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ON THE ABSORPTION PROBLEM FOR RANDOM DISPERSIONS

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The paper is devoted to the steady-state problem of absorption of a diffusing species (say, irradiation defects) in a random dispersion of spheres. The defects are created at a constant rate throughout the medium and are absorbed afterward, with different sink strengths, by the matrix and by the inclusions. One is to find the random diffusing species field and, in particular, the effective sink strength of the dispersion, having assumed the statistics of the spheres known. The problem is modelled by a Helmhotz equation with a random coefficient (the randomly fluctuating sink strength of the dispersion). The statistical solution of the latter is explicitly constructed, in a simple form, by means of the so-called factorial functional series, recently introduced by one of the authors. In particular, analytical formulae, correct to the order "square of sphere fraction", are obtained for the effective sink strength of the dispersion and for the two-point correlation function of the diffusing species field. An effective numerical procedure, allowing to specify these quantities, is described and numerical results are finally presented and discussed.

1. Introduction

Imagine a diffusing species is created with a rate K in a heterogeneous, say, two-phase medium and is simultaneously absorbed with a certain sink strength that has different values in each of the constituents. In what follows, the particular case of a random dispersion of equi-sized and nonoverlapping spheres will be considered only. The reason is that this case is of primary importance for both application and theory as a reasonable model for real particulate media with small or moderate filler fraction. In the steady-state limit, which will be treated here, the diffusing species concentration $\varphi(\mathbf{x})$ is governed by the equation

$$\Delta\varphi(\mathbf{x}) - k^2(\mathbf{x})\varphi(\mathbf{x}) + K = 0. \tag{1.1}$$

(We neglect the variance of the diffusion coefficient in the medium since Eq. (1.1) already reflects the most intriguing feature of the situation under study—the competition between the constituents for absorbing the diffusing species.) Note that the species may represent the defects appearing in a solid due to irradiation, see, e.g., the extensive survey of Brailsford and Bullough[3] for references and more details. The absorption coefficient (sink strength) $k^2(\mathbf{x})$ is a given random field, assumed nonnegative and statistically homogeneous and isotropic. In the case of a dispersion the field $k^2(\mathbf{x})$ has the form

$$k^2(\mathbf{x}) = \begin{cases} k_m^2, & \text{if } \mathbf{x} \in \text{matrix}, \\ k_f^2, & \text{if } \mathbf{x} \in \text{spheres}. \end{cases} \tag{1.2}$$

From a theoretical point of view, the basic problem we are interested in consists in evaluating, in statistical sense,[2] the random field $\varphi(\mathbf{x})$, i.e., all its multipoint correlations and the joint correlations of $\varphi(\mathbf{x})$ and $k^2(\mathbf{x})$ and, in particular, in finding the mean diffusing species concentration $\langle\varphi(\mathbf{x})\rangle$; the brackets $\langle\cdot\rangle$ hereafter denote ensemble averaging. The latter value would allow us to obtain the effective absorption coefficient, k^{*2}, of the medium defined by the relation

$$k^{*2} = \frac{K}{\langle\varphi(\mathbf{x})\rangle}. \tag{1.3}$$

The coefficient k^{*2} enters the "homogenized" equation

$$\Delta\langle\varphi(\mathbf{x})\rangle - k^{*2}\langle\varphi(\mathbf{x})\rangle + K = 0, \tag{1.4}$$

obtained from Eq. (1.1) after averaging. Eq. (1.4) describes the overall behaviour of the medium; it means that from a macroscopical point of view the heterogeneous medium absorbs the diffusing species as if it were homogeneous, with a constant (due to the assumed statistical homogeneity) sink strength k^{*2}. In this sense the absorption problem (1.1) belongs to the wide class of homogenization problems, extensively dealt with in the literature, especially in the context of scalar conductivity and elasticity, see, e.g., Beran's book,[2] the survey of Willis[24] or the recent revue of Torquato.[23] An effective method for treating these problems for particulate solids and for dispersions of spheres, in particular, has been recently introduced, applied to a number of specific problems and discussed by Christov and Markov, see Ref. 14 for more details and bibliography. The essence of the method is the utilization of functional (Volterra-Wiener) series with certain unknown kernels for representing the needed random fields. The kernels are to be specified by means of the respective governing equations. In turn, a special regrouping of the series allows to obtain, after truncation, approximations for these fields with a known accuracy. The regrouped series are called factorial due to the special properties of the fields that generate them. An essential feature of the method is that it allows to obtain not only the average values of the needed fields but all multipoint correlations as well. The application of this technique for the heat conduction problem in random dispersions of spheres (which could even overlap) is thoroughly described in Ref. 14.

Details of the general procedure and some rigorous proofs are given, e.g., in Ref. 13; as an illustration, Eq. (1.1) has also been treated there, but in rather cumbersome a way, without any numerical results and discussion.

Note that the theory of diffusion-controlled reaction is modelled by the limiting ("degenerate") case of Eq. (1.1) for a dispersion of spheres. In this theory the spheres are perfect absorbers ($k_f^2 = \infty$) while the matrix does not absorb at all ($k_m^2 = 0$). That is why in the literature much attention has been paid to this case only and a number of approximate schemes and variational estimates have been proposed for k^{*2}, see, e.g.,[6,16,18,21–23] and the references therein. On the other hand, the only detailed study of the random equation (1.1) from the point of view of its homogenization, for an arbitrary function $k^2(\mathbf{x})$, is due, as far as we know, to Talbot and Willis,[21] who considered certain approximate schemes for evaluating k^{*2} and then derived bounds on this quantity upon introducing a variational principle of Hashin-Shtrikman's type. More recently the authors proposed and investigated in[15] certain three-point bounds on k^{*2} of the type of those of Beran[2] for scalar conductivity and specified them afterward[11] for random dispersions of spheres.

The aim of the present study is a detailed study of the absorption problem (1.1) for dispersion of spheres, with an arbitrary absorption field (1.2), by means of the factorial series approach.

The outline of the paper is as follows. We first recall very briefly (Section 2) the statistical description of a random dispersion of spheres, as well as the definition and the basic properties of the factorial fields which play the central role in the sequel (for more details, see, e.g., Refs. 12-14, 20 et al.). In Section 3 we outline the arguments that allow us to represent the solution of Eq. (1.1) as a factorial series—a functional series with respect to the factorial fields of the dispersion, with certain unknown kernels.[13] A simple procedure for identifying the kernels is proposed in Section 4 which yields, in particular, the full statistical solution of Eq. (1.1), correct to the order c^2, where c is the volume fraction of the spheres, provided the solutions of the respective one- and two-sphere absorption problems are known. The similarity with the cluster expansion technique as developed by Matern and Felderhof[16] is pointed out. The procedure is illustrated by evaluating in a closed integral form the effective sink strength for the dispersion and the covariance function of the diffusing species concentration $\varphi(\mathbf{x})$ to the same order c^2. To get numerical results for those quantities, a twin expansion solution for the two-sphere absorption problem is developed in Section 5. The obtained numerical results for the aforementioned quantities—the effective sink strength and covariance—are discussed in the final Section 6.

2. Statistics of the Dispersion and the Factorial Fields

Let $\mathbf{x}_\alpha$ be the system of random points that serve as centers of the spheres. The statistics of the system $\mathbf{x}_\alpha$ is conveniently represented by its multipoint distribution densities $f_p = f_p(\mathbf{y}_1, \dots, \mathbf{y}_p)$. We assume the dispersion statistically isotropic and homogeneous; then, in particular, $f_p = f_p(\mathbf{y}_{2,1}, \dots, \mathbf{y}_{p,1})$, where $\mathbf{y}_{j,i} = \mathbf{y}_j - \mathbf{y}_i$.

Moreover, $f_1 = n$, where n is the number density of the spheres, so that $c = nV_a$ is their volume fraction; $V_a = \frac{4}{3}\pi a^3$ and a is the radius of the spheres.

Let us imagine now that by means of a certain manufacturing process we produce random dispersions with different number densities n. The statistics of the systems $\mathbf{x}_\alpha$ will then depend on n as a parameter, i.e., $f_p = f_p(\mathbf{y}_1, \dots, \mathbf{y}_p; n)$. We shall assume, as usual, that $f_p \sim n^p$, i.e., f_p has the asymptotic order n^p at $n \to 0$, $p = 1, 2, \dots$. In particular, for the two-point distribution density f_2 we have

$$f_2(\mathbf{y}_1, \mathbf{y}_2) = n^2 g(r), \quad g(r) = g_0(r) + O(n), \tag{2.1}$$

$r = |\mathbf{y}_2 - \mathbf{y}_1|$; $g_0(r)$ is the zero-density limit of the radial distribution function $g(r)$ for the system $\mathbf{x}_\alpha$.

Next we introduce the so-called random density field[20]

$$\psi(\mathbf{x}) = \sum_\alpha \delta(\mathbf{x} - \mathbf{x}_\alpha). \tag{2.2}$$

Its moments are uniquely defined by the probability densities of the set $\mathbf{x}_\alpha$. The respective formulas read:

$$\begin{aligned} \langle \psi(\mathbf{y}) \rangle &= f_1(\mathbf{y}) = n, \\ \langle \psi(\mathbf{y}_1)\psi(\mathbf{y}_2) \rangle &= n\delta(\mathbf{y}_{1,2}) + f_2(\mathbf{y}_1, \mathbf{y}_2), \\ \langle \psi(\mathbf{y}_1)\psi(\mathbf{y}_2)\psi(\mathbf{y}_3) \rangle &= n\delta(\mathbf{y}_{1,2})\delta(\mathbf{y}_{1,3}) \\ &+3\big\{\delta(\mathbf{y}_{1,2}) f_2(\mathbf{y}_{1,3})\big\}_s + f_3(\mathbf{y}_1, \mathbf{y}_2, \mathbf{y}_3), \end{aligned} \tag{2.3}$$

etc., where $\{\cdot\}_s$ means symmetrization with respect to all different combinations of indices in the braces, see, e.g., Ref. 20.

Let

$$\begin{aligned} \Delta_\psi^{(0)}(\mathbf{y}) = 1,\ &\Delta_\psi^{(1)}(\mathbf{y}) = \psi(\mathbf{y}), \\ \Delta_\psi^{(k)}(\mathbf{y}_1, \dots, \mathbf{y}_k) &= \psi(\mathbf{y}_1)[\psi(\mathbf{y}_2) - \delta(\mathbf{y}_{2,1})] \dots \\ \times\ [\psi(\mathbf{y}_k) - \delta(\mathbf{y}_{k,1}) - &\dots - \delta(\mathbf{y}_{k,k-1})], \quad k = 2, 3, \dots , \end{aligned} \tag{2.4}$$

be the so-called factorial fields for the set $\mathbf{x}_\alpha$. The factorial fields have two basic properties. First,

$$\Delta_\psi^{(k)}(\mathbf{y}_1, \dots, \mathbf{y}_k) = \begin{cases} \psi(\mathbf{y}_1) \dots \psi(\mathbf{y}_k), & \text{if } \mathbf{y}_i \neq \mathbf{y}_j, \\ 0, & \text{if } \mathbf{y}_i = \mathbf{y}_j \text{ for a pair } i \neq j, \end{cases} \tag{2.5}$$

(which explains to a certain extent the term factorial). Second,

$$\left\langle \Delta_\psi^{(k)}(\mathbf{y}_1, \dots, \mathbf{y}_k) \right\rangle = f_k(\mathbf{y}_1, \dots, \mathbf{y}_k), \quad k = 0, 1, \dots , \tag{2.6}$$

see Refs. 5, 13, 14 for details and rigorous proofs.

Note also the formula

$$\Delta_\psi^{(1)}(\mathbf{y}_1)\Delta_\psi^{(1)}(\mathbf{y}_2) = \Delta_\psi^{(2)}(\mathbf{y}_1, \mathbf{y}_2) + \Delta_\psi^{(1)}(\mathbf{y}_1)\delta(\mathbf{y}_{1,2}), \tag{2.7a}$$

or, more generally,

$$\Delta_\psi^{(p)}(\mathbf{y}_1,\dots,\mathbf{y}_p)\Delta_\psi^{(1)}(\mathbf{y}_{p+1}) = \Delta_\psi^{(p+1)}(\mathbf{y}_1,\dots,\mathbf{y}_p,\mathbf{y}_{p+1})$$

$$+\Delta_\psi^{(p)}(\mathbf{y}_1,\dots,\mathbf{y}_p)\Big\{\sum_{i=1}^{p}\delta(\mathbf{y}_{i,p+1})\Big\}, \quad p=2,3,\dots\,, \tag{2.7b}$$

which follow immediately from the definition (2.4). More general formulae for products of the factorials are given in Ref. 13, but we shall need in what follows only the fact that

$$\Big\langle \Delta_\psi^{(k_1)}\dots\Delta_\psi^{(k_m)}\Big\rangle = O(n^M), \tag{2.8}$$

$M=\max(k_1,\dots,k_m)$, which holds under the assumption $f_p\sim n^p$.

3. Factorial Series Solution of the Absorption Problem

It could be easily shown, using the variational formulation of the problem (1.1) and some facts from the convex analysis, that the problem (1.1) possesses solution which is unique (see Ref. 15). Thus Eq. (1.1) defines implicitly a nonlinear operator $\mathcal{F}$ which transforms the known random field of the sink strength $k^2(\mathbf{x})$ into the random field $\varphi(\mathbf{x})$ of the diffusing species concentration:

$$\mathcal{F}: k^2(\cdot)\longrightarrow\varphi(\cdot). \tag{3.1}$$

Using the terminology of the general system theory,[19] we can consider $k^2(\mathbf{x})$ as the "input" in a "black box" whose "output" is $\varphi(\mathbf{x})$. Following the general idea of this theory, we develop the operator $\mathcal{F}$ into the so-called functional (Volterra-Wiener) series

$$\varphi(\mathbf{x}) = \mathcal{F}\big[k^2(\cdot)\big] = \Phi_0(\mathbf{x}) + \int\Phi_1(\mathbf{x}-\mathbf{y})k^2(\mathbf{y})\,d^3\mathbf{y}$$

$$+\iint\Phi_2(\mathbf{x}-\mathbf{y}_1,\mathbf{x}-\mathbf{y}_2)k^2(\mathbf{y}_1)k^2(\mathbf{y}_2)\,d^3\mathbf{y}_1d^3\mathbf{y}_2+\cdots \tag{3.2}$$

with certain nonrandom kernels Φ_0, Φ_1, ... (Hereafter the integrals are over R^3 if the integration domain is not explicitly indicated.)

In turn, the random absorption field (1.2) of the dispersion has a simple integral representation of the form

$$k^2(\mathbf{x}) = k_m^2 + [k^2]\int h(\mathbf{x}-\mathbf{y})\Delta_\psi^{(1)}(\mathbf{y})\,d^3\mathbf{y}, \tag{3.3}$$

$[k^2]=k_f^2-k_m^2$, where $h(\mathbf{y})$ is the characteristic function of a single sphere of radius a, located at the origin.

Let us now rearrange the series (3.1), inserting there Eq. (3.3) and replacing the products of the ψ's by the factorials (2.4):

$$\varphi(\mathbf{x}) = \Phi_0(\mathbf{x}) + \int\Phi_1(\mathbf{x}-\mathbf{y})\Delta_\psi^{(1)}(\mathbf{y})\,d^3\mathbf{y}$$

$$+\iint \Phi_2(\mathbf{x}-\mathbf{y}_1,\mathbf{x}-\mathbf{y}_2)\Delta_\psi^{(2)}(\mathbf{y}_1,\mathbf{y}_2)\,d^3\mathbf{y}_1 d^3\mathbf{y}_2+\cdots. \tag{3.4}$$

The new kernels Φ_p can be expressed through those in Eq. (3.2) (for brevity we use the same notation Φ_p for the former).

After[13] we call the series of the type (3.4) factorial. The central result of Ref. 13 states that for the class of point sets $\mathbf{x}_\alpha$ which comply with the assumption $f_p \sim n^p$, the factorial series (3.4) are virial in the following sense.

Let us denote by $\varphi^{(N)}(\mathbf{x})$ the series (3.4) truncated after the N-tuple term. Then

$$\varphi(\mathbf{x})=\varphi^{(N)}(\mathbf{x})+o(c^N) \tag{3.5a}$$

in a statistical sense, i.e., all multipoint correlation functions of the fields $\varphi(\mathbf{x})$ and $\varphi^{(N)}(\mathbf{x})$ differ by infinitesimals of the order $o(c^N)$ at $c\to 0$. The same holds true for the field $\kappa(\mathbf{x})\varphi(\mathbf{x})$, i.e.,

$$\kappa(\mathbf{x})\varphi(\mathbf{x})=\kappa(\mathbf{x})\varphi^{(N)}(\mathbf{x})+o(c^N). \tag{3.5b}$$

Thus the solution of the absorption problem (1.1), asymptotically correct to the order c^N, requires specification of the first $N+1$ kernels of the factorial series (3.4). The needed to this end technique is discussed in Refs. 13, 14 et al.: the truncated series (3.4) (after a certain "orthogonalization") are introduced in (1.1), the result is multiplied by the factorials $\Delta_\psi^{(k)}(\mathbf{y}_1,\ldots,\mathbf{y}_k)$, $k=0,1,\ldots,N$, and then averaged. This procedure leads to a complicated system of integro-differential equations for the unknown kernels even in the case $N=2$, see Ref. 13. Here we shall employ a much simpler method, based on the multiplication formulae (2.7) for the factorial fields.

4. Explicit Solution of the Absorption Problem to the Order $\mathbf{c^2}$

According to the aforesaid (Section 3), for the solution of the random problem (1.1), asymptotically correct to the order c^2, it suffices to specify the three kernels Φ_0, Φ_1 and Φ_2 in the truncated factorial series:

$$\varphi(\mathbf{x})\sim\varphi^{(2)}(\mathbf{x})=\Phi_0+\int\Phi_1(\mathbf{x}-\mathbf{y})\Delta_\psi^{(1)}(\mathbf{y})\,d^3\mathbf{y}$$
$$+\iint \Phi_2(\mathbf{x}-\mathbf{y}_1,\mathbf{x}-\mathbf{y}_2)\Delta_\psi^{(2)}(\mathbf{y}_1,\mathbf{y}_2)\,d^3\mathbf{y}_1 d^3\mathbf{y}_2. \tag{4.1}$$

(It is easily seen that $\Phi_0(\mathbf{x})=\Phi_0$ is a constant due to the statistical homogeneity of the dispersion.) To this end we insert the representations (3.3) and (4.1) of $k^2(\mathbf{x})$ and $\varphi(\mathbf{x})$, respectively, into Eq. (1.1) and group the terms, containing the factorials $\Delta_\psi^{(p)}$ for one and the same p, $p=0,1,2$, using the formulae (2.7a) and (2.7b) at $p=2$. (The term, containing $\Delta_\psi^{(3)}$, that appears due to Eq. (2.7b), is neglected since it contributes quantities of the order c^3 or higher, as it follows from Eq. (2.8) and the assumption $f_3\sim n^3$.) The eventual result of such a regrouping reads:

$$\left\{-k_m^2\Phi_0(\mathbf{x})+K\right\}\Delta_\psi^{(0)}$$

$$+\int\Big\{\Delta\Phi_1(\mathbf{x}-\mathbf{y})-k_0^2(\mathbf{x}-\mathbf{y})\Phi_1(\mathbf{x}-\mathbf{y})-[k^2]h(\mathbf{x}-\mathbf{y})\Phi_0\Big\}\Delta_\psi^{(1)}(\mathbf{y})\,d^3\mathbf{y}$$

$$+\int\!\!\int\Big\{\Delta\Phi_2(\mathbf{x}-\mathbf{y}_1,\mathbf{x}-\mathbf{y}_2)-\Big[k_0^2(\mathbf{x}-\mathbf{y}_1)+[k^2]h(\mathbf{x}-\mathbf{y}_2)\Big]\Phi_2(\mathbf{x}-\mathbf{y}_1,\mathbf{x}-\mathbf{y}_2)$$

$$-\frac{1}{2}[k^2]\Big[h(\mathbf{x}-\mathbf{y}_1)\Phi_1(\mathbf{x}-\mathbf{y}_2)+h(\mathbf{x}-\mathbf{y}_2)\Phi_1(\mathbf{x}-\mathbf{y}_1)\Big]\Big\}\Delta_\psi^{(2)}(\mathbf{y}_1,\mathbf{y}_2)\,d^3\mathbf{y}_1d^3\mathbf{y}_2=0. \tag{4.2}$$

(Recall that $\Delta_\psi^{(0)}=1$.) Here

$$k_0^2(\mathbf{x})=k_m^2+[k^2]h(\mathbf{x})=\begin{cases}k_f^2, & \text{at} \quad |\mathbf{x}|<a,\\ k_m^2, & \text{at} \quad |\mathbf{x}|\geq a.\end{cases} \tag{4.3}$$

The equation (4.2) is satisfied if the multipliers in the braces in front of the factorials $\Delta_\psi^{(0)}$, $\Delta_\psi^{(1)}$ and $\Delta_\psi^{(2)}$ vanish. This leads us immediately to the following system of equations

$$k_m^2\Phi_0(\mathbf{x})=K, \quad \text{i.e.,} \quad \Phi_0(\mathbf{x})=\frac{K}{k_m^2}=\text{const}\,, \tag{4.4a}$$

$$\Delta\Phi_1(\mathbf{x})-k_0^2(\mathbf{x})\Phi_1(\mathbf{x})-[k^2]h(\mathbf{x})\Phi_0=0, \tag{4.4b}$$

$$2\Delta\Phi_2(\mathbf{x}-\mathbf{z},\mathbf{x})-2\Big[k_0^2(\mathbf{x})+[k^2]h(\mathbf{x}-\mathbf{z})\Big]\Phi_2(\mathbf{x}-\mathbf{z},\mathbf{x})$$
$$-[k^2]\big\{h(\mathbf{x})\Phi_1(\mathbf{x}-\mathbf{z})+h(\mathbf{x}-\mathbf{z})\Phi_1(\mathbf{x})\big\}=0, \tag{4.4c}$$

from which the kernels Φ_0, Φ_1 and Φ_2 will be determined. (In Eq. (4.4c) the differentiation is with respect to $\mathbf{x}$, and $\mathbf{z}$ plays the role of a parameter.)

Denote by $H^{(1)}(\mathbf{x})$ the solution of the equation

$$\Delta H^{(1)}(\mathbf{x})-k_0^2(\mathbf{x})H^{(1)}(\mathbf{x})-[k^2]h(\mathbf{x})=0, \tag{4.5}$$

which is continuous in the whole R^3, and its normal derivative is continuous on the sphere $|\mathbf{x}|=a$. According to Eqs. (4.3) and (4.5), $H^{(1)}(\mathbf{x})$ is the steady-state concentration of the diffusing species in an unbounded medium with sink strength k_m^2, generated at the rate $-[k^2]$ in a spherical inhomogeneity of sink strength k_f^2. Due to this interpretation Eq. (4.5) represents the single-sphere problem for Eq. (1.1).

Simple arguments, using the radial symmetry of the function $H^{(1)}(\mathbf{x})$ and the continuity conditions allow us to find it explicitly:

$$H^{(1)}(\mathbf{x})=\frac{[k^2]}{k_f^2}\begin{cases}A_1\dfrac{a_f\sinh r_f}{r_f\sinh a_f}-1, & \text{at} \quad |\mathbf{x}|<a,\\ A_2\dfrac{a_m}{r_m}e^{a_m-r_m}, & \text{at} \quad |\mathbf{x}|\geq a,\end{cases} \tag{4.6a}$$

where

$$A_1=\frac{1+a_m}{a_m+a_f\coth a_f}, \; A_2=\frac{1-a_f\coth a_f}{a_m+a_f\coth a_f}, \tag{4.6b}$$

$$a_m=ak_m, \; a_f=ak_f, \; r_m=rk_m, \; r_f=rk_f, \tag{4.6c}$$

are dimensionless parameters and $r=|\mathbf{x}|$.

From Eq. (4.4b) it is obvious that

$$\Phi_1(\mathbf{x}) = \frac{K}{k_m^2} H^{(1)}(\mathbf{x}), \tag{4.7}$$

i.e., the kernel of the one-tuple integral term of the series (4.1) is proportional to the solution of the respective single-sphere problem.

Consider finally Eq. (4.4c) for the kernel Φ_2. It is easily seen that if we set

$$\Phi_2(\mathbf{x}-\mathbf{z},\mathbf{x}) = \frac{K}{k_m^2} H_{20}(\mathbf{x}-\mathbf{z},\mathbf{x}), \tag{4.8}$$

then

$$2H_{20}(\mathbf{x}-\mathbf{z},\mathbf{x}) = H^{(2)}(\mathbf{x};\mathbf{z}) - H^{(1)}(\mathbf{x}) - H^{(1)}(\mathbf{x}-\mathbf{z}), \tag{4.9}$$

where $H^{(2)}(\mathbf{x};\mathbf{z})$ is the bounded and continuous everywhere solution of the equation

$$\Delta H^{(2)}(\mathbf{x};\mathbf{z}) - \big\{k_m^2 + [k^2]\big(h(\mathbf{x}) + h(\mathbf{x}-\mathbf{z})\big)\big\} H^{(2)}(\mathbf{x};\mathbf{z}) - [k^2]\big\{h(\mathbf{x}) + h(\mathbf{x}-\mathbf{z})\big\} = 0. \tag{4.10}$$

Similarly to the single-sphere solution, we require also that the normal derivatives of the solution of Eq. (4.10) be continuous on the spheres $|\mathbf{x}| = a$ and $|\mathbf{x}-\mathbf{z}| = a$.

Obviously, the field $H^{(2)}(\mathbf{x};\mathbf{z})$ is the two-sphere solution for our problem (1.1), since it gives the concentration of the diffusing species in an unbounded matrix, generated with the rate $-[k^2]$, from sources located into two spherical inhomogeneities of radius a, one centered at the origin, the other—at the point $\mathbf{z}$, $|\mathbf{z}| > 2a$, see Eq. (4.10).

The above results (4.1)–(4.10) indicate the strong resemblance between the factorial series approach advocated here and the cluster expansion technique as developed by Matern and Felderhof, see Ref. 16 and the citations therein. Recall that such a resemblance showed up in the scalar conductivity context as well, when heat conduction problem through a random dispersion was treated in Refs. 13, 14 by means of the factorial series; the eventual result was shown[14] to coincide with the appropriate cluster expansion developed in this context by Felderhof et al.[7]. It can be suggested therefore that the factorial series approach represents a rigorous formalization of the more intuitive and physically appealing cluster expansions method, but such a discussion goes beyond the scope of this paper and should be done elsewhere.

According to Eq. (4.9) the kernel Φ_2 in the expansion (4.1) is, to the order $o(c^2)$, proportional to the field that should be added to the single-sphere solutions, $H^{(1)}(\mathbf{x})$ and $H^{(1)}(\mathbf{x}-\mathbf{z})$, generated by the two spheres (centered at the origin and at the point $\mathbf{z}$ respectively), in order to obtain the two-sphere solution $H^{(2)}(\mathbf{x};\mathbf{z})$. A similar result holds in the problems of scalar conductivity and elasticity of random dispersions, see, e.g., Refs. 7, 8, 14 et al.

The relations (4.4a), (4.6a) and (4.7)–(4.10) determine the kernels in Eq. (4.1). Thus the solution to the random absorption problem (1.1), correct to the order $o(c^2)$, is obtained as the truncated factorial series (4.1). This allows to evaluate all the statistical characteristics we are interested in. For instance, in virtue of Eqs. 2.6), (4.4a) and (4.6)–(4.9), one gets

$$\langle \varphi(\mathbf{x}) \rangle = \frac{K}{k_m^2}(1 + a_1 c + a_2 c^2) + o(c^2), \tag{4.11a}$$

where

$$a_1 = -\frac{[k^2]}{k_f^2}\left\{1 - 3\frac{[k^2]}{k_m^2}\,\frac{(1+a_m)(1-a_f\coth a_f)}{a_f^2(a_m + a_f\coth a_f)}\right\}, \tag{4.11b}$$

$$a_2 = -\frac{1}{V_a^2}\frac{[k^2]}{k_m^2}\int h(\mathbf{y}_1)\left\{\int g_0(\mathbf{y}_2 - \mathbf{y}_1)\big[H^{(1)}(\mathbf{y}_2) + 2H_{20}(\mathbf{y}_1,\mathbf{y}_2)\big]\, d^3\mathbf{y}_2\right\} d^3\mathbf{y}_1, \tag{4.11c}$$

and $g_0(r)$s the zero-density limit of the radial distribution function for the set $\mathbf{x}_\alpha$ of sphere centers, see Eq. (2.1).

In turn, the definition (1.2) of the effective sink strength together with Eqs. (4.11), yields

$$\frac{k^{*2}}{k_m^2} = 1 + b_1 c + b_2 c^2 + o(c^2),$$
$$b_1 = -a_1, \quad b_2 = a_1^2 - a_2. \tag{4.12}$$

It is noted that to the order $O(c)$ the formula (4.12) coincides with the prediction of the self-consistent theory of Brailsford and Bullough.[3]

Let $k_m^2 \to 0$ and $k_f^2 \to \infty$ which corresponds to the case of ideally absorbing spherical sinks distributed throughout a nonabsorbing matrix. This case, let us recall is of primary importance for the theory of diffusion-controlled reactions, see Section 1 and the references cited therein. Taking these two limits in Eq. (4.11b), one easily gets the well-known Smoluchowski formula

$$k^{*2} = \frac{3}{a^2}c + o(c) = k_s n + o(n),$$

where $k_s = 4\pi a$.

However, having taken the same limits $k_m^2 \to 0$ and $k_f^2 \to \infty$ in Eq. (4.11c), we find that $|a_2| = \infty$ and thus the c^2-coefficient $b_2 = \infty$ as well. This means that the dependence of the effective sink strength on the sphere fraction, $k^{^2} = k^{*2}(c)$, has in the case under study the form

$$k^{*2} = \frac{3}{a^2}\Big(c + f(c)\Big), \quad f(c) = o(c),$$

where $f(c)$ is a nonanalytic function of c in the vicinity of the point $c = 0$. This conclusion is corroborated by the results of Felderhof and Deutch[6], Matern and Felderhof[16] and Talbot and Willis[22] who argued, using different kinds of arguments, that

$$f(c) = c\sqrt{3c} + o(c^{3/2}).$$

It is an open question how the factorial series approach can be modified in order to cover properly the limiting situation under discussion in general, and the nonanalytic dependence of the statistical characteristics upon sphere fraction in particular. Note that certain approximations of self-consistent type can be naturally introduced within the frame of the cluster expansion technique[16]; they yield the first few terms of above indicated nonanalytic function $k^{*2}(c)$ for dilute sphere fractions $c \ll 1$.

Consider next the covariance function for the diffusing species field $\varphi(\mathbf{x})$, i.e., the function $M_2^{\varphi}(\mathbf{x}) = \langle \varphi'(\mathbf{0})\varphi'(\mathbf{x})\rangle$, where $\varphi'(\mathbf{x}) = \varphi(\mathbf{x}) - \langle \varphi(\mathbf{x})\rangle$ is the fluctuating part of $\varphi(\mathbf{x})$. From Eq. (4.1) and the relations

$$\left\langle \Delta_{\psi}^{(2)}(\mathbf{y}_1,\mathbf{y}_2)\Delta_{\psi}^{(1)}(\mathbf{y}_3)\right\rangle = n^2 g_0(\mathbf{y}_{1,2})[\delta(\mathbf{y}_{1,3}) + \delta(\mathbf{y}_{2,3})] + o(n^2),$$

$$\left\langle \Delta_{\psi}^{(2)}(\mathbf{y}_1,\mathbf{y}_2)\Delta_{\psi}^{(2)}(\mathbf{y}_3,\mathbf{y}_4)\right\rangle = n^2 g_0(\mathbf{y}_{1,2})[\delta(\mathbf{y}_{1,3})\delta(\mathbf{y}_{2,4}) + \delta(\mathbf{y}_{1,4})\delta(\mathbf{y}_{2,3})] + o(n^2),$$

which easily follow from Eqs. (2.4) and (2.3), one gets

$$M_2^{\varphi}(\mathbf{x}) = \frac{K^2}{k_m^4}F(\mathbf{x}), \quad F(\mathbf{x}) = cF_1(\mathbf{x}) + c^2 F_2(\mathbf{x}) + o(c^2), \tag{4.13a}$$

where

$$F_1(\mathbf{x}) = \frac{1}{V_a}\int H^{(1)}(\mathbf{y})H^{(1)}(\mathbf{x}-\mathbf{y})\, d^3\mathbf{y}, \tag{4.13b}$$

$$\begin{aligned} F_2(\mathbf{x}) = -\frac{1}{V_a^2}\Bigg\{ &\iint H^{(1)}(\mathbf{y})H^{(1)}(\mathbf{x}+\mathbf{t}-\mathbf{y})R_0(\mathbf{t})\, d^3\mathbf{t}\, d^3\mathbf{y} \\ &+ \iint \Big[H^{(1)}(\mathbf{y}) + H^{(1)}(\mathbf{y}-\mathbf{t}) - H^{(2)}(\mathbf{y};\mathbf{t})\Big]\Big[H^{(1)}(\mathbf{x}+\mathbf{y}-\mathbf{t}) \\ &+ \frac{1}{2}\Big(H^{(1)}(\mathbf{x}+\mathbf{t}-\mathbf{y}) + H^{(2)}(\mathbf{y}-\mathbf{x};\mathbf{t}) - H^{(1)}(\mathbf{y}-\mathbf{x})\Big)\Big] g_0(\mathbf{t})\, d^3\mathbf{t}\, d^3\mathbf{y}\Bigg\}. \end{aligned} \tag{4.13c}$$

With the same ease other statistical characteristics, pertaining to the problem (1.1), can be obtained in a closed form. Their evaluation needs, however, a convenient for numerical implementation solution of the two-sphere problem (4.10).

5. Twin Expansion Solution of the Two-Sphere Problem

In order to find the solution of the two-sphere problem, we shall use the so-called twin expansion method. We shall expose here only the main idea and the results, see Ref. 10 for more details. It is to be noted also that a fully similar in spirit method has been successfully employed already in the respective two-sphere problems for heat conduction,[7,8] elasticity,[4] diffraction,[9] etc.

Let us introduce two Cartesian systems and two systems of spherical coordinates as shown in Fig. 1. Both spheres are of radius a, the origins of the systems are at the centers of the spheres, the χ-coordinate is common for them and the distance between the centers, $|O_1O_2|$, is denoted by R, so that

$$x_1 = x_2, \quad y_1 = y_2, \quad z_1 = z_2 + R.$$

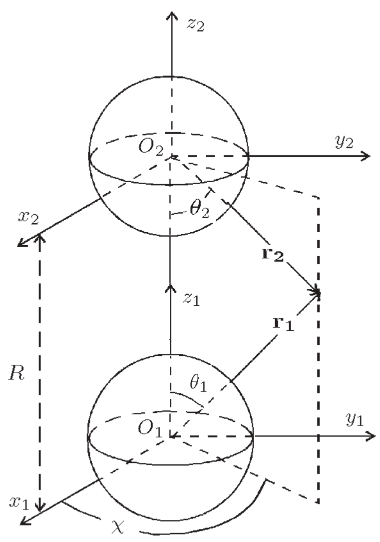

Fig. 1.

Then, obviously,

$$
\begin{aligned}
x_1 &= r_1 \sin\theta_1 \cos\chi, \quad & x_2 &= r_2 \sin\theta_2 \cos\chi, \\
y_1 &= r_1 \sin\theta_1 \sin\chi, \quad & y_2 &= r_2 \sin\theta_2 \sin\chi, \\
z_1 &= r_1 \cos\theta_1, \quad & z_2 &= -r_2 \cos\theta_2,
\end{aligned}
$$

where $0 \le r_1,\, r_2 < \infty$, $0 \le \theta_1,\, \theta_2 \le \pi$, $0 \le \chi < 2\pi$.

We start with the formula

$$
\frac{1}{\sqrt{k_m r_2}} K_{n+\frac{1}{2}}(k_m r_2) P_n(\cos\theta_2) = (-1)^n \sqrt{\frac{2\pi}{k_m R}} \frac{1}{\sqrt{k_m r_1}}
$$

$$
\times \sum_{s=0}^{\infty} \frac{(-1)^s}{N_{0s}} I_{s+\frac{1}{2}}(k_m r_1) P_s(\cos\theta_1) \left[\sum_{\sigma=|s-n|}^{s+n} (-1)^\sigma b_\sigma^{(s0n0)} K_{\sigma+\frac{1}{2}}(k_m R) \right] \tag{5.1}
$$

which connects the spherical wave functions given in the two spherical coordinate systems in Fig. 1. Here $I_{s+\frac{1}{2}}$ and $K_{\sigma+\frac{1}{2}}$ denote the respective modified Bessel functions, $N_{0s} = \dfrac{2}{2s+1}$, $P_s(x)$ are the Legendre polynomials and

$$
b_n^{(n_1 m_1 n_2 m_2)} = (-1)^{m_2} \sqrt{\frac{(n_1+m_1)!(n_2+m_2)!(n-m_1+m_2)!}{(n_1-m_1)!(n_2-m_2)!(n+m_1-m_2)!}}
$$

$$
\times (n_1 n_2 00|n0)(n_1 n_2 m_1, -m_2|n, m_1-m_2),
$$

with $(n_1 n_2 m_1 m_2|n, m_1+m_2)$ denoting the Clebsh-Gordon coefficients, see Refs. 1, 8, 9 for details. We recall that a spherical wave function is a particular solution of

Eq. (1.1) obtained after separating the variables in a spherical coordinate system. Here, moreover, these solutions will not depend on the coordinate χ due to the obvious symmetry of the two-sphere configuration (Fig. 1).

We look for the solution $H^{(2)}(\mathbf{x};\mathbf{z})$ of the Eq. (1) in the following form:

—inside the sphere "i" as

$$H^{(2)}_{(i)} = -\frac{[k^2]}{k_f^2} + \sum_{n=0}^{\infty} A_n \sqrt{\frac{a}{r_i}} I_{n+\frac{1}{2}}(k_f r_i) P_n(\cos\theta_i), \quad i = 1, 2; \tag{5.2a}$$

—outside the spheres as

$$H^{(2)}_{(\mathrm{out})} = \sum_{n=0}^{\infty} \Big\{ C_n^{(1)} \sqrt{\frac{a}{r_1}} K_{n+\frac{1}{2}}(k_m r_1) P_n(\cos\theta_1) + C_n^{(2)} \sqrt{\frac{a}{r_2}} K_{n+\frac{1}{2}}(k_m r_2) P_n(\cos\theta_2) \Big\}. \tag{5.2b}$$

Due to the obvious symmetry of the problem under study we have $C_n = C_n^{(1)} = C_n^{(2)}$, $n = 0, 1, \ldots$. Thus

$$H^{(2)}_{(\mathrm{out})} = \sum_{n=0}^{\infty} C_n \left\{ \sqrt{\frac{a}{r_1}} K_{n+\frac{1}{2}}(k_m r_1) P_n(\cos\theta_1) + \sqrt{\frac{a}{r_2}} K_{n+\frac{1}{2}}(k_m r_2) P_n(\cos\theta_2) \right\}. \tag{5.3}$$

The coefficients A_n and C_n are to be found from the boundary conditions—the continuity of the field $H^{(2)}(\mathbf{x};\mathbf{z})$ and its normal derivative across each spherical interface (with respect to $\mathbf{x}$; recall that $\mathbf{z}$ plays the role of a parameter).

According to Eq. (5.1), we recast the solution (5.2b) of Eq. (1) outside the spheres as

$$H^{(2)}_{(\mathrm{out})} = \sqrt{\frac{a}{r_1}} \sum_{n=0}^{\infty} P_n(\cos\theta_1) \left\{ C_n K_{n+\frac{1}{2}}(k_m r_1) + \sqrt{\frac{2\pi}{k_m R}} \frac{1}{N_{0n}} I_{n+\frac{1}{2}}(k_m r_1) \times \sum_{s=0}^{\infty} (-1)^{n+s} C_s \left[\sum_{\sigma=|s-n|}^{s+n} (-1)^{\sigma} b_{\sigma}^{(s0n0)} K_{\sigma+\frac{1}{2}}(k_m R) \right] \right\}. \tag{5.4}$$

Making use of the above mentioned boundary conditions, the orthogonality of the Legendre polynomials and the fact that $b^{(s000)} = 1$, $s = 0, 1, \ldots$, see Ref. 9, we find the following relations between the unknown coefficients

$$A_0 I_{\frac{1}{2}}(a_f) - \frac{[k^2]}{k_f^2} = C_0 K_{\frac{1}{2}}(a_m) + \frac{1}{N_{00}} I_{\frac{1}{2}}(a_m) \sqrt{\frac{2\pi}{k_m R}} \sum_{s=0}^{\infty} C_s K_{s+\frac{1}{2}}(k_m R), \tag{5.5a}$$

$$A_n I_{n+\frac{1}{2}}(a_f) = C_n K_{n+\frac{1}{2}}(a_m) + \frac{1}{N_{0n}} I_{n+\frac{1}{2}}(a_m) \sqrt{\frac{2\pi}{k_m R}}$$

$$\times \sum_{s=0}^{\infty} C_s \Big[\sum_{\sigma=|s-n|}^{s+n} (-1)^{n+s+\sigma} b_{\sigma}^{(s0n0)} K_{\sigma+\frac{1}{2}}(k_m R) \Big], \quad n = 1, 2, \dots ; \tag{5.5b}$$

$$A_0 \left[2a_f I'_{\frac{1}{2}}(a_f) - I_{\frac{1}{2}}(a_f) \right] = C_0 \left[2a_m K'_{\frac{1}{2}}(a_m) - K_{\frac{1}{2}}(a_m) \right]$$

$$+ \frac{1}{N_{00}} \sqrt{\frac{2\pi}{k_m R}} \left[2a_m I'_{\frac{1}{2}}(a_m) - I_{\frac{1}{2}}(a_m) \right] \sum_{s=0}^{\infty} C_s K_{s+\frac{1}{2}}(k_m R), \tag{5.6b}$$

$$A_n \left[2a_f I'_{n+\frac{1}{2}}(a_f) - I_{n+\frac{1}{2}}(a_f) \right] = C_n \left[2a_m K'_{n+\frac{1}{2}}(a_m) - K_{n+\frac{1}{2}}(a_m) \right]$$

$$+ \frac{1}{N_{0n}} \sqrt{\frac{2\pi}{k_m R}} \left[2a_m I'_{n+\frac{1}{2}}(a_m) - I_{n+\frac{1}{2}}(a_m) \right]$$

$$\times \sum_{s=0}^{\infty} C_s \left[\sum_{\sigma=|s-n|}^{s+n} (-1)^{n+s+\sigma} b_{\sigma}^{(s0n0)} K_{\sigma+\frac{1}{2}}(k_m R) \right], \quad n = 1, 2, \dots . \tag{5.6b}$$

Simple manipulations, employing the well-known properties

$$I'_s(x) = \frac{1}{2} \big[I_{s+1}(x) + I_{s-1}(x) \big], \quad K'_s(x) = -\frac{1}{2} [K_{s+1}(x) + K_{s-1}(x)]$$

of the modified Bessel functions, allow to exclude A_n from Eqs. (5.5) so that Eq. (5.6) yields the needed equations for the coefficients C_n:

$$C_0 U_0 + V_0 \sum_{s=0}^{\infty} C_s K_{s+\frac{1}{2}}(k_m R) = -2 \frac{[k^2]}{k_f^2} a_f I_{\frac{3}{2}}(a_f),$$

$$C_n U_n + V_n \sum_{s=0}^{\infty} C_s \left[\sum_{\sigma=|s-n|}^{s+n} (-1)^{n+s+\sigma} b_{\sigma}^{(s0n0)} K_{\sigma+\frac{1}{2}}(k_m R) \right] = 0, \tag{5.7}$$

$n = 1, 2, \dots$, where

$$U_n = a_f K_{n+\frac{1}{2}}(a_m) \Big[I_{n-\frac{1}{2}}(a_f) + I_{n+\frac{3}{2}}(a_f) \Big]$$

$$+ a_m I_{n+\frac{1}{2}}(a_f) \Big[K_{n-\frac{1}{2}}(a_m) + K_{n+\frac{3}{2}}(a_m) \Big], \tag{5.8a}$$

$$V_n = \frac{1}{N_{on}} \sqrt{\frac{2\pi}{k_m R}} \Big\{ a_f I_{n+\frac{1}{2}}(a_m) \Big[I_{n-\frac{1}{2}}(a_f) + I_{n+\frac{3}{2}}(a_f) \Big]$$

$$- a_m I_{n+\frac{1}{2}}(a_f) \Big[I_{n-\frac{1}{2}}(a_m) + I_{n+\frac{3}{2}}(a_m) \Big] \Big\}, \tag{5.8b}$$

and $a_f = ak_f$, $a_m = ak_m$ are dimensionless parameters, see Eq. (4.6c).

For the coefficients A_n we get in turn:

$$A_0 = \frac{1}{I_{\frac{1}{2}}(a_f)} \left\{ \frac{[k^2]}{k_f^2} + C_0 K_{\frac{1}{2}}(a_m) + \frac{1}{2} I_{\frac{1}{2}}(a_m) \sqrt{\frac{2\pi}{k_m R}} \sum_{s=0}^{\infty} C_s K_{s+\frac{1}{2}}(k_m R) \right\}, \tag{5.9a}$$

$$A_n = \frac{1}{I_{n+\frac{1}{2}}(a_f)} \left\{ C_n K_{n+\frac{1}{2}}(a_m) + \frac{1}{N_{0n}} I_{n+\frac{1}{2}}(a_m) \sqrt{\frac{2\pi}{k_m R}} \right.$$

$$\left. \times \sum_{s=0}^{\infty} C_s \left[\sum_{\sigma=|s-n|}^{s+n} (-1)^{n+s+\sigma} b_\sigma^{(s0n0)} K_{\sigma+\frac{1}{2}}(k_m R) \right] \right\}, \quad n \;=\; 1, 2, \ldots . \qquad (5.9\text{b})$$

In this way the two-sphere problem (4.10) is reduced to the solution of the infinite system of linear equations (5.7) for any given separation distance R between the spheres.

Note that if the spheres are well apart, the following asymptotic formula for the coefficient A_0 holds

$$A_0 = \frac{[k^2]}{k_f^2} \sqrt{\frac{\pi a_f}{2}} \frac{1 + a_m}{a_f \cosh a_f + a_m \sinh a_f} + o\left(\frac{1}{\sqrt{R_m}} e^{-R_m} \right), \quad R_m \gg 1 \qquad (5.10)$$

which is a simple consequence of Eqs. (5.7), (5.8a), (5.9a) and the well known asymptotic behaviour of the spherical Bessel functions; $R_m = Rk_m$ is the dimensionless distance.

The natural numerical procedure to solve the system (5.7) is the method of truncation. Namely, assuming $C_n = 0$ at $n > N$ in Eq. (5.7), we get a linear system of $N + 1$ equations for the first $N + 1$ coefficients C_n, $n = 0, 1, \ldots$. Solving the latter, we find the approximate values $C_n^{(N)}$ of these coefficients. Then, at $N \to \infty$, the approximations $C_n^{(N)}$ will converge to the exact values C_n. The proof of this fact, i.e., the justification of the truncation method, needs a bit more detailed investigation of the asymptotic behaviour of the coefficients in the system (5.7), which is performed in Ref. 10.

Due to the exponential decay of the modified Bessel functions $K_{n+\frac{1}{2}}(x)$, $n = 0, 1, \ldots,$ the series solution developed in the present section converges very rapidly when the spheres are well apart. For instance, to obtain the values of the coefficients A_n, C_n and the field $H^{(2)}(\mathbf{x}; \mathbf{z})$ with three decimal digits, it suffices to take $N = 10$ if $R/a \geq 3$. However, as the spheres approach each other, more equations should be kept in the truncated system, e.g., at $2.1 < a/R < 3$ we should take $N = 20$ in order to have the same three decimal digits correct.

6. Results and Discussion

Having described an effective procedure for solving the two-sphere problem (4.10) (Section 5), we are now in a position to evaluate numerically all statistical characteristics connected with the solution of Eq. (1.1). We start with the simplest one—the effective sink strength k^{*2} of the dispersion. Since the c-coefficient b_1 is given in Eqs. (4.13b) and (4.11b) in a closed form, we are interested in the c^2-coefficient only. Upon inserting the solutions (4.6) and (5.2) for the one- and two-sphere problems, respectively, into Eq. (4.11c) and using the orthogonality of the Legendre polynomials, we find

$$b_2 = b_1^2 + 9 \frac{[k^2]}{k_m^2} \frac{I_{\frac{3}{2}}(a_f)}{a_f a_m^3} \int_{2a_m}^{\infty} R_m^2 g_0(R_m/k_m) G_0(R_m)\, dR_m,$$

$$G_0(R_m) = A_0(R_m) - \frac{[k^2]}{k_f^2}\sqrt{\frac{\pi a_f}{2}}\frac{1+a_m}{a_f \cosh a_f + a_m \sinh a_f}, \tag{6.1}$$

where $R_m = Rk_m$, $A_0(R_m) = A_0$ is the value of the coefficient A_0 in the spherical wave expansion (5.2a) for the function $H^{(2)}(\mathbf{x};\mathbf{z})$. According to Eq. (5.10), the function G_0 decays exponentially at infinity which guarantees the absolute convergence of the integral in Eq. (6.1). The truncation method for the system (5.7) is then used together with the Simpson formula for numerical integration. The number of equations N in the truncated system is so chosen as to guarantee three correct decimal digits for the values of A_0 (see the comments at the end of Section 5). The integral in Eq. (6.1) is replaced by one over the finite interval $(0, R_m^0)$; using the asymptotic (5.10), the value R_m^0 is so chosen, in turn, as to make the contribution of the integral over the interval (R_m^0, ∞) less than 0.0001. The step in the Simpson integration formula for the remaining integral over the finite interval $(0, R_m^0)$ is appropriately decreased until the three decimal digits of the values of the integral become stable. The obtained numerical values of the coefficient b_2 are given in the Table 1 for the simplest case when $g_0(r) = 1$ at $r \geq 2a$ and vanishes otherwise.

TABLE 1

Values of the c^2-coefficient b_2 of the sink strength of the dispersion at $a_m = ak_m = 1, 10$ and various $a_f = ak_f$.

a_f/a_m	0.01	0.1	0.25	0.5	1	2	5	10	100
$a_m = 1$	0.792	0.748	0.650	0.381	0	1.801	10.219	15.927	22.624
$a_m = 10$	4.492	3.402	2.604	1.729	0	0.248	0.390	0.426	0.430

Some comments, concerning the values of b_2, as shown in Table 1, are warranted. Let us recall the result of Papanicolaou, see, e.g., Ref. 17, concerned with the limiting cases of Eq. (1.1). First, if $a_f, a_m \ll 1$, then $k^{*2} \approx ck_f^2 + (1-c)k_m^2$, i.e., $\dfrac{k^{*2}}{k_m^2} = 1 + c\dfrac{[k^2]}{k_m^2}$. This means that $b_2 \ll 1$ if $a_f \ll 1$ and $a_f/a_m < 1$.

Second, if $a_f, a_m \gg 1$, then

$$\frac{1}{k^{*2}} \approx \frac{c}{k_f^2} + \frac{1-c}{k_m^2}, \quad \text{i.e.,} \quad \frac{k^{*2}}{k_m^2} \approx \frac{1}{1 + c\dfrac{[k^2]}{k_f^2}},$$

so that

$$b_2 \approx \left(\frac{[k^2]}{k_f^2}\right)^2 = \left(\frac{(a_f/a_m)^2 - 1}{a_f^2/a_m^2}\right)^2 < 1$$

at $a_m \gg 1$ and $a_f/a_m > 1$, approaching 1 as $a_f/a_m \to \infty$.

Considerable values of b_2 are this to be expected only when a_m and a_f have widely different magnitudes, say, $a_m \leq 1$ and $a_f \gg 1$, or $a_f = 1$, $a_m \gg 1$. The numerical results, shown in Table 1, confirm this conclusion.

TABLE 2

Autovariance $F(\mathbf{0})$; $a_m = 1$;
$a_f = 0.1, 10$; $c = 0.05, 0.10, 015$.

c	0.05	0.10	0.15
$a_f = 0.1$	0.006	0.013	0.022
$a_f = 10$	0.154	0.420	0.799

The numerical evaluation of the covariance function $M_2^\varphi(\mathbf{x})$, as given in Eq. (4.13), is performed in the same manner as that of b_2. In Table 2 the values of the (dimensionless) autovariance $F(\mathbf{0}) = \dfrac{k_m^4}{K^2} M_2^\varphi(\mathbf{0})$ are first shown, for $c = 0.05,\ 0.10$ and 0.15. (Since Eq. (4.14) is correct to the order c^2 only, the values it predicts for $c > 0.15$ are already unreliable.)

In turn, the values of the correlation function

$$\rho_2^\varphi(r) = \frac{M_2^\varphi(r)}{M_2^\varphi(0)} = \frac{F(r)}{F(0)}, \quad r = |\mathbf{x}|,$$

are given in Tables 3 and 4 as functions of the dimensionless length r/a for the same values of a_m, a_f and c. The results shown in these tables indicate that the two-point correlation is not very sensitive to the sphere fraction c, especially at $a_f > a_m$ (Table 4). Thus we can approximately factorize $M_2^\varphi(r)$

$$M_2^\varphi(r) \approx M_2^\varphi(0) m(r/a),$$

where $m(r/a)$ does not depend on the volume fraction c; the fraction c influences strongly only the autovariance function $M_2^\varphi(0)$, see Table 2.

TABLE 3

Dimensionless two-point correlation function $\rho_2^\varphi(r/a)$;
$a_m = 1$, $a_f = 0.1$; sphere fractions $c = 0.05, 0.10, 015$.

x/a	0	1	2	3	4	5	6	7
$c = 0.05$	1	0.707	0.379	0.155	0.052	0.017	0.006	0.001
$c = 0.10$	1	0.735	0.402	0.167	0.061	0.023	0.008	0.002
$c = 0.15$	1	0.757	0.420	0.180	0.067	0.025	0.009	0.003

TABLE 4

The same as in Table 3 for $a_f = 10$.

x/a	0	1	2	3	4	5	6	7
$c = 0.05$	1	0.755	0.423	0.194	0.078	0.031	0.012	0.005
$c = 0.10$	1	0.776	0.451	0.211	0.088	0.036	0.014	0.005
$c = 0.15$	1	0.788	0.464	0.221	0.094	0.039	0.015	0.006

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