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ON THE EFFECTIVE CONDUCTIVITY OF A CLASS OF RANDOM DISPERSIONS

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A new class of random dispersions is considered in which not only the location of the spheres is random, but their conductivity is random as well. The classical variational principles are employed in which classes of trial fields in the form of suitably truncated functional series are introduced. In this way three-point variational bounds on the effective conductivity of the dispersion are derived and discussed in more detail for some particular statistical distributions of sphere conductivity. A rigorous formula for the effective conductivity, correct to the order square of sphere fraction, is finally obtained which contains only absolutely convergent integrals.

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1. INTRODUCTION

Consider a dispersion of *homogeneous* non-overlapping spheres of *random* conductivity $\widetilde{\kappa}_f$, immersed at random into an unbounded matrix of conductivity κ_m. For convenience of notations hereafter we represent the conductivity $\widetilde{\kappa}_f$ in the form $\widetilde{\kappa}_f = K_f \widetilde{s}$, where $K_f = \langle \widetilde{\kappa}_f \rangle$ is the mean conductivity of the sphere, embedded into the matrix. Then $\widetilde{s}$ represents their "non-dimensional conductivity" for which $\langle \widetilde{s} \rangle = 1$.

Let $\{\mathbf{x}_j\}$ be the random system of sphere's centers and at the position $\mathbf{x}_j$ a sphere with conductivity s_j, random as well, is centered. Thus a set of marked random points $\{\mathbf{x}_j, s_j\}$ is defined whose statistical description suffices for the dispersion. A similar marked random system was considered by Christov and Markov

[1, 2] in the study of dispersions of spheres with random radii $\widetilde{a}$. (For the general definition of sets of marked random points see [3].) We assume henceforth, for the sake of simplicity solely, that the spheres possess a fixed and non-random radius a. Then the random conductivity field $\kappa(\mathbf{x})$ of the dispersion has the form

$$\kappa(\mathbf{x}) = \kappa_m + \sum_j (K_f s_j - \kappa_m) h(\mathbf{x} - \mathbf{x}_j), \tag{1.1}$$

where $h(\mathbf{x})$ is the characteristic function for a single sphere located at the origin. In Sec. 2.1 we briefly discuss the statistical description of the system of marked random points $\{\mathbf{x}_j, s_j\}$, similar to that used in [1, 2].

For definiteness we shall deal with the problem of heat conduction through the random dispersion as a simple representative of a wide class of similar transport phenomena. The governing equations of the problem, in the absence of body sources, are

$$\nabla \cdot \mathbf{q}(\mathbf{x}) = 0, \quad \mathbf{q}(\mathbf{x}) = \kappa(\mathbf{x}) \nabla\theta(\mathbf{x}), \quad \langle \nabla\theta(\mathbf{x}) \rangle = \mathbf{G}, \tag{1.2}$$

where $\theta(\mathbf{x})$ is the random temperature field, $\mathbf{q}(\mathbf{x})$ — the heat flux vector, $\mathbf{G}$ is the prescribed macroscopic value of the temperature gradient, the brackets $\langle \cdot \rangle$ denote statistical averaging. Hereafter the media are assumed statistically homogeneous and isotropic. The solution of Eqs. (1.2) is understood in a statistical sense, so that one is to evaluate all multipoint moments (correlation functions) of $\theta(\mathbf{x})$ and the joint moments of $\kappa(\mathbf{x})$ and $\theta(\mathbf{x})$, see, e.g., [4]. Among the latter is the one-point moment

$$\langle \kappa(\mathbf{x}) \nabla\theta(\mathbf{x}) \rangle = \kappa^* \langle \nabla\theta(\mathbf{x}) \rangle = \kappa^* \mathbf{G}, \tag{1.3}$$

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

As argued by Christov and Markov [5], the solution $\theta(\mathbf{x})$ of the random problem (1.2) can be expanded as a functional (Volterra-Wiener) series, generated by the conductivity field $\kappa(\mathbf{x})$, namely,

$$\theta(\mathbf{x}) = \mathbf{G} \cdot \mathbf{x} + \int K_1(\mathbf{x} - \mathbf{y}) \kappa'(\mathbf{y})\, d^3\mathbf{y}$$

$$+ \iint K_2(\mathbf{x} - \mathbf{y}_1, \mathbf{x} - \mathbf{y}_2) \left[\kappa'(\mathbf{y}_1)\kappa'(\mathbf{y}_2) - M_2^{\kappa}(\mathbf{y}_1 - \mathbf{y}_2) \right] d^3\mathbf{y}_1 d^3\mathbf{y}_2 + \cdots, \tag{1.4}$$

with certain non-random kernels T_i, $i = 1, 2, \ldots$ They also proposed to truncate this series afterwards. In Eq. (1.4) $\kappa'(\mathbf{x}) = \kappa(\mathbf{x}) - \langle \kappa \rangle$, $M_2^{\kappa}(\mathbf{x} - \mathbf{y}) = \langle \kappa'(\mathbf{x}) \kappa'(\mathbf{y}) \rangle$. (Hereafter the integrals with respect to spatial variables are over the whole $\mathbb{R}^3$ if the integration domain is not explicitly indicated.) Two types of applications for such truncated series could be envisaged. The first is to use them as approximate, in a certain sense, solutions of the problem (1.2). This possibility was discussed in more detail and worked out in the case of random dispersions of spheres by Markov [6, 7] and Markov and Christov [2]. For the dispersion under study this kind of application will be explained and worked out in Sec. 2.2. The second is to use such truncated series as classes of trial fields for the variational principles

[8, 12]. This idea was developed by Markov [8] on the base of the classical principle, corresponding to the problem (1.2), namely,

$$W_A[\theta(\cdot)] = \left\langle \kappa(\mathbf{x})|\nabla\theta(\mathbf{x})|^2 \right\rangle \longrightarrow \min, \quad \langle\nabla\theta(\mathbf{x})\rangle = \mathbf{G}, \tag{1.5}$$

$\min W_A = \kappa^* G^2$, see, e.g., [4]. For example, the simplest non-trivial class is obtained when the functional series (1.4) is truncated after the single integral term, i.e.

$$\mathcal{K}_A^{(1)} = \left\{ \theta(\mathbf{x}) \mid \theta(\mathbf{x}) = \mathbf{G}\cdot\mathbf{x} + \int K_1(\mathbf{x}-\mathbf{y})\kappa'(\mathbf{y})\, d^3\mathbf{y} \right\}, \tag{1.6}$$

where $K_1(\mathbf{x})$ is an adjustable kernel. This class was introduced and discussed in detail by Markov [8], where it was shown that minimizing $W_A[\theta(\cdot)]$ over the class $\mathcal{K}_A^{(1)}$ gives the best three-point upper bound $\kappa^{(3)}$ on the effective conductivity κ^*, i.e. the most restrictive one which uses three-point statistical information for the medium. In order to obtain the appropriate three-point lower bound on κ^*, it is necessary to consider the classical dual variational principle for the problem (1.2) formulated with respect to the heat flux $\mathbf{q}(\mathbf{x}) = \nabla \times \mathbf{\Phi}(\mathbf{x})$,

$$W_B[\mathbf{\Phi}(\cdot)] = \left\langle k(\mathbf{x})|\nabla \times \mathbf{\Phi}(\mathbf{x})|^2 \right\rangle \longrightarrow \min, \quad \langle\mathbf{q}(\mathbf{x})\rangle = \mathbf{Q}, \tag{1.7}$$

$\min W_B = k^* Q^2$ (here $k(\mathbf{x}) = 1/\kappa(\mathbf{x})$ and $k^* = 1/\kappa^*$), over a class of the kind (1.6). In Sec. 3.1 we shall derive the optimal three-point bounds for the dispersion making use of an alternative variational procedure successfully applied in the monodisperse case, see [8, 9, 12].

Moreover, Markov [8] showed how the earlier proposed variational techniques could be put into this general frame. For example, the Beran method [13] is a Ritz type procedure in which the kernel K_1 in (1.6) is chosen to be proportional to the fixed (Beran's) kernel K_B:

$$K_1(\mathbf{x}) = \lambda K_B(\mathbf{x}), \quad K_B(\mathbf{x}) = \mathbf{G}\cdot\nabla\frac{1}{4\pi|\mathbf{x}|}, \tag{1.8}$$

where $\lambda \in \mathbb{R}$ is an adjustable parameter. The question of the optimality of Beran's procedure for the dispersion under study will be discussed in Sec. 3.2. It will be shown that it is not optimal even to the order c, where c is the volume fraction of the spheres. Finally, in Sec. 4, using some of the author's ideas of his recent work [14], an exact c^2-formula for the effective conductivity κ^* of the dispersion under study will be found in a variational way.

2. STATISTICAL DESCRIPTION OF THE DISPERSION AND FACTORIAL FUNCTIONAL EXPANSION

2.1. STATISTICAL DESCRIPTION OF THE DISPERSION

The system of marked random points $\{\mathbf{x}_j, s_j\}$ can be considered as a set of points randomly distributed in the four-dimensional domain $\mathbb{R}^3 \times \mathbb{U}$, where $\mathbb{U} = (0, +\infty)$. Similarly to the monodisperse case, this system is fully described

by the multipoint probability densities $F_n(\mathbf{y}_1, \ldots, \mathbf{y}_n; s_1, \ldots, s_n)$, see, e.g., [1, 2, 8]. The latter define the probability

$$dP = F_n(\mathbf{y}_1, \ldots, \mathbf{y}_n; s_1, \ldots, s_n)\, d^3\mathbf{y}_1 \ldots d^3\mathbf{y}_n\, ds_1 \ldots ds_n \tag{2.1}$$

to find simultaneously a center of sphere within the infinitesimal volumes

$$\mathbf{y}_i \le \mathbf{y} < \mathbf{y}_i + d\mathbf{y}_i \tag{2.2a}$$

of the spatial positions $\mathbf{y}_i$ with conductivities $\widetilde{s}_1, \ldots, \widetilde{s}_n$ in the vicinities

$$s_i \le \widetilde{s} < s_i + ds_i \tag{2.2b}$$

of the values $s_1, \ldots, s_n$, respectively, $i = 1, \ldots, n$.

The functions $F_n(\mathbf{y}_1, \ldots, \mathbf{y}_n; s_1, \ldots, s_n)$ define too rich a class of dispersions whose study seems very complicated in general. That is why, if our aim is to reach certain tangible results, one must narrow this class. The following arguments lead in a natural way to such a simplification. Let dP_Y be the probability to find simultaneously a center of sphere in each of the volumes (2.2a), regardless to the conductivity of the latter. Obviously, $dP \le dP_Y$ and

$$dP_Y = f_n(\mathbf{y}_1, \ldots, \mathbf{y}_n)\, d^3\mathbf{y}_1 \ldots d^3\mathbf{y}_n, \tag{2.3}$$

where the functions $f_n(\mathbf{y}_1, \ldots, \mathbf{y}_n)$ are the multipoint probability densities for the system of non-marked random points $\mathbf{x}_j$, i.e. they are the same that appear in the monodisperse case, see, e.g., [8]. Then $dP = dP_Y dP^*$, where dP^* is the conditional probability, namely, the probability to find simultaneously a center of sphere in the volumes (2.2a) with conductivities $\widetilde{s}_1, \ldots, \widetilde{s}_n$ in the regions (2.2b) respectively, provided a center of sphere is found in the volumes (2.2a). Hence

$$dP^* = \eta_n(s_1, \ldots, s_n \mid \mathbf{y}_1, \ldots, \mathbf{y}_n)\, ds_1 \ldots ds_n,$$

where

$$F_n(\mathbf{y}_1, \ldots, \mathbf{y}_n; s_1, \ldots, s_n) = f_n(\mathbf{y}_1, \ldots, \mathbf{y}_n)\, \eta_n(s_1, \ldots, s_n \mid \mathbf{y}_1, \ldots, \mathbf{y}_n), \tag{2.4}$$

$n = 1, 2 \ldots$ Obviously, the dependence of functions η_n upon $\mathbf{y}_1, \ldots, \mathbf{y}_n$ reflects the "selectivity" of these sphere's locations toward spheres of certain conductivities. The consideration of dispersions in the general case, when such a "selectivity" is arbitrary, seems a hopeless problem. That is why we adopt now the following simplifying assumption concerning the structure of the dispersions: *There exist no locations in the space* $\mathbb{R}^3$ *which possess selectivity toward spheres of certain conductivities.* Hence we assume that

$$\eta_n(s_1, \ldots, s_n \mid \mathbf{y}_1, \ldots, \mathbf{y}_n) = P_n(\mathbf{y}_1, \ldots, \mathbf{y}_n)$$

or, according to (2.4),

$$F_n(\mathbf{y}_1, \ldots, \mathbf{y}_n; s_1, \ldots, s_n) = f_n(\mathbf{y}_1, \ldots, \mathbf{y}_n)\, P_n(s_1, \ldots, s_n), \tag{2.5}$$

which means, as a matter of fact, that there is no correlation between location and conductivity of the spheres. The functions $P_n(s_1, \ldots, s_n)$ are the multivariate probability densities of conductivities of spheres, regardless to the spatial positions of the latter; they give the probability dP_S^* of n arbitrarily chosen spheres of the dispersion, having conductivities in the vicinities (2.2b), to be $dP_S^* = P_n(s_1, \ldots, s_n)\, ds_1 \ldots ds_n$.

Since the dispersions under study are assumed statistically homogeneous and isotropic, the system $\{\mathbf{x}_j\}$ has the same properties. Hence, in particular, $f_1 = n$ and $f_k = f_k(\mathbf{y}_{2,1}, \ldots, \mathbf{y}_{k,1})$, where $\mathbf{y}_{i,j} = \mathbf{y}_j - \mathbf{y}_i$ and n denotes the number density, i.e. the mean number of points $\mathbf{x}_j$ per unit volume. Obviously, $n = c/V_a$, where $V_a = \frac{4}{3}\pi a^3$ is the volume of a single sphere. Moreover, we shall assume, as usual, that $f_k \sim n^k$, i.e. f_k has the asymptotic order n^k at $n \to 0$, $k = 1, 2, \ldots$, see [8]. We shall note also that the assumption of non-overlapping of spheres yields

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

Taking into account this assumption and (2.5) for the first pair of probability densities F_1 and F_2 we have

$$F_1(\mathbf{y}; s) = nP(s), \quad F_2(\mathbf{y}_1, \mathbf{y}_2; s_1, s_2) = n^2 g_0(r) P_2(s_1, s_2), \tag{2.6}$$

where $P(s) = P_1(s)$, $r = |\mathbf{y}_2 - \mathbf{y}_1|$ and g_0 is the zero-density limit of the well-known radial distribution function $g(r) = f_2(r)/n^2$, i.e. $g(r) = g_0(r) + O(n)$.

Let

$$\psi(\mathbf{x}; s) = \sum_j \delta(\mathbf{x} - \mathbf{x}_j)\delta(s - s_j) \tag{2.7}$$

be the Stratonovich random density field generated by the system of marked random points $\{\mathbf{x}_j, s_j\}$ (see [15, 2]). According to Eq. (1.1) the field $\kappa(\mathbf{x})$ can be written then as

$$\kappa(\mathbf{x}) = \langle\kappa\rangle + \int\!\!\int (K_f s - \kappa_m) h(\mathbf{x} - \mathbf{y}) \psi'(\mathbf{y}; s)\, d^3\mathbf{y}\, ds, \tag{2.8}$$

where $\psi'(\mathbf{y}; s)$ is the fluctuating part of the field $\psi(\mathbf{y}; s)$. (Hereafter the integrals with respect to the mark s are over the semiaxis $(0, +\infty)$.) The random field $\psi(\mathbf{x}; s)$ is uniquely defined by the random set $\{\mathbf{x}_j, s_j\}$ and vice versa. In particular, the multipoint moments of $\psi(\mathbf{x}; s)$ can easily be expressed by means of the probability densities F_k:

$$\begin{gathered}
\langle\psi(\mathbf{y}; s)\rangle = F_1(\mathbf{y}; s) = nP(s), \\
\langle\psi(\mathbf{y}_1; s_1)\psi(\mathbf{y}_2; s_2)\rangle = F_1(\mathbf{y}_1; s_1)\delta(\mathbf{y}_{1,2})\delta(s_{1,2}) + F_2(\mathbf{y}_1, \mathbf{y}_2; s_1, s_2), \\
\langle\psi(\mathbf{y}_1; s_1)\psi(\mathbf{y}_2; s_2)\psi(\mathbf{y}_3; s_3)\rangle = F_1(\mathbf{y}_1; s_1)\delta(\mathbf{y}_{1,2})\delta(s_{1,2})\delta(\mathbf{y}_{1,3})\delta(s_{1,3}) \\
+3\{\delta(\mathbf{y}_{1,3})\delta(s_{1,3})F_2(\mathbf{y}_1, \mathbf{y}_2; s_1, s_2)\}_s + F_3(\mathbf{y}_1, \mathbf{y}_2, \mathbf{y}_3; s_1, s_2, s_3),
\end{gathered} \tag{2.9}$$

etc., see [1, 2], where $\{\cdot\}_s$ denotes symmetrization with respect to all different combinations of indices in the brackets, $s_{i,j} = s_j - s_i$.

Similarly to the considerations in [6, 7] (for monodisperse case) and [2] (for the dispersion of spheres with random radii), it is reasonable to develop the random temperature field $\theta(\mathbf{x})$ in the following functional series

$$\theta(\mathbf{x}) = T_0(\mathbf{x}) + \int\int T_1(\mathbf{x}-\mathbf{y}, s)\Delta_\psi^{(1)}(\mathbf{y}; s)\, d^3\mathbf{y} ds$$

$$+\int\int\int\int T_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2, s_1, s_2)\Delta^{(2)}(\mathbf{y}_1, \mathbf{y}_2; s_1, s_2)\, d^3\mathbf{x}_1\, d^3\mathbf{y}_2\, ds_1 ds_2 + \cdots, \quad (2.10)$$

where

$$\Delta_\psi^{(0)} = 1, \quad \Delta_\psi^{(1)}(\mathbf{y}; s) = \psi(\mathbf{y}; s),$$

$$\Delta_\psi^{(k)}(\mathbf{y}_1, \ldots, \mathbf{y}_k; s_1, \ldots, s_k) = \psi(\mathbf{y}_1; s_1)[\psi(\mathbf{y}_2; s_2) - \delta(\mathbf{y}_{2,1})\delta(s_{2,1})] \quad (2.11)$$

$$\ldots[\psi(\mathbf{y}_k; s_k) - \delta(\mathbf{y}_{k,1})\delta(s_{k,1}) - \cdots - \delta(\mathbf{y}_{k,k-1})\delta(s_{k,k-1})], \quad k = 2, 3, \ldots,$$

are the random fields, generated by the random density field $\psi(\mathbf{x}; s)$, and called in [7] factorial fields. The kernel T_k in (2.10) can be easily expressed by means of the first k kernels of the series (1.3). According to a basic result of [7], the series (2.10) is virial in the sense that the truncation after the p-tuple term of its gives results for all multipoint moments of the solution $\theta(\mathbf{x})$ to the random problem (1.2), which are correct to the order c^p provided the first kernels T_i, $i = 0, \ldots, p$, are properly identified. A general procedure for the identification of the kernels T_i is described in [2, 6, 7].

Since our aim is the evaluation of the effective conductivity κ^* to the order c^2, we are interested in the solution of the problem (1.2) to the same order. To simplify the analysis, after [2, 6, 7] we render the series (2.10) n^2-orthogonal in the sense that the averaged value of the product of any pair of its different terms has the order $o(n^2)$. To this end we introduce the following linear combinations of the factorial fields (2.11):

$$D_\psi^{(0)} = 1, \quad D_\psi^{(1)}(\mathbf{y}; s) = \Delta_\psi^{(1)}(\mathbf{y}; s) - nP(s) = \psi'(\mathbf{y}; s),$$

$$D_\psi^{(2)}(\mathbf{y}_1, \mathbf{y}_2; s_1, s_2) = \Delta_\psi^{(2)}(\mathbf{y}_1, \mathbf{y}_2; s_1, s_2) - n^2 g_0(\mathbf{y}_{2,1})P_2(s_1, s_2)$$

$$-n^2 g_0(\mathbf{y}_{2,1})P_2(s_1, s_2)[D_\psi^{(1)}(\mathbf{y}_1; s_1)/P(s_1) + D_\psi^{(1)}(\mathbf{y}_2; s_2)/P(s_2)],$$

$$D_\psi^{(k)}(\mathbf{y}_1, \ldots, \mathbf{y}_j; s_1, \ldots, s_k) = \Delta_\psi^{(k)}(\mathbf{y}_1, \ldots \mathbf{y}_k; s_1, \ldots, s_k), \quad (2.12)$$

$k = 3, 4, \ldots$ As a consequence of Eqs. (2.9) and (2.11) it can be easily verified that

$$\left\langle D_\psi^{(1)}(\mathbf{y}; s)\right\rangle = 0, \quad \left\langle D_\psi^{(2)}(\mathbf{y}_1, \mathbf{y}_2; s_1, s_2)\right\rangle = o(n^2), \quad (2.13a)$$

$$\left\langle D_\psi^{(1)}(\mathbf{y}_1; s_1) D_\psi^{(2)}(\mathbf{y}_2, \mathbf{y}_3; s_2, s_3)\right\rangle = o(n^2). \quad (2.13b)$$

Since the series (2.10) is virial, these relations suffice to claim that the fields (2.12) form an n^2-orthogonal system. Then let us truncate the series (2.10) after the four-tuple integral term. Thus we obtain the kind of the c^2-solution of the random problem (1.2) for the dispersion. In the truncated series we rearrange the terms in such a manner that only the n^2-orthogonal fields $D_\psi^{(1)}$ and $D_\psi^{(2)}$ enter:

$$\theta(\mathbf{x}) = \mathbf{G}\cdot\mathbf{x} + \int\int T_1(\mathbf{x}-\mathbf{y},s) D_\psi^{(1)}(\mathbf{y};s)\, d^3\mathbf{y}\, ds$$

$$+ \int\int\int\int T_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2, s_1, s_2) D_\psi^{(2)}(\mathbf{y}_1,\mathbf{y}_2;s_1,s_2)\, d^3\mathbf{y}_1 d^3\mathbf{y}_2\, ds_1 ds_2. \quad (2.14)$$

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 (2.10). The zeroth-order term in (2.14) is indeed $\mathbf{G}\cdot\mathbf{x}$, since $D_\psi^{(1)}$ and $D_\psi^{(2)}$ are centered and $\langle\nabla\theta(\mathbf{x})\rangle = \mathbf{G}$, see Eqs. (2.13) and (1.2).

The identification of the kernels T_1 and T_2 can be performed by a procedure, proposed originally by Christov and Markov [5], see also [2, 6, 7]. It consists in inserting the truncated series (2.14) into the random equation (1.2), multiplying the result by the fields $D_\psi^{(p)}$, $p = 0, 1, 2$, and averaging the results. In this way a certain system of integral-differential equations for the needed kernels of the truncated series can be straightforwardly derived. Here we employ an alternative method, recently proposed in [14] for the monodisperse case. Namely, the truncated series (2.14) will be inserted into the classical variational principle (1.5) as a class of trial fields, varying the kernels. Since this class contains the actual temperature field to the order c^2, the obtained equations for the optimal kernels T_1 and T_2 are the same as those for the needed kernels in (2.14). In particular, this procedure leads in passing to the exact determination of the effective conductivity to the order c^2.

In what follows we shall need also the following formulae for the moments of the field (2.12):

$$\left\langle D_\psi^{(1)}(\mathbf{y}_1;s_1) D_\psi^{(1)}(\mathbf{y}_2;s_2)\right\rangle = nP(s_1)\delta(\mathbf{y}_{1,2})\delta(s_{1,2}) - n^2\mathcal{R}_0(\mathbf{y}_{1,2};s_1,s_2), \quad (2.15a)$$

$$\left\langle D_\psi^{(1)}(\mathbf{y}_1;s_1) D_\psi^{(1)}(\mathbf{y}_2;s_2) D_\psi^{(1)}(\mathbf{y}_3;s_3)\right\rangle = nP(s_1)\delta(\mathbf{y}_{1,2})\delta(s_{1,2})\delta(\mathbf{y}_{1,3})\delta(s_{1,3})$$
$$-n^2 3\{\delta(\mathbf{y}_{1,2})\delta(s_{1,2})\mathcal{R}_0(\mathbf{y}_{2,3};s_2,s_3)\}_s, \quad (2.15b)$$

$$\left\langle D_\psi^{(2)}(\mathbf{y}_1,\mathbf{y}_2;s_1,s_2) D_\psi^{(1)}(\mathbf{y}_3;s_3) D_\psi^{(1)}(\mathbf{y}_4;s_4)\right\rangle$$
$$= \left\langle D_\psi^{(2)}(\mathbf{y}_1,\mathbf{y}_2;s_1,s_2) D_\psi^{(2)}(\mathbf{y}_3,\mathbf{y}_4;s_3,s_4)\right\rangle = n^2 g_0(\mathbf{y}_{2,1}) P_2(s_1,s_2) \quad (2.15c)$$
$$\times[\delta(\mathbf{y}_{3,1})\delta(s_{3,1})\delta(\mathbf{y}_{4,2})\delta(s_{4,2}) + \delta(\mathbf{y}_{4,1})\delta(s_{4,1})\delta(\mathbf{y}_{3,2})\delta(s_{3,2})],$$

$$\left\langle D_\psi^{(2)}(\mathbf{y}_1,\mathbf{y}_2;s_1,s_2) D_\psi^{(2)}(\mathbf{y}_3,\mathbf{y}_4;s_3,s_4) D_\psi^{(1)}(\mathbf{y}_5;s_5)\right\rangle$$
$$= n^2 g_0(\mathbf{y}_{2,1}) P_2(s_1,s_2)[\delta(\mathbf{y}_{5,1})\delta(s_{5,1}) + \delta(\mathbf{y}_{5,2})\delta(s_{5,2})] \quad (2.15d)$$
$$\times[\delta(\mathbf{y}_{3,1})\delta(s_{3,1})\delta(\mathbf{y}_{4,2})\delta(s_{4,2}) + \delta(\mathbf{y}_{4,1})\delta(s_{4,1})\delta(\mathbf{y}_{3,2})\delta(s_{3,2})],$$

where

$$\mathcal{R}_0(\mathbf{y}_{2,1}; s_1, s_2) = P(s_1)P(s_2) - g_0(|\mathbf{y}_{2,1}|)P_2(s_1, s_2)\,; \qquad (2.16)$$

they are correct to the order n^2 and represent straightforward consequences of Eqs. (2.9), (2.11) and (2.12).

3. VARIATIONAL THREE-POINT BOUNDS

3.1. THE OPTIMAL THREE-POINT BOUNDS FOR THE DISPERSION

It is natural to begin the consideration of the classical variational principle (1.5) on the simpler class of trial fields that it yields when the factorial series (2.14) is truncated after the one-tuple integral term. Namely, we introduce the class

$$\mathcal{T}_A^{(1)} = \left\{ \theta(\mathbf{x}) \;\middle|\; \theta(\mathbf{x}) = \mathbf{G}\cdot\mathbf{x} + \int\!\!\int T_1(\mathbf{x}-\mathbf{y}, s) D_\psi^{(1)}(\mathbf{y}; s)\, d^3\mathbf{y}\, ds \right\}, \qquad (3.1)$$

where $T_1(\mathbf{x}, s)$ is an adjustable kernel. Obviously, this class contains the actual temperature field to the order c only. That is why one can obtain the exact value of effective conductivity κ^* to the same order only, together with certain bounds on κ^* for the higher order of c.

This class is the counterpart of the class (1.6). Due to Eq. (2.8), the classes (1.6) and (3.1) coincide: if a transition from $\kappa'(\mathbf{x})$ to $\psi'(\mathbf{x})$ is performed according to Eq. (2.8), the kernel $K_1(\mathbf{x})$ is transformed into the kernel $T_1(\mathbf{x}, s)$ by means of the convolution with the characteristic function $h(\mathbf{x})$:

$$T_1(\mathbf{x}, s) = (K_f s - \kappa_m)\,(h * K_1)\,(\mathbf{x}) = (K_f s - \kappa_m) \int h(\mathbf{x}-\mathbf{y}) K_1(\mathbf{y})\, d^3\mathbf{y}. \qquad (3.2)$$

Consequently, the upper bound on κ^*, obtained from the restriction of the functional W_A over the class $\mathcal{T}_A^{(1)}$ coincides with the optimal third-order bound $\kappa^{(3)}$, see Sec. 1. Moreover, due to Eqs. (2.8) and (2.9), we can claim that the bound $\kappa^{(3)}$ is the best one for the dispersion which employs the statistical information provided by the two- and three-point probability densities F_2 and F_3.

Making use of Eq. (2.8) and the formulae (2.15) for the moments of the fields $D_\psi^{(1)}$, the restriction $W_A^{(1)}[T_1(\cdot)]$ of the functional W_A over the class (3.1) becomes

$$W_A^{(1)}[T_1(\cdot)] = W_A\,\Big|_{\mathcal{T}_A^{(1)}} = \langle\kappa\rangle\, G^2 + n\,\langle\kappa\rangle \left\{ \int\!\!\int |\nabla T_1(\mathbf{z}, s)|^2 P(s)\, d^3\mathbf{z}\, ds \right.$$

$$\left. - n \int\!\!\int\!\!\int\!\!\int \mathcal{R}_0(\mathbf{z}_1 - \mathbf{z}_2; s_1, s_2) \nabla T_1(\mathbf{z}_1, s_1) \cdot \nabla T_1(\mathbf{z}_2, s_2)\, d^3\mathbf{z}_1\, d^3\mathbf{z}_2\, ds_1\, ds_2 \right\}$$

$$+ 2n\,\mathbf{G} \cdot \left\{ \int\!\!\int (K_f s - \kappa_m) h(\mathbf{z}) \nabla T_1(\mathbf{z}, s) P(s)\, d^3\mathbf{z}\, ds \right.$$

$$\left. - n \int\!\!\int\!\!\int (K_f s_1 - \kappa_m) \mathcal{F}_0(\mathbf{z}; s_1, s_2) \nabla T_1(\mathbf{z}, s_2)\, d^3\mathbf{z}\, ds_1\, ds_2 \right\}$$

$$+n\left\{\int(K_f s-\kappa_m)h(\mathbf{z})|\nabla T_1(\mathbf{z},s)|^2P(s)\,d^3\mathbf{z}\,ds\right.$$

$$-n\left[2\iiint\int(K_f s_1-\kappa_m)h(\mathbf{z}_1)\mathcal{R}_0(\mathbf{z}_{2,1},s_1,s_2)\nabla T_1(\mathbf{z}_1,s_1)\right.$$

$$\cdot\nabla T_1(\mathbf{z}_2,s_2)\,d^3\mathbf{z}_1 d^3\mathbf{z}_2\,ds_1 ds_2$$

$$\left.\left.+\iiint\mathcal{F}_0(\mathbf{z};s_1,s_2)|\nabla T_1(\mathbf{z},s_2)|^2\,d^3\mathbf{z}\,ds_1\,ds_2\right]\right\}+o(n^2), \tag{3.3}$$

where

$$\mathcal{F}_0(\mathbf{z};s_1,s_2)=\int h(\mathbf{y})\mathcal{R}_0(\mathbf{z}-\mathbf{y};s_1,s_2)\,d\mathbf{y}. \tag{3.4}$$

Hereafter the differentiation is with respect to the appropriate spatial variable.

The optimal kernel $T_1(\mathbf{x},s)$, i.e. the solution of the Euler-Lagrange equation for the functional $W_A^{(1)}$, is looked for in the virial form

$$T_1(\mathbf{x},s)=T_1(\mathbf{x},s;n)=T_{1,0}(\mathbf{x},s)+T_{1,1}(\mathbf{x},s)\,n+\cdots \tag{3.5}$$

This representation of $T_1(\mathbf{x},s)$ induces the appropriate virial expansion of the functional (3.3):

$$W_A^{(1)}[T_1(\cdot)]=\langle\kappa\rangle G^2+W_A^{(1,1)}[T_{1,0}(\cdot)]n+W_A^{(1,2)}[T_{1,0}(\cdot),T_{1,1}(\cdot)]n^2+\cdots \tag{3.6}$$

The functionals $W_A^{(1,1)}$ and $W_A^{(1,2)}$ depend on the indicated virial coefficients as follows:

$$W_A^{(1,1)}[T_{1,0}(\cdot)]=\kappa_m\iint|\nabla T_{1,0}(\mathbf{x},s)|^2P(s)\,d^3\mathbf{x}\,ds$$

$$+\iint(K_f s-\kappa_m)h(\mathbf{x})[\,\nabla T_{1,0}(\mathbf{x},s)+2\mathbf{G}\,]\cdot\nabla T_{1,0}(\mathbf{x},s)P(s)\,d^3\mathbf{x}\,ds, \tag{3.7}$$

$$W_A^{(1,2)}[T_{1,0}(\cdot),T_{1,1}(\cdot)]=\overline{W}_A^{(1,2)}[T_{1,0}(\cdot)]+2\int P(s)\,ds$$

$$\times\int\nabla\cdot\{\kappa_m\nabla T_{1,0}(\mathbf{x},s)+(K_f s-\kappa_m)h(\mathbf{x})[\,\mathbf{G}+\nabla T_{1,0}(\mathbf{x},s)\,]\}\,T_{1,1}(\mathbf{x},s)\,d^3\mathbf{x}, \tag{3.8a}$$

$$\overline{W}_A^{(1,2)}[T_{1,0}(\cdot)]=(K_f-\kappa_m)V_a\iint|\nabla T_{1,0}(\mathbf{x},s)|^2P(s)\,d^3\mathbf{x}\,ds$$

$$-\iiint\int(K_f s_1-\kappa_m)h(\mathbf{x}_1)|\nabla T_{1,0}(\mathbf{x}_2,s_2)|^2\mathcal{R}_0(\mathbf{x}_1-\mathbf{x}_2;s_1,s_2)\,d^3\mathbf{x}_1\,d^3\mathbf{x}_2\,ds_1\,ds_2$$

$$+\kappa_m\iiint\int\nabla T_{1,0}(\mathbf{x}_1,s_1)\cdot\nabla T_{1,0}(\mathbf{x}_2,s_2)\mathcal{R}_0(\mathbf{x}_1-\mathbf{x}_2;s_1,s_2)\,d^3\mathbf{x}_1\,d^3\mathbf{x}_2\,ds_1\,ds_2$$

$$-2\iint\{\kappa_m\nabla T_{1,0}(\mathbf{x}_1,s_1)+(K_f s_1-\kappa_m)h(\mathbf{x}_1)[\,\mathbf{G}+\nabla T_{1,0}(\mathbf{x}_1,s_1)\,]\}\,d^3\mathbf{x}_1\,ds_1$$

$$\cdot \iint \nabla T_{1,0}(\mathbf{x}_2, s_2) \mathcal{R}_0(\mathbf{x}_1 - \mathbf{x}_2; s_1, s_2)\, d^3\mathbf{x}_2\, ds_2. \tag{3.8b}$$

The optimal kernel $T_1(\mathbf{x}, s)$ satisfies the equation $\delta W_A^{(1)} = 0$, so that we have, in particular,

$$\delta W_A^{(1,1)}[T_{1,0}(\cdot)] = 0, \quad \delta W_A^{(1,2)}[T_{1,0}(\cdot), T_{1,1}(\cdot)] = 0. \tag{3.9}$$

The first of these equations yields straightforwardly

$$P(s)\nabla \cdot \{\kappa_m \nabla T_{1,0}(\mathbf{x}, s) + (K_f s - \kappa_m) h(\mathbf{x})[\mathbf{G} + \nabla T_{1,0}(\mathbf{x}, s)]\} = 0, \tag{3.10}$$

which is just the equation for the disturbance, $T^{(1)}(\mathbf{x}, s)$, to the temperature field $\mathbf{G} \cdot \mathbf{x}$ in an unbounded matrix, introduced by a single spherical inhomogeneity of conductivity $K_f s$, located at the origin. The analytic form of this disturbance is well-known:

$$T_{1,0}(\mathbf{x}, s) = T^{(1)}(\mathbf{x}, s) = 3\beta(s)\, \mathbf{G} \cdot \nabla \varphi(\mathbf{x}), \quad \beta(s) = \frac{K_f s - \kappa_m}{K_f s + 2\kappa_m}; \tag{3.11}$$

here

$$\varphi(\mathbf{x}) = h * \frac{1}{4\pi|\mathbf{x}|}, \quad \text{i.e.} \quad \varphi(\mathbf{x}) = \int \frac{h(\mathbf{y})}{4\pi|\mathbf{x} - \mathbf{y}|}\, d^3\mathbf{y}$$

is the Newtonian potential for a single sphere of radius a, located at the origin. (We assume, obviously enough, that $P(s) \neq 0$.)

With $T_{1,0}(\mathbf{x}, s)$ already found, one should vary only $T_{1,1}(\mathbf{x}, s)$ in the functional (3.8) in order to derive the Euler-Lagrange equation for the latter. However, this is not possible, because Eq. (3.10) yields

$$W_A^{(1,2)}[T_{1,0}(\cdot), T_{1,1}(\cdot)] = \overline{W}_A^{(1,2)}[T_{1,0}(\cdot)]. \tag{3.12}$$

Hence, according to Eq. (3.6) for the optimal upper three-point bound $\kappa^{(3)}$ we have

$$\kappa^* G^2 \leq \kappa^{(3)} G^2 = \langle \kappa \rangle G^2 + \frac{1}{V_a} W_A^{(1,1)}[T_{1,0}(\cdot)]c + \frac{1}{V_a} \overline{W}_A^{(1,2)}[T_{1,0}(\cdot)]c^2 + o(c^2). \tag{3.13}$$

The foregoing reasoning has two implications. First, we can conclude that the optimal upper bound $\kappa^{(3)}$ to the order c^2 depends only on the field $T^{(1)}(\mathbf{x}, s)$; the explicit form of $T_{1,1}(\mathbf{x}, s)$ is not required at all, see Eq. (3.13). Second, the kernel $T_1(\mathbf{x}, s)$ is optimal to the order c^2 if its leading coefficient $T_{1,0}(\mathbf{x}, s)$ in the virial expansion (3.5) is proportional to the single-sphere disturbance field $T^{(1)}(\mathbf{x}, s)$. In this connection it is to be noted that the known Ritz type procedure of Torquato [16] leads to the optimal bound to the order c^2. (For the latter the kernel $T_1(\mathbf{x}, s)$ in (3.1) should be chosen as $T_1(\mathbf{x}, s) = \lambda T^{(1)}(\mathbf{x}, s)$, where λ is an adjustable parameter.) This fact holds also for dispersions of radial inhomogeneous spheres with random radii, see [17]. To the order c^p at $p > 2$, however, the cluster bounds of Torquato are not optimal even for the monodisperse case, see [9, 12].

Repeating the above arguments with respect to the dual principle (1.7) leads to a fully similar conclusion for the optimal lower bound, namely, that to the order c^2 the latter is fully determined by the disturbance $\mathbf{q}^{(1)}(\mathbf{x}, s)$ to the heat flux $\mathbf{Q}$ in an unbounded matrix, introduced by a single spherical inhomogeneity of conductivity $K_f s$, located at the origin.

Let

$$\frac{\kappa^*}{\kappa_m} = 1 + a_{1\kappa}c + a_{2\kappa}c^2 + \cdots \tag{3.14}$$

be the virial expansion for the effective conductivity of the dispersion. Making use of Eqs. (3.10) and (3.11), the connection of the disturbances $T^{(1)}(\mathbf{x}, s)$ and $\mathbf{q}^{(1)}(\mathbf{x}, s)$, and the relation $h(\mathbf{x})\nabla T^{(1)}(\mathbf{x}, s) = -\beta(s)h(\mathbf{x})\mathbf{G}$, we easily get as a consequence of (3.7), (3.13) and their counterparts for the dual variational principle (1.7), that

$$a_{1\kappa} = 3N, \quad N = N[P(\cdot)] = \langle \beta(\tilde{s}) \rangle = \int_0^\infty \beta(s)P(s)\,ds, \tag{3.15}$$

so that the upper and lower bounds coincide to the order c, as it should have been expected. After simple algebra, based on Eqs. (3.8), (3.10), (3.11), (3.13) and their counterparts for the lower bound, we get the following inequalities for the c^2-coefficient $a_{2\kappa}$:

$$a^l_{2\kappa} \leq a_{2\kappa} \leq a^u_{2\kappa}, \tag{3.16a}$$

$$a^l_{2\kappa} = 3\left\{ N^2 + \int_0^\infty \frac{K_f s_1 - \kappa_m}{K_f s_1} ds_1 \int_0^\infty \beta^2(s_2)\mathcal{M}_2(s_1, s_2) ds_2 \right\}, \tag{3.16b}$$

$$a^u_{2\kappa} = 3\left\{ N^2 + \int_0^\infty \frac{K_f s_1 - \kappa_m}{\kappa_m} ds_1 \int_0^\infty \beta^2(s_2)\mathcal{M}_2(s_1, s_2) ds_2 \right\}, \tag{3.16c}$$

where

$$\mathcal{M}_2(s_1, s_2) = -\frac{1}{2\pi} \int_0^\infty \frac{1}{r^3} \frac{\partial}{\partial r} \mathcal{F}_0(r; s_1, s_2)\, dr \tag{3.17}$$

is a statistical parameter for the dispersion; the function $\mathcal{F}_0(r; s_1, s_2)$ is defined in Eq. (3.4), $r = |x|$.

The formula (3.15) clearly indicates that the effective conductivity κ^* depends on the statistical distribution of conductivity of spheres even to the order c. (Let us recall that the c-coefficient $a_{1\kappa}$ is independent of the size distribution for a dispersion of spheres of random radii, see [1, 17, 18].) Moreover, it is to be noted that in general $\langle \beta(\tilde{s}) \rangle \neq \beta(\langle \tilde{s} \rangle) = \beta(1) = (K_f - \kappa_m)/(K_f + 2\kappa_m)$, so that the dispersion is *not equivalent* to a monodisperse dispersion of sphere (with the same sphere fraction c) of the mean conductivity K_f even to the order c, see Eq. (3.15).

According to (3.2), the Beran's kernel $K_B(\mathbf{x})$, see (1.8), is transformed into the kernel

$$T_B(\mathbf{x}, s) = (K_f s - \kappa_m)\,\mathbf{G} \cdot \nabla\varphi(\mathbf{x}) \tag{3.18}$$

at the transition from $\kappa'(\mathbf{x})$ to $\psi'(\mathbf{x}; s)$. Due to Eq. (3.11), the kernel $K_1(\mathbf{x}) = \lambda K_B(\mathbf{x})$, $\lambda \in \mathbb{R}$, will be optimal to the order c and consequently to the order c^2 also (see (3.12)), if and only if $T^{(1)}(\mathbf{x}, s) = \lambda T_B(\mathbf{x}, s)$ for a certain $\lambda \in \mathbb{R}$ and *for all* s such that $P(s) \neq 0$. It is shown, however, that it is possible only if $P(s) = \delta(s - s_0)$, i.e., if the probability to find a sphere of conductivity different of $K_f s_0$ is equal to zero; in other words, for the usually considered dispersions of spheres possessing one and the same conductivity. Hence, we can conclude that the Beran's bounds are not optimal even to the order c for the considered dispersions.

The above arguments imply the following simple way for a generalization of the Beran's procedure. Namely, if we choose the kernel $K_1(\mathbf{x})$ in the form $K_1(\mathbf{x}) = \lambda(s) K_B(\mathbf{x})$, see (1.8), where now $\lambda(s)$ is an adjustable function, then the optimal bounds to the order c^2 will be obtained.

Let us note that the Beran's bounds are more complicated for the dispersion under study. For example, the minimization of the functional (3.7) at $T_{1,0}(\mathbf{x}, s) = \lambda T_B(\mathbf{x}, s)$ with respect to $\lambda \in \mathbb{R}$ leads to the following upper bound $a^u_{1\kappa(B)}$ on the c-coefficient $a_{1\kappa}$:

$$a_{1\kappa} \leq a^u_{1\kappa(B)}, \quad a^u_{1\kappa(B)} = \frac{K_f}{\kappa_m} - 1 - \frac{\left\langle (K_f \tilde{s} - \kappa_m)^2 \right\rangle^2}{\kappa_m \left\langle (K_f \tilde{s} - \kappa_m)^2 (K_f \tilde{s} + 2\kappa_m) \right\rangle}; \tag{3.19}$$

the equality sign, $a_{1\kappa} = a^u_{1\kappa(B)}$, being achieved at $P(s) = \delta(s - s_0)$ only, i.e. if the spheres have one and the same conductivity.

3.3. EXAMPLES

We shall illustrate the influence of the statistical distribution of conductivities of spheres on the obtained c^2-bounds (3.15), (3.16). First, we note that if we adopt the assumption of statistical independence of the conductivities of each two spheres, i.e.

$$P_2(s_1, s_2) = P(s_1) P(s_2), \tag{3.20}$$

which sounds reasonable enough (at least in the dilute case under study), then the form of the bounds (3.16) becomes more or less similar to that in the monodisperse case. Namely, in the frame of this assumption

$$\mathcal{R}_0(\mathbf{x}_{12}; s_1, s_2) = P(s_1) P(s_2) R(\mathbf{x}_{12}), \quad \mathcal{F}_0(\mathbf{y}; s_1, s_2) = P(s_1) P(s_2) F_0(\mathbf{y}),$$

where

$$R(\mathbf{x}_{12}) = 1 - g_0(\mathbf{x}_{12}), \quad F_0(\mathbf{y}) = \int h(\mathbf{x}) R(\mathbf{x} - \mathbf{y})\, d^3\mathbf{x}$$

are the same functions that appeared in [8] when dealing with the monodisperse case. Then the formulae (3.16) simplify:

$$a^l_{2\kappa} = 3\left\{N^2 + \langle\beta^2(\widetilde{s})\rangle\left\langle\frac{K_f\widetilde{s}-\kappa_m}{K_f\widetilde{s}}\right\rangle m_2\right\},$$

$$a^u_{2\kappa} = 3\left\{N^2 + \langle\beta^2(\widetilde{s})\rangle\frac{K_f-\kappa_m}{\kappa_m}m_2\right\}, \tag{3.21}$$

where

$$m_2 = -\frac{1}{2\pi}\int_0^\infty \frac{F_0'(r)}{r^3}\,dr = 2\int_2^\infty \frac{\lambda^2}{(\lambda^2-1)^3}g_0(\lambda a)\,d\lambda \tag{3.22}$$

is the same statistical parameter as in the monodisperse case, see [8]. In particular, if the spheres have non-random conductivity $\kappa_f = K_f$, then

$$a^l_{2\kappa} = 3\beta^2\left\{1+\frac{[\kappa]}{\kappa_f}m_2\right\},\quad a^u_{2\kappa} = 3\beta^2\left\{1+\frac{[\kappa]}{\kappa_m}m_2\right\}, \tag{3.23}$$

where $[\kappa] = \kappa_f - \kappa_m$, $\beta = \beta(1) = [\kappa]/(\kappa_f + 2\kappa_m)$, see [8] again, which coincides with the monodisperse result of Markov [8]. Under the assumption (3.20) we shall consider the following two examples.

3.3.1. *"Triangular" distribution.* Since the conductivity $\widetilde{\kappa}_f = K_f\widetilde{s} \geq 0$, it is impossible to adopt the popular Gaussian distribution. That is way we consider the "triangular" (Simpson) distribution of $\widetilde{\kappa}_f$ in the interval $[K_1, K_2]$ as a certain counterpart of the Gaussian one. Then

$$P(s) = \begin{cases} \dfrac{2K_f}{K_2-K_1}\left[1-\dfrac{|K_1+K_2-2K_fs|}{K_2-K_1}\right] & \text{at} \quad K_fs\in[K_1,K_2], \\ 0 \qquad \text{otherwise,} \end{cases} \tag{3.24}$$

where $K_f = (K_1+K_2)/2$. After simple algebra, based of Eqs. (3.15) and (3.24), we get

$$a_{1\kappa} = 3N,\quad N = 1+\frac{6}{\gamma^2\omega^2}\Big[4(\gamma+2)\ln(2\gamma+4)$$

$$-(\gamma(2-\omega)+4)\ln(\gamma(2-\omega)+4)-(\gamma(2+\omega)+4)\ln(\gamma(2+\omega)+4)\Big], \tag{3.25}$$

where $\gamma = K_f/k_m$ and $\omega = (K_2-K_1)/K_f$ is, so to say, the "divergence" of the non-dimensional sphere conductivity. Since $K_2 \geq K_1 \geq 0$, then $\gamma \geq 0$ and $0 \leq \omega \leq 2$.

Similarly, with Eq. (3.20) taken into account, the bounds (3.16) read

$$a^l_{2\kappa} = 3\left\{N^2+\Upsilon(\gamma,\omega)\Lambda(\gamma,\omega)m_2\right\},\quad a^u_{2\kappa} = 3\left\{N^2+(\gamma-1)\Lambda(\gamma,\omega)m_2\right\}, \tag{3.26a}$$

where

$$\Upsilon(\gamma,\omega) = 1-\frac{2}{\gamma\omega^2}\Big[(2-\omega)\ln(2-\omega)+(2+\omega)\ln(2+\omega)-4\ln 2\Big], \tag{3.26b}$$

$$\Lambda(\gamma,\omega) = 1 + \frac{12}{\gamma^2\omega^2}\Big[2(2\gamma+7)\ln(2\gamma+4)$$
$$-(\gamma(2-\omega)+7)\ln(\gamma(2-\omega)+4) - (\gamma(2+\omega)+7)\ln(\gamma(2+\omega)+4)\Big]. \quad (3.26c)$$

The quantities $a_{1\kappa}$, $a^u_{1\kappa(B)}$, $a^l_{2\kappa}$ and $a^u_{2\kappa}$ as functions of the parameter ω are shown in Fig. 1 and 2 for $\gamma = 5$. The "well-stirred" case $g_0(r) = 1$ at $r > 2a$ is considered, when $m_2 = \frac{5}{18} - \frac{1}{8}\ln 2 \approx 0.14045$, see [8]. In Fig. 1 the value of approximation $\widetilde{a}_{1\kappa} = 3(\gamma-1)/(\gamma+2)$ for $a_{1\kappa}$ is also given, which corresponds to the rough assumption that the dispersion is replaced with a monodisperse one of

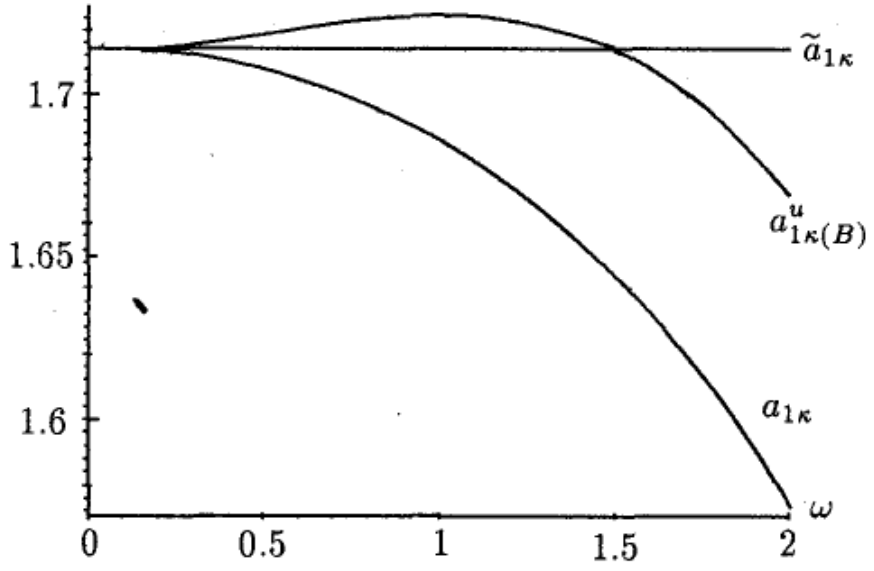

Fig. 1. The variations of the c-coefficient $a_{1\kappa}$ of the effective conductivity of the dispersion with "divergence" ω in the "triangular" case ($\gamma = K_f/\kappa_m = 5$); $a_{1\kappa}$ — the exact value (3.15); $a^u_{1\kappa(B)}$ — the Beran's upper bound, see (3.27); $\widetilde{a}_{1\kappa}$ — the "monodisperse" approximation

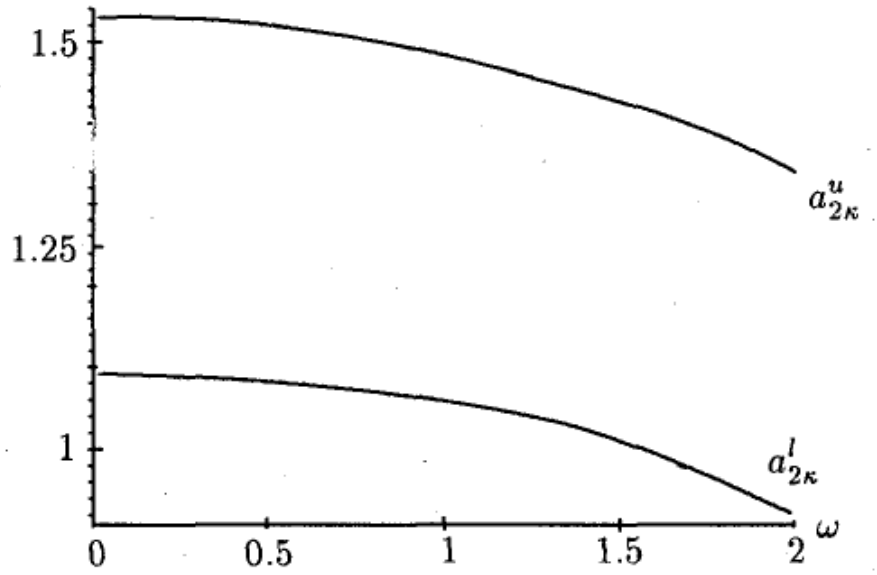

Fig. 2. The variations of the c^2-bounds $a^l_{2\kappa}$ and $a^u_{2\kappa}$ of the effective conductivity of the dispersion with "divergence" ω in the "triangular" case ($\gamma = K_f/\kappa_m = 5$)

sphere's conductivity that equals the mean value K_f (the "monodisperse" approximation). It is seen that this approximation is non-realistic; it is only justified at the limit case $\omega \to 0$. The dependence on ω of the upper Beran's bound $a^u_{1\kappa(B)}$, which now has the form

$$a^u_{1\kappa(B)} = \gamma - 1 - \frac{1}{72}\frac{(\gamma^2\omega^2 + 24(\gamma-1)^2)^2}{\gamma^3\omega^2 + 8(\gamma-1)^2(\gamma+2)}; \tag{3.27}$$

is plotted as well in Fig. 1.

3.3.2. *A Dispersion Containing Two Kinds of Spheres.* Consider the case when there exist only two kinds of spheres in the dispersion, having the conductivities $\kappa_f^{(1)}$, $\kappa_f^{(2)}$ and volume fractions c_1, c_2, respectively, $c = c_1 + c_2$. Then

$$P(s) = p_1\delta(s - s^{(1)}) + p_2\delta(s - s^{(2)}),$$

where $s^{(i)} = \kappa_f^{(i)}/K_f$, $p_i = c_i/c$, $i = 1, 2$, $K_f = p_1\kappa_f^{(1)} + p_2\kappa_2^{(2)}$. In this case the c-coefficient $a_{1\kappa}$ becomes

$$a_{1\kappa} = 3(p_1\beta_1 + p_2\beta_2),$$

where

$$\beta_i = \beta(s^{(i)}) = \frac{\kappa_f^{(i)} - \kappa_m}{\kappa_f^{(i)} + 2\kappa_m} = \frac{\alpha_i - 1}{\alpha_i + 2}, \quad \alpha_i = \frac{\kappa_f^{(i)}}{\kappa_m}, \quad i = 1, 2.$$

Similarly, the bounds (3.16) on the c^2-coefficient $a_{2\kappa}$ read

$$a^l_{2\kappa} = 3\sum_{i=1}^{2}\left(p_i\beta_i + \left(1 - \frac{p_1}{\alpha_1} - \frac{p_2}{\alpha_2}\right)\beta_i^2 m_2\right),$$

$$a^u_{2\kappa} = 3\sum_{i=1}^{2}\left(p_i\beta_i + (p_1\alpha_1 + p_2\alpha_2 - 1)\beta_i^2 m_2\right).$$

Let us note that the dispersion under study represents a three-phase medium: in the matrix two types of spherical particles of different conductivities are distributed. The generation of the above formulae for n-phase media of this kind is straightforward.

4. VARIATIONAL DERIVATION OF c^2-FORMULA FOR THE EFFECTIVE CONDUCTIVITY OF THE DISPERSION

Consider now the series (2.14) as a class of trial fields:

$$T_A^{(2)} = \left\{\theta(\mathbf{x}) \mid \theta(\mathbf{x}) = \mathbf{G}\cdot\mathbf{x} + \int\!\!\int T_1(\mathbf{x}-\mathbf{y}, s)D_\psi^{(1)}(\mathbf{y}; s)\, d^3\mathbf{y}\, ds\right.$$

$$\left. + \int\!\!\int\!\!\int\!\!\int T_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2, s_1, s_2)D_\psi^{(2)}(\mathbf{y}_1, \mathbf{y}_2; s_1, s_2)\, d^3\mathbf{y}_1\, d^3\mathbf{y}_2\, ds_1\, ds_2\right\}, \tag{4.1}$$

where now the kernels $T_1(\mathbf{x}, s)$ and $T_2(\mathbf{x}, \mathbf{y}, s_1, s_2)$ are adjustable. Using the formulae for the moments of the fields $D_\psi^{(1)}$ and $D_\psi^{(2)}$, see Eqs. (2.15), the restriction $W_A^{(2)}[T_1(\cdot), T_2(\cdot,\cdot)]$ of the functional W_A over this class becomes

$$W_A^{(2)}[T_1(\cdot), T_2(\cdot,\cdot)] = W_A\big|_{T_A^{(2)}} = W_A^{(1)}[T_1(\cdot)] + \widetilde{W}_A^{(2)}[T_1(\cdot), T_2(\cdot,\cdot)],$$

where

$$\widetilde{W}_A^{(2)}[T_1(\cdot), T_2(\cdot,\cdot)] = n^2\kappa_m \int\int\int\int g_0(\mathbf{y}_{2,1}) P_2(s_1, s_2) \Big[|\nabla_x T_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2, s_1, s_2)|^2$$

$$+ \nabla_x T_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2, s_1, s_2) \cdot \nabla_x T_2(\mathbf{x}-\mathbf{y}_2, \mathbf{x}-\mathbf{y}_1, s_2, s_1) \Big]\, d^3\mathbf{y}_1\, d^3\mathbf{y}_2\, ds_1\, ds_2$$

$$+ 2n^2 \int\int\int\int g_0(\mathbf{y}_{2,1}) P_2(s_1, s_2) \Big[(K_f s_1 - \kappa_m) h(\mathbf{x}-\mathbf{y}_1) \nabla T_1(\mathbf{x}-\mathbf{y}_2, s_2)$$

$$+ (K_f s_2 - \kappa_m) h(\mathbf{x}-\mathbf{y}_2) \nabla T_1(\mathbf{x}-\mathbf{y}_1, s_1) \Big] \cdot \nabla_x T_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2, s_1, s_2)\, d^3\mathbf{y}_1\, d^3\mathbf{y}_2\, ds_1\, ds_2$$

$$+ n^2 \int\int\int\int g_0(\mathbf{y}_{2,1}) P_2(s_1, s_2) \big[(K_f s_1 - \kappa_m) h(\mathbf{x}-\mathbf{y}_1) + (K_f s_2 - \kappa_m) h(\mathbf{x}-\mathbf{y}_2) \big]$$

$$\times \big[|\nabla_x T_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2, s_1, s_2)|^2 + \nabla_x T_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2, s_1, s_2)$$

$$\cdot \nabla_x T_2(\mathbf{x}-\mathbf{y}_2, \mathbf{x}-\mathbf{y}_1, s_2, s_1) \big]\, d^3\mathbf{y}_1\, d^3\mathbf{y}_2\, ds_1\, ds_2 + o(n^2).$$

The optimal kernels $T_1(\mathbf{x}, s)$ and $T_2(\mathbf{x}, \mathbf{y}, s_1, s_2)$ are looked for again in the virial form (3.5) for T_1 and

$$T_2(\mathbf{x}, \mathbf{y}, s_1, s_2) = T_2(\mathbf{x}, \mathbf{y}, s_1, s_2; n)$$

$$= T_{2,0}(\mathbf{x}, \mathbf{y}, s_1, s_2) + T_{2,1}(\mathbf{x}, \mathbf{y}, s_1, s_2)\, n + \cdots$$

for T_2, which implies the respective virial expansion of the functional $W_A^{(2)}$, namely,

$$W_A^{(2)}[T_1(\cdot), T_2(\cdot,\cdot)] = \langle\kappa\rangle G^2 + W_A^{(1,1)}[T_{1,0}(\cdot)]\, n$$

$$+ W_A^{(2,2)}[T_{1,0}(\cdot), T_{1,1}(\cdot), T_{2,0}(\cdot,\cdot)]\, n^2 + o(n^2), \tag{4.2}$$

where

$$W_A^{(2,2)}[T_{1,0}(\cdot), T_{1,1}(\cdot), T_{2,0}(\cdot,\cdot)]$$

$$= W_A^{(1,2)}[T_{1,0}(\cdot), T_{1,1}(\cdot)] + \widetilde{W}_A^{(2)}[T_{1,0}(\cdot), T_{2,0}(\cdot,\cdot)]; \tag{4.3}$$

here $W_A^{(1,1)}$ and $W_A^{(1,2)}$ are the virial coefficients from Eq. (3.6) for which, let us recall, Eqs. (3.11) and (3.12) hold. Hence, the minimization of the functional $W_A^{(2)}$ is reduced to that of the functional

$$\widetilde{W}_A^{(2)\dagger}[T_{2,0}(\cdot,\cdot)] = \widetilde{W}_A^{(2)}[T^{(1)}(\cdot), T_{2,0}(\cdot,\cdot)].$$

The Euler-Lagrange equation for the latter is

$$P_2(s_1,s_2)\Big\{\kappa_m(\nabla_{\mathbf{z}_1}+\nabla_{\mathbf{z}_2})\cdot\Big[g_0(\mathbf{z}_1-\mathbf{z}_2)(\nabla_{\mathbf{z}_1}+\nabla_{\mathbf{z}_2})\tilde{T}_{2,0}(\mathbf{z}_1,\mathbf{z}_2,s_1,s_2)\Big]$$

$$+(\nabla_{\mathbf{z}_1}+\nabla_{\mathbf{z}_2})\cdot\Big[g_0(\mathbf{z}_1-\mathbf{z}_2)\big[(K_f s_1-\kappa_m)h(\mathbf{z}_1)\nabla T^{(1)}(\mathbf{z}_2,s_2)$$

$$+(K_f s_2-\kappa_m)h(\mathbf{z}_2)\nabla T^{(1)}(\mathbf{z}_1,s_1)\big]\Big]$$

$$+(\nabla_{\mathbf{z}_1}+\nabla_{\mathbf{z}_2})\cdot\Big[g_0(\mathbf{z}_1-\mathbf{z}_2)\big[(K_f s_1-\kappa_m)h(\mathbf{z}_1)+(K_f s_2-\kappa_m)h(\mathbf{z}_2)\big]$$

$$+(\nabla_{\mathbf{z}_1}+\nabla_{\mathbf{z}_2})\tilde{T}_{2,0}(\mathbf{z}_1,\mathbf{z}_2,s_1,s_2)\Big]\Big\}=0 \tag{4.4}$$

with the notation

$$\tilde{T}_{2,0}(\mathbf{z}_1,\mathbf{z}_2,s_1,s_2)=T_{2,0}(\mathbf{z}_1,\mathbf{z}_2,s_1,s_2)+T_{2,0}(\mathbf{z}_2,\mathbf{z}_1,s_2,s_1).$$

Taking into account that $(\nabla_{\mathbf{z}_1}+\nabla_{\mathbf{z}_2})g_0(\mathbf{z}_1-\mathbf{z}_2)=0$, an appropriate change of variables allows to recast Eq. (4.4) as

$$g_0(\mathbf{z})P_2(s_1,s_2)\Big\{\kappa_m\Delta_x\tilde{T}_{2,0}(\mathbf{x},\mathbf{x}-\mathbf{z},s_1,s_2)$$

$$+\nabla_x\cdot\big[(K_f s_1-\kappa_m)h(\mathbf{x})\nabla T^{(1)}(\mathbf{x}-\mathbf{z},s_2)+(K_f s_2-\kappa_m)h(\mathbf{x}-\mathbf{z})\nabla T^{(1)}(\mathbf{x},s_1)\big] \tag{4.5}$$

$$+\nabla_x\cdot\Big[\big[(K_f s_1-\kappa_m)h(\mathbf{x})+(K_f s_2-\kappa_m)h(\mathbf{x}-\mathbf{z})\big]\nabla_x\tilde{T}_{2,0}(\mathbf{x},\mathbf{x}-\mathbf{z},s_1,s_2)\Big]\Big\}=0.$$

Similarly to the monodisperse case [5], the solution of Eq. (4.5) is

$$\tilde{T}_{2,0}(\mathbf{x},\mathbf{x}-\mathbf{z},s_1,s_2)=T^{(2)}(\mathbf{x},s_1;\mathbf{z},s_2)-T^{(1)}(\mathbf{x},s_1)-T^{(1)}(\mathbf{x}-\mathbf{z},s_2)\,, \tag{4.6}$$

where $T^{(2)}(\mathbf{x},s_1;\mathbf{y},s_2)$ is the disturbance to the temperature field $\mathbf{G}\cdot\mathbf{x}$ in an unbounded matrix of conductivity κ_m, generated by two spherical inhomogeneities: one of conductivity $K_f s_1$ located at the origin, and the other of conductivity $K_f s_2$ located at the point $\mathbf{y}$.

Making use of Eq. (4.4), the minimum value of the functional $\widetilde{W}_A^{(2)\dagger}$ can be recast now in the form in which the field $T_{2,0}(\mathbf{x},\mathbf{y},s_1,s_2)$ enters linearly:

$$\min\widetilde{W}_A^{(2)\dagger}\,[T_{2,0}(\cdot,\cdot)]=n^2\iiiint g_0(\mathbf{z}_1-\mathbf{z}_2)P_2(s_1,s_2)$$

$$\times\big[(K_f s_1-\kappa_m)h(\mathbf{z}_1)\nabla T^{(1)}(\mathbf{z}_2,s_2)+(K_f s_2-\kappa_m)h(\mathbf{z}_2)\nabla T^{(1)}(\mathbf{z}_1,s_1)\big]$$

$$\cdot(\nabla_{\mathbf{z}_1}+\nabla_{\mathbf{z}_2})T_{2,0}(\mathbf{z}_1,\mathbf{z}_2,s_1,s_2)\,d^3\mathbf{z}_1\,d^3\mathbf{z}_2\,ds_1\,ds_2$$

$$=n^2\iiiint P_2(s_1,s_2)(K_f s_1-\kappa_m)g_0(\mathbf{y})h(\mathbf{x})\nabla T^{(1)}(\mathbf{x}-\mathbf{y},s_2)$$

$$\cdot\big[\nabla_x T^{(2)}(\mathbf{x},s_1;\mathbf{y},s_2)-\nabla T^{(1)}(\mathbf{x},s_1)-\nabla T^{(1)}(\mathbf{x}-\mathbf{y},s_2)\big]\,d^3\mathbf{x}\,d^3\mathbf{y}\,ds_1\,ds_2. \tag{4.7}$$

Taking into account Eqs. (4.2), (4.3), (3.11), (3.12) and the formulae

$$\int h(\mathbf{x})\, d^3\mathbf{x} \int g_0(\mathbf{y}) \nabla T^{(1)}(\mathbf{x}-\mathbf{y}, s_2) \cdot \nabla T^{(1)}(\mathbf{x}, s_1)\, d^3\mathbf{y} = 0,$$

$$P_2(s_1, s_2) \int h(\mathbf{x})\, d^3\mathbf{x} \int g_0(\mathbf{y}) |\nabla T^{(1)}(\mathbf{x}-\mathbf{y}, s_2)|^2\, d^3\mathbf{y} = 3\beta^2(s_2) \mathcal{M}_2(s_1, s_2) V_a^2,$$

one finds for the c^2-coefficient

$$a_{2\kappa} = 3N^2 + a'_{2\kappa},$$

where

$$a'_{2\kappa} = \frac{1}{V_a^2} \int\int P_2(s_1, s_2) \frac{K_f s_1 - \kappa_m}{\kappa_m}\, ds_1 ds_2$$

$$\times \int h(\mathbf{x})\, d^3\mathbf{x} \int g_0(\mathbf{y}) \nabla_x T^{(1)}(\mathbf{x}-\mathbf{y}, s_2) \cdot \nabla_x T^{(2)}(\mathbf{x}, s_1; \mathbf{y}, s_2)\, d^3\mathbf{y}. \qquad (4.8)$$

Let us recall that the latter result follows from the fact that the solution of the random problem (1.2), asymptotically valid to the order c^2, is one of the trial fields from the class $\mathcal{T}_A^{(2)}$, see Sec. 2.2, over which the energy functional $W_A[\theta(\cdot)]$ is minimized. The formula (4.8) is the counterpart of the formula (3.9) in [14] in the monodisperse case. Note that the formula (4.8) contains absolutely convergent integrals only, see [10, 11] for details, so that no "renormalization" is needed, similar to that used by Jeffrey [19].

Finally, it is to be noted that the coefficient $T_{1,1}(\mathbf{x}, s)$ in the expansion (3.5) cannot be found within the frame of the above performed variational n^2-analysis. For the full solution of the random problem (1.2) to the order c^2 in the explained above sense it is necessary, however, to know the virial coefficients $T_{1,0}(\mathbf{x}, s)$, $T_{1,1}(\mathbf{x}, s)$ and $T_{2,0}(\mathbf{x}, \mathbf{y}, s_1, s_2)$: for example, when evaluating the two-point correlation function $\langle \theta'(\mathbf{x}) \theta'(\mathbf{y}) \rangle$, the convolution $\int T_{1,0}(\mathbf{x}-\mathbf{y}, s) T_{1,1}(\mathbf{y}, s)\, d^3\mathbf{y}$ appears, see [6] for details. That is why, in order to obtain function $T_{1,1}(\mathbf{x}, s)$ and as a consequence the full statistical solution of problem (1.2) to the order c^2, either the higher degrees of n in the virial expansion of the functional W_A should be considered or the procedure of Christov and Markov [5] should be employed instead. In the latter, however, conditionally convergent integral in the formula for the effective conductivity will show up with a correct mode of integration extracted in the course of the appropriate solution, see again [2, 6, 7] for details.

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