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On the Heat Propagation Problem for Random Dispersions of Spheres[1]

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The problem of heat conduction through a random dispersion of nonoverlapping spheres is considered. The full stochastic description of the random temperature field, $\theta(\boldsymbol{x})$, in the dispersion is obtained in the form of a factorial functional series. Such series, recently introduced in [1], have the important property that their truncations after the p-tuple term give results for all needed multipoint correlation functions which are correct to the order c^p ; here c is the volume fraction of the spheres, $p = 1, 2, \ldots$. The procedure of identification of the kernels in the series is considered in detail for the case $p = 2$ so that the full stochastic description of $\theta(\boldsymbol{x})$, correct to the order c^2, is obtained in a closed form. In particular, the effective conductivity of the dispersion is found to the same order and shown to coincide with the known formula of D. Jeffrey [2].

1. Introduction

Let us consider an infinite heterogeneous medium of random constitution. For definiteness, we shall deal with the problem of heat conduction through the medium as a simple representative of a wide class of similar transport phenomena, conveniently tabulated and discussed in [3].

Let $\kappa(\boldsymbol{x})$ be the random field of thermal conductivity of the medium which we assume to be statistically homogeneous and isotropic. The temperature field, $\theta(\boldsymbol{x})$, in the medium, at the absence of body sources, is governed by the equations

$$\nabla\theta(\boldsymbol{x}) = 0, \quad \theta(\boldsymbol{x}) = \kappa(\boldsymbol{x})\nabla\theta(\boldsymbol{x}), \tag{1.1a}$$

where $\theta(\boldsymbol{x})$ is the opposite heat flux vector. We prescribe also the mean value of the temperature gradient to be constant

$$\langle\nabla\theta(\boldsymbol{x})\rangle = \boldsymbol{G}, \tag{1.1b}$$

which plays the role of a boundary condition for the random equation (1.1a). The brackets $\langle\cdot\rangle$ denote ensemble averaging.

The random problem (1.1) is the typical one for the theory of heterogeneous composite materials of random constitution. This is obviously a nonlinear problem, because the known field

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$\kappa(\boldsymbol{x})$ multiplies the unknown $\theta(\boldsymbol{x})$, and this strong nonlinearity is the source of all difficulties in the theory of such materials. The uniqueness and existence theorem for the random problem (1.1) was proved in [4] under the natural condition $0 < K_1 \leq \kappa(\boldsymbol{x}) \leq K_2 < \infty$.

Let us recall now [5] that to solve the random problem (1.1) means to determine all r-point moments (or equivalently the correlation functions) of the temperature field $\theta(\boldsymbol{x})$, i.e. the averages

$$\langle \theta(\boldsymbol{x}_1) \dots \theta(\boldsymbol{x}_r) \rangle \,, \tag{1.2a}$$

and the joint moments of $\theta(\boldsymbol{x})$ and $\kappa(\boldsymbol{x})$, i.e. the averages

$$\langle \theta(\boldsymbol{x}_1) \dots \theta(\boldsymbol{x}_s) \kappa(\boldsymbol{x}_{s+1}) \dots \kappa(\boldsymbol{x}_{s+l}) \rangle \,, \tag{1.2b}$$

$r, s, l = 1, 2, \dots$, by means of the known moments

$$\langle \kappa(\boldsymbol{x}_1) \dots \kappa(\boldsymbol{x}_p) \rangle \,, \tag{1.3}$$

$p = 1, 2, \dots$, of the given conductivity field. In particular, among the joint moments (1.2b) one has to evaluate the one-point moment

$$\langle \kappa(\boldsymbol{x}) \nabla\theta(\boldsymbol{x}) \rangle = \kappa^* \langle \nabla\theta(\boldsymbol{x}) \rangle = \kappa^* \boldsymbol{G}, \tag{1.4}$$

where κ^* is the effective conductivity of the medium.

The parameter κ^* and its elastic counterparts are, as a matter of fact, the only characteristics of random media which has been extensively studied in the literature, see, e.g., [6] or the survey [7]. The reasons for the attention paid to κ^* are twofold: first, κ^* is the simplest nontrivial scalar characteristics of the random solution $\theta(\boldsymbol{x})$ of the problem (1.1); second, and perhaps more important, κ^* prescribes the effective macroscopical response of the random medium. However, it is well acknowledged now, due to the pioneering work of W. Brown [8], that to calculate κ^* rigorously one needs the full statistical description of the random medium, i.e., all multipoint moments (1.3) of the conductivity field $\kappa(\boldsymbol{x})$. Thus both problems — the particular one of calculating κ^* and the much more general one of solving (1.1) in statistical sense, i.e., of finding the random temperature field $\theta(\boldsymbol{x})$, — require one and the same amount of statistical information. In the author's view this fact implies that, except for some simplest cases, the said two problems are of one and the same level of difficulty. Moreover, any attempt to calculate κ^* rigorously can be successful only if it is somehow incorporated within the solution of the more general problem (1.1). Or, to put it differently, the calculation of κ^* cannot, in general, be torn away from the full stochastic solution of the problem (1.1). This means, in particular, that everyone of statistical characteristics (1.2) of the field $\theta(\boldsymbol{x})$, and not only κ^*, should be of importance when studying macroscopic response of a random medium. May be somewhat similar arguments stimulated M. Beran to consider evaluation and bounding, in certain simple cases, of some other statistical characteristics in random media, e.g., the variances (in our context the quantities $\langle |\nabla\theta'(0)|^2 \rangle$ and $\langle |\boldsymbol{q}'(0)|^2 \rangle$, see [9] and the references therein; hereafter the prime will denote the fluctuating part of the respective random variable.

We shall illustrate here this point of view on the classical example of a stationary random dispersion of equisized nonoverlapping spheres. Such a random medium has been considered by many authors starting perhaps with J. Maxwell [10] who gave the famous heuristic formula for its effective conductivity

$$\frac{\kappa^*}{\kappa_m} = 1 + \frac{3\beta c}{1 - \beta c} = 1 + 3\beta c + 3\beta c^2 + \dots \,; \tag{1.5}$$

here $\beta = [\kappa]/(k_f + 2k_m)$, $[\kappa] = k_f - k_m$, k_f and k_m are the conductivities of the spheres and of the matrix respectively and c is the volume fraction of the spheres.

A number of approaches and models borrowed from physics were also proposed to evaluate κ^* among them the ideas of self-consistency, effective field, coherent potential approximation, etc., see again [6, 7] et al. As is well known, these models look very reasonable; the problem with them is that they are not extracted from a rigorous statistical analysis and therefore, as a rule, it is rarely clear what kind of random constitution, if any, lies behind the respective formulae for the effective properties to which the models lead. Moreover, some negative results were recently obtained by the author [11], e.g., it appeared that the Maxwell formula (1.5) is not realizable for a dispersion of nonoverlapping spheres even to the order c^2.

The difficulties in calculating κ^* have made many authors consider a simpler problem which consists in the following. Let

$$\frac{\kappa^*}{\kappa_m} = 1 + a_1 c + a_2 c^2 + \cdots \tag{1.6}$$

be the so-called virial expansion of the effective conductivity of the dispersion, i.e. the expansion in powers of the volume fraction c of the spheres. As noted by D. Jeffrey [2], the coefficient a_1 is, as a matter of fact, the only thing rigorously calculated by J. Maxwell: $a_1 = 3\beta$, see (1.5); it does not depend on the statistics of the dispersion. The c^2-coefficient a_2 is however strongly influenced by sphere's distribution and therefore its evaluation should be much more complicated.

The problem of evaluating a_2 for a given statistics of the dispersion has been extensively treated in the literature after the famous work of A. Einstein, [12] who obtained the c-term in the effective viscosity of a fluid dispersion containing rigid spheres. Many attempts to extend the Einstein formula to the order c^2 have been made, see, e.g., [13] for a detailed survey of the work done prior to 1956 along this line. The basic idea underlying the up-to-date calculation of the coefficient a_2 is the so-called cluster (or group) expansion. This idea, introduced by Finkel'berg [14], and later on employed and/or elaborated in many papers, to mention only [2, 15, 16], consists, loosely speaking, in representing κ^* as a sum of consecutive terms that result from interactions within successively larger groups of spherical inhomogeneities. Without entering a more lengthy discussion, we shall only mention the following. (i) The cluster expansions concern the effective properties only and not the full statistical description of the random fields under study. (ii) They are usually introduced on the base of a certain heuristic reasoning unconvincing, in the author's view, in general. (Moreover, some arguments of D. Jeffrey and J. McCoy [17, p.21], clearly indicate that there exist hidden difficulties in justification of the cluster expansion ideology even on heuristic level.) (iii) The formula for a_2, derived by means of the cluster expansion, contains nonabsolutely convergent integrals which should somehow be unambiguously defined. To this end G. Batchelor [18] and D. Jeffrey [2] devised the so-called "renormalization procedure" in order to render the respective integrals absolutely convergent. A detailed and very careful analysis of this procedure was performed by D. Jeffrey [19] who assigned a specific physical meaning of the various terms that appear, e.g., the renormalizing quantities were interpreted as a result of the long-rang interactions in the dispersions.

Another approach was introduced by J. Willis and J. Acton [20] (in the elastic context) who started with an integral equation for the so-called polarization field. In this equation, however, the polarization was replaced by its fluctuation, in order to make the integrals absolutely

convergent; such a replacement also represents a kind of "renormalization procedure" adopted in [20] without any comment. The approach of E. HINCH [21] consists in deriving an infinite hierarchy of equations for the averaged field quantities like multipoint moments and to decouple it asymptotically for dilute concentrations; making use again of a certain "renormalization" he reached the c^2-formula of G. BATCHELOR and J. GREEN [22] for the viscosity of a fluid suspension.

The foregoing remarks suffice to explain why one should look for a different and more rigorous, than cluster expansions with renormalization, approach when evaluating a_2, in particular, and in the statistical theory of random dispersions, in general.

As it will be seen below, for a wide class of dispersions the evaluation of a_2 and the evaluation of the random temperature field $\theta(\boldsymbol{x})$ to the order c^2 both require one and the same amount of statistical information, namely, the zero-density limit, $g_0(\boldsymbol{x})$, of the radial distribution function $g_0(\boldsymbol{x})$ for the set of sphere's centers. Similarly to the above said, concerning the evaluation of κ^* and the solution of the problem (1.1), this fact implies in the author's view that the problem of evaluation of the c^2-coefficient a_2 cannot be satisfactorily and rigorously solved unless it is incorporated into the solution of the more general problem of full statistical solution of (1.1), correct to the order c^2, i.e., of evaluation of all multipoint moments (1.2) to this order of accuracy.

Following this line of arguments, we shall explicitly construct in this paper the full statistical solution of the problem (1.1) for dispersions of nonoverlapping spheres in the form of factorial functional series. The definition and basic features of such series are recalled in §2. The procedure of identification of the kernels in the factorial series for the temperature field $\theta(\boldsymbol{x})$ is detailed in the case $p = 2$ (§3), and the needed kernels of the truncated factorial series in this case are explicitly found in §4. In this way we can obtain in a closed form, correct to the order c^2, all multipoint moments (1.2) for the random dispersion of spheres (§5). In the simplest particular case, when calculating κ^* to the order c^2, we shall derive the renormalized formula of G. BATCHELOR and D. JEFFREY in a rigorous way.

2. Factorial functional series

To accomplish our aim of solving the problem (1.1) for random dispersions we shall utilize the functional series approach. This approach seems to be novel in mechanics of random heterogeneous solids; it was recently introduced in [23, 24]. The basic idea that underlies the approach consists in the following. The random problem (1.1) defines implicitly a nonlinear operator which transforms the random conductivity field $\kappa(\boldsymbol{x})$ — the "input" — into the random temperature field $\theta(\boldsymbol{x})$ — the "output":

$$\theta(\cdot) = \mathcal{L}[\kappa(\cdot)].$$

Following a general idea of system theory, see, e.g., [25], we shall expand the operator $\mathcal{L}$ as a functional series generated by the input $\kappa(\boldsymbol{x})$, i.e.

$$\theta(\boldsymbol{x}) = K_0(\boldsymbol{x}) + \int K_1(\boldsymbol{x}-\boldsymbol{y})\kappa(\boldsymbol{y})\,\mathrm{d}\boldsymbol{y}$$

$$+\iint K_2(\boldsymbol{x}-\boldsymbol{y}_1, \boldsymbol{x}-\boldsymbol{y}_2)\kappa(\boldsymbol{y}_1)\kappa(\boldsymbol{y}_2)\,\mathrm{d}\boldsymbol{y}_1\mathrm{d}\boldsymbol{y}_2 + \cdots, \tag{2.1}$$

with certain nonrandom kernels K_0, $K_1, \ldots$. Hereafter, if the integration domain is not explicitly indicated, the integrals are taken over the whole R^3.

Having represented the field $\theta(\boldsymbol{x})$ as the functional series (2.1), it is natural to truncate the latter after the p-th tuple term in order to obtain certain approximations, $\theta^{(p)}(\boldsymbol{x})$, for $\theta(\boldsymbol{x})$. Such a truncation immediately brings forth two basic questions, common for all problems in which functional series are employed,

(i) How to rearrange the terms of the series so as the said truncations "converge" to the field $\theta(\boldsymbol{x})$

$$\theta^{(p)}(\boldsymbol{x}) \to \theta(\boldsymbol{x}), \quad p \to \infty \tag{2.2}$$

(ii) In what sense the convergence in (2.2) is to be understood.

For a wide class of random dispersions the answer to these two questions was given in [1] so that we should first recall briefly some of the basic results of this paper.

Let $\boldsymbol{x}_j$ be the random point set comprising all sphere's centers. The statistics of the dispersions is conveniently described by means of the probability density functions $f_k = f_k(\boldsymbol{y}_1, \dots, \boldsymbol{y}_k)$ which give the probability dP to find simultaneously a point from the set $\boldsymbol{x}_j$ per each of the infinitesimal volumes $\boldsymbol{y}_i < \boldsymbol{y} < \boldsymbol{y}_i + \mathrm{d}\boldsymbol{y}_i$, $i = 1, \dots, k$, to be

$$dP = f_k(\boldsymbol{y}_1, \dots, \boldsymbol{y}_k)\, \mathrm{d}\boldsymbol{y}_1 \dots \mathrm{d}\boldsymbol{y}_k, \quad k = 1, 2, \dots ,$$

see [26]. The assumption of nonoverlapping yields

$$f_k(\boldsymbol{y}_1, \dots, \boldsymbol{y}_k) = 0, \quad \text{if } |\boldsymbol{y}_i - \boldsymbol{y}_j| < 2a, \quad \text{for a pair } i \neq j. \tag{2.3}$$

Hereafter we assume all random sets and fields statistically homogeneous and isotropic; then, in particular, we have $f_1(\boldsymbol{y}) = n$, where n is the number density of the set $\boldsymbol{x}_j$, i.e., the mean number of spheres per unit volume. Obviously, $n = c/V_a$, where $V_a = \frac{4}{3}\pi a$ is the volume of a single sphere. Also $f_k(\boldsymbol{y}_1, \dots, \boldsymbol{y}_k) = f_k(\boldsymbol{y}_{k1}, \dots, \boldsymbol{y}_{kk-1}$, where $\boldsymbol{y}_{ij} = \boldsymbol{y}_i - \boldsymbol{y}_j$, $k > 1$.

Let us imagine that by means of a certain manufacturing process one produces dispersions with different number densities n. The statistics of the dispersions will then depend on n as a parameter, i.e. $f_k = f_k(\boldsymbol{Y}_k; n)$, $\boldsymbol{Y}_k = (\boldsymbol{y}_1, \dots, \boldsymbol{y}_k)$. If the manufacturing process is "smooth" enough, we may write

$$f_k(\boldsymbol{Y}_k; n) = \sum_{l=1}^{\infty} f_{kl}(\boldsymbol{Y}_k) n^l, \quad k \geq 1. \tag{2.4}$$

After [11] we adopt now the following basic assumption concerning the constitution of the dispersions which are produced

$$\begin{gathered} \text{The distance between the nearest spheres tends to infinity} \\ \text{as } n \to 0, \text{ i.e. for dilute sphere fraction.} \end{gathered} \tag{2.5}$$

This means that the process does not put spheres in rigid complexes, say, in dumb-bells. The assumption (2.5) is thus fully natural when we speak of dispersions of spheres; otherwise we should have spoken of dispersions of dumb-bells or other more complicated sphere complexes.

A simple analysis carried out by the author [12, p.II] shows that the assumption (2.2) yields $f_{kl} = 0$ at $l < k$, and therefore

$$f_k(\boldsymbol{Y}_k; n) = n^k f_{kk}(\boldsymbol{Y}_k) + o(n^k), \tag{2.6}$$

i.e. $f_k \sim n^k$, $k = 1, 2, \ldots$, which is the usual assumption for the probability densities f_k. We, however, prefer to start with the assumption (2.6) because it seems mote clear intuitively.

A very convenient characteristics of the random dispersion is the so-called random density field

$$\psi(\boldsymbol{x}) = \sum_j \delta(\boldsymbol{x} - \boldsymbol{x}_j), \tag{2.7}$$

generated by the set $\boldsymbol{x}_j$ of sphere's centers [26]. Since the field $\psi(\boldsymbol{x})$is uniquely defined by the set $\boldsymbol{x}_j$, its moments can be expressed by means of the densities functions f_k which describe the set $\boldsymbol{x}_j$ statistically. The respective formulae are given by R. Stratonovich [26]; they read:

$$\langle \psi(\boldsymbol{y}) \rangle = f_1(\boldsymbol{y}) = n,$$

$$\langle \psi(\boldsymbol{y}_1)\psi(\boldsymbol{y}_2) \rangle = f_1(\boldsymbol{y}_1)\delta(\boldsymbol{y}_1 - \boldsymbol{y}_2) + f_2(\boldsymbol{y}_1, \boldsymbol{y}_2),$$

$$\langle \psi(\boldsymbol{y}_1)\psi(\boldsymbol{y}_2)\psi(\boldsymbol{y}_3) \rangle = f_1(\boldsymbol{y}_1)\delta(\boldsymbol{y}_1 - \boldsymbol{y}_2)\delta(\boldsymbol{y}_1 - \boldsymbol{y}_3)$$

$$+3\{\delta(\boldsymbol{y}_1 - \boldsymbol{y}_2) f_2(\boldsymbol{y}_1, \boldsymbol{y}_3)\}_s + f_3(\boldsymbol{y}_1, \boldsymbol{y}_2, \boldsymbol{y}_3), \tag{2.8}$$

etc., where $\langle \cdot \rangle_s$ means symmetrization with respect to all different combinations of indices in the brackets. Relations (2.8) show that, vice versa, the multipoint moments of $\psi(\boldsymbol{x})$ define uniquely the multipoint distribution densities f_k. Thus the random density field $\psi(\boldsymbol{x})$ provides an alternative form of characterization of the random set $\boldsymbol{x}_j$ and, in turn, of the random dispersion of spheres.

The usefulness of the field $\psi(\boldsymbol{x})$ in the study of particulate media was demonstrated in [22,23]; it stems from the fact that the random conductivity field $\kappa(\boldsymbol{x})$ for the dispersions under study has a simple integral representation by means $\mathrm{j}(\boldsymbol{x})$, namely,

$$\kappa(\boldsymbol{x}) = k_m + [\kappa] \int h(\boldsymbol{x} - \boldsymbol{y}) \psi(\boldsymbol{y}) \, \mathrm{d}\boldsymbol{y}, \tag{2.9}$$

where $h(\boldsymbol{x})$ is the characteristic function for a single sphere located at the origin, i.e., $h(\boldsymbol{x}) = 1$, if $|\boldsymbol{x}| < a$ and vanishes otherwise.

Let us introduce now the set of random fields, generated by the random density field $\psi(\boldsymbol{x})$:

$$\Delta_\psi^{(0)} = 1, \quad \Delta_\psi^{(1)}(\boldsymbol{y}) = \psi(\boldsymbol{y}), \ldots,$$

$$\Delta_\psi^{(k)}(\boldsymbol{y}_1, \cdots, \boldsymbol{y}_k) = \psi(\boldsymbol{y}_1)[\psi(\boldsymbol{y}_1)\delta(\boldsymbol{y}_2 - \boldsymbol{y}_1)] \tag{2.11}$$

$$\cdots [\psi(\boldsymbol{y}_k) - \delta(\boldsymbol{y}_k - \boldsymbol{y}_1) - \cdots - \delta(\boldsymbol{y}_k - \boldsymbol{y}_{k-1})],$$

$k = 2, 3, \cdots$, which we call the factorial fields or, briefly, the factorials for the set $\boldsymbol{x}_j$. To the best of our knowledge, the fields (2.10) in this form were first introduced by C. Christov [27] whose aim was to obtain a simple general formula for the multivariate Charlier polynomials. The name factorials stems from the formula

$$\Delta_\psi^{(k)}(\boldsymbol{y}_1, \cdots, \boldsymbol{y}_k) = \begin{cases} f_k(\boldsymbol{y}_1, \cdots, \boldsymbol{y}_k)\psi(\boldsymbol{y}_1) \cdots \psi(\boldsymbol{y}_k), & \text{if } \boldsymbol{y}_i \neq \boldsymbol{y}_j, \\ 0, & \text{if } \boldsymbol{y}_i = \boldsymbol{y}_j \text{ for a pair } i \neq j. \end{cases} \tag{2.11}$$

whose proof is given in [1].

Since the field $\psi(\boldsymbol{x})$ provides an exhaustive statistical description of the random dispersion, we can use it as an input in a functional expansion similar to (2.1). In turn, we can replace in this expansion the products $\psi(\boldsymbol{y}_1)\dots\psi(\boldsymbol{y}_k)$ by the factorial fields (2.11):

$$\theta(\boldsymbol{x}) = T_0(\boldsymbol{x}) + \int T_1(\boldsymbol{x}-\boldsymbol{y})\Delta_\psi^{(1)}(\boldsymbol{y})\,\mathrm{d}\boldsymbol{y}$$

$$+\iint T_2(\boldsymbol{x}-\boldsymbol{y}_1,\boldsymbol{x}-\boldsymbol{y}_2)\Delta_\psi^{(2)}(\boldsymbol{y}_1,\boldsymbol{y}_2)\,\mathrm{d}\boldsymbol{y}_1\mathrm{d}\boldsymbol{y}_2+\cdots\ . \tag{2.12}$$

The kernels T_p in (2.12) can be easily expressed by means of the kernels of the series (2.1); no new notations are used for them. Series of the type (2.12) are called in [1] factorial. The basic result of [1] states that for the class of dispersions that comply with the assumption (2.5) and thus with (2.6), the series (2.12) is virial. This means that the convergence in (2.2) is virial in the sense that the truncations $\theta^{(p)}(\boldsymbol{x})$ of the series (2.12) give results for all multipoint moments (1.2) of the solution $\theta(\boldsymbol{x})$ to the random problem (1.1) which are correct to the order c^p, provided the kernels T_i are properly identified. The performance of the factorial series was illustrated in [1] on the steady-state diffusion problem in a random lossy dispersion and the first three kernels T_0, T_1, T_2 were asymptotically found yielding a c^2-solution to the respective random problem. Here we shall turn to the application of the factorial series (2.12) for solving the random problem (1.1) for the dispersions that satisfy the condition (2.5).

3. Equations for the kernels of the factorial series

The identification of the kernels T_i in the factorial series (2.12) can be performed by means of the procedure, successfully employed in [23, 24, 28] et al. in some particular situations. The procedure consists in the following [1]. Suppose we want to determine the field $\theta(\boldsymbol{x})$ to the order n^p, i.e., c^p, only, $p = 1, 2, \dots$. Then the truncation $\boldsymbol{q}(\boldsymbol{x})$ is solely needed and thus the kernels $T_0, T_1, \dots, T_p$ are to be specified. Consider the equation (1.1a) that governs $\theta(\boldsymbol{x})$, insert there $\theta^{(p)}(\boldsymbol{x})$ instead of $\theta(\boldsymbol{x})$, and $\kappa(\boldsymbol{x})$ according to (2.1), multiply by multiply by 1, $\Delta_\psi^{(1)}(0), \dots, \Delta_\psi^{(p)}(0, \boldsymbol{z}_1, \dots, \boldsymbol{z}_{p-1})$ and average the results, keeping in mind (1.1b) as well. Making use of the formulae for the average values of the respective products of the factorials (they are straightforward consequences of (2.8) and (2.10), see, e.g., (3.4) below), and truncating them to the same order n^p, we get a system of $p+1$ equations for the needed kernels T_0 to T_p.

In what follows we shall consider in detail the case $p = 2$ only, making use of some ideas and results, sketched briefly in the author's lecture [29]. The reasons for such a choice of p are threefold: First, this suffices to demonstrate well the technique to be used for an arbitrary p. Second, the length of calculations is kept within reasonable limits and some tangible results are achieved. Third, the obtained solution is valid to the order c^2 — a case, as already mentioned in §1, considered by many authors when calculating the effective conductivity which provides a number of known results for comparison and discussion.

The n^2-analysis below is facilitated if an n^2-orthogonal system of basic fields is employed instead of the factorials. The n^2-orthogonality means that the average values of any pair of different basic fields is of order higher than n^2 and thus it could be neglected within the frame of the n^2-approximation under study. Let us point out that this is a particular case of the notion of virial orthogonality, introduced by the author in [29, 1], for functionals generated by sets of random points. This notion is weaker than the Wiener notion of orthogonality in

stochastic sense [30], but unlike the latter, virial orthogonality can be accomplished for any random system $\boldsymbol{x}_j$ that complies with the assumption (2.5), see [1, 23, 29] for comments and details.

The n^2-orthogonal system comprises the fields [29]:

$$D_\psi^{(0)} = 1, \quad D_\psi^{(1)}(\boldsymbol{y}) = \Delta_\psi^{(1)}(\boldsymbol{y}) - n = \psi'(\boldsymbol{y}),$$

$$D_\psi^{(2)}(\boldsymbol{y}_1, \boldsymbol{y}_2) = \Delta_\psi^{(2)}(\boldsymbol{y}_1, \boldsymbol{y}_2) - n g_0(\boldsymbol{y}_{12}) \left[D_\psi^{(1)}(\boldsymbol{y}_1) + D_\psi^{(1)}(\boldsymbol{y}_2) \right] - n^2 g_0(\boldsymbol{y}_{12}),$$

$$D_\psi^{(p)}(\boldsymbol{y}_1, \ldots, \boldsymbol{y}_p) = \Delta_\psi^{(p)}(\boldsymbol{y}_1, \ldots, \boldsymbol{y}_p), \quad p = 3, 4, \ldots. \tag{3.1}$$

Here $\psi'(\boldsymbol{y})$ is the fluctuating part of $\psi(\boldsymbol{y})$ and

$$g_0(\boldsymbol{y}) = f_{22}(\boldsymbol{y}) = g(\boldsymbol{y}) + O(n), \tag{3.2}$$

so that $g_0(\boldsymbol{y})$ is the leading term, i.e., the zero-density limit, in the virial expansion of the usual radial distribution function $g(\boldsymbol{y}) = f_2(\boldsymbol{y})/n^2$ for the random set $\boldsymbol{x}_j$ of sphere's centers. The representation (3.2) follows from (2.6) at $k = 2$.

As a consequence of (3.1), (2.8) and (2.10) it can be readily verified that

$$\left\langle D_\psi^{(1)}(\boldsymbol{y}) \right\rangle = 0, \quad \left\langle D_\psi^{(2)}(\boldsymbol{y}_1, \boldsymbol{y}_2) \right\rangle = o(n^2),$$

$$\left\langle D_\psi^{(1)}(\boldsymbol{y}_1) D_\psi^{(2)}(\boldsymbol{y}_2, \boldsymbol{y}_3) \right\rangle = o(n^2). \tag{3.3}$$

Since the series (2.12) is virial, (3.3) suffices to claim that the fields (3.1) do form an n^2-orthogonal system.

In what follows we shall also need the following formulae:

$$\left\langle D_\psi^{(1)}(\boldsymbol{y}_1) D_\psi^{(1)}(\boldsymbol{y}_2) \right\rangle = n\delta(\boldsymbol{y}_{12}) - n^2 R_0(\boldsymbol{y}_{21})$$

$$\left\langle D_\psi^{(1)}(\boldsymbol{y}_1) D_\psi^{(1)}(\boldsymbol{y}_2) D_\psi^{(1)}(\boldsymbol{y}_3) \right\rangle = n\delta(\boldsymbol{y}_{21})\delta(\boldsymbol{y}_{31}) - n^2 3\{\delta(\boldsymbol{y}_{21}) R_0(\boldsymbol{y}_{21})\}_s,$$

$$\left\langle D_\psi^{(2)}(\boldsymbol{y}_1, \boldsymbol{y}_2) D_\psi^{(1)}(\boldsymbol{y}_3) D_\psi^{(1)}(\boldsymbol{y}_4) \right\rangle = \left\langle D_\psi^{(2)}(\boldsymbol{y}_1, \boldsymbol{y}_2) D_\psi^{(2)}(\boldsymbol{y}_3, \boldsymbol{y}_4) \right\rangle$$

$$= n^2 g_0(\boldsymbol{y}_{21}) \left[\delta(\boldsymbol{y}_{31})\delta(\boldsymbol{y}_{42}) + \delta(\boldsymbol{y}_{32})\delta(\boldsymbol{y}_{41}) \right], \tag{3.4}$$

$$\left\langle D_\psi^{(2)}(\boldsymbol{y}_1, \boldsymbol{y}_2) D_\psi^{(2)}(\boldsymbol{y}_3, \boldsymbol{y}_4) D_\psi^{(1)}(\boldsymbol{y}_5 \right\rangle$$

$$= n^2 g_0(\boldsymbol{y}_{12}) [\delta(\boldsymbol{y}_{51}) + \delta(\boldsymbol{y}_{52})][\delta(\boldsymbol{y}_{31})\delta(\boldsymbol{y}_{42}) + \delta(\boldsymbol{y}_{41})\delta(\boldsymbol{y}_{32})],$$

$\boldsymbol{y}_{ij} = \boldsymbol{y}_i - \boldsymbol{y}_j$, $R_0(\boldsymbol{y}) = 1 - g_0(\boldsymbol{y})$; they are correct to the order n^2 and represent straightforward consequences of (2.8), (2.10), (3.1) and (3.3).

Let us truncate the series (2.12) after the two-tuple term. Due to the virial property of this series we can obtain in this way the c^2-solution of the random problem (1.1) for the dispersion

provided the kernels are properly identified. In the truncated series we rearrange the terms and introduce the n^2-orthogonal fields $D_\psi^{(1)}$ and $D_\psi^{(2)}$, given in (3.1), instead of $\Delta_\psi^{(1)}$, $\Delta_\psi^{(2)}$:

$$\theta(\boldsymbol{x}) = \boldsymbol{G}\cdot\boldsymbol{x} + \int T_1(\boldsymbol{x}-\boldsymbol{y}) D_\psi^{(1)}(\boldsymbol{y})\,\mathrm{d}\boldsymbol{y}$$

$$+\iint T_2(\boldsymbol{x}-\boldsymbol{y}_1, \boldsymbol{x}-\boldsymbol{y}_2) D_\psi^{(2)}(\boldsymbol{y}_1,\boldsymbol{y}_2)\,\mathrm{d}\boldsymbol{y}_1\mathrm{d}\boldsymbol{y}_2 + \cdots. \tag{3.5}$$

The new kernels T_1 and T_2 here (no new notations are used for them) are linear combinations of the kernels T_0,T_1 and T_2 of the series (2.12). The kernels T_1 and T_2 depend also on the number density n of the spheres. The kernel T_2 is a symmetric function of its arguments; moreover, it can be shown that the condition of nonoverlapping yields

$$T_2(\boldsymbol{z}_1,\boldsymbol{z}_2) = 0, \quad \text{if } |\boldsymbol{z}_1-\boldsymbol{z}_2| < 2a. \tag{3.6}$$

The zeroth-order term in (3.5) is indeed $\boldsymbol{G}\cdot\boldsymbol{x}$ since $D_\psi^{(1)}$ and $D_\psi^{(2)}$ are centered, cf. (3.3), and since $\theta(\boldsymbol{x})$ should satisfy (1.1b).

We thus conclude that the c^2-solution of the basic random problem (1.1) for the dispersion requires that the nonrandom kernels T_1 and T_2 of the truncated series (3.5) be identified. To this end we employ the above mentioned scheme: We insert (2.9) and (3.5) into (1.1a), multiply by $D_\psi^{(1)}(0)$ and $D_\psi^{(2)}(0,\boldsymbol{z})$ and average the results. In virtue of (3.3) and (3.4) we get eventually

$$k_m\Delta S(\boldsymbol{x}) + [\kappa]\nabla\cdot\Big\{h(\boldsymbol{x})[\boldsymbol{G}+\nabla S(\boldsymbol{x})] - nF_0(\boldsymbol{x})G$$

$$+n[V_a - F_0(\boldsymbol{x})]\nabla S(\boldsymbol{x}) - n\int \nabla S(\boldsymbol{x}-\boldsymbol{y})h(\boldsymbol{x}-\boldsymbol{y})R_0(\boldsymbol{y})\,\mathrm{d}\boldsymbol{y}$$

$$+2ng_0(\boldsymbol{y})h(\boldsymbol{x}-\boldsymbol{y})\nabla T_2(\boldsymbol{x}-\boldsymbol{y},\boldsymbol{x})\,\mathrm{d}\boldsymbol{y}\Big\} = 0, \tag{3.7a}$$

$$[1-R_0(\boldsymbol{z})]\nabla\cdot\Big\{2[\kappa_m + [\kappa](h(\boldsymbol{x})+h(\boldsymbol{x}-\boldsymbol{z})]\nabla T_2(\boldsymbol{x},\boldsymbol{x}-\boldsymbol{z})$$

$$+[\kappa][h(\boldsymbol{x})\nabla T_1(\boldsymbol{x}-\boldsymbol{z}) + h(\boldsymbol{x}-\boldsymbol{z})\nabla T_1(\boldsymbol{x})]\Big\} = 0, \tag{3.7b}$$

where

$$S(\boldsymbol{x}) = T_1(\boldsymbol{x}) - n\int T_1(\boldsymbol{x}-\boldsymbol{y})R_0(\boldsymbol{y})\,\mathrm{d}\boldsymbol{y}, \tag{3.8}$$

$$F_0(\boldsymbol{x}) = \int h(\boldsymbol{x}-\boldsymbol{y})R_0(\boldsymbol{y})\,\mathrm{d}\boldsymbol{y}. \tag{3.9}$$

Everywhere in (3.7) the differentiation is with respect to $\boldsymbol{x}$, $\nabla = \nabla_x$, $\boldsymbol{z}$ plays the role of a parameter.

4. Virial solution of the system (3.7)

We shall look for the solution of the system (3.7) in the truncated virial form

$$T_1(\boldsymbol{x}) = T_1(\boldsymbol{x};n) = T_{10}(\boldsymbol{x}) + nT_{11}(\boldsymbol{x}),$$

$$T_2(\boldsymbol{y}_1, \boldsymbol{y}_2) = T_2(\boldsymbol{y}_1, \boldsymbol{y}_2; n) = T_{20}(\boldsymbol{y}_1, \boldsymbol{y}_2); \tag{4.1}$$

we underline that the kernels T_1 and T_2 depend on the number density n as well. This form is justified in our c^2-analysis because in all formulae for averaged statistical quantities T_1 appears multiplied by n and T_2 — by n^2. The virial coefficients T_{10}, T_{11} and T_{22} do not already depend on the number density n; they comply with the system

$$\kappa_m \Delta S(\boldsymbol{x}) + [\kappa]\nabla \cdot \Big\{ h(\boldsymbol{x})[\boldsymbol{G} + \nabla S_0(\boldsymbol{x})\Big\} = 0, \tag{4.2a}$$

$$\kappa_m \Delta S(\boldsymbol{x}) + [\kappa]\nabla \cdot \Big\{ h(\boldsymbol{x}) \nabla S_1(\boldsymbol{x}) + [V_a - F_0(\boldsymbol{x})]\nabla S_0(\boldsymbol{x})$$
$$- \int \nabla S_0(\boldsymbol{x} - \boldsymbol{y}) h(\boldsymbol{x} - \boldsymbol{y}) R_0(\boldsymbol{y})\, \mathrm{d}\boldsymbol{y} + 2I_{20}\boldsymbol{x}) \Big\} = 0, \tag{4.2b}$$

$$[1 - R_0(\boldsymbol{z})]\nabla \cdot \Big\{ 2[\kappa_m + [\kappa](h(\boldsymbol{x}) + h(\boldsymbol{x} - \boldsymbol{z}))]\nabla T_{20}(\boldsymbol{x}, \boldsymbol{x} - \boldsymbol{z})$$
$$+ [\kappa]\left[h(\boldsymbol{x}) \nabla T_{10}(\boldsymbol{x} - \boldsymbol{z}) + h(\boldsymbol{x} - \boldsymbol{z}) \nabla T_{10}(\boldsymbol{x}) \right] \Big\} = 0, \tag{4.2c}$$

which follows straightforwardly if one inserts (4.1) into (3.7). Here

$$I_{20}(\boldsymbol{x}) = \int g_0(\boldsymbol{y}) h(\boldsymbol{x} - \boldsymbol{y}) \nabla T_{20}(\boldsymbol{x} - \boldsymbol{y}, \boldsymbol{x})\, \mathrm{d}\boldsymbol{y} \tag{4.3}$$

and $S(\boldsymbol{x}) = S_0(\boldsymbol{x}) + nS_1(\boldsymbol{x})$, so that S_0 and S_1 are the first two virial coefficients of the function $S(\boldsymbol{x})$, defined in (3.8); obviously

$$S_0(\boldsymbol{x}) = T_{11}(\boldsymbol{x}), \tag{4.4}$$

$$(4.5) S_1(\boldsymbol{x}) = T_{11}(\boldsymbol{x}) - \int T_{10}(\boldsymbol{x} - \boldsymbol{y}) R_0(\boldsymbol{y})\, \mathrm{d}\boldsymbol{y}. \tag{4.5}$$

Unlike the basic system (3.7), the system (4.2) for the needed virial coefficients T_{10}, T_{11}, T_{22} is already decoupled[3] and can be analytically solved. Indeed, Eqn. (4.2a) is nothing but the equation for the disturbance $T^{(1)}(\boldsymbol{x})$ to the temperature field in an unbounded matrix introduced by a single spherical inhomogeneity, when the temperature gradient at infinity is $\boldsymbol{G}$. Its solution is thus well known to be

$$T_{10}(\boldsymbol{x}) = S_0(\boldsymbol{x}) = \begin{cases} -\beta \boldsymbol{G} \cdot \boldsymbol{x}, & \text{if } r < a, \\ -\beta a^3 \boldsymbol{G} \cdot \boldsymbol{x}/r^3, & \text{if } r > a, \end{cases} \tag{4.6}$$

where $r = |\boldsymbol{x}|$ and $\beta = [\kappa]/(zk_m + 2\kappa_f)$; therefore $T_{10}(\boldsymbol{x})$ does not depend on the statistics of the dispersion.

[3]Let us note that E. Hinch [21] has used a similar idea of asymptotical decoupling, at $c \to 0$, of his hierarchy of equations for the averaged field quantities in a suspension. This seems to be, however, the only point of resemblance between our approach and that of Hinch's.

Due to (4.6), Eqn. (4.2b) can be recast as

$$\kappa_m \Delta S_1(\boldsymbol{x}) + [\kappa]\nabla \cdot \Big\{ h(\boldsymbol{x})\nabla S_1(\boldsymbol{x}) + [V_a - F_0(\boldsymbol{x})]\nabla T_1(\boldsymbol{x})$$

$$+(\beta - 1)F_0(\boldsymbol{x})\boldsymbol{G} + 2I_{20}(\boldsymbol{x})\Big\} = 0, \tag{4.7}$$

which allows us to specify S_1 provided we know the function T_{20} and thus the integral I_{20}, defined in (4.3).

Consider the equation (4.2c) for T_{20}. Similarly to (4.2a) it does not depend on the statistics of the medium and therefore it coincides with that derived in [23] for the particular case of a well-stirred dispersion (for which $g(\boldsymbol{y}) = g_0(\boldsymbol{y}) = 1$ at $|\boldsymbol{y}| > 2a$). As is shown in [23], the solution of (4.2c) has the form

$$2T_{20}(\boldsymbol{x} - \boldsymbol{z}, \boldsymbol{x}) = T^{(2)}(\boldsymbol{x}; \boldsymbol{z}) - T_{10}(\boldsymbol{x}) - T_{10}(\boldsymbol{x} - \boldsymbol{z}), \tag{4.8}$$

where $T^{(2)}(\boldsymbol{x}; \boldsymbol{z})$ is the disturbance to the temperature field in an unbounded matrix, introduced by a pair of identical spherical inhomogeneities with centers at the origin and at the point $\boldsymbol{z}$, $|\boldsymbol{z}| \geq 2a$, when the temperature gradient at infinity equals $\boldsymbol{G}$. Each one of these inclusions, if it were alone, would disturb the temperature field in the homogeneous matrix by $T_{10}(\boldsymbol{x})$ and $T_{10}(\boldsymbol{x} - \boldsymbol{z})$, respectively. Thus the field $T_{20}(\boldsymbol{x} - \boldsymbol{z}, \boldsymbol{x})$ is proportional to the field which should be added to the single-inclusion disturbances $T_{10}(\boldsymbol{x})$ and $T_{10}(\boldsymbol{x} - \boldsymbol{z})$ in order to obtain the double-inclusion disturbance $T^{(2)}(\boldsymbol{x}; \boldsymbol{z})$. Note that the foregoing interpretation of the virial coefficients T_{10} and T_{20} is fully similar to that of the respective coefficients in the steady-state diffusion problem [1].

It is important to point out that a simple asymptotical analysis of Eqn. (4.2c) yields

$$2T_{20}(\boldsymbol{x} - \boldsymbol{z}, \boldsymbol{x}) = \beta a^3 \nabla T_{10}(\boldsymbol{x} - \boldsymbol{z}) \cdot \nabla \frac{1}{\boldsymbol{x}} + o(|\boldsymbol{z}|^{-3}), \quad |\boldsymbol{z}| \gg 1. \tag{4.9}$$

The field $T^{(2)}(\boldsymbol{x}; \boldsymbol{z})$ can be analytically found, e.g., by means of the method of twin expansions [14, 2, 15], and therefore we shall think it known. Hence we know, at least in principle, the functions T_{10} and T_{20}, both independent of the statistics of the dispersion and therefore only Eqn. (4.7) for the function S_1, and thus for T_{11}, remains to be solved. This needs first of all an evaluation of the integral I_{20} in (4.3) which seems a very difficult tusk if we try to do it directly. We shall encompass this difficulty by replacing I_{20} with another integral which is much easier to be calculated. We shall argue as follows.

Let us rewrite Eqn. (4.2c) in the form

$$[1 - R_0(\boldsymbol{z})]\nabla \cdot \Big\{ 2[\kappa] h(\boldsymbol{x} - \boldsymbol{z}) \nabla T_{20}(\boldsymbol{x} - \boldsymbol{z}, \boldsymbol{x}) \Big\}$$

$$= -[1 - R_0(\boldsymbol{z})]\nabla \cdot \Big\{ 2[\kappa_m + [\kappa] h(\boldsymbol{x})] \nabla T_{20}(\boldsymbol{x} - \boldsymbol{z}, \boldsymbol{x}) \tag{4.10}$$

$$-[\kappa][h(\boldsymbol{x})\nabla T_{10}(\boldsymbol{x} - \boldsymbol{z}) + h(\boldsymbol{x} - \boldsymbol{z})\nabla T_{10}(\boldsymbol{x})] \Big\}.$$

Due to the presence of $h(\boldsymbol{x} - \boldsymbol{z})$, the left-hand side of (4.10) is absolutely integrable with respect to $\boldsymbol{z}$ in the region $Z_{2a} = \{|\boldsymbol{z}| \mid |\boldsymbol{z}| > 2a\}$ and thus the same holds for the right-hand side. (Let us remind that the differentiation everywhere is with respect to $\boldsymbol{x}$ so that it commutes

with the integration with respect to $\boldsymbol{z}$.) That is why we can choose the mode of integration as we wish. We choose, the reason to become clear a bit later, the following mode of integration

$$\int \cdot \, \mathrm{d}\boldsymbol{z} = \lim_{R\to\infty} \int_{Z_{2a,R}} \cdot \, \mathrm{d}\boldsymbol{z}; \quad \int_{Z_{2a,R}} \cdot \, \mathrm{d}\boldsymbol{z} = \int_{2a}^{R} r^2 \, \mathrm{d}r \int_{\Omega} \cdot \, \mathrm{d}\Omega, \tag{4.11}$$

where $Z_{2a,R} = \{|\boldsymbol{z}| \mid 2a < |\boldsymbol{z}| \le R\}$. This means that in the integrals over $Z_{2a,R}$ we first integrate with respect to the angular coordinates, i.e., on the unit sphere $\Omega = \{|\boldsymbol{z}| \mid |\boldsymbol{z}| = 1\}$, and then with respect to the radial coordinate $r = |\boldsymbol{z}|$.

Let us introduce now the functions

$$L_{10}(\boldsymbol{x}) = \int_{Z_{2a}} g_0(\boldsymbol{z}) \nabla T_{10}(\boldsymbol{x} - \boldsymbol{z}) \, \mathrm{d}\boldsymbol{z}, \tag{4.12}$$

$$L_{20}(\boldsymbol{x}) = \int_{Z_{2a}} g_0(\boldsymbol{z}) T_{20}(\boldsymbol{x} - \boldsymbol{z}, \boldsymbol{x}) \, \mathrm{d}\boldsymbol{z}. \tag{4.13}$$

Due to (4.6) and (4.9), both integrals in (4.12) and (4.13) are not absolutely convergent. However, they exist in the sense (4.11) since when integrating first with respect to the angular coordinates the contribution of the "bad" leading term $1/|\boldsymbol{z}|^3$ vanishes; the latter is well seen from (4.6) and from the asymptotics (4.9) for the function T_{20}. The remaining terms are already of the order $1/|\boldsymbol{z}|^m$, $m \ge 4$, as $|\boldsymbol{z}| \to \infty$, so that they are absolutely integrable. Let us point out that the existence of the integrals (4.12) and (4.13) in the sense (4.11) is just the reason why this mode of integration is chosen.

Let us integrate now (4.10) with respect to $\boldsymbol{z}$ over the region Z_{2a} taking the integrals in the sense (4.11) when needed

$$2\kappa_m \Delta L_{20}(\boldsymbol{x}) + [\kappa] \nabla \cdot \Big\{ 2h(\boldsymbol{x}) L_{20}(\boldsymbol{x}) + 2I_{20}(\boldsymbol{x}) \\ + h(\boldsymbol{x}) L_{10}(\boldsymbol{x}) + \nabla T_{10}(\boldsymbol{x}) \int g_0(\boldsymbol{z}) h(\boldsymbol{x} - \boldsymbol{z}) \, \mathrm{d}\boldsymbol{z} \Big\} = 0.$$

Thus, having introduced the conditionally convergent integrals (4.12) and (4.13), we have combined them in (4.14) in such a manner which cancels out the contributions of the non convergent parts of the two integrals, in order to obtain an expression, (4.14), containing the absolutely convergent integral $I_{20}(\boldsymbol{x})$. As noted by D. Jeffrey [2], a similar combination of conditionally convergent integrals appeared in the classical Einstein work [12] on effective viscosity of suspensions.

In virtue of (3.9) we have

$$\int_{Z_{2a}} g_0(\boldsymbol{z}) h(\boldsymbol{x} - \boldsymbol{z}) \, \mathrm{d}\boldsymbol{z} = V_a - F_0(\boldsymbol{x}). \tag{4.15}$$

Also, it may be easily shown that

$$h(\boldsymbol{x}) L_{10}(\boldsymbol{x}) = 0, \quad \text{i.e.} \quad L_{10}(\boldsymbol{x}) = 0 \text{ at } \boldsymbol{x} < a. \tag{4.16}$$

Keeping in mind (4.15) and (4.16), we recast Eqn. (4.14) as

$$\kappa_m \Delta L_{20}(\boldsymbol{x}) + [\kappa] \nabla \cdot \Big\{ 2h(\boldsymbol{x}) \nabla L_{20}(\boldsymbol{x}) + I_{20}(\boldsymbol{x}) \\ + [V_a - F_0(\boldsymbol{x})] \nabla T_{10}(\boldsymbol{x})(\boldsymbol{x}) \Big\} = 0. \tag{4.17}$$

We next subtract (4.17) from (4.7) and write down the result in the form

$$\kappa_m \Delta H(\boldsymbol{x}) + [\kappa]\nabla \cdot \Big\{ h(\boldsymbol{x}) \nabla H(\boldsymbol{x}) + (\beta - 1) F_0(\boldsymbol{x}) G \Big\} = 0, \tag{4.18}$$

where

$$H(\boldsymbol{x}) = S_1(\boldsymbol{x}) - 2L_{20}(\boldsymbol{x}) \tag{4.19}$$

is the new unknown function instead of $S_1(\boldsymbol{x})$. Thus we have eliminated the integral $I_{20}(\boldsymbol{x})$ from our analysis, replacing it by the integral $L_{20}(\boldsymbol{x})$ which, though conditionally convergent (in the sense (4.11)) is much easier to be evaluated. And what is more important, Eqn. (4.18) can be readily solved in a closed form.

Eqn. (4.18) first appeared in [23] in fully different context (in the frame of the so-called singular approximation in the theory of effective conductivity) in the particular case of a well-stirred dispersion. In this case the solution of (4.19) was obtained in [23] only within the sphere $|\boldsymbol{x}| < a$. For an arbitrary function $g_0(\boldsymbol{y})$ the solution of (4.19) is

$$H(\boldsymbol{x}) = \begin{cases} \beta(1-\beta) V_a \boldsymbol{G} \cdot \boldsymbol{x}, & \text{if } r < a, \\ \left(-\beta^2 a^3 V_a + 3\beta \displaystyle\int_0^r \rho^2 F_0(\rho)\, \mathrm{d}\rho \right) \boldsymbol{G} \cdot \boldsymbol{x}/r^3, & \text{if } r > a, \end{cases} \tag{4.20}$$

which can be easily found if one considers separately Eqn. (4.19) in the regions $|\boldsymbol{x}| < a$ and $|\boldsymbol{x}| > a$ and then use the respective conditions of continuity for H($\boldsymbol{x}$) and its normal flux of $\kappa(\boldsymbol{x})\nabla H(\boldsymbol{x})$ on the surface $|\boldsymbol{x}| = a$.

The formula (4.20) terminates the solution, correct to the order c^2, of the basic random problem (1.1) for the dispersions under study. Indeed, Eqns. (4.19) and (4.20) define $S_1(\boldsymbol{x})$ and $T_{11}(\boldsymbol{x})$ can be then calculated by means of (4.5). The functions $T_{10}(\boldsymbol{x})$ and $T_{20}(\boldsymbol{x} - \boldsymbol{z}, \boldsymbol{x})$ are already determined through the solutions of the one- and two-sphere problems respectively, see (4.6) and (4.8). Upon introducing T_{10}, T_{11} and T_{22} into (4.1), we shall explicitly obtain the kernels $T_1(\boldsymbol{x})$ and $T_2(\boldsymbol{y}_1, \boldsymbol{y}_2)$ of the truncated functional series (3.5) which gives the full stochastic solution of the problem (1.1) to the order c^2, as already explained. In this way all statistical characteristics (1.2) of the random temperature field can be evaluated to the order c^2 in a closed analytical form using the zero-density limit, $g_0(\boldsymbol{y})$, of the radial distribution function for the set $\boldsymbol{x}_j$ of sphere's centers and the solutions $T^{(1)}(\boldsymbol{x})$ and $T^{(2)}(\boldsymbol{x}; \boldsymbol{z})$ for the one- and two-sphere problems respectively.

Eqns (4.18) and (4.19) reveal aslo one of the reasons why non-absolutely convergent integrals kept appearing in the c^2-theory of effective properties of dispersions, staring with the classical work of Lord Rayleigh [31]: namely, because the solution of the basic stochastic problem (1.1) in this case can be represented in a natural and convenient way by means of such integrals.

5. Calculation of the statistical characteristics in the dispersion to the order c^2

To illustrate the performance of the above obtained c^2-solution of the random problem (1.1), let us evaluate, e.g., the two-point correlation function for the temperature field $\theta(\boldsymbol{x})$. Keeping in mind (3.3) to (3.5), we have

$$M_2^{\theta}(\boldsymbol{x}) = \big\langle \theta'(0)\theta'(\boldsymbol{x}) \big\rangle = c M_{21}^{\theta}(\boldsymbol{x}) + c^2 M_{21}^{\theta}(\boldsymbol{x}) + o(c^2), \tag{5.1}$$

where $\theta'(\boldsymbol{x})$ is the fluctuating part of the temperature and

$$M_{21}^{\theta}(\boldsymbol{x}) = \frac{1}{V_a}\int T_{10}(\boldsymbol{x}-\boldsymbol{y})T_{10}(\boldsymbol{y})\,\mathrm{d}\boldsymbol{y}, \tag{5.2}$$

$$M_{22}^{\theta}(\boldsymbol{x}) = \frac{1}{V_a^2}\Big\{\int T_{10}(\boldsymbol{x}-\boldsymbol{y})T_{11}(\boldsymbol{y})\,\mathrm{d}\boldsymbol{y}$$

$$-\iint T_{10}(\boldsymbol{x}-\boldsymbol{y}_1)T_{10}(\boldsymbol{y}_2)R_0(\boldsymbol{y}_1-\boldsymbol{y}_2)\,\mathrm{d}\boldsymbol{y}_1\,\mathrm{d}\boldsymbol{y}_2 \tag{5.3}$$

$$+2\iint T_{20}(\boldsymbol{x}-\boldsymbol{y}_1,\boldsymbol{x}-\boldsymbol{y}_2)T_{20}(\boldsymbol{y}_1,\boldsymbol{y}_2)\,\mathrm{d}\boldsymbol{y}_1\,\mathrm{d}\boldsymbol{y}_2\Big\}.$$

With the same ease the rest of the multipoint correlation functions (1.2) can be obtained in a similar form. Of course, the explicit evaluation of these functions needs an explicit expression for the function $L_{20}(\boldsymbol{x})$, defined in (4.13), and evaluation of integrals of the type of those that appeared in (5.2) and (5.3). This is not an easy task; however, the difficulties to be encountered concern already deterministic problems and have purely analytical nature.

Consider now the simpler case of one-point statistical characteristics of the type (1.4), i.e., the effective conductivity κ^* of the dispersion. In virtue of (2.9) and (3.3) to (3.5) we have, to the order c^2,

$$\kappa^*\boldsymbol{G} = \langle\kappa(\boldsymbol{x})\nabla\theta(\boldsymbol{x})\rangle = \langle\kappa\rangle\,\boldsymbol{G} + n[\kappa]\int h(\boldsymbol{x})\nabla S(\boldsymbol{x})\,\mathrm{d}\boldsymbol{x}, \tag{5.4}$$

where $S(\boldsymbol{x})$ is given in (3.8). Thus only the values of $\nabla S(\boldsymbol{x})$ within the sphere $V_a = \{\boldsymbol{x} \mid |\boldsymbol{x}| < a\}$ are needed when calculating κ^*. In turn, making use of (4.4) to (4.6), (4.8), (4.13), (4.19) and (4.20), we find the c- and c^2-coefficients in the virial expansion (1.6) of the effective conductivity κ^* to be

$$\frac{\kappa^*}{\kappa_m} = 1 + 3\beta c + (3\beta^2 + a_2')c^2 + o(c^2), \tag{5.5}$$

$$a_2'\boldsymbol{G} = \frac{[\kappa]}{\kappa_m}\,\frac{2}{V_a^2}\int h(\boldsymbol{x})L_{20}(\boldsymbol{x})\,\mathrm{d}\boldsymbol{x}$$

$$= \frac{[\kappa]}{\kappa_m}\,\frac{2}{V_a^2}\int h(\boldsymbol{x})\,\mathrm{d}\boldsymbol{x}\int g_0(\boldsymbol{z})\nabla T_{20}(\boldsymbol{x}-\boldsymbol{z},\boldsymbol{x})\,\mathrm{d}\boldsymbol{z}. \tag{5.6}$$

(Note that a_2' is just the c^2-deviation of the effective conductivity of the dispersion from the Maxwell formula (1.5).)

Due to (4.8) and (4.16), we get the eventual formula for a_2'

$$a_2'\boldsymbol{G} = \frac{[\kappa]}{\kappa_m}\,\frac{2}{V_a^2}\int h(\boldsymbol{x})\,\mathrm{d}\boldsymbol{x}\int_{Z_{2a}} g_0(\boldsymbol{z})\Big[\nabla T^{(2)}(\boldsymbol{x};\boldsymbol{z}) - \nabla T^{(1)}(\boldsymbol{x})\Big]\mathrm{d}\boldsymbol{z}. \tag{5.7}$$

The integral with respect to $\boldsymbol{z}$ in the right-hand side of (5.7) is not absolutely convergent; however, it should be understood in the sense (4.11) so that it exists and gives finite and unambiguous results for a_2', see §4.

Due to the statistical isotropy of the dispersion, Eqns. (5.6) and (5.7) can be recast as

$$a_2' = 3\beta^2 + \int_2^\infty \Phi(\lambda;\beta) g_0(\lambda a)\ \mathrm{d}\lambda, \tag{5.8}$$

where

$$(5.9)\Phi(\lambda;\beta) = \frac{[\kappa]}{\kappa_m}\,\frac{8\pi}{V_a^2}\,\lambda^2 a^2 \int h(\boldsymbol{x})\nabla T_{20}(\boldsymbol{x}-\boldsymbol{z},\boldsymbol{x})\,\mathrm{d}\boldsymbol{x}, \tag{5.9}$$

$\lambda = |\boldsymbol{z}|/a$; we underline here that the kernel Φ depends on the parameter $\beta = [\kappa]/(\kappa_f + 2\kappa_m)$ as well.

It is noteworthy that the integrand in (5.5) can be made absolutely integrable if the above mentioned (§1) "renormalization" is performed. The latter consists in adding to the "bad" integrands quantities which make them absolutely integrable but, at the same time, contribute nothing to the values of the integrals. Afterward certain justification of the procedure is looked for see [2, 3, 18, 19, 22] et al. For Eqn. (5.7), such a quantity, as suggested by D. Jeffrey [2], can be devised using the asymptotical behaviour of the integrand. Indeed, due to (4.9) and (4.16), it can be chosen as $\beta\nabla T^{(1)}(\boldsymbol{x}-\boldsymbol{z})$ which, when added to the integrand does not alter the value of the integral in (5.7):

$$a_2'\boldsymbol{G} = \frac{[\kappa]}{\kappa_m}\,\frac{1}{V_a^2}\int h(\boldsymbol{x}) \int \Big[\nabla T^{(2)}(\boldsymbol{x},\boldsymbol{z}) - \nabla T^{(1)}(\boldsymbol{x}) - \nabla T^{(1)}(\boldsymbol{x}-\boldsymbol{z})\Big]\,\mathrm{d}\boldsymbol{z}. \tag{5.10}$$

On the other hand, the integrand in (5.10) is already absolutely integrable, having the asymptotics $o(|\boldsymbol{z}|^{-3})$ at $|\boldsymbol{z}| \gg 1$. (A more detailed analysis, performed in [2], shows that the asymptotics of this integrand is $O(|\boldsymbol{z}|^{-6})$. This fact follows also from the estimates (5.13) below.) Eqn. (5.10) is just the renormalized formula of D. Jeffrey [2]. The same expression in an equivalent form was derived as well by B. Felderhof et al. [15].

It is clear, however, that the above renormalization is by no means necessary in our analysis, because (4.11) defines unambiguously the mode of integration in (5.7) and makes the value of the integral there finite. The renormalization, within the frame of our approach, can be viewed only as a computational device which may help evaluate numerically the respective integrals.

For a well-stirred dispersion, the integral in (5.7) or, equivalently, in (5.10), was first evaluated by D. Jeffrey and later on by B. Felderhof et al. [15]. Thus the values of κ^* as a function of the ratio $\alpha = \kappa_f/\kappa_m$ or, which is the same, $\beta = (\alpha-1)/(\alpha+2)$, are now available for the well-stirred case. In particular, it appears that

$$a_2 \to 4.506 \quad \text{at} \quad \kappa_f/\kappa_m \to \infty,$$

$$a_2 \to 0.588 \quad \text{at} \quad \kappa_f/\kappa_m \to 0. \tag{5.11}$$

To the best of our knowledge no such results exist for an arbitrary function $g_0(\boldsymbol{y})$. Only recently the author [11, p.II] obtained the following bounds for the c^2-coefficient $a_2 = 3\beta^2 + a_2'$ in Eqn. (5.5)

$$3\beta^2\left(1 + \frac{[\kappa]}{\kappa_f}\,m_2\right) \le a_2 \le 3\beta^2\left(1 + \frac{[\kappa]}{\kappa_f}\,m_2\right), \tag{5.12a}$$

where

$$m_2 = 2\int_2^\infty \frac{\lambda^2}{(\lambda^2-1)^3}\, g_0(\lambda a)\, \mathrm{d}\lambda \tag{5.12b}$$

is a certain statistical parameter for the dispersion.

The bounds (5.11) imply the following estimates for the kernel $\Phi(\lambda;\beta)$ in (5.8):

$$\Phi^l(\lambda;\beta) \le \Phi(\lambda;\beta) \le \Phi^u(\lambda;\beta),$$

$$\Phi^l(\lambda;\beta) = 18\frac{\beta^3}{1+2\beta}\,\frac{\lambda^2}{(\lambda^2-1)^3}\,, \tag{5.13}$$

$$\Phi^u(\lambda;\beta) = 18\frac{\beta^3}{1-\beta}\,\frac{\lambda^2}{(\lambda^2-1)^3}\,.$$

The bounds (5.12) are close only if the conductivities of the spheres and of the matrix do not differ much, i.e., if $\beta \ll 1$. In the two limiting cases $\kappa_f/\kappa_m \to \infty$, i.e. $\beta \to 1$, and $\kappa_f/\kappa_m \to 0$, i.e. $\beta \to -0.5$, one of the bounds (5.12) degenerates.

We can use, however, the nondegenerating bound together with the numerically found values (5.11) in these two cases in order to propose the following approximation for the integrand $\Phi(\lambda;\beta)$ in (5.8):

$$\Phi(\lambda;\beta) = 18\beta^3\,\frac{\lambda^2}{(\lambda^2-1)^3}\begin{cases}\dfrac{b''}{1+2\beta}\,, & \text{if } \kappa_f/\kappa_m > 1,\\[2ex] \dfrac{b''}{1-\beta}\,, & \text{if } \kappa_f/\kappa_m < 1.\end{cases} \tag{5.14a}$$

where

$$b' = 3.57423, \quad b'' = 3.07583. \tag{5.14b}$$

The approximate kernel in (5.14) is proportional to the bound (5.13) which does not degenerate in the respective limiting case, being multiplied by the constants b' or b''. These constants are as chosen as to secure both numerical values (5.11) in these limiting cases for the well-stirred case (for which $m_2 = 0.14045$).

For a well-stirred dispersion the formulae (5.5) and (5.7) for the coefficient a_2 coincide with the formula, proposed by J. Peterson and J. Hermans [32]. These authors employed certain heuristic arguments, tantamount to cluster expansion ideology of Finkel'berg [14] et al. and tacitly adopted the mode of integration (4.11) without giving any reason. They proposed also a useful computational technique for the evaluation of the respective conditionally convergent integrals in (5.7) in the two-dimensional counterpart of the dispersion under study, i.e. for an array of equisized parallel cylinders of radius a subject to mean temperature gradient orthogonal to their axes. In this case κ^* is just the transverse effective conductivity of such a fiber-reinforced medium. The technique of the said authors [32] allows us to obtain the following formula for the transverse conductivity κ^*

$$\frac{\kappa^*}{\kappa_m} = 1 + 2\beta c + 2\beta^2 c^2(1 + 2\beta M(\beta) + o(c^2), \tag{5.15}$$

where $c = nS_a$ is the volume fraction of the fibers, $S_a = \pi a^2$, $\beta = [\kappa]/(\kappa_f + \kappa_m)$ in the two-dimensional case here, and

$$M(\beta) = 16 \sum_{l=1}^{\infty} l \int_0^{\infty} g_0(2\cosh\tau) \frac{\cosh\tau \sinh^3\tau}{1 - \beta^2 \mathrm{e}^{-4l\tau}} \,\mathrm{d}\tau = \sum_{p=0}^{\infty} M_p \beta^{2p}, \tag{5.16}$$

so that

$$M_p = 16 \sum_{l=1}^{\infty} l \int_0^{\infty} g_0(2\cosh\tau) \cosh\tau \sinh^3\tau \, \mathrm{e}^{2l(3+2p)} \,\mathrm{d}\tau, \tag{5.17a}$$

or, equivalently,

$$M_p = \int_2^{\infty} \lambda g_0(\lambda a) \left\{ \sum_{k=0}^{p} (-1)^k C^k_{2+2p-k} \lambda^{2(p-k+1)} \right\}^{-2} \mathrm{d}\lambda. \tag{5.17b}$$

The derivation of (5.15) to (5.17) will be considered in detail elsewhere[4], together with the problem of accuracy for approximations if the type (5.14).

6. Concluding remarks

In this paper we have presented a systematic statistical theory of simple transport phenomena in a wide class of random dispersions of spheres making use, for the sake of definiteness, of the context of heat propagation. Obviously, the theory can serve as a pattern in the studies of similar or more complicated linear or nonlinear stochastic problems concerning elasticity, permeability, etc., for such dispersions or for more general particulate media of random constitution. The success of the theory could be attributed to two things. The first is that we have not narrowed the scope of the analysis looking for the effective conductivity κ^* only. Instead, our goal was much broader — the full stochastic solution of the basic random problem (1.1), whose very particular case is the calculation of κ^*. In this way we appeared able to obtain rigorous and unambiguous results for the multipoint correlation functions of interest and, in particular, for the effective conductivity. The second thing is the appropriate application of functional series which thus once again proved to be highly advantageous for random heterogeneous media and which offer unique possibilities when relating micro- and macro-properties of the media. The decisive point in their application here is the notion of virial convergence and its implementation through the factorial fields of the random sets of sphere' centers.

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