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

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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 η_{eff} of suspension with its structure on the microscopic level. His result $\eta_{\text{eff}}/\eta = 1 + 5\phi/2$, where η denotes the viscosity of solvent and ϕ 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 [3].

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 Fig. 1. 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 [4].

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 [5], 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 [6]. The first systematic extension of Einstein's work for more concentrated suspensions was performed by Peterson and Fixman [7]. They obtained a virial expansion of the effective viscosity up to the second order, which includes the two-body hydrodynamic interactions. It

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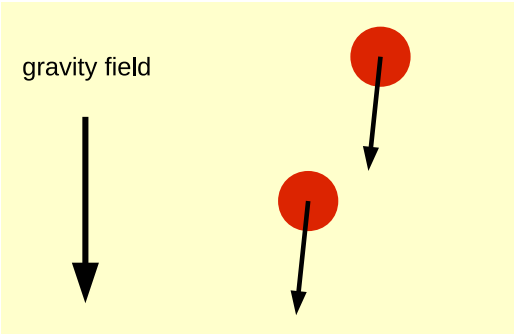


FIG. 1. (Color online) Example of hydrodynamic interactions: two particles sedimenting in the gravity field influence their mutual motion (the two vectors show the velocities of the particles).

was an approach in which the transport coefficient in the second order was given by absolutely convergent integrals. Despite 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 [8]. 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 Brown's approach, who obtained a similar result limited to the lowest terms in the single-particle polarizability expansion of a dielectric constant [9]. Felderhof, Ford, and Cohen also gave the microscopic explanation of the famous Clausius-Mossotti formula [10], which is an analog of the Saito formula [5] for the effective viscosity in the physics of suspensions. It is worth mentioning here the effective medium approaches [11,12] and their extensions including the two-body hydrodynamic interactions in a more accurate way [13,14].

Nowadays, the most prominent statistical physics approach to the short-time transport properties of suspensions is the Beenakker-Mazur method [15–17], which was developed and applied for different suspensions [18–23]. 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 [24–26] and from numerical simulations [27,28] 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 hydrodynamic 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 [8]. 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) [10]. 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 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 [25,26]. 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 dominant contributions is presented in the Ref. [25]. This idea is developed in the present article.

The outline of this article is as follows. In the second section, we describe the suspension on the microscopic level and discuss the macroscopic characteristics of suspensions. In the third section, we repeat the analysis of Felderhof, Ford, and Cohen leading to the cluster expansion of the macroscopic characteristics. In the fourth section, we introduce the ring expansion of the macroscopic characteristics, which is a rigorous result. The ring expansion is further used in the 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 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 η. 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 [29]. The stationary Stokes equations for the problem of the suspension of N spheres, centered at positions $X \equiv \mathbf{R}_1, \ldots, \mathbf{R}_N$, freely moving in ambient flow $\mathbf{v}_0(\mathbf{r})$, under action of external force density field $\mathbf{f}_{\text{ext}}(\mathbf{r})$, can be represented in the following integral form

[30,31]:

$$\mathbf{U}_i(X,\mathbf{r}) = \int d^3r' \mathbf{M}_0(\mathbf{R}_i,\mathbf{r},\mathbf{r}')\mathbf{f}_{\text{ext}}(\mathbf{r}') + \int d^3r' \mathbf{M}_<(\mathbf{R}_i,\mathbf{r},\mathbf{r}') \left[\mathbf{v}_0(\mathbf{r}') + \sum_{\substack{j=1 \\ j\neq i}}^{N} \int d^3r'' \mathbf{G}_0(\mathbf{r}',\mathbf{r}'')\mathbf{f}_j\big(X;\mathbf{r}''\big) \right],$$

$$\mathbf{f}_i(X,\mathbf{r}) = \int d^3r' \hat{\mathbf{M}}(\mathbf{R}_i,\mathbf{r},\mathbf{r}') \left[\mathbf{v}_0(\mathbf{r}') + \sum_{\substack{j=1 \\ j\neq i}}^{N} \int d^3r'' \mathbf{G}_0(\mathbf{r}',\mathbf{r}'')\mathbf{f}_j(X;\mathbf{r}'') \right] + \int d^3r' \mathbf{M}_>(\mathbf{R}_i,\mathbf{r},\mathbf{r}')\mathbf{f}_{\text{ext}}(\mathbf{r}). \quad (1)$$

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

$$\mathbf{U}_i(X;\mathbf{r}) = \mathbf{V}_i(X) + \boldsymbol{\Omega}_i(X) \times (\mathbf{r}-\mathbf{R}_i) \quad \text{for} \quad |\mathbf{r}-\mathbf{R}_i| \leqslant a, \quad (2)$$

with translational $\mathbf{V}_i$ and rotational $\boldsymbol{\Omega}_i$ velocity of the particles. Moreover, $\mathbf{f}_i(X;\mathbf{r})$ describes the force density [32–34] acting on the fluid by the surface of the particle number i and is defined by

$$\mathbf{f}_i(X;\mathbf{r}) = -\sigma(\mathbf{r}) \cdot \hat{\mathbf{n}}_i(\mathbf{r})\, \delta(|\mathbf{r}-\mathbf{R}_i| - a), \quad (3)$$

where σ represents the stress tensor in the fluid, $\hat{\mathbf{n}}_i(\mathbf{r}) = (\mathbf{r}-\mathbf{R}_i)/|\mathbf{r}-\mathbf{R}_i|$ is a vector normal to the surface of the sphere i, whereas $\delta(x)$ stands for the one-dimensional Dirac delta function. $\mathbf{G}_0(\mathbf{r})$ in Eqs. (1) is the Oseen tensor

$$\mathbf{G}_0(\mathbf{r}) = (\mathbf{1} + \hat{\mathbf{r}}\hat{\mathbf{r}})/(8\pi\eta|\mathbf{r}|), \quad (4)$$

with $\hat{\mathbf{r}} = \mathbf{r}/|\mathbf{r}|$. The Oseen tensor is a Green function of the Stokes equations [35], hence flow of the whole suspension $\mathbf{v}(\mathbf{r})$ is given by

$$\mathbf{v}(\mathbf{r}) = \mathbf{v}_0(\mathbf{r}) + \sum_{i=1}^{N} \int d^3r' \mathbf{G}_0(\mathbf{r}-\mathbf{r}')\mathbf{f}_i(X;\mathbf{r}'). \quad (5)$$

Equations (1) are linear both in the ambient flow $\mathbf{v}_0$ and in the external force density $\mathbf{f}_{\text{ext}}$. Therefore, to describe the response operators $\mathbf{M}_0$, $\mathbf{M}_<$, $\hat{\mathbf{M}}$, and $\mathbf{M}_>$, it is sufficient and convenient to consider special cases of a single-particle problem. $\mathbf{M}_0$ in Eqs. (1), in the case of the single-particle problem in the external force density field $\mathbf{f}_{\text{ext}}(\mathbf{r})$, and in absence of the ambient flow $\mathbf{v}_0 = 0$, yields the velocity field of the particle

$$\mathbf{U}_1(\mathbf{R}_1;\mathbf{r}) = \int d^3r' \mathbf{M}_0(\mathbf{R}_1,\mathbf{r},\mathbf{r}')\mathbf{f}_{\text{ext}}(\mathbf{r}'). \quad (6)$$

The single-particle operator $\mathbf{M}_<$ gives the particle velocity field $\mathbf{U}_1$, when the particle is placed in the ambient flow $\mathbf{v}_0$:

$$\mathbf{U}_1(\mathbf{R}_1;\mathbf{r}) = \int d^3r' \mathbf{M}_<(\mathbf{R}_1,\mathbf{r},\mathbf{r}')\mathbf{v}_0(\mathbf{r}'). \quad (7)$$

$\hat{\mathbf{M}}(\mathbf{R},\mathbf{r},\mathbf{r}')$, called the single-particle convective friction kernel, yields the force density $\mathbf{f}_1(\mathbf{R}_1;\mathbf{r})$ on the surface of the single particle at the position $\mathbf{R}_1$, when it is placed in the ambient flow $\mathbf{v}_0(\mathbf{r})$:

$$\mathbf{f}_1(\mathbf{R}_1;\mathbf{r}) = \int d^3r' \hat{\mathbf{M}}(\mathbf{R}_1,\mathbf{r},\mathbf{r}')\mathbf{v}_0(\mathbf{r}'). \quad (8)$$

Finally, $\mathbf{M}_>$ describes the force density $\mathbf{f}_1(\mathbf{R}_1;\mathbf{r})$ on the surface of the single particle at the position $\mathbf{R}_1$, under the action of the external force $\mathbf{f}_{\text{ext}}$:

$$\mathbf{f}_1(\mathbf{R}_1;\mathbf{r}) = \int d^3r' \mathbf{M}_>(\mathbf{R}_1,\mathbf{r},\mathbf{r}')\mathbf{f}_{\text{ext}}(\mathbf{r}'). \quad (9)$$

In this article, we investigate Eqs. (1) mostly without referring to the specific form of the response operators $\mathbf{M}_0$, $\mathbf{M}_<$, $\mathbf{M}_>$, $\hat{\mathbf{M}}$. For their detail form, we refer the reader to Refs. [31,36].

To facilitate further analysis of Eqs. (1), we omit integral variables in those equations, writing them in the following form:

$$\mathbf{U}_i(X) = \mathbf{M}_0(i)\mathbf{f}_{\text{ext}} + \mathbf{M}_<(i) \left[\mathbf{v}_0 + \sum_{\substack{j=1 \\ j\neq i}}^{N} \mathbf{G}_0\mathbf{f}_j(X) \right],$$

$$\mathbf{f}_i(X) = \hat{\mathbf{M}}(i) \left[\mathbf{v}_0 + \sum_{\substack{j=1 \\ j\neq i}}^{N} \mathbf{G}_0\mathbf{f}_j(X) \right] + \mathbf{M}_>(i)\mathbf{f}_{\text{ext}}. \quad (10)$$

For the position of the particle i in the single-particle response operators $\mathbf{M}_0$, $\mathbf{M}_<$, $\hat{\mathbf{M}}$, $\mathbf{M}_>$, we also use the following abbreviation: $i \equiv \mathbf{R}_i$. Finally, we write the above equations as follows [31]:

$$\begin{bmatrix} \mathbf{U}_i(X) \\ \mathbf{f}_i(X) \end{bmatrix} = \mathbf{M}(i) \left(\begin{bmatrix} \mathbf{f}_{\text{ext}} \\ \mathbf{v}_0 \end{bmatrix} + \sum_{\substack{j=1 \\ j\neq i}}^{N} \mathbf{G} \begin{bmatrix} \mathbf{U}_j(X) \\ \mathbf{f}_j(X) \end{bmatrix} \right), \quad (11)$$

introducing 6×6 dimensional matrices $\mathbf{M}$ and $\mathbf{G}$ defined by the equations

$$\mathbf{M}(\mathbf{R},\mathbf{r},\mathbf{r}') = \begin{bmatrix} \mathbf{M}_0(\mathbf{R},\mathbf{r},\mathbf{r}') & \mathbf{M}_<(\mathbf{R},\mathbf{r},\mathbf{r}') \\ \mathbf{M}_>(\mathbf{R},\mathbf{r},\mathbf{r}') & \hat{\mathbf{M}}(\mathbf{R},\mathbf{r},\mathbf{r}') \end{bmatrix} \quad (12)$$

and

$$\mathbf{G}(\mathbf{r},\mathbf{r}') = \begin{bmatrix} \mathbf{0} & \mathbf{0} \\ \mathbf{0} & \mathbf{G}_0(\mathbf{r},\mathbf{r}') \end{bmatrix}. \quad (13)$$

A. Scattering series

To solve Eq. (11), several methods can be used. One of the possible approaches is the method of reflections [3]. It relies

on taking successive iterations of Eq. (11) which leads to the following formula:

$$\begin{bmatrix}\mathbf{U}_i(X)\\ \mathbf{f}_i(X)\end{bmatrix} = \mathbf{M}(i)\begin{bmatrix}\mathbf{f}_{\text{ext}}\\ \mathbf{v}_0\end{bmatrix} + \sum_{\substack{j=1,\\ j\neq i}}^{N} \mathbf{M}(i)\mathbf{G}\mathbf{M}(j)\begin{bmatrix}\mathbf{f}_{\text{ext}}\\ \mathbf{v}_0\end{bmatrix} + \sum_{\substack{j=1,\\ j\neq i}}^{N}\sum_{\substack{k=1,\\ k\neq j}}^{N} \mathbf{M}(i)\mathbf{G}\mathbf{M}(j)\mathbf{G}\mathbf{M}(k)\begin{bmatrix}\mathbf{f}_{\text{ext}}\\ \mathbf{v}_0\end{bmatrix} + \dots . \tag{14}$$

The above expression of the force densities $\mathbf{f}_i(X)$ and the velocities of the particles $\mathbf{U}_i(X)$ has a form of a multiple scattering series. It means that $\mathbf{f}_i(X)$ and $\mathbf{U}_i(X)$ are given by the sum of the scattering sequences, for example,

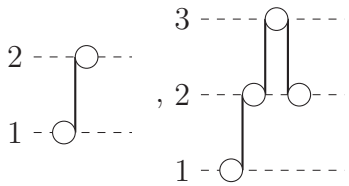
$$\mathbf{M}(1)\mathbf{G}\mathbf{M}(2) \tag{15}$$

and

$$\mathbf{M}(1)\mathbf{G}\mathbf{M}(2)\mathbf{G}\mathbf{M}(3)\mathbf{G}\mathbf{M}(2). \tag{16}$$

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

It is convenient and useful to represent the scattering sequences graphically [30]. The above two sequences can be represented, respectively, by



. (17)

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

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

$$\mathbf{T}_{ij}(X) = \mathbf{M}(i)\delta_{ij} + \mathbf{M}(i)\mathbf{G}\mathbf{M}(j)(1-\delta_{ij}) + \sum_{\substack{k=1\\ k\neq i, k\neq j}}^{N} \mathbf{M}(i)\mathbf{G}\mathbf{M}(k)\mathbf{G}\mathbf{M}(j) + \dots . \tag{18}$$

Therefore, the velocity $\mathbf{U}_i(X)$ and the force density $\mathbf{f}_i(X)$ in the expression (14) are given by the formula

$$\begin{bmatrix}\mathbf{U}_i(X)\\ \mathbf{f}_i(X)\end{bmatrix} = \sum_{j=1}^{N} \mathbf{T}_{ij}(X)\begin{bmatrix}\mathbf{f}_{\text{ext}}\\ \mathbf{v}_0\end{bmatrix}. \tag{19}$$

B. Macroscopic response

To describe properties of suspension on the macroscopic level, we consider an ensemble of configurations of particles $X \equiv \mathbf{R}_1, \dots, \mathbf{R}_N$, which is described by a probability distribution function $p(X)$. We also introduce the average force density defined by the equation

$$\langle \mathbf{f}(\mathbf{R},\mathbf{r})\rangle = \left\langle \sum_{i=1}^{N} \delta(\mathbf{R}-i)\mathbf{f}_i(X,\mathbf{r})\right\rangle \tag{20}$$

and the average particle velocity field

$$\langle \mathbf{U}(\mathbf{R},\mathbf{r})\rangle = \left\langle \sum_{i=1}^{N} \delta(\mathbf{R}-i)\mathbf{U}_i(X,\mathbf{r})\right\rangle, \tag{21}$$

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

$$\begin{bmatrix}\langle \mathbf{U}(\mathbf{R},\mathbf{r})\rangle\\ \langle \mathbf{f}(\mathbf{R},\mathbf{r})\rangle\end{bmatrix} = \int d^3R' d^3r' \mathbf{T}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}')\begin{bmatrix}\mathbf{f}_{\text{ext}}(\mathbf{r}')\\ \mathbf{v}_0(\mathbf{r}')\end{bmatrix}, \tag{22}$$

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

$$\mathbf{T}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}') = \left\langle \sum_{i=1}^{N}\sum_{j=1}^{N} \delta(\mathbf{R}-i)\mathbf{T}_{ij}(X,\mathbf{r},\mathbf{r}')\delta(\mathbf{R}'-j)\right\rangle. \tag{23}$$

Notice that in the above operator $\mathbf{T}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}')$, the Dirac delta functions fix positions of the first (i) and the last (j) particles in the scattering series $\mathbf{T}_{ij}$ at the positions $\mathbf{R}$ and $\mathbf{R}'$, respectively. The average flow of the suspension $\langle \mathbf{v}(\mathbf{r})\rangle$ is a combination of the ambient flow $\mathbf{v}_0(\mathbf{r})$, in which the particles are immersed and of flow generated by the presence of the particles

$$\langle \mathbf{v}(\mathbf{r})\rangle = \mathbf{v}_0(\mathbf{r}) + \int d^3R \int d^3r' \mathbf{G}_0(\mathbf{r},\mathbf{r}')\langle \mathbf{f}(\mathbf{R},\mathbf{r}')\rangle, \tag{24}$$

which is obtained by averaging the formula (5). We eliminate the flow $\mathbf{v}_0$ from Eqs. (24) and (22), which leads to the formula

$$\begin{bmatrix}\langle \mathbf{U}\rangle\\ \langle \mathbf{f}\rangle\end{bmatrix} = \mathbf{T}\begin{bmatrix}\mathbf{f}_{\text{ext}}\\ \langle \mathbf{v}\rangle\end{bmatrix} - \mathbf{T}\mathbf{G}\begin{bmatrix}\langle \mathbf{U}\rangle\\ \langle \mathbf{f}\rangle\end{bmatrix}, \tag{25}$$

in which we also facilitate the notation by omitting the integral variables. Its subsequent iterations lead to a relation of the average particle velocity $\langle \mathbf{U}\rangle$ and the force density $\langle \mathbf{f}\rangle$ to the external force density $\mathbf{f}_{\text{ext}}$ and the average flow of suspension $\langle \mathbf{v}\rangle$:

$$\begin{bmatrix}\langle \mathbf{U}(\mathbf{R},\mathbf{r})\rangle\\ \langle \mathbf{f}(\mathbf{R},\mathbf{r})\rangle\end{bmatrix} = \int d^3R' d^3r' \mathbf{T}^{\text{irr}}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}')\begin{bmatrix}\mathbf{f}_{\text{ext}}(\mathbf{r}')\\ \langle \mathbf{v}(\mathbf{r}')\rangle\end{bmatrix}, \tag{26}$$

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

$$\mathbf{T}^{\text{irr}} = \mathbf{T}(1+\mathbf{G}\mathbf{T})^{-1}. \tag{27}$$

Equation (26) is directly related to the macroscopic properties of the suspension. For example, the effective viscosity η_{eff} can be inferred from the relation between the average force density $\langle \mathbf{f}(\mathbf{R},\mathbf{r})\rangle$ and the average suspension flow $\langle \mathbf{v}(\mathbf{r})\rangle$, when no external forces act on the particles, $\mathbf{f}_{\text{ext}} = 0$. The relation between $\langle \mathbf{f}(\mathbf{R},\mathbf{r})\rangle$ and $\langle \mathbf{v}(\mathbf{r})\rangle$ in this situation results from Eq. (26), after projecting it into the lower half of the

double vectors $[\langle \mathbf{U}\rangle, \langle \mathbf{f}\rangle]$ and $[\mathbf{f}_{\rm ext}, \langle \mathbf{v}\rangle]$. In order to do that, we introduce a projector P_L defined by

$$P_L \begin{bmatrix} \langle \mathbf{U}\rangle \\ \langle \mathbf{f}\rangle \end{bmatrix} = \langle \mathbf{f}\rangle, \tag{28}$$

with its transposition P_L^T. After projection, Eq. (26) reads as

$$\langle \mathbf{f}(\mathbf{R},\mathbf{r})\rangle = \int d^3R' d^3r' P_L \mathbf{T}^{\rm irr}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}') P_L^T \langle \mathbf{v}(\mathbf{r}')\rangle. \tag{29}$$

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

$$X_{\alpha\beta\delta\gamma}(\mathbf{R},\mathbf{R}') = \int d^3r \int d^3r' (\mathbf{r}-\mathbf{R})_\alpha \times \left[P_L \mathbf{T}^{\rm irr}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}') P_L^T\right]_{\beta\delta} (\mathbf{r}'-\mathbf{R}')_\gamma, \tag{30}$$

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

$$X^{dd}_{\alpha\beta\delta\gamma}(\mathbf{R},\mathbf{R}') = \tfrac{1}{4}[X_{\alpha\beta\delta\gamma}(\mathbf{R},\mathbf{R}') + X_{\beta\alpha\delta\gamma}(\mathbf{R},\mathbf{R}') + X_{\alpha\beta\gamma\delta}(\mathbf{R},\mathbf{R}') + X_{\beta\alpha\gamma\delta}(\mathbf{R},\mathbf{R}')], \tag{31}$$

we obtain the effective viscosity $\eta_{\rm eff}$, using the formula [29,30]

$$\eta_{\rm eff} = \eta + \lim_\infty \frac{1}{10N} \sum_{\alpha,\beta=1}^{3} \int d^3R \int d^3R' X^{dd}_{\alpha\beta\beta\alpha}(\mathbf{R},\mathbf{R}'). \tag{32}$$

Thermodynamic limit $\lim_\infty$ is performed in the above equation.

Apart from the short-time effective viscosity $\eta_{\rm eff}$, 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

$$\mathbf{F}_{\rm ext}(\mathbf{r}) = F_0 \hat{\mathbf{q}}\, {\rm Re}\exp(-i\mathbf{q}\mathbf{R}). \tag{33}$$

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

$$\langle \mathbf{V}(\mathbf{r})\rangle = \left\langle \sum_{i=1}^{N} \delta(\mathbf{R}-\mathbf{R}_i)\mathbf{V}_i(X)\right\rangle, \tag{34}$$

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

$$\langle \mathbf{V}(\mathbf{r})\rangle = V(q)\hat{\mathbf{q}}\, {\rm Re}\exp(-i\mathbf{q}\mathbf{R}). \tag{35}$$

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 $H(q) \to 1$ in the limit of a diluted suspension, i.e., when the volume fraction $\phi \to 0$. As we also discuss in Appendix A, the microscopic expression for the hydrodynamic function $H(q)$ has the following form [30]:

$$H(q) = \frac{1}{\mu_0}\frac{1}{3}{\rm Tr}\left[\int d^3R\, e^{-i\mathbf{q}\cdot\mathbf{R}} Y(\mathbf{r})\right], \tag{36}$$

where 3×3 matrix $Y(\mathbf{r})$ is defined by the equation

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

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

$$P^U \begin{bmatrix} \langle \mathbf{U}\rangle \\ \langle \mathbf{f}\rangle \end{bmatrix} = \langle \mathbf{U}\rangle. \tag{38}$$

P_U^T denotes transposition of P_U.

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,

$$K = H(q=0) \tag{39}$$

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

$$D_c = D_0 H(q=0), \tag{40}$$

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

$$D_s = D_0 H(q\to\infty). \tag{41}$$

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

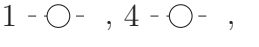
Both the effective viscosity and the hydrodynamic function can be inferred from the $\mathbf{T}^{\rm irr}$ operator. It is shown by the expressions (30)–(32) for the effective viscosity $\eta_{\rm eff}$ and by Eqs. (36) and (37) for the wave dependent sedimentation coefficient $H(q)$. Therefore, $\mathbf{T}^{\rm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

III. FELDERHOF, FORD, AND COHEN ANALYSIS OF $\mathbf{T}^{\rm irr}$

In the first stage of our analysis of $\mathbf{T}^{\rm irr}$ defined by Eq. (26), we follow the idea of Felderhof, Ford, and Cohen. They obtained the microscopic expression for $\mathbf{T}^{\rm irr}$ for the dielectric system in the form of a cluster expansion [8]. The application of their idea to the physics of suspensions is straightforward because the governing equations are similar for suspensions and dielectrics [30,37]. To perform the cluster expansion of the operator $\mathbf{T}^{\rm irr}$ on the basis of the expression (27), Felderhof, Ford, and Cohen introduced the cluster expansion of the operator $\mathbf{T}$.

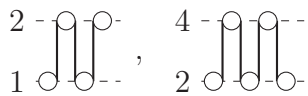
A. Cluster expansion of T

In the expression (23), $\mathbf{T}_{ij}(\mathbf{r},\mathbf{r}';X)$ 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.,



(42)

two-particle scattering sequences, e.g.,



(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:

$$\begin{aligned}&\mathbf{T}^{(s)}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C)\\&\quad=\text{all } s\text{-particle scattering sequences from}\\&\qquad\times\sum_{i=1}^{N}\sum_{j=1}^{N}\delta(\mathbf{R}-i)\mathbf{T}_{ij}(\mathbf{r},\mathbf{r}';X)\delta(\mathbf{R}'-j),\\&\quad\text{which include all particles from } s\text{-particle group } C.\end{aligned}\tag{44}$$

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

$$\begin{aligned}&\sum_{i=1}^{N}\sum_{j=1}^{N}\delta(\mathbf{R}-i)\mathbf{T}_{ij}(\mathbf{r},\mathbf{r}';X)\delta(\mathbf{R}'-j)\\&\quad=\sum_{s=1}^{N}\sum_{C\subset X,|C|=s}\mathbf{T}^{(s)}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C).\end{aligned}\tag{45}$$

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 Eq. (45) over the probability distribution function leads to the cluster expansion for the average scattering series $\mathbf{T}$ given by Eq. (23):

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

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 $\binom{N}{s}$. It yields

$$\mathbf{T}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}')=\left\langle\sum_{s=1}^{N}\binom{N}{s}\mathbf{T}^{(s)}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||1\ldots s)\right\rangle.\tag{47}$$

Introducing s-particle distribution functions defined by

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

we obtain the cluster expansion of the response operator $\mathbf{T}$ in the following form:

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

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

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 $\mathbf{T}^{\text{irr}}$ operator, Felderhof, Ford, and Cohen used the relation (27), which may be represented in the form

$$\mathbf{T}^{\text{irr}}=\mathbf{T}-\mathbf{TGT}+\mathbf{TGTGT}-\ldots.\tag{51}$$

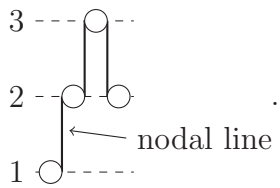
Let us look at the second term, i.e., $\mathbf{TGT}$. Representing the $\mathbf{T}$ by the cluster expansion (49), produces many terms, each of the form

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

In the expression $\mathbf{T}^{(s_1)}\mathbf{G}\mathbf{T}^{(s_2)}$, the scattering sequences between s_1 particles appearing in $\mathbf{T}^{(s_1)}$ are "connected" by the Green function $\mathbf{G}$ with the scattering sequences consisted of s_2 particles appearing in $\mathbf{T}^{(s_2)}$. Altogether, $\mathbf{T}^{(s_1)}\mathbf{G}\mathbf{T}^{(s_2)}$ forms s_1+s_2-particle scattering sequences. The scattering sequences built from the $\mathbf{T}^{(s_1)}\mathbf{G}\mathbf{T}^{(s_2)}$ are of a special type, i.e., there is a line $\mathbf{G}$ connecting a particle from $\mathbf{T}^{(s_1)}$ to a particle from $\mathbf{T}^{(s_2)}$. This line $\mathbf{G}$ is the only "connection" between the particles from $\mathbf{T}^{(s_1)}$ and $\mathbf{T}^{(s_2)}$. It is critical to distinguish the lines $\mathbf{G}$, which are the only connections between some groups of the particles in a scattering sequence. Those $\mathbf{G}$ are called the nodal lines [30]. It is described by the following examples. In the scattering sequence given by expression (15), there is one propagator $\mathbf{G}$. 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 $\mathbf{G}$. The underlined propagator

$$\mathbf{M}(1)\underline{\mathbf{G}}\mathbf{M}(2)\mathbf{G}\mathbf{M}(3)\mathbf{G}\mathbf{M}(2)\tag{53}$$

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 as



.(54)

It is easy to identify a nodal line in diagrammatic language: if cutting a line of a propagator $\mathbf{G}$ divides the diagram into two separate pieces, then the propagator $\mathbf{G}$ 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 $\mathbf{T}^{\mathrm{irr}}$ is the notion of the nodal line. Therefore, we perform further division of the scattering series $\mathbf{T}$, by specifying the nodal lines in the scattering sequences.

In agreement with the definition (44), $\mathbf{T}^{(s)}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||1\ldots s)$ 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 $\bar{\mathbf{T}}^{(s)}(\mathbf{R},\mathbf{r};\mathbf{R}'',\mathbf{r}''||1\ldots s)$ the scattering sequences with specified groups of particles $C_1,\ldots,C_g$ separated by nodal lines, defining

$$\begin{aligned}&\bar{\mathbf{T}}^{(s)}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C_1|\ldots|C_g)\\ &\quad= \text{all scattering sequences from}\\ &\qquad\mathbf{T}^{(s)}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C_1\ldots C_g) \text{ with } g-1 \text{ nodal lines}\\ &\qquad\text{separating particles from the groups } C_1,\ldots,C_g. \end{aligned}\tag{55}$$

With the above definition, the s-particle scattering sequences can be divided as follows:

$$\begin{aligned}\mathbf{T}^{(s)}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C) = &\sum_{g=1}^{s}\sum_{s_1+\ldots+s_g=s}\sum_{\substack{C_1,\ldots,C_g\subset C,\\ |C_1|+\ldots+|C_g|=s}}\\ &\times\ \bar{\mathbf{T}}^{(s)}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C_1|\ldots|C_g).\end{aligned}\tag{56}$$

Here, $\sum_{\substack{C_1,\ldots,C_g\subset C,\\ |C_1|=s_1;\ldots;|C_g|=s_g}}$ 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 $\bar{\mathbf{T}}^{(s)}(C)$ in the expression (56), i.e., the term with $g=1$, which have no nodal lines. The definition (55) implies that $\bar{\mathbf{T}}^{(s)}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C) =$ all scattering sequences from $\mathbf{T}^{(s)}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C)$ without nodal lines. The above scattering sequences without nodal lines play a significant role. They are called the irreducible scattering sequences [30].

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

$$\sum_{s_1+s_2=s}\sum_{\substack{C_1,C_2\subset C,\\ |C_1|+|C_2|=s}}\bar{\mathbf{T}}^{(s)}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C_1|C_2).\tag{57}$$

From the definition (55), it follows that $\bar{\mathbf{T}}^{(s)}(C_1|C_2)$ has one nodal line $\mathbf{G}$ separating the particles from the groups C_1 and C_2. Therefore, all scattering sequences in $\bar{\mathbf{T}}^{(s)}(C_1|C_2)$ have the following structure: first, there are some reflections between the particles from the group C_1, then there is exactly one reflection $\mathbf{G}$ 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 $\bar{\mathbf{T}}^{(s)}(C_1|C_2)$ has the following structure:

$$\bar{\mathbf{T}}^{(s)}(C_1|C_2) = \bar{\mathbf{T}}^{(s_1)}(C_1)\mathbf{G}\bar{\mathbf{T}}^{(s_2)}(C_2),\tag{58}$$

in which the irreducible scattering sequences $\bar{\mathbf{T}}^{(s)}(C)$ 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),

$$\bar{\mathbf{T}}^{(s)}(C_1|\ldots|C_g) = \bar{\mathbf{T}}^{(s_1)}(C_1)\mathbf{G}\ldots\mathbf{G}\bar{\mathbf{T}}^{(s_g)}(C_g),\tag{59}$$

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 $\bar{\mathbf{T}}^{(s_i)}(C_i)$ of the scattering sequences $\bar{\mathbf{T}}^{(s)}(C_1|\ldots|C_g)$. Each irreducible section $\bar{\mathbf{T}}^{(s_i)}(C_i)$ 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:

$$\mathbf{S}_I(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C) = \frac{1}{s!}\bar{\mathbf{T}}^{(s)}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C),\tag{60}$$

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

$$\begin{aligned}\mathbf{T}^{(s)}(C) = &\sum_{g=1}^{s}\sum_{s_1+\ldots+s_g=s}\sum_{\substack{C_1,\ldots,C_g\subset C,\\ |C_1|+\ldots+|C_g|=s}} s_1!\ldots s_g!\\ &\times\ \mathbf{S}_I(C_1)\mathbf{G}\ldots\mathbf{G}\mathbf{S}_I(C_g).\end{aligned}\tag{61}$$

The above representation used in the cluster expansion of $\mathbf{T}$ operator represented by Eqs. (49) and (50), after a simple algebra, leads to the expression

$$\begin{aligned}\mathbf{T} = &\sum_{g=1}^{\infty}\sum_{C_1,\ldots,C_g}\int dC_1\ldots dC_g\, n(C_1,\ldots,C_g)\\ &\times\ \mathbf{S}_I(C_1)\mathbf{G}\ldots\mathbf{G}\mathbf{S}_I(C_g).\end{aligned}\tag{62}$$

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:

$$\begin{aligned}&\sum_{C_1,\ldots,C_g}\int dC_1\ldots dC_g\, f(C_1,\ldots,C_g)\\ &\quad=\sum_{n_1=1}^{\infty}\ldots\sum_{n_g=1}^{\infty}\int d^3R_1^1\ldots d^3R_{n_1}^1\ldots d^3R_1^g\ldots d^3R_{n_g}^g\\ &\qquad\times\ f\left(\mathbf{R}_1^1,\ldots,\mathbf{R}_{n_1}^1,\ldots,\mathbf{R}_1^g,\ldots,\mathbf{R}_{n_g}^g\right).\end{aligned}\tag{63}$$

D. Cluster expansion of $\mathbf{T}^{\mathrm{irr}}$

We go back to the expression (51),

$$\mathbf{T}^{\mathrm{irr}} = \mathbf{T} - \mathbf{TGT} + \mathbf{TGTGT} - \ldots, \tag{64}$$

in order to derive the cluster expansion of the $\mathbf{T}^{\mathrm{irr}}$ operator. As we discussed before, the average scattering series $\mathbf{T}$ includes all possible scattering sequences. $\mathbf{TGT}$ in Eq. (64) produces scattering sequences with at least one nodal line, $\mathbf{TGTGT}$ 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 Eq. (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 $\mathbf{T}$ of Eq. (64) because the higher terms $\mathbf{TGT}$, $\mathbf{TGTGT}$, ... include at least one nodal line. In the expression (62) for $\mathbf{T}$, the irreducible scattering sequences come only from the term $g = 1$. Therefore, all terms on the right-hand side of Eq. (64) with the block structure C_1, are of the form

$$n(C_1)\mathbf{S}_I(C_1). \tag{65}$$

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

$$[n(C_1,C_2) - n(C_1)n(C_2)]\mathbf{S}_I(C_1)\mathbf{G}\mathbf{S}_I(C_2). \tag{66}$$

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 form

$$\begin{aligned}[n(C_1C_2C_3) - n(C_1C_2)n(C_3) - n(C_1)n(C_2C_3)\\ + n(C_1)n(C_2)n(C_3)]\mathbf{S}_I(C_1)\mathbf{G}\mathbf{S}_I(C_2)\mathbf{G}\mathbf{S}_I(C_3).\end{aligned} \tag{67}$$

In such manner, analysis of all block structures $C_1|\ldots|C_g$ is possible. Functions appearing along with the block structures of the form $\mathbf{S}_I(C_1)\mathbf{G}\ldots\mathbf{G}\mathbf{S}_I(C_g)$, are denoted by $b(C_1|\ldots|C_g)$ and are called the block distribution functions [8]. Therefore, we have

$$b(C_1) = n(C_1), \tag{68}$$

$$b(C_1|C_2) = n(C_1,C_2) - n(C_1)n(C_2), \tag{69}$$

$$\begin{aligned}b(C_1|C_2|C_3) = n(C_1C_2C_3) - n(C_1C_2)n(C_3)\\ - n(C_1)n(C_2C_3) + n(C_1)n(C_2)n(C_3)\end{aligned} \tag{70}$$

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 [30,38]:

$$b(C) = n(C), \tag{71a}$$

$$\begin{aligned}b(C_1|\ldots|C_kC_{k+1}|\ldots|C_g) = b(C_1|\ldots|C_k)b(C_{k+1}|\ldots|C_g)\\ + b(C_1|\ldots|C_k|C_{k+1}|\ldots|C_g).\end{aligned} \tag{71b}$$

The above analysis leads to the following cluster expansion of the $\mathbf{T}^{\mathrm{irr}}$ operator:

$$\begin{aligned}\mathbf{T}^{\mathrm{irr}} = \sum_{g=1}^{\infty}\sum_{C_1,\ldots,C_g}\int dC_1\ldots dC_g b(C_1|\ldots|C_g)\\ \times\ \mathbf{S}_I(C_1)\mathbf{G}\ldots\mathbf{G}\mathbf{S}_I(C_g).\end{aligned} \tag{72}$$

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

E. Self-scattering sequences

There are phenomena in the physics of suspensions in which only a part of the scattering series $\mathbf{T}$, given by Eq. (23), plays a role. An example of the above is the self-diffusion coefficient. It is related only to those scattering sequences in $\mathbf{T}$, 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 $\mathbf{S}_I^{\mathrm{self}}$ are related to the irreducible scattering sequences $\mathbf{S}_I$, as follows:

$$\begin{aligned}&\mathbf{S}_I^{\mathrm{self}}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C)\\ &\quad= \text{all scattering sequences from } \mathbf{S}_I(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C),\\ &\quad\text{which start and end at the same particle.}\end{aligned} \tag{73}$$

The average of the self-scattering sequences will be denoted by $\mathbf{B}$:

$$\mathbf{B}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}') = \sum_{C_1}\int dC_1\, n(C_1)\mathbf{S}_I^{\mathrm{self}}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||C_1). \tag{74}$$

With the above quantity, the scattering sequences in $\mathbf{T}$ 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 $\mathbf{B}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}')$, the latter are denoted by $\mathbf{T}_{\mathrm{off}}$. Therefore,

$$\mathbf{T} = \mathbf{B} + \mathbf{T}_{\mathrm{off}}. \tag{75}$$

Operator $\mathbf{T}^{\mathrm{irr}}$ can be divided in a similar manner:

$$\mathbf{T}^{\mathrm{irr}} = \mathbf{B} + \mathbf{T}_{\mathrm{off}}^{\mathrm{irr}}. \tag{76}$$

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

It is worth noting that the self-scattering sequences $\mathbf{B}$ can be calculated from off-scattering sequences $\mathbf{T}_{\mathrm{off}}$ as

follows [39]:

$$\mathbf{B}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}') = n_1\delta(\mathbf{R}-\mathbf{R}')\mathbf{M}(\mathbf{R},\mathbf{r},\mathbf{r}')+\delta(\mathbf{R}-\mathbf{R}') \times [\mathbf{T}_{\text{off}}\mathbf{G}\mathbf{M}](\mathbf{R},\mathbf{r};\mathbf{R},\mathbf{r}'). \quad (77)$$

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 $\mathbf{T}_{\text{off}}$ to some particle, and then they come back with one reflection $\mathbf{G}$ 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 $\mathbf{T}^{\text{irr}}$

The cluster expansion (72) of the response operator $\mathbf{T}^{\text{irr}}$ was derived by Felderhof, Ford, and Cohen about three decades ago [8]. 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 $\mathbf{G}_{\text{eff}}$ defined by

$$\mathbf{G}_{\text{eff}} = \mathbf{G} + \mathbf{G}\mathbf{T}\mathbf{G}. \quad (78)$$

In the case of homogeneous system in the thermodynamic limit, the effective Green function $\mathbf{G}_{\text{eff}}$ of suspension is related to the Green function $\mathbf{G}$ of pure fluid according to the following formula [30]:

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

The above formula describes the asymptotic decay of the effective propagator $\mathbf{G}_{\text{eff}}(\mathbf{r},\mathbf{r}')$ for the large distances between the points $\mathbf{r}$ and $\mathbf{r}'$. 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 $\mathbf{T}$ and $\mathbf{T}^{\text{irr}}$, which can be inverted and then represented in the following way:

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

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

$$\mathbf{T}_{\text{off}} = \mathbf{T}^{\text{irr}}_{\text{off}} + \mathbf{T}^{\text{irr}}\mathbf{G}_{\text{eff}}\mathbf{T}^{\text{irr}}, \quad (81)$$

obtained with the application of the formulas (76) and (75). Let us notice that $\mathbf{T}_{\text{off}}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}') = 0$ for overlapping configurations, i.e., for $|\mathbf{R}-\mathbf{R}'| < 2a$. 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 $\mathbf{T}_{\text{off}}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}')$ for the overlapping configurations has a consequence in the last equation. It reduces to

$$f(\mathbf{R}-\mathbf{R}')\mathbf{T}^{\text{irr}}_{\text{off}}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}') = -f(\mathbf{R}-\mathbf{R}')[\mathbf{T}^{\text{irr}}\mathbf{G}_{\text{eff}}\mathbf{T}^{\text{irr}}](\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'), \quad (82)$$

after its multiplication by a function $f(\mathbf{R}-\mathbf{R}')$ defined by

$$f(\mathbf{R}-\mathbf{R}') = \begin{cases} 1 & \text{for } |\mathbf{R}-\mathbf{R}'| < 2a, \\ 0 & \text{for } |\mathbf{R}-\mathbf{R}'| \geqslant 2a, \end{cases} \quad (83)$$

which equals 0 for nonoverlapping and 1 for overlapping configurations of the two particles centered at $\mathbf{R}$ and $\mathbf{R}'$.

Equation (82) shows that in $\mathbf{T}^{\text{irr}}$, there are some contributions with the effective propagator $\mathbf{G}_{\text{eff}}$. It suggests that the propagator $\mathbf{G}$ appearing in the cluster expansion (72) of Felderhof, Ford, and Cohen can be renormalized. In other words, $\mathbf{T}^{\text{irr}}$ can be given by the formula

$$\mathbf{T}^{\text{irr}} = \sum_{d=1}^{\infty}\sum_{C_1\ldots C_d}\int dC_1\ldots dC_d H(C_1|\ldots|C_d)\mathbf{S}_I(C_1) \times \mathbf{G}_{\text{eff}}\ldots\mathbf{G}_{\text{eff}}\mathbf{S}_I(C_d), \quad (84)$$

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

Following, 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 $\mathbf{S}_I(C_1)$, i.e., the block structure without a nodal line. In the expression (84), the block structure $\mathbf{S}_I(C_1)$ without a nodal line appears only in the term $d = 1$. In the expression (72), the block structure $\mathbf{S}_I(C_1)$ 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 $\mathbf{S}_I(C_1)$, is possible if we assume

$$b(C_1) = H(C_1). \quad (85)$$

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 $\mathbf{G}_{\text{eff}}$,

$$\mathbf{G}_{\text{eff}} = \sum_{g=0}^{\infty}\sum_{C_1,\ldots,C_g}\int dC_1\ldots dC_g\, n(C_1,\ldots,C_g) \times \mathbf{G}\mathbf{S}_I(C_1)\mathbf{G}\ldots\mathbf{G}\mathbf{S}_I(C_g)\mathbf{G}, \quad (86)$$

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 $\mathbf{S}_I(C_i)\mathbf{G}_{\rm eff}\mathbf{S}_I(C_j)$. Therefore, the scattering sequences $\mathbf{S}_I(C_1)\mathbf{G}\mathbf{S}_I(C_2)\mathbf{G}\mathbf{S}_I(C_3)\mathbf{G}\mathbf{S}_I(C_4)$ appear in the ring expansion (84) in the term corresponding to $d = 2$,

$$H(C_{i_1}|C_{i_2})\mathbf{S}_I(C_{i_1})\mathbf{G}_{\rm eff}\mathbf{S}_I(C_{i_2}), \tag{87}$$

in the term $d = 3$,

$$H(C_{i_1}|C_{i_2}|C_{i_3})\mathbf{S}_I(C_{i_1})\mathbf{G}_{\rm eff}\mathbf{S}_I(C_{i_2})\mathbf{G}_{\rm eff}\mathbf{S}_I(C_{i_3}), \tag{88}$$

and in the term $d = 4$,

$$H(C_{i_1}|C_{i_2}|C_{i_3}|C_{i_4})\mathbf{S}_I(C_{i_1})\mathbf{G}_{\rm eff}\mathbf{S}_I(C_{i_2})\mathbf{G}_{\rm eff}\mathbf{S}_I(C_{i_3})\mathbf{G}_{\rm eff}\mathbf{S}_I(C_{i_4}). \tag{89}$$

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 $d \geqslant 5$ 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 $n(C_2C_3)\mathbf{G}\mathbf{S}_I(C_2)\mathbf{G}\mathbf{S}_I(C_3)\mathbf{G}$ in the expansion (86) produces the block structure $C_1|C_2|C_3|C_4$, yielding

$$H(C_1|C_4)n(C_2C_3)\mathbf{S}_I(C_1)\mathbf{G}\mathbf{S}_I(C_2)\mathbf{G}\mathbf{S}_I(C_3)\mathbf{G}\mathbf{S}_I(C_4). \tag{90}$$

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 $\mathbf{G}_{\rm eff}$ 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

$$H(C_1|C_2|C_4)n(C_3)\mathbf{S}_I(C_1)\mathbf{G}\mathbf{S}_I(C_2)\mathbf{G}\mathbf{S}_I(C_3)\mathbf{G}\mathbf{S}_I(C_4). \tag{91}$$

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

$$H(C_1|C_3|C_4)n(C_2)\mathbf{S}_I(C_1)\mathbf{G}\mathbf{S}_I(C_2)\mathbf{G}\mathbf{S}_I(C_3)\mathbf{G}\mathbf{S}_I(C_4). \tag{92}$$

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 $\mathbf{G}_{\rm eff}$ introduce only one nodal line $\mathbf{G}$. In this case, we have $i_1 = 1, i_2 = 2, i_3 = 3, i_4 = 4$ and obtain the following contribution:

$$H(C_1|C_2|C_3|C_4)\mathbf{S}_I(C_1)\mathbf{G}\mathbf{S}_I(C_2)\mathbf{G}\mathbf{S}_I(C_3)\mathbf{G}\mathbf{S}_I(C_4). \tag{93}$$

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 Eq. (72), lead to equality

$$\begin{aligned} b(C_1|C_2|C_3|C_4) = {} & H(C_1|C_4)n(C_2C_3)+H(C_1|C_2|C_4)n(C_3) \\ & + H(C_1|C_3|C_4)n(C_2)+H(C_1|C_2|C_3|C_4). \end{aligned} \tag{94}$$

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

$$b(C_1|C_2) = H(C_1|C_2), \tag{95}$$

$$b(C_1|C_2|C_3) = H(C_1|C_2|C_3) + H(C_1|C_3)n(C_2). \tag{96}$$

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 $C_1|\ldots|C_g$ consisting of $g \geqslant 2$ 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 $\mathbf{S}_I(C_1)\mathbf{G}_{\rm eff}\ldots\mathbf{G}_{\rm eff}\mathbf{S}_I(C_d)$. 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 $\mathbf{S}_I(C_1)\mathbf{G}_{\rm eff}\ldots\mathbf{G}_{\rm eff}\mathbf{S}_I(C_d)$ 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 $\mathbf{S}_I$ in the expression $\mathbf{S}_I(C_1)\mathbf{G}_{\rm eff}\ldots\mathbf{G}_{\rm eff}\mathbf{S}_I(C_d)$. 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 $\mathbf{S}_I(C_1)\mathbf{G}_{\rm eff}\ldots\mathbf{G}_{\rm eff}\mathbf{S}_I(C_d)$, which produces the block structure $\mathbf{S}_I(C_1)\mathbf{G}\ldots\mathbf{G}\mathbf{S}_I(C_g)$. Comparison of all terms in the expansions (84) and (72), producing the block structure $\mathbf{S}_I(C_1)\mathbf{G}\ldots\mathbf{G}\mathbf{S}_I(C_g)$, yields

$$\begin{aligned} b(C_1|\ldots|C_g) = \sum_{d=2}^{g} \sum_{1=i_1<i_2<\ldots<i_d=g} & H(C_{i_1}|\ldots|C_{i_d}) \\ & \times n(\{C_{i_1}\ldots C_{i_2}\}\backslash\{C_{i_1}C_{i_2}\})\ldots \\ & \times n(\{C_{i_{d-1}}\ldots C_{i_d}\}\backslash\{C_{i_{d-1}}C_{i_d}\}), \end{aligned} \tag{97}$$

which is valid for $g \geqslant 2$. The symbol "\" denotes a difference of sets of the particles, e.g., $\{12567\}\backslash\{56\} = \{127\}$. We assume that for the empty set $n(\varnothing) = 1$. 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 $\mathbf{T}^{\rm irr}$. 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 $\mathbf{S}_I$ connected by the propagators ($\mathbf{G}$ or $\mathbf{G}_{\rm eff}$) are averaged over configurations of particles, weighted with the distribution functions (b or H). From that perspective, and due to the fact that the effective Green function $\mathbf{G}_{\rm eff}$ appears in the ring expansion instead of $\mathbf{G}$, 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 $\mathbf{G}$, which appears in the cluster expansion (72), includes only information concerning liquid. On the other hand, the effective Green function $\mathbf{G}_{\rm eff}$ in the expression (84) contains macroscopic information about suspension. It is exhibited by the appearance of the effective viscosity $\eta_{\rm eff}$ in the asymptotic form of the effective propagator for large distances showed in the formula (79). The effective viscosity $\eta_{\rm eff}$ of hard-sphere suspension may significantly differ from the viscosity η of pure liquid, especially for the higher volume fractions ϕ.

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:

$$n(C_1C_2) \to n(C_1)n(C_2). \tag{98}$$

A use of the above cluster property of the distribution functions n in Eq. (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:

$$b(C_1|C_2|C_3) \longrightarrow b(C_1|C_3)b(C_2). \tag{99}$$

The cluster property of the distribution functions n applied in Eq. (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:

$$H(C_1|C_2|C_3) \longrightarrow 0. \tag{100}$$

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 $\mathbf{S}_I(C_i)$ and $\mathbf{S}_I(C_{i+1})$ in the ring expansion (84) are "connected" by both the effective propagator $\mathbf{G}_{\text{eff}}$ 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 $\mathbf{S}_I(C_i)$ and $\mathbf{S}_I(C_{i+1})$ forms a "ring."

The ring expansion of $\mathbf{T}^{\text{irr}}$ represented by Eq. (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 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 $\mathbf{G}_{\text{eff}}$ 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 method of calculations of transport properties of suspensions.

The above derivation of the ring expansion is presented in the shorthand notation which emphasizes the idea of the underlying physics. It is worth presenting the ring expansion without the shorthand notation. Following the expression (63), the lowest two terms of the ring expansion (84) are given by

$$\begin{aligned}\mathbf{T}^{\text{irr}}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}') = {}& \sum_{n_1=1}^{\infty}\int d^3R_1^1\ldots d^3R_{n_1}^1\, H\left(\mathbf{R}_1^1,\ldots,\mathbf{R}_{n_1}^1\right)\mathbf{S}_I\left(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}'||\mathbf{R}_1^1,\ldots,\mathbf{R}_{n_1}^1\right)\\ &+\sum_{n_1=1}^{\infty}\sum_{n_2=1}^{\infty}\int d^3R_1^1\ldots d^3R_{n_1}^1 d^3R_1^2\ldots d^3R_{n_2}^2 d^3r'' d^3r''' d^3R'' d^3R'''\, H\left(\mathbf{R}_1^1,\ldots,\mathbf{R}_{n_1}^1|\mathbf{R}_1^2,\ldots,\mathbf{R}_{n_2}^2\right)\\ &\times\ \mathbf{S}_I\left(\mathbf{R},\mathbf{r};\mathbf{R}'',\mathbf{r}''||\mathbf{R}_1^1,\ldots,\mathbf{R}_{n_1}^1\right)\mathbf{G}_{\text{eff}}(\mathbf{r}'',\mathbf{r}''')\mathbf{S}_I\left(\mathbf{R}''',\mathbf{r}''';\mathbf{R}',\mathbf{r}'||\mathbf{R}_1^2,\ldots,\mathbf{R}_{n_2}^2\right)+\ldots.\end{aligned} \tag{101}$$

V. RENORMALIZATION OF CLAUSIUS-MOSSOTTI APPROXIMATION

After the derivation of the cluster expansion (72) of the $\mathbf{T}^{\text{irr}}$ operator, Felderhof, Ford, and Cohen gave the microscopic explanation of the Clausius-Mossotti formula [10]. 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 $\mathbf{T}^{\text{irr}}$, which leads to the Clausius-Mossotti relation.

Going along the line of the explanations of Felderhof, Ford, and Cohen [10] for a dielectric system, the Clausius-Mossotti relation is obtained, when the operator $\mathbf{T}^{\text{irr}}_{\text{CM}}$ defined with the formula

$$\mathbf{T}^{\text{irr}}_{\text{CM}} = \mathbf{T}^{\text{irr}}(1+[h\mathbf{G}]\mathbf{T}^{\text{irr}})^{-1} \tag{102}$$

is approximated by the single-particle term

$$\mathbf{T}^{\text{irr}}_{\text{CM}}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}') \approx n_1\delta(\mathbf{R}-\mathbf{R}')\mathbf{M}(\mathbf{R},\mathbf{r},\mathbf{r}'). \tag{103}$$

In the above definition (102), a superposition between the quantities $\mathbf{T}^{\text{irr}}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}')$ and $[h\mathbf{G}](\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}') = h(\mathbf{R},\mathbf{R}')\mathbf{G}(\mathbf{r},\mathbf{r}')$ appears. $h(\mathbf{r},\mathbf{r}')$ stands for the two-particle correlation function. It is worth noting that in Ref. [10], instead of the two-particle correlation function $h(\mathbf{r},\mathbf{r}')$, function $-f(\mathbf{r},\mathbf{r}')$ with f defined by the formula (83) appears. Both possibilities lead to the Clausius-Mossotti relation. We call the operator $\mathbf{T}^{\text{irr}}_{\text{CM}}$ 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,

$$\frac{\eta_{\text{eff}}}{\eta} = \frac{1+\frac{3}{2}\phi}{1-\phi}. \tag{104}$$

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

$$\mathbf{T}^{\text{irr}}_{\text{RCM}} = \mathbf{T}^{\text{irr}}(1+[h\mathbf{G}_{\text{eff}}]\mathbf{T}^{\text{irr}})^{-1}, \tag{105}$$

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

$$\mathbf{T}^{\text{irr}}_{\text{RCM}} \approx \mathbf{B}, \tag{106}$$

in analogy to the approximation (103). In the latter equation, instead of the single-particle response operator $\mathbf{M}$ appearing in the Clausius-Mossotti approximation, we take into account all self-scattering sequences $\mathbf{B}$, introduced before with the formula (74). Equations (105), (27), (78), (75), and (77) along with the approximation (106) define the renormalized Clausius-Mossotti approximation. Those equations form a close system of equations for operators $\mathbf{B}$, $\mathbf{T}^{\text{irr}}_{\text{RCM}}$, $\mathbf{T}^{\text{irr}}$, $\mathbf{G}_{\text{eff}}$, $\mathbf{T}$, and $\mathbf{T}_{\text{off}}$. The system can be solved for given volume fraction ϕ (or the single-particle density n_1) and for given two-particle correlation function $h(\mathbf{r},\mathbf{r}')$. 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 $\mathbf{T}^{\text{irr}}$ found within the renormalized Clausius-Mossotti approximation, one can calculate further the short-time transport characteristics, such as the effective viscosity η_{eff} from Eqs. (30)–(32) and the wave dependent hydrodynamic function $H(q)$ with the collective diffusion D_c and the self-diffusion D_s coefficient from Eqs. (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 $\mathbf{T}$ operator [see Eq. (22)] obtained within the Clausius-Mossotti approximation denoted by $\mathbf{T}^{\text{CM}}$. The Clausius-Mossotti approximation (103), by Eqs. (102) and (27), leads to the expression

$$\mathbf{T}^{\text{CM}} = n_1\mathbf{M}[1-[g\mathbf{G}]n_1\mathbf{M}]^{-1}, \tag{107}$$

where g is the radial distribution function related to the correlation function h as follows: $g = 1+h$. The above formula for $\mathbf{T}$ can be interpreted in terms of scattering sequences. The $\mathbf{T}$ operator, which on rigorous level is given by the sum of all possible scattering sequences as Eq. (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), lead to the following formula for the $\mathbf{T}$ operator (denoted by $\mathbf{T}^{\text{RCM}}$):

$$\mathbf{T}^{\text{RCM}} = \mathbf{B}[1-([g\mathbf{G}]+h[\mathbf{GTG}])\mathbf{B}]^{-1}. \tag{108}$$

The above equation differs from the expression (107), in particular by the term $h[\mathbf{GTG}]$. This term contains the dominant terms in the virial expansion on three-particle level for the sedimentation coefficient (cf. b_4 coefficient in Ref. [25]) and for the effective viscosity (cf. ν_2 coefficient in Ref. [26]). 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 ϕ and the two-body correlation function $h(\mathbf{r},\mathbf{r}')$ are given. Within the renormalized Clausius-Mossotti approximation, we perform calculations for the volume fractions $\phi = 0.05, 0.15, 0.25, 0.35$, 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. 2–6, 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 η_{eff}, respectively.

In Figs. 2–6 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

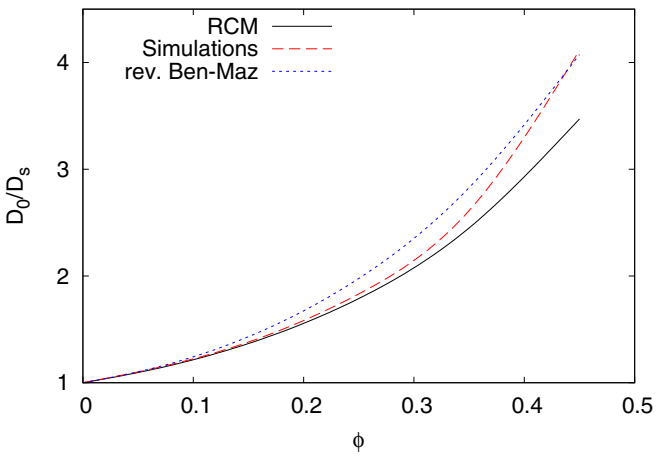


FIG. 2. (Color online) Inverse of the translational short-time self-diffusion coefficient D_s [Eq. (41)] normalized by the self-diffusion coefficient of a single particle $D_0 = k_BT/(6\pi\eta a)$ as a function of volume fraction ϕ for monodisperse suspension of hard spheres in equilibrium. Black (solid 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].

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].

Our results for the effective viscosity η_{eff} and the hydrodynamic function $H(q)$ (with its low and high q behavior) presented in Figs. 2–6 obtained within the renormalized Clausius-Mossotti approximation, when compared with the numerical simulations and the revised 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 the 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 $H(q)$ (see Figs. 4 and 5). 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 Eq. (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. The first variant is given by Eqs. (103) and (102), and is used in this article. The second variant is also given by Eqs. (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 $H_{\text{CM}}(q)$, because

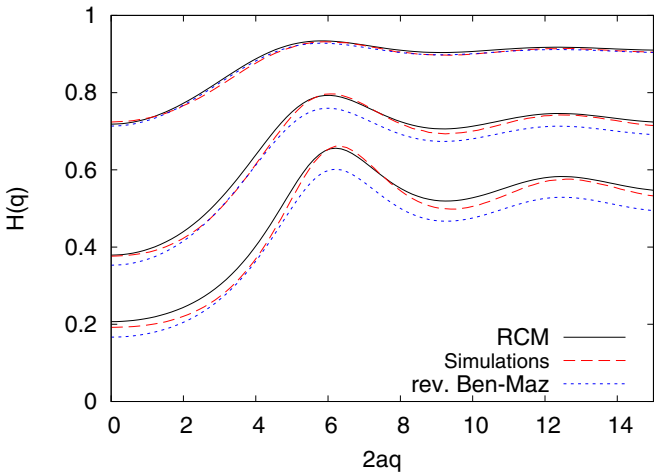


FIG. 4. (Color online) The hydrodynamic function $H(q)$ [Eq. (36)] as a function of wave vector for volume fractions $\phi = 0.05, 0.15$, and 0.25, for monodisperse suspension of hard spheres in equilibrium. Black (solid line): the renormalized Clausius-Mossotti approximation introduced in this article; red (long-dashed) line: numerical simulations [43]; blue (short-dashed) line: the revised Beenakker-Mazur method [44].

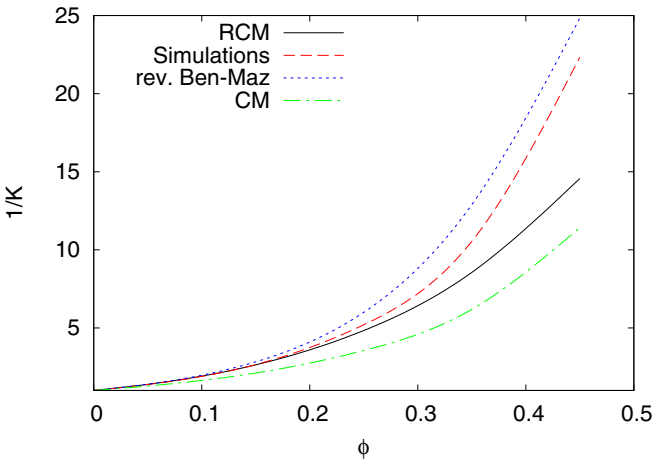


FIG. 3. (Color online) Inverse of the sedimentation coefficient K [Eq. (39)] as a function of volume fraction ϕ for monodisperse suspension of hard spheres in equilibrium. Black (solid line): the renormalized Clausius-Mossotti approximation introduced in this article; red (long-dashed) line: numerical simulations of Abade *et al.* [43]; blue (short-dashed) line: the revised Beenakker-Mazur method [44]; green (dotted-dashed) line: Clausius-Mossotti approximation defined by Eq. (103).

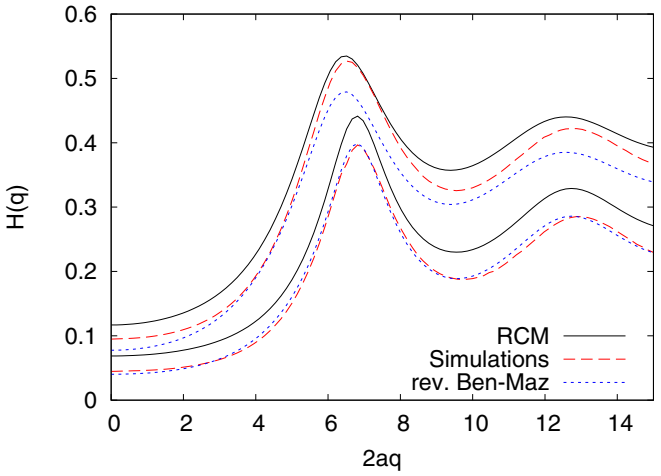


FIG. 5. (Color online) The hydrodynamic function $H(q)$ [Eq. (36)] as a function of wave vector for volume fractions $\phi = 0.35$ and 0.45, for monodisperse suspension of hard spheres in equilibrium. Black (solid line): the renormalized Clausius-Mossotti approximation introduced in this article; red (long-dashed) line: numerical simulations [43]; blue (short-dashed) line: the revised Beenakker-Mazur method [44].

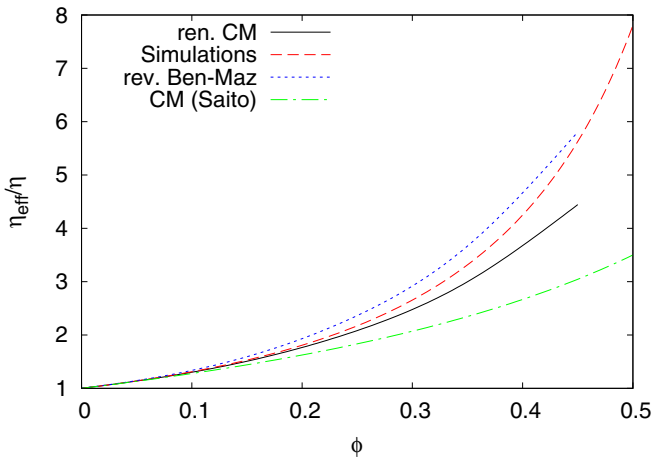


FIG. 6. (Color online) The relative effective viscosity η_{eff}/η (high frequency, low shear) [Eq. (32)] as a function of volume fraction ϕ for monodisperse suspension of hard spheres in equilibrium. Black (solid line): the renormalized Clausius-Mossotti approximation introduced in this article; red (long-dashed) line: numerical simulations of Ladd [45]; blue (short-dashed) line: the revised Beenakker-Mazur method [44]; green (dotted-dashed) line: Saito formula [5].

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:

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

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 Fig. 3.

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 to 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 $\mathbf{A}_{\gamma_0}$, which depends on the volume fraction ϕ, but does not depend on the distribution of particles, e.g., the two-body correlation function [17]. Therefore, this propagator $\mathbf{A}_{\gamma_0}$ is the same for hard-sphere suspension and for suspension of charged particles in equilibrium. On the other hand, the effective propagator $\mathbf{G}_{\text{eff}}$ 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 [22,58]. We are going to address the above questions in our further work.

ACKNOWLEDGMENTS

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APPENDIX A: WAVE DEPENDENT SEDIMENTATION

The wave dependent sedimentation coefficient $H(q)$ describes response of quiescent suspension $\mathbf{v}_0 = 0$ [cf. Eq. (11)] under action of a sinusoidal external force density, e.g.,

$$\mathbf{f}_{\text{ext}}(\mathbf{r}) = f_0 \hat{\mathbf{e}}_z \text{Re}\exp(-iq\hat{\mathbf{e}}_z \cdot \mathbf{r})\exp(-\eta|z|). \tag{A1}$$

The above external force density corresponds to the plane wave in the direction of the wave vector $\mathbf{q} = q\hat{\mathbf{e}}_z$. 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 $\mathbf{f}_{\text{ext}}(\mathbf{r}) = \hat{\mathbf{e}}_z f_{\text{ext}}(z)$. For homogeneous and isotropic suspension that property induces the same form for the average velocity

field, i.e., $\langle \mathbf{v}(\mathbf{r})\rangle = \hat{\mathbf{e}}_z\langle v(z)\rangle$. Incompressibility condition for the average velocity field $\langle \mathbf{v}(\mathbf{r})\rangle = \hat{\mathbf{e}}_z\langle v(z)\rangle$ implies $\langle v(z)\rangle =$ const. The force given by Eq. (A1) has also the mirror symmetry in z direction $f_{\rm ext}(z) = -f_{\rm ext}(-z)$, 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 $\langle v(z)\rangle =$ const, we obtain that the average velocity field vanishes in the whole suspension $\langle \mathbf{v}(\mathbf{r})\rangle = 0$.

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

$$\langle \mathbf{U}(\mathbf{R},\mathbf{r})\rangle = \int d^3R' d^3r' P_U \mathbf{T}^{\rm irr}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}') P_U^T \mathbf{f}_{\rm ext}(\mathbf{r}'), \quad \text{(A2)}$$

where projector P_U (with its transposition P_U^T) by definition projects on the upper half of the double vector $[\langle \mathbf{U}\rangle, \langle \mathbf{f}\rangle]$, as follows from the formula (38). From now on, we will take the limit $\eta \to 0^+$, in which the zero net flux condition, $\langle \mathbf{v}(\mathbf{r})\rangle = 0$ remains.

The external force density given by Eq. (A1) is torque free because the torque acting on the particle at position $\mathbf{R}$ vanishes, $\int d^3r\, \theta(|\mathbf{r}-\mathbf{R}|-a)(\mathbf{r}-\mathbf{R}) \times \mathbf{f}_{\rm ext}(\mathbf{r}) = 0$, 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 Eq. (9), for such external force density for hard spheres, can be written as

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

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

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

Equation (A3) is a straightforward consequence of the form of $\mathbf{M}_>$ given in Ref. [31]. Similar holds for the single-particle $\mathbf{M}_0$ operator:

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

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

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

In the problem of sedimentation, we consider the average translational velocity of particles $\langle \mathbf{V}(\mathbf{r})\rangle = \langle \sum_{i=1}^N \delta(\mathbf{R}-\mathbf{R}_i)\mathbf{V}_i(X)\rangle$. 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

$$\langle \mathbf{V}(\mathbf{r})\rangle = \frac{1}{\frac{4}{3}\pi a^3} \int d^3r \langle \mathbf{U}(\mathbf{R},\mathbf{r})\rangle, \quad \text{(A7)}$$

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

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

where

$$\mathbf{Y}(\mathbf{R},\mathbf{R}') = \frac{1}{\left(\frac{4}{3}\pi a^3\right)^2} \int d^3r \int d^3r' P_U \mathbf{T}^{\rm irr}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}') P_U^T. \quad \text{(A9)}$$

For homogeneous suspension, the above kernel $\mathbf{Y}(\mathbf{R},\mathbf{R}')$ depends only on the difference of positions, therefore, we can introduce

$$\mathbf{Y}(\mathbf{R}-\mathbf{R}') \equiv \mathbf{Y}(\mathbf{R},\mathbf{R}'). \quad \text{(A10)}$$

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

$$\mathbf{F}_{\rm ext}(\mathbf{r}) = F_0 \hat{\mathbf{q}}\, {\rm Re}\exp(-i\mathbf{q}\cdot\mathbf{R}), \quad \text{(A11)}$$

the average translational velocity has also the plane-wave form

$$\langle \mathbf{V}(\mathbf{r})\rangle = V(q)\hat{\mathbf{q}}\, {\rm Re}\exp(-i\mathbf{q}\mathbf{R}), \quad \text{(A12)}$$

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

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

with hydrodynamic function given by Eq. (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 $\mathbf{B}$, $\mathbf{T}^{\rm irr}_{\rm RCM}$, $\mathbf{G}_{\rm eff}$, $\mathbf{T}^{\rm irr}$, $\mathbf{T}$, $\mathbf{T}_{\rm off}$. Each of those quantities is a fourfold 3×3 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 $\mathbf{T}(\mathbf{R},\mathbf{r},\mathbf{R}',\mathbf{r}')_{u\alpha u',\alpha'}$. It has two indexes $u,u' = U,P$, which denote upper or lower part of the double vector, and another two Cartesian indices $\alpha,\alpha' = 1,2,3$. In our calculations, we represent those quantities as multipole hydrodynamic matrices [59]. In Ref. [31], 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 Ref. [31] is adopted also in this article, therefore, we do not repeat that material. In the same way as in Ref. [31], we introduce the multipole picture of the hydrodynamic matrices $\mathbf{B}$, $\mathbf{T}^{\rm irr}_{\rm RCM}$, $\mathbf{G}_{\rm eff}$, $\mathbf{T}^{\rm irr}$, $\mathbf{T}$, and $\mathbf{T}_{\rm off}$. In the multipole picture, all of the above quantities become infinite dimensional hydrodynamic multipole matrices, e.g., $\mathbf{T}(\mathbf{R},\mathbf{r};\mathbf{R}',\mathbf{r}')_{u\alpha u',\alpha'} \to [T(\mathbf{R},\mathbf{R}')]_{ulm\sigma,u'l'm'\sigma'}$. Therefore, the variables $\mathbf{R},\mathbf{R}'$ and Cartesian indices are transformed into multipole numbers l, m, and σ 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, $[T(\mathbf{R}-\mathbf{R}')]_{ulm\sigma,u'l'm'\sigma'} \equiv [T(\mathbf{R},\mathbf{R}')]_{ulm\sigma,u'l'm'\sigma'}$. We also use isotropy of the system, which allow to calculate the multipole matrix $T(\mathbf{R})$ for any vector $\mathbf{R}$ when the T for the z direction $\mathbf{R} = R\mathbf{e}_z$ is known.

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

$$\hat{T}(\mathbf{k}) = \int d^3R\, \exp[-i\mathbf{k}\mathbf{R}]T(\mathbf{R}), \tag{B1}$$

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

$$\hat{T}(\mathbf{k}) = \hat{B} + \hat{T}_{\rm off}(\mathbf{k}), \tag{B2}$$

$$\hat{T}(\mathbf{k}) = \hat{T}^{\rm irr}(\mathbf{k})[1 - \hat{G}(\mathbf{k})\hat{T}^{\rm irr}(\mathbf{k})]^{-1}, \tag{B3}$$

$$\hat{G}_{\rm eff}(\mathbf{k}) = \hat{G}(\mathbf{k}) + \hat{G}(\mathbf{k})\hat{T}(\mathbf{k})\hat{G}(\mathbf{k}), \tag{B4}$$

$$\hat{T}^{\rm irr}_{\rm RCM}(\mathbf{k}) \approx \hat{B}, \tag{B5}$$

$$\hat{T}^{\rm irr}(\mathbf{k}) = \hat{T}^{\rm irr}_{\rm RCM}(\mathbf{k})\big[1 - \widehat{[hG_{\rm eff}]}(\mathbf{k})\hat{T}^{\rm irr}_{\rm RCM}(\mathbf{k})\big]^{-1}, \tag{B6}$$

$$\hat{B} = n_1 M + \int d^3R\, T_{\rm off}(-\mathbf{R})G(\mathbf{R})M, \tag{B7}$$

where

$$\widehat{[hG_{\rm eff}]}(\mathbf{k}) = \int d^3R \exp[-i\mathbf{k}\mathbf{R}]h(\mathbf{R})G_{\rm eff}(\mathbf{R}). \tag{B8}$$

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 spaces. 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.,$[\hat{T}(\mathbf{k})]_{ulm\sigma,u'l'm'\sigma'}$. The truncation is characterized by L, which is the highest multipole used in the calculations: we consider matrix elements with $l,l' \leqslant L$ only. The calculations were performed for different parameters truncation $L = 4,\ldots,10$. The dependence of the effective viscosity coefficient $\eta_{\rm eff}/\eta$ on the function $1/(L\ln^3 L)$ containing a natural logarithm of the truncation parameter L is presented in Fig. 7. The figure shows that even for the highest truncation parameter $L = 10$ [which corresponds to $1/(L\ln^3 L) \approx 0.008$], 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 10 in Fig. 7, 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., $T(R\mathbf{e}_z)$. Points $R = \xi_0,\ldots,\xi_N$, with $\xi_0 = 0$ and $\xi_n = \xi_1 \exp[\alpha(i-1)]$ were considered. The parameter α was determined from the assumption that the first and the last section are equal, i.e., $\xi_1 = \xi_N - \xi_{N-1}$. Therefore, ξ_1 and N determine a set of points,

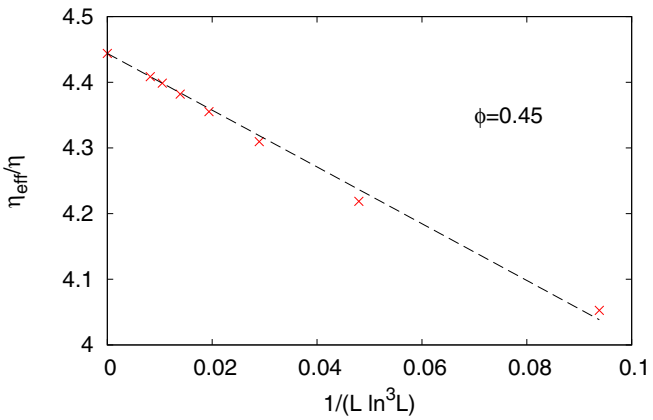

FIG. 7. (Color online) The relative effective viscosity coefficient $\eta_{\rm eff}/\eta$ as a function of the logarithm of the multipole truncation L obtained by the renormalized Clausius-Mossotti approximation for suspension of volume fraction $\phi = 0.45$.

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 ξ_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 $\xi_1/(2a) = \frac{1}{2},\frac{1}{3},\frac{1}{4},\frac{1}{5}$ 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

$$\sup_q |H_i(q) - H_{i-1}(q)| \approx \sup_q |H_{i+1}(q) - H_i(q)|\Delta, \tag{B9}$$

where $H_i(q)$ denotes the hydrodynamic function after the ith 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 [31]. 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.

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