g)$, are denoted by $b(C_1|...|C_g)$ and are called the block distribution functions [7]. Therefore we have

$$b(C_1) = n(C_1),$$
$$b(C_1|C_2) = n(C_1, C_2) - n(C_1)n(C_2),$$
$$b(C_1|C_2|C_3) = n(C_1C_2C_3) - n(C_1C_2)n(C_3) - n(C_1)n(C_2C_3) + n(C_1)n(C_2)n(C_3),$$

for the block structures up to three groups. Expressions for the block distribution functions for higher number of groups are more and more complicated. Nevertheless, the block distribution functions may be calculated from the following recursive formula [29, 38]

$$b(C) = n(C)$$
$$b(C_1|...|C_k|C_{k+1}|...|C_g) = b(C_1|...|C_k)b(C_{k+1}|...|C_g) + b(C_1|...|C_k|C_{k+1}|...|C_g).$$

The above analysis leads to the following cluster expansion of $T_{irr}$ operator:

$$T_{irr} = \sum_{g=1}^{\infty} \sum_{C_1,...,C_g} \int dC_1...dC_gb(C_1|...|C_g)S_I(C_1)G...GS_I(C_g).$$

It is worth noting, that the structure of $T_{irr}$ operator is similar to the structure of $T$ operator given by the expression (62). The only difference lies in the distribution functions: in $T_{irr}$ - the block distribution functions $b(C_1|...|C_g)$ appear, whereas in $T$ operator - the standard s-particle distribution functions $n(C_1...C_g)$ appear.

E. Self scattering sequences

There are phenomena in the physics of suspensions, in which only a part of the scattering series $T$, given by equation (23), plays a role. An example of the above is the self-diffusion coefficient. It is related only to those scattering sequences in $T$, which start and end at the same particle. The scattering sequences, which start and end at the same particle, we call the self-scattering sequences. The self-scattering sequences are irreducible, because there are no nodal lines in any scattering sequence, which starts and ends at the same particle. Therefore,
the self-scattering sequences $S^\text{self}_I$ are related to the irreducible scattering sequences $S_I$, as follows,

$$S^\text{self}_I (\mathbf{R}, \mathbf{r}; \mathbf{R}', \mathbf{r}'|\mathbf{C}) = \text{all scattering sequences from } S_I(\mathbf{R}, \mathbf{r}; \mathbf{R}', \mathbf{r}'|\mathbf{C}),$$

which start and end at the same particle. (73)

The average of the self-scattering sequences will be denoted by $B$,

$$B (\mathbf{R}, \mathbf{r}; \mathbf{R}', \mathbf{r}') = \sum_{C_1} \int dC_1 \ n(C_1) S^\text{self}_I (\mathbf{R}, \mathbf{r}; \mathbf{R}', \mathbf{r}'|C_1).$$

(74)

With the above quantity, the scattering sequences in $T$ can be divided into the self-scattering sequences and the scattering sequences, which start and end at different particles (off-scattering sequences). The former are given by $B (\mathbf{R}, \mathbf{r}; \mathbf{R}', \mathbf{r}')$, the latter are denoted by $T_{\text{off}}$. Therefore,

$$T = B + T_{\text{off}}.$$  

(75)

Operator $T^{\text{irr}}$ can be divided in a similar manner,

$$T^{\text{irr}} = B + T^{\text{irr}}_{\text{off}}.$$  

(76)

As shown in the above formulas, the self-part of both operators $T$ and $T^{\text{irr}}$ are the same.

It is worth noting, that the self-scattering sequences $B$ can be calculated from off-scattering sequences $T_{\text{off}}$ as follows [39]

$$B (\mathbf{R}, \mathbf{r}; \mathbf{R}', \mathbf{r}') = n_1 \delta (\mathbf{R} - \mathbf{R}') \ M (\mathbf{R}, \mathbf{r}, \mathbf{r}') + \delta (\mathbf{R} - \mathbf{R}') \ [T_{\text{off}} \ G M] (\mathbf{R}, \mathbf{r}; \mathbf{R}, \mathbf{r}') .$$

(77)

The second term in the above expression corresponds to scattering sequences of the following structure. The sequences start at the particle at position $\mathbf{R}$, then go with all possible scattering sequences $T_{\text{off}}$ to some particle, and then they come back with one reflection $G$ to the starting particle at $\mathbf{R}$. Moreover, in the above equation $n_1$ stands for the one-particle distribution function.

**IV. RING EXPANSION OF $T^{\text{irr}}$**

The cluster expansion (72) of the response operator $T^{\text{irr}}$ was derived by Felderhof, Ford and Cohen about three decades ago [7]. This achievement allows to express the transport
coefficients of dispersive media, such as suspensions and dielectrics, by absolutely convergent integrals. Despite this important step done by the three scientists, no reasonable statistical physics method of calculation of transport properties of suspensions can be found in current literature. We have already discussed this point in the introduction.

In order to give a motivation of our approach introduced further in this article, we invoke the effective Green function $G_{\text{eff}}$ defined by

$$G_{\text{eff}} = G + GTG. \quad (78)$$

In the case of homogeneous system in the thermodynamic limit, the effective Green function $G_{\text{eff}}$ of suspension is related to the Green function $G$ of pure fluid according to the following formula, [29]

$$G_{\text{eff}}(r, r') \approx \frac{\eta}{\eta_{\text{eff}}(\phi)} G(r - r'), \quad \text{for } |r - r'| \to \infty. \quad (79)$$

The above formula describes the asymptotic decay of the effective propagator $G_{\text{eff}}(r, r')$ for the large distances between the points $r$ and $r'$. The decay is governed by the effective viscosity of suspension $\eta_{\text{eff}}(\phi)$, which depends on the volume fraction $\phi = 4\pi n_1 a^3/3$, with $n_1$ standing for the number density of the particles in suspension.

Let us consider relation (27) between $T$ and $T^{\text{irr}}$, which can be inverted and then represented in the following way,

$$T = T^{\text{irr}} + T^{\text{irr}} G_{\text{eff}} T^{\text{irr}}, \quad (80)$$

where the formula for the effective propagator $G_{\text{eff}} = G \left(1 - T^{\text{irr}}G\right)^{-1}$ is used. Considering only the off-scattering sequences in the above expression, we receive the equation

$$T_{\text{off}} = T_{\text{off}}^{\text{irr}} + T^{\text{irr}} G_{\text{eff}} T^{\text{irr}}, \quad (81)$$

obtained with the application of the formulas (76) and (75). Let us notice, that $T_{\text{off}}(R, r; R', r') = 0$ for overlapping configurations, i.e. for $|R - R'| < 2a$. It results from the assumption, that the hard spheres in suspension cannot overlap and in the expression (62), the distribution function $n(C)$ vanishes for the overlapping configurations. The vanishing of $T_{\text{off}}(R, r; R', r')$ for the overlapping configurations has a consequence in the last equation. It reduces to

$$f(R - R') T_{\text{off}}^{\text{irr}}(R, r; R', r') = -f(R - R') \left[T^{\text{irr}} G_{\text{eff}} T^{\text{irr}}\right](R, r; R', r'), \quad (82)$$
after its multiplication by a function \( f(R - R') \) defined by

\[
\begin{align*}
  f(R - R') &= \begin{cases} 
    1 & \text{for } |R - R'| < 2a \\
    0 & \text{for } |R - R'| \geq 2a, 
  \end{cases} 
\end{align*}
\]

which equals 0 for nonoverlapping and 1 for overlapping configurations of the two particles centered at \( R \) and \( R' \).

The equation (82) shows, that in \( T_{\text{irr}} \), there are some contributions with the effective propagator \( G_{\text{eff}} \). It suggests, that the propagator \( G \) appearing in the cluster expansion (72) of Felderhof, Ford and Cohen can be renormalized. In other words, \( T_{\text{irr}} \) can be given by the following formula,

\[
T_{\text{irr}} = \sum_{d=1}^{\infty} \sum_{C_1 \ldots C_d} \int dC_1 \ldots dC_d H(C_1 \ldots |C_d) S_I(C_1) G_{\text{eff}} \ldots G_{\text{eff}} S_I(C_d),
\]

with yet unknown functions \( H(C_1 \ldots |C_d) \), which we call the block correlation functions. The above expression for \( T_{\text{irr}} \) has the same structure as expression (72), but contains the effective propagator \( G_{\text{eff}} \), instead of the propagator \( G \) and also contains the block correlation functions \( H(C_1 \ldots |C_d) \), instead of the block distribution functions \( b(C_1 \ldots |C_g) \). We call the expression (84) for \( T_{\text{irr}} \) operator, the ring expansion, in order to differentiate it from the cluster expansion (72) of this operator introduced by Felderhof, Ford and Cohen.

Below, we prove the ring expansion (84) and derive a formula for the block correlation functions \( H(C_1 \ldots |C_d) \). We will use the similar approach, as in the derivation of the Felderhof, Ford and Cohen's formula (72) in the previous section. We will consider the right-hand sides of both expressions (72) and (84), considering scattering sequences with a given block structure \( C_1 \ldots |C_g \).

We start with the block structure \( S_I(C_1) \), i.e. the block structure without a nodal line. In the expression (84), the block structure \( S_I(C_1) \) without a nodal line appears only in the term \( d = 1 \). In the expression (72), the block structure \( S_I(C_1) \) also appears in the lowest order term \( g = 1 \) only. Therefore, equality of the expressions (72) and (84), on the level of the irreducible scattering sequences \( S_I(C_1) \), is possible if we assume

\[
b(C_1) = H(C_1).
\]

Before further considerations for a general block structure, let us find all terms in the ring expansion (84), which have the block structure \( C_1 |C_2|C_3|C_4 \). For the purpose of the
above, we need the cluster expansion of the effective Green function \( \mathbf{G}_{\text{eff}} \).

\[
\mathbf{G}_{\text{eff}} = \sum_{g=0}^{\infty} \sum_{C_1, \ldots, C_g} \int dC_1 \ldots dC_g \ n(C_1, \ldots, C_g) \mathbf{G}_{I}(C_1) \mathbf{G} \ldots \mathbf{G}_{I}(C_g) \mathbf{G},
\]

which is a straightforward consequence of the expressions (78) and (62). In the above formula, the term with \( g = 0 \) corresponds to the Oseen tensor \( \mathbf{G} \). The effective Green function introduces one, two, and more nodal lines in the block structure \( \mathbf{S}_{I}(C_i) \mathbf{G}_{\text{eff}} \mathbf{S}_{I}(C_j) \).

Therefore, the scattering sequences \( \mathbf{S}_{I}(C_1) \mathbf{G}_{I}(C_2) \mathbf{G}_{I}(C_3) \mathbf{G}_{I}(C_4) \) appear in the ring expansion (84) in the term corresponding to \( d = 2 \),

\[
H(C_{i_1}|C_{i_2}) \mathbf{S}_{I}(C_{i_1}) \mathbf{G}_{\text{eff}} \mathbf{S}_{I}(C_{i_2}),
\]

in the term \( d = 3 \),

\[
H(C_{i_1}|C_{i_2}|C_{i_3}) \mathbf{S}_{I}(C_{i_1}) \mathbf{G}_{\text{eff}} \mathbf{S}_{I}(C_{i_2}) \mathbf{G}_{\text{eff}} \mathbf{S}_{I}(C_{i_3}),
\]

and in the term \( d = 4 \),

\[
H(C_{i_1}|C_{i_2}|C_{i_3}|C_{i_4}) \mathbf{S}_{I}(C_{i_1}) \mathbf{G}_{\text{eff}} \mathbf{S}_{I}(C_{i_2}) \mathbf{G}_{\text{eff}} \mathbf{S}_{I}(C_{i_3}) \mathbf{G}_{\text{eff}} \mathbf{S}_{I}(C_{i_4}).
\]

Other terms in the ring expansion (84) do not contain the scattering sequences with three nodal lines, because the term \( d = 1 \) contains no nodal lines, whereas the terms corresponding to \( d \geq 5 \) contain at least four nodal lines.

Each of the terms in the expressions (87-89) contains many different scattering sequences produced by the cluster expansion (86) of the effective Green function. In the case of the expression (87), only the term \( n(C_2C_3) \mathbf{G}_{I}(C_2) \mathbf{G}_{I}(C_3) \mathbf{G} \) in the expansion (86) produces the block structure \( C_1|C_2|C_3|C_4 \), yielding

\[
H(C_1|C_4) n(C_2C_3) \mathbf{S}_{I}(C_1) \mathbf{G}_{I}(C_2) \mathbf{G}_{I}(C_3) \mathbf{G}_{I}(C_4).
\]

In order to obtain this, we assume \( i_1 = 1, i_2 = 4 \) in the expression (87). In the case of the expression (88), there are two possibilities leading to the block structure \( C_1|C_2|C_3|C_4 \).

The first possibility corresponds to the situation, when the first (left) propagator \( \mathbf{G}_{\text{eff}} \) in the expression (88) introduces one nodal line and the second propagator introduces two nodal lines. Therefore, we assume \( i_1 = 1, i_2 = 2, i_3 = 4 \) and the contribution of the term given by the expression (88) is

\[
H(C_1|C_2|C_4) n(C_3) \mathbf{S}_{I}(C_1) \mathbf{G}_{I}(C_2) \mathbf{G}_{I}(C_3) \mathbf{G}_{I}(C_4).
\]
The second possibility corresponds to an opposite situation, when the first propagator in expression (88) introduces two nodal lines and the second propagator introduces one nodal line. Here, we assume $i_1 = 1$, $i_2 = 3$, $i_3 = 4$ and the contribution is

$$H (C_1|C_3|C_4) n (C_2) S_I (C_1) GS_I (C_2) GS_I (C_3) GS_I (C_4). \quad (92)$$

In the case of expression (89), there is only one possibility to obtain the block structure $C_1|C_2|C_3|C_4$, i.e. when all propagators $G_{\text{eff}}$ introduce only one nodal line $G$. In this case, we have $i_1 = 1$, $i_2 = 2$, $i_3 = 3$, $i_4 = 4$ and obtain the following contribution,

$$H (C_1|C_2|C_3|C_4) S_I (C_1) GS_I (C_2) GS_I (C_3) GS_I (C_4). \quad (93)$$

Finally, all terms in the expression (84), which have the block structure $C_1|C_2|C_3|C_4$ (i.e. terms given by expressions (90-93)), after comparison with the term containing the same block structure $C_1|C_2|C_3|C_4$ from the equation (72), lead to equality

$$b (C_1|C_2|C_3|C_4) = H (C_1|C_4) n (C_2C_3) + H (C_1|C_2|C_4) n (C_3) + H (C_1|C_3|C_4) n (C_2) + H (C_1|C_2|C_3|C_4). \quad (94)$$

Similar considerations for the block structures $C_1|C_2$ and $C_1|C_2|C_3$ lead to the expressions

$$b (C_1|C_2) = H (C_1|C_2), \quad (95)$$

$$b (C_1|C_2|C_3) = H (C_1|C_2|C_3) + H (C_1|C_3) n (C_2). \quad (96)$$

The above considerations for the block structure $C_1|C_2|C_3|C_4$, leading to the formula (94), can be generalized to the case of a block structure $C_1|\ldots|C_g$ consisted of $g \geq 2$ groups. The block structures $C_1|\ldots|C_g$ appear in the ring expansion (84) in the terms $d = 2, \ldots, g$ only, i.e. the terms of the form $S_I (C_1) G_{\text{eff}} \ldots G_{\text{eff}} S_I (C_d)$. As the term (88) for the case $g = 4$ introduces the block structure $C_1|C_2|C_3|C_4$ in two ways, each of $S_I (C_1) G_{\text{eff}} \ldots G_{\text{eff}} S_I (C_d)$ can introduce the block structure $C_1|\ldots|C_g$ in several ways. All terms can be uniquely classified by specification of $d$ groups $C_{i_1}|\ldots|C_{i_d}$ among $C_1|\ldots|C_g$, which come from the blocks $S_I$ in the expression $S_I (C_1) G_{\text{eff}} \ldots G_{\text{eff}} S_I (C_d)$. The edge groups must be the same, therefore $i_1 = 1$ and $i_d = g$. Each set of numbers $1 = i_1 < i_2 < \ldots < i_{d-1} < i_d = g$ corresponds to a single term in the expression $S_I (C_1) G_{\text{eff}} \ldots G_{\text{eff}} S_I (C_d)$, which produces
the block structure $S_I(C_1)G \ldots GS_I(C_g)$. Comparison of all terms in the expansion (84) and (72), producing the block structure $S_I(C_1)G \ldots GS_I(C_g)$ yields

$$b(C_1|\ldots|C_g) =$$

$$\sum_{d=2}^{g} \sum_{1=i_1<i_2<\ldots<i_d=g} H(C_{i_1}|\ldots|C_{i_d}) \times$$

$$n\left(\{C_{i_1} \ldots C_{i_d}\} \setminus \{C_{i_1}, C_{i_2}\}\right) \ldots n\left(\{C_{i_{d-1}} \ldots C_{i_d}\} \setminus \{C_{i_{d-1}}, C_{i_d}\}\right),$$

(97)

which is valid for $g \geq 2$. The symbol ‘\’ denotes a difference of sets of the particles, e.g. $\{12567\} \setminus \{56\} = \{127\}$. We assume that for the empty set $n(\emptyset) = 1$. The above formula is a recursive expression for the block correlation functions $H$.

**A. Comparison of ring and cluster expansion**

The ring expansion (84) introduced in the previous section is a rigorous expression for the response operator $T_{irr}$. This is an alternative formula to the cluster expansion (72) of Felderhof, Ford and Cohen. The cluster expansion and the ring expansion have the same structure: the irreducible scattering sequences $S_I$ connected by the propagators ($G$ or $G_{eff}$), are averaged over configurations of particles, weighted with the distribution functions ($b$ or $H$). From that perspective, and due to fact, that the effective Green function $G_{eff}$ appears in the ring expansion instead of $G$, the formula (84) can be seen as the renormalized cluster expansion.

There are two important differences between the ring and the cluster expansion. The first difference lies in the propagators. The propagator $G$, which appears in the cluster expansion (72), includes only information concerning liquid. On the other hand, the effective Green function $G_{eff}$ in the expression (84), contains macroscopic information about suspension. It is exhibited by the appearance of the effective viscosity $\eta_{eff}$ in the asymptotic form of the effective propagator for large distances showed in the formula (79). The effective viscosity $\eta_{eff}$ of hard-sphere suspension may significantly differ from the viscosity $\eta$ of pure liquid - especially for the higher volume fractions $\phi$.

The second difference between the ring and the cluster expansion lies in the distribution functions. The block distribution functions $b$ appearing in the cluster expansion are given with the formula (71). The block correlation functions $H$, which appeared in the ring
expansion, are defined with the expression (97). There is an essential difference between \( b \) and \( H \). It is related to the cluster property of the distribution functions \( n \), which we assume in this article. The cluster property relies on the factorization of the distribution function \( n(C_1C_2) \) in the limit of large distance between the groups \( C_1 \) and \( C_2 \),

\[
n(C_1C_2) \rightarrow n(C_1)n(C_2).
\]

(98)

A use of the above cluster property of the distribution functions \( n \) in the equation (70), when the group \( C_2 \) in the middle of the block structure \( C_1|C_2|C_3 \) goes away from the other groups, leads to the following factorization of the block distribution function

\[
b(C_1|C_2|C_3) \rightarrow b(C_1|C_3)b(C_2).
\]

(99)

The cluster property of the distribution functions \( n \) applied in the equation (96) for \( H(C_1|C_2|C_3) \) in the same limit - when the group \( C_2 \) goes away - results in the following decay of the block correlation function,

\[
H(C_1|C_2|C_3) \rightarrow 0.
\]

(100)

The above asymptotic decay is a motivation for the name of the block correlation functions \( H \). The above property is also a motivation for the name of the expression (84) - i.e. ring expansion. Two subsequent blocks, \( S_I(C_i) \) and \( S_I(C_{i+1}) \), in the ring expansion (84) are 'connected' by, both, the effective propagator \( G_{\text{eff}} \) and by the correlation function \( H(\ldots|C_i|C_{i+1}|\ldots) \) - both 'connections' vanish, when \( C_i \) goes away from \( C_{i+1} \). We imagine, that such double-connection of the \( S_I(C_i) \) and \( S_I(C_{i+1}) \) form a 'ring'.

The ring expansion of \( T^{\text{irr}} \) represented by the equation (84) along with the expression (97) for the block correlation functions is the main analytical result of this article. It is an alternative expression to the Felderhof, Ford and Cohen’s cluster expansion represented by the formula (72). Our ring expansion appears as a result of a resummation performed on the level of the cluster expansion. This resummation procedure leads to the ring expansion which has similar structure as the structure of the cluster expansion. The role of the Oseen tensors in the cluster expansion - after resummation - is played by the effective Green function \( G_{\text{eff}} \) given by the formula (78). The effective Green function has a physical interpretation because it relates the force (generating the ambient flow) with the velocity field of the suspension - in contrast to the Oseen tensor which relates the force with the velocity field of a pure liquid.
Moreover, the effective Green function is related to the effective viscosity, as the expression (79) shows. Because the resummation procedure leads to a similar structure as the structure of the starting expression, we call this procedure the renormalization. Consequently, the effective Green function may also be called the renormalized (effective) Green function. The ring expansion is further used in the next section to introduce a new method of calculations of transport properties of suspensions.

The above derivation of the ring expansion is presented in the short-hand notation which emphasizes the idea of the underlying physics. It is worth presenting the ring expansion without the short-hand notation. Following the expression (63) the lowest two terms of the ring expansion are given by

\[
    T_{\text{irr}}(R, r; R', r') = \\
    \sum_{n_1=1}^{\infty} \int d^3 R_1^1 \ldots d^3 R_{n_1}^1 H(R_1^1, \ldots, R_{n_1}^1) S_I(R, r; R', r'|R_1^1, \ldots, R_{n_1}^1) \\
    + \sum_{n_1=1}^{\infty} \sum_{n_2=1}^{\infty} \int d^3 R_1^1 \ldots d^3 R_{n_1}^1 d^3 R_1^2 \ldots d^3 R_{n_2}^2 d^3 r'' d^3 r''' d^3 R''' H(R_1^1, \ldots, R_{n_1}^1; R_1^2, \ldots, R_{n_2}^2) \\
    \times S_I(R, r; R'', r''') S_I(R'', r'''; R', r'|R_1^2, \ldots, R_{n_2}^2) \\
    + \ldots
\]

V. RENORMALIZATION OF CLAUSIUS-MOSSOTTI APPROXIMATION

After the derivation of the cluster expansion (72) of the \(T_{\text{irr}}\) operator, Felderhof, Ford and Cohen gave the microscopic explanation of the Clausius-Mossotti formula [9]. It is an expression for the relative dielectric constant of a nonpolar dielectric system. It may be derived using the macroscopic considerations [40]. Felderhof, Ford and Cohen explained the Clausius-Mossotti formula on the microscopic level, showing a class of terms in the cluster expansion of \(T_{\text{irr}}\), which leads to the Clausius-Mossotti relation.

Going along the line of the explanations of Felderhof, Ford, and Cohen [9] for a dielectric system, the Clausius-Mossotti relation is obtained, when the operator \(T_{\text{CM}}^{\text{irr}}\) defined with the following formula

\[
    T_{\text{CM}}^{\text{irr}} = T_{\text{irr}} (1 + [hG] T_{\text{irr}})^{-1},
\]

is approximated by the single particle term,

\[
    T_{\text{CM}}^{\text{irr}}(R, r; R', r') \approx n_1 \delta(R - R') M(R, r, r').
\]
In the above definition (102), a superposition between the quantities \( T_{\text{irr}} (R, r; R', r') \) and \([hG (R, r; R', r')] = h (R, R') G (r, r')\) appears. \( h (R, R')\) stands for the two-particle correlation function. It is worth noting, that in the reference [9], instead of the two-particle correlation function \( h (R, R')\), function \(-f (R, R')\) with \( f\) defined by the formula (83) appears. Both possibilities lead to the Clausius-Mossotti relation. We call the operator \( T_{\text{CM}}\) - the Clausius-Mossotti operator - because it is a straightforward generalization of the Clausius-Mossotti function. In case of suspensions, the above procedure leads to the Saito formula for the effective viscosity [41], namely

\[
\eta_{\text{eff}} = \frac{1 + \frac{3}{2} \phi}{1 - \phi}.
\] (104)

The Clausius-Mossotti approximation is expressed by the approximated formula (103), applied to the Clausius-Mossotti operator, defined by the equation (102). In this equation, the Oseen tensor \( G\) appears. In the previous section, we show that the propagators \( G\) in the cluster expansion (72) can be renormalized and, as a result, \( T_{\text{irr}}\) operator can be represented by the ring expansion (84), with the effective Green function \( G_{\text{eff}}\) appearing instead of \( G\). This suggests to define renormalized Clausius-Mossotti operator \( T_{\text{RCM}}\), as follows,

\[
T_{\text{RCM}} = T_{\text{irr}} (1 + [hG_{\text{eff}}] T_{\text{irr}})^{-1}.
\] (105)

in analogy to the formula (102), and to generalize the Clausius-Mossotti approximation by

\[
T_{\text{RCM}} \approx B,
\] (106)

in analogy to the approximation (103). In the latter equation, instead of the single particle response operator \( M\) appearing in the Clausius-Mossotti approximation, we take into account all self-scattering sequences \( B\), introduced before with the formula (74). The equations (105), (27), (78), (75), (77) along with the approximation (106) define the renormalized Clausius-Mossotti approximation. Those equations form a close system of equations for operators \( B, T_{\text{RCM}}, T_{\text{irr}}, G_{\text{eff}}, T,\) and \( T_{\text{off}}\). The system can be solved for given volume fraction \( \phi\) (or the single particle density \( n_1\)) and for given two-particle correlation function \( h (R, R')\). We solve those equations numerically. Not to interrupt our line of reasoning, we refer the reader to the appendix B containing the technical details of our numerical calculations. From \( T_{\text{irr}}\) found within the renormalized Clausius-Mossotti approximation, one can calculate further the short-time transport characteristics, such as the effective viscosity \( \eta_{\text{eff}}\) from the
equations (30-32) and the wave dependent hydrodynamic function $H(q)$ with the collective diffusion $D_c$ and the self-diffusion $D_s$ coefficient from the equations (36-41).

Before presenting in the next section the results for the transport characteristics calculated within the generalized Clausius-Mossotti approximation, in what follows we comment on its physical meaning. To this end we discuss the $T$ operator (see equation (22)) obtained within the Clausius-Mossotti approximation denoted by $T^{CM}$. The Clausius-Mossotti approximation (103) - by equations (102) and (27) - leads to the expression

$$T^{CM} = n_1 M [1 - [gG] n_1 M]^{-1},$$

(107)

where $g$ is the radial distribution function related to the correlation function $h$ as follows, 

$$g = 1 + h.$$ 

The above formula for $T$ can be interpreted in terms of scattering sequences. The $T$ operator - which on rigorous level is given by the sum of all possible scattering sequences as the equation (18) shows - in the Clausius-Mossotti approximation is given by a sum of scattering sequences in which the reflections never go back to a particle which already reflected the flow. Moreover, there are correlations $g$ only between neighboring particles in the scattering sequences. On the other hand the generalized Clausius-Mossotti approximation (106) - along with the rigorous equations (105), (78) and (27) - leads to the following formula for the $T$ operator (denoted by $T^{RCM}$),

$$T^{RCM} = B [1 - (gG + h[GTG]) B]^{-1}.$$ 

(108)

The above equation differs from the expression (107), in particular by the term $h[GTG]$. This term contains the dominant terms in the virial expansion on three-particle level for the sedimentation coefficient (cf. $b_4$ coefficient in the reference [24]) and for the effective viscosity (cf. $\nu_2$ coefficient in the reference [25]). Therefore we expect that the renormalized (generalized) Clausius-Mossotti approximation will give more accurate results than the original Clausius-Mossotti approximation.

VI. RESULTS AND DISCUSSION

The renormalized Clausius-Mossotti approximation introduced in the previous paragraph allows to calculate the short-time transport properties of suspension, when the volume fraction $\phi$ and the two-body correlation function $h(R, R')$ are given. Within the renormalized Clausius-Mossotti approximation, we perform calculations for the volume fractions
FIG. 1. Inverse of the translational short-time self-diffusion coefficient $D_s$, equation (41), normalized by the self-diffusion coefficient of a single particle, $D_0 = k_B T / (6\pi \eta a)$, as a function of volume fraction $\phi$ for monodisperse suspension of hard-spheres in equilibrium. Black (solid line) - the renormalized Clausius-Mossotti approximation introduced in this article, red (long-dashed) line - numerical simulations of Abade et al. [43], blue (short-dashed) line - the revised Beenakker-Mazur method [44].

$\phi = 0.05, 0.15, 0.25, 0.35,$ and $0.45$. For each volume fraction, we use the two-particle correlation function in the Percus-Yevick approximation for the hard-sphere potential [42]. Our results are presented in figs. 1-5, which show the translational short-time self-diffusion coefficient $D_s$, the sedimentation coefficient $K$, the hydrodynamic function $H(q)$, and the effective viscosity coefficient $\eta_{eff}$, respectively.

In the figs. 1-5 we also present results of the numerical simulations and the (revised) second order Beenakker-Mazur method [44], which is nowadays the most comprehensive theoretical scheme for calculations of the short-time transport properties of suspensions. At this point it is worth noting, that it is difficult to realize experimentally a monodisperse suspension of hard-spheres [46] and to measure its volume fraction precisely [47]. However, if the experimental conditions satisfy the assumptions underlying our theoretical model (such as monodisperse, hard-sphere potential, regime of the zero Reynolds number), then this suspension would have characteristics consistent with the precise numerical simulations. Therefore, in this article, we assess the theoretical methods for monodisperse hard-sphere suspensions by comparing them with numerical simulations, instead of comparing with the experimental works [48–56].
FIG. 2. Inverse of the sedimentation coefficient $K$, equation (39), as a function of volume fraction $\phi$ for monodisperse suspension of hard-spheres in equilibrium. Black (solid line) - the renormalized Clausius-Mossotti approximation introduced in this article, red (long-dashed) line - numerical simulations of Abade at al. [43], blue (short-dashed) line - the revised Beenakker-Mazur method [44], green (dot-dashed) line - Clausius-Mossotti approximation defined by the equation (103).

FIG. 3. The hydrodynamic function $H(q)$, equation (36), as a function of wave vector for volume fractions $\phi = 0.05$, $\phi = 0.15$ and $\phi = 0.25$, for monodisperse suspension of hard-spheres in equilibrium. Black (solid line) - the renormalized Clausius-Mossotti approximation introduced in this article, red (long-dashed) line - numerical simulations [43], blue (short-dashed) line - the revised Beenakker-Mazur method [44].

Our results for the effective viscosity $\eta_{\text{eff}}$ and the hydrodynamic function $H(q)$ (with its low and high $q$ behavior) presented in figs. 1-5 obtained within the renormalized Clausius-Mossotti approximation, when compared with the numerical simulations and the revised
FIG. 4. The hydrodynamic function $H(q)$, equation (36), as a function of wave vector for volume fractions $\phi = 0.35$ and $\phi = 0.45$, for monodisperse suspension of hard-spheres in equilibrium. Black (solid line) - the renormalized Clausius-Mossotti approximation introduced in this article, red (long-dashed) line - numerical simulations [43], blue (short-dashed) line - the revised Beenakker-Mazur method [44].

FIG. 5. The relative effective viscosity $\eta_{\text{eff}}/\eta$ (high frequency, low shear), equation (32), as a function of volume fraction $\phi$ for monodisperse suspension of hard-spheres in equilibrium. Black (solid line) - the renormalized Clausius-Mossotti approximation introduced in this article, red (long-dashed) line - numerical simulations of Ladd [45], blue (short-dashed) line - the revised Beenakker-Mazur method [44], green (dot-dashed) line - Saito formula [4].

second order Beenakker-Mazur method - can be summarized as follows. For the volume fractions $\phi \lesssim 30\%$, the relative error (with respect to the numerical simulations) of the renormalized Clausius-Mossotii approximation is a few times less or comparable with the relative
error of the Beenakker-Mazur method - it holds for the effective viscosity, the sedimentation coefficient, the self-diffusion coefficient, and for almost whole range of the wave vectors $q$ of the hydrodynamic function. The only exception is a range of the wave vectors $2 < 2aq < 5$ placed between $q = 0$ and $q$ in the principal maximum of the hydrodynamic function $H(q)$ (vide figs. 3 and 4). Here, the Beenakker-Mazur method is consistent with numerical simulations. For volume fractions above $\phi \approx 30\%$, the Beenakker-Mazur method leads to better agreement with numerical simulations than the renormalized Clausius-Mossotti approximation, for the effective viscosity and for most of the wave vectors $q$ of hydrodynamic function.

Before comparison of the results of our renormalized Clausius-Mossotti approximation defined by the equation (106) with the Clausius-Mossotti approximation defined by the formula (103), it has to be emphasized that the term Clausius-Mossotti approximation may refer to two different variants of approximation. First variant is given by the equations (103) and (102), and is used in this article. The second variant is also given by the equations (103) and (102), but instead of the two-body correlation function $h$, its lowest virial term, i.e. the Mayer function for hard spheres [57], is used. The Clausius-Mossotti approximation in case of the effective viscosity leads to the Saito formula (104), whereas in case of the hydrodynamic function denoted in this approximation by $H_{CM}(q)$ - because of the fact that for the large wave vectors hydrodynamic function is related to the self-scattering sequences and the Clausius-Mossotti approximation takes into account only one single particle-term among self-scattering sequences - it gives the following result

$$\lim_{q \to \infty} H_{CM}(q) = 1.$$ (109)

Therefore the self-diffusion coefficient in the Clausius-Mossotti approximation does not depend on the volume fraction of suspension. The opposite limit of the zero wave vector related to the sedimentation coefficient is presented in the fig. 2.

It is worth shedding light on the results of our article - the derivation of the ring expansion (84) and the formulation of the renormalized Clausius-Mossotti approximation - from the perspective of the hydrodynamic interactions and statistical physics. The Beenakker-Mazur method is currently the most comprehensive statistical physics method to calculate the short-time transport properties of suspensions. With this article, we introduce another method - the renormalized Clausius-Mossotti approximation. Neither of the above approximations take the two-body hydrodynamic interactions fully into account. It could
be verified by a simple analysis of relevant equations on the two-body level. Consequently, the strong hydrodynamic interactions of close particles in suspensions are disregarded in both approaches. Therefore, to construct a satisfactory method of calculations of transport properties of suspensions, which would take the two-body hydrodynamic interactions into consideration, remains an open problem of statistical physics. It is worth noting here, that to take the two-body hydrodynamic interactions fully into account in the Beenakker-Mazur renormalized fluctuation expansion, a resummation up infinite order is needed. An extension of the renormalized Clausius-Mossotti approximation to take the two-body hydrodynamic interactions fully into consideration can be done, e.g. by modification of the approximation (106), adding the two-body contributions. This type of extension is natural, because it goes along the line of a usual, systematic generalization of the Clausius-Mossotti approximation [41]. In order to fully grasp the two-body effect in the Beenakker-Mazur method, one needs to consider all orders of the fluctuation expansion. It is an important difference between the renormalized Clausius-Mossotti approximation and the Beenakker-Mazur method.

There is another intriguing point from the perspective of statistical physics. The renormalized Clausius-Mossotti approximation and the Beenakker-Mazur second order approach are similar, because an important element of both methods is an effective propagator. In the Beenakker-Mazur method, its role is played by the quantity $A_{\gamma_0}$, which depends on the volume fraction $\phi$, but does not depend on the distribution of particles, e.g. the two-body correlation function [16]. Therefore, this propagator $A_{\gamma_0}$ is the same for hard-sphere suspension and for suspension of charged particles in equilibrium. On the other hand, the effective propagator $G_{eff}$ in the ring expansion, on which the renormalized Clausius-Mossotti approximation is constructed, depends both, on the hydrodynamic interactions, and the distribution of particles. This confrontation gives rise to the question concerning sensitivity of both methods to the change in structure of suspension - when, for example, an electrostatic interparticle repulsion increases in a suspension and the volume fraction remains unchanged. This question in the case of the Beenakker-Mazur method has been answered in some situations: the Beenakker-Mazur method is rather insensitive to the change in the structure of suspension [21, 58]. We are going to address the above questions in our further work.
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Appendix A: Wave dependent sedimentation

The wave dependent sedimentation coefficient $H(q)$ describes response of quiescent suspension, $v_0 = 0$, cf. equations (11), under action of a sinusoidal external force density, e.g.

$$f_{\text{ext}}(r) = f_0 \hat{e}_z \text{Re} \exp(-iq\hat{e}_z \cdot r) \exp(-\eta |z|).$$  

(A1)

The above external force density corresponds to the plane wave in the direction of the wave vector $q = q\hat{e}_z$. For a moment, we also introduce the damping factor $\exp(-\eta |z|)$ and later we will take the limit $\eta \to 0^+$. The above external force is translationally invariant in $x$ and $y$ directions, $f_{\text{ext}}(r) = \hat{e}_z f_{\text{ext}}(z)$. For homogeneous and isotropic suspension that property induces the same form for the average velocity field, i.e. $\langle v(r) \rangle = \hat{e}_z \langle v(z) \rangle$. Incompressibility condition for the average velocity field $\langle v(r) \rangle = \hat{e}_z \langle v(z) \rangle$ implies $\langle v(z) \rangle = \text{const}$. The force given by equation (A1) has also the mirror symmetry in $z$ direction, $f_{\text{ext}}(z) = -f_{\text{ext}}(-z)$, which, for homogeneous and isotropic suspension, induces the same symmetry for the velocity field $\langle v(z) \rangle = -\langle v(-z) \rangle$. Along with the incompressibility condition $\langle v(z) \rangle = \text{const}$, we obtain, that the average velocity field vanishes in the whole suspension, $\langle v(r) \rangle = 0$.

This property of the zero net flux, $\langle v(r) \rangle = 0$, simplify the equation (26). In this situation, the upper component of the vector $[(\langle U \rangle, \langle f \rangle)]$ in the equation (26) is given by

$$\langle U(R, r) \rangle = \int d^3 R' d^3 r' P_U T^\text{intr}(R, r; R', r') P_U^T f_{\text{ext}}(r'),$$  

(A2)

were projector $P_U$ (with its transposition $P_U^T$), by definition projects on the upper half of the double vector $[(\langle U \rangle, \langle f \rangle)]$, as follows from the formula (38). From now on, we will take the limit $\eta \to 0^+$, in which the zero net flux condition, $\langle v(r) \rangle = 0$, remains.
The external force density given by equation (A1) is torque-free, because the torque acting on the particle at position $\mathbf{R}$ vanishes,

$$\int d^3 r \theta (|\mathbf{r} - \mathbf{R}| - a) (\mathbf{r} - \mathbf{R}) \times \mathbf{f}_{\text{ext}} (\mathbf{r}) = 0,$$

for any position $\mathbf{R}$. The Heaviside function $\theta (|\mathbf{r} - \mathbf{R}| - a)$ used here, vanishes outside the particle centered at $\mathbf{R}$ and is equal to 1 inside the volume of the particle. It is very simple to show, that in the case of the torque free external force density, action of $\mathbf{M}_>$ operator given by equation (9), for such external force density for hard spheres, can be written as

$$f_i (\mathbf{R}_1; \mathbf{r}) = \frac{1}{4 \pi a^3} \int d^3 r' \mathbf{M}_> (\mathbf{R}_1, \mathbf{r}, \mathbf{r}') \mathbf{F}_{\text{ext}} (\mathbf{R}_1), \quad (A3)$$

where $\mathbf{F}_{\text{ext}} (\mathbf{R})$ is the total force acting on a particle centered at position $\mathbf{R}$,

$$\mathbf{F}_{\text{ext}} (\mathbf{R}) = \int d^3 r \theta (|\mathbf{r} - \mathbf{R}| - a) \mathbf{f}_{\text{ext}} (\mathbf{r}). \quad (A4)$$

The equation (A3) is a straightforward consequence of the form of $\mathbf{M}_>$ given in the reference [30]. Similar holds for the single particle $\mathbf{M}_0$ operator,

$$U_1 (\mathbf{R}_1; \mathbf{r}) = \frac{1}{4 \pi a^3} \int d^3 r' \mathbf{M}_0 (\mathbf{R}_1, \mathbf{r}, \mathbf{r}') \mathbf{F}_{\text{ext}} (\mathbf{R}_1). \quad (A5)$$

Therefore, action of the operators $\mathbf{M}_>$ and $\mathbf{M}_0$ on the force density $\mathbf{f}_{\text{ext}}$ in equations (10), can be replaced using (A3) and (A5). It has the following implication in the equation (A2),

$$\langle U (\mathbf{R}, \mathbf{r}) \rangle = \frac{1}{4 \pi a^3} \int d^3 R' \int d^3 r' P_U T^{\text{irr}} (\mathbf{R}, \mathbf{r}; \mathbf{R}', \mathbf{r}') P_U^T \mathbf{F}_{\text{ext}} (\mathbf{R}'). \quad (A6)$$

In the problem of sedimentation, we consider the average translational velocity of particles,

$$\langle \mathbf{V} (\mathbf{R}) \rangle = \left\{ \sum_{i=1}^N \delta (\mathbf{R} - \mathbf{R}_i) \mathbf{V}_i (X) \right\}. \quad (A7)$$

For hard spheres, $\langle \mathbf{V} (\mathbf{R}) \rangle$ is related to the average velocity field of particles $\langle \mathbf{U} (\mathbf{R}, \mathbf{r}) \rangle$, by the formula

$$\langle \mathbf{V} (\mathbf{R}) \rangle = \frac{1}{4 \pi a^3} \int d^3 r \langle \mathbf{U} (\mathbf{R}, \mathbf{r}) \rangle,$$

which follows from the relation (2).

The expression (A6), after passing from the particle velocity field $\langle \mathbf{U} (\mathbf{R}, \mathbf{r}) \rangle$ for hard spheres, to the average translation velocity $\langle \mathbf{V} (\mathbf{R}) \rangle$, yields

$$\langle \mathbf{V} (\mathbf{R}) \rangle = \int d^3 R' \mathbf{Y} (\mathbf{R}, \mathbf{R}') \mathbf{F}_{\text{ext}} (\mathbf{R}'), \quad (A8)$$

where

$$\mathbf{Y} (\mathbf{R}, \mathbf{R}') = \frac{1}{(4 \pi a^3)^2} \int d^3 r \int d^3 r' P_U T^{\text{irr}} (\mathbf{R}, \mathbf{r}; \mathbf{R}', \mathbf{r}') P_U^T. \quad (A9)$$
For homogeneous suspension, the above kernel $\mathbf{Y} (\mathbf{R}, \mathbf{R}')$ depends only on the difference of positions, therefore we can introduce

$$\mathbf{Y} (\mathbf{R} - \mathbf{R}') \equiv \mathbf{Y} (\mathbf{R}, \mathbf{R}').$$  \hspace{1cm} (A10)

Moreover, isotropy implies, that $\mathbf{Y} (\mathbf{R})$ is of the form $y_0 (\mathbf{R}) \mathbf{1} + y_2 (\mathbf{R}) \hat{\mathbf{R}} \hat{\mathbf{R}}$. This form of $\mathbf{Y} (\mathbf{R})$, along with simple algebraic manipulations of equations (A8), leads to the following conclusion. For the external force given by a plane wave,

$$\mathbf{F}_{\text{ext}} (\mathbf{R}) = F_0 \hat{\mathbf{q}} \Re \exp (-i \mathbf{q} \cdot \mathbf{R}),$$  \hspace{1cm} (A11)

the average translational velocity has also the plane-wave form,

$$\langle \mathbf{V} (\mathbf{R}) \rangle = V (q) \hat{\mathbf{q}} \Re \exp (-i \mathbf{qR}),$$  \hspace{1cm} (A12)

where the coefficient $V (q)$ is given by the formula

$$V (q) = H (q) \mu_0 F_0,$$

with hydrodynamic function given by equation (36). The stokes coefficient $\mu_0 = 1/(6\pi \eta a)$.

**Appendix B: Calculations within renormalized Clausius-Mossotti approximation**

In this appendix we give some details of our calculations within the renormalized Clausius-Mossotti approximation. It demands to solve the set of equations (106), (105), (27), (78), (75) and (77) for the quantities $\mathbf{B}$, $\mathbf{T}_{\text{irr}}$, $\mathbf{G}_{\text{eff}}$, $\mathbf{T}_{\text{irr}}$, $\mathbf{T}$, $\mathbf{T}_{\text{off}}$. Each of those quantities is a fourfold $3 \times 3$ matrix, as it may be inferred for example from the formula (22) for the $\mathbf{T}$ operator. Therefore the $\mathbf{T}$ operator written with all variables and indexes is represented by $\mathbf{T} (\mathbf{R}, \mathbf{r}, \mathbf{R}', \mathbf{r}')_{u u', \alpha \alpha'}$. It has two indexes $u, u' = U, P$, which denotes upper or lower part of the double vector, and another two Cartesian indexes $\alpha, \alpha' = 1, 2, 3$. In our calculations we represent those quantities as multipole hydrodynamic matrices [59]. In the reference [30] the reader can find how to introduce a multipole picture for the forces and velocities and how to represent the hydrodynamic matrices $\mathbf{M}$ and $\mathbf{G}$ in the multipole picture. The notation used in the reference [30] is adopted also in this article, therefore we do not repeat that material. In the same way as in the reference [30] we introduce the multipole picture of the hydrodynamic matrices $\mathbf{B}$, $\mathbf{T}_{\text{irr}}$, $\mathbf{G}_{\text{eff}}$, $\mathbf{T}_{\text{irr}}$, $\mathbf{T}$ and $\mathbf{T}_{\text{off}}$. In the multipole picture, all
of the above quantities become infinite dimensional hydrodynamic multipole matrices, e.g. \( T(R, r; R', r')_{\alpha u'\sigma'} \to [T(R, R')]_{u\sigma, u'l'm'\sigma'} \). Therefore the variables \( r, r' \) and Cartesian indexes are transformed into multipole numbers \( l, m \) and \( \sigma \) having the following range: \( l = 1, 2, \ldots, \infty \), \( m = -l, -l+1, \ldots, l \), whereas \( \sigma = 0, 1, 2 \). An important role in our calculations is played by the homogeneity of the system, because it implies that the matrices depend on the difference of positions only, for example \([T(R - R')]_{u\sigma, u'l'm'\sigma'} \equiv [T(R, R')]_{u\sigma, u'l'm'\sigma'}\). We also use isotropy of the system, which allow to calculate the multipole matrix \( T(R) \) for any vector \( R \) when the \( T \) for \( z \) direction \( R = Re_z \) is known.

Using also the Fourier space, with the Fourier transformation given by

\[
\hat{T}(k) = \int d^3R \exp[-iKR]T(R), \quad (B1)
\]

with similar definition for other multipole hydrodynamic matrices, we represent the set of equations (106), (105), (27), (78), (75) and (77) as follows

\[
\hat{T}(k) = \hat{B} + \hat{T}_{\text{off}}(k), \quad (B2)
\]

\[
\hat{T}(k) = \hat{T}_{\text{irr}}(k) \left( 1 - \hat{G}(k)\hat{T}_{\text{irr}}(k) \right)^{-1}, \quad (B3)
\]

\[
\hat{G}_{\text{eff}}(k) = \hat{G}(k) + \hat{G}(k)\hat{T}(k)\hat{G}(k), \quad (B4)
\]

\[
\hat{T}_{\text{RCM}}(k) \approx \hat{B}, \quad (B5)
\]

\[
\hat{T}_{\text{irr}}(k) = \hat{T}_{\text{RCM}}(k) \left( 1 - [\hat{h}G_{\text{eff}}(k)]\hat{T}_{\text{RCM}}(k) \right)^{-1}, \quad (B6)
\]

\[
\hat{B} = n_1M + \int d^3R T_{\text{off}}(-R)G(R)M, \quad (B7)
\]

where

\[
[\hat{h}G_{\text{eff}}](k) = \int d^3R \exp[-iKR]h(R)G_{\text{eff}}(R). \quad (B8)
\]

In the above equations there appear superpositions of the multipole hydrodynamic matrices, inversion of matrices, and the matrices appear both in the positional and the Fourier space. We created numerical code to solve these equations of the renormalized Clausius-Mossotti approximation. Three aspects appear here.

First, in our numerical calculations, we truncate the hydrodynamic matrices, e.g. \([\hat{T}(k)]_{u\sigma, u'l'm'\sigma'}\). The truncation is characterized by \( L \), which is the highest multipole used in the calculations - we consider matrix elements with \( l, l' \leq L \) only. The calculations were performed for different parameters truncation, \( L = 4, \ldots, 10 \). The dependence of the effective viscosity
coefficient \( \eta_{\text{eff}}/\eta \) on the function \( 1/(L \log^3 L) \) of the truncation parameter \( L \) is presented in the fig. 6. The figure shows, that even for the highest truncation parameter \( L = 10 \) (which corresponds to \( 1/(L \log^3 L) \approx 0.008 \)) the effective viscosity coefficient is still sensitive to the truncation \( L \). Therefore, we extrapolate the coefficient up to \( L \to \infty \). The extrapolated value of the coefficient is given by intersection of a straight line, passing through the points corresponding to \( L = 9 \) and \( L = 10 \) in the fig. 6, with the vertical axis. Similar procedure of the extrapolation is carried in the case of the other transport characteristics.

The second numerical aspect is related to a discretization of distance \( R \) for hydrodynamic matrices, e.g. \( T(Re_z) \). Points \( R = \xi_0, \ldots, \xi_N \), with \( \xi_0 = 0 \) and \( \xi_n = \xi_1 \exp \left[ \alpha (i - 1) \right] \) were considered. The parameter \( \alpha \) was determined from the assumption, that the first and the last section are equal, i.e., \( \xi_1 = \xi_N - \xi_{N-1} \). Therefore, \( \xi_1 \) and \( N \) determine a set of points, in which the hydrodynamic matrices were considered in the code. Other values - if needed - were calculated by interpolation or from the asymptotic expansion (e.g. for \( R \) larger than \( \xi_N \)). It is worth noting that the exponential mesh is convenient to calculate the three dimensional Fourier transform of the hydrodynamic matrices. In our calculations the three dimensional Fourier transform of hydrodynamic matrices in the formula (B1) was first reduced to the one dimensional Hankel transform [60] in a similarity to a dielectric system [61]. Then the exponential mesh is used to perform Hankel transform with the use of numerical procedures for fast Fourier transform. We performed calculations for \( \xi_1/(2a) = 1/2, 1/3, 1/4, 1/5 \) and \( N = 512, 1024, 2048, 4096 \) respectively. Larger \( N \) correspond to a denser mesh. We found the mesh characterized by \( N = 4096 \) sufficient and it is used to obtain the results presented in this article.

The third aspect of the numerical calculations is related to the fact, that the system of equations was solved iteratively. We observed, that up to the volume fractions \( \phi \approx 45\% \), after a few iterations, difference between hydrodynamic functions in subsequent iterations decays as it happens in a geometric series. It may be written as follows

\[
\sup_q |H_i(q) - H_{i-1}(q)| \approx \sup_q |H_{i+1}(q) - H_i(q)| \Delta,
\]  

where \( H_i(q) \) denotes the hydrodynamic function after the \( i \)-th iteration and symbol \( \sup_q f(q) \) stands for maximal value of a function \( f \). The highest observed value of \( \Delta \approx 0.7 \). The iteration procedure was stopped when the following condition \( \sup_q |H_i(q) - H_{i-1}(q)| < 10^{-4} \) was satisfied.
It is worth mentioning, that the computer time and memory to solve the equations iteratively is comparable with the calculations, which we performed in the case of the revised Beenakker-Mazur method [30]. The numerical results within the renormalized Clausius-Mossotti approximation presented in this article were calculated with a use of a desktop computer within one day.

![Graph showing the relative effective viscosity coefficient $\eta_{eff}/\eta$ as a function of multipole truncation $L$ obtained by the renormalized Clausius-Mossotti approximation for suspension of volume fraction $\phi = 0.45$.](image)

**FIG. 6.** The relative effective viscosity coefficient $\eta_{eff}/\eta$ as a function of multipole truncation $L$ obtained by the renormalized Clausius-Mossotti approximation for suspension of volume fraction $\phi = 0.45$.

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