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ON THE FIELD FLUCTUATIONS IN A RANDOM DISPERSION

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In this note the variances of the basic random fields — temperature and heat flux — in a dilute dispersion of spheres with a small volume fraction $c \ll 1$, subjected to a constant macroscopic temperature gradient are studied. The basic result is an estimate on the c^2-term of these variances, which includes the well-known c^2-term of the effective conductivity, extensively studied in the literature.

Keywords: random dispersion of spheres, effective conductivity, variance of random fields

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1. INTRODUCTION

The aim of this note is to report some preliminary results concerning field fluctuation in a dispersion of nonoverlapping spheres. The heat conduction context is chosen, above all, for the sake of simplicity. A similar study of any transport phenomenon through the medium in the linear case can be performed along the same line.

Let us recall first how the problem is stated. Assume we have an unbounded matrix material of conductivity κ_m throughout which filler particles of conductivity κ_f are distributed in a statistically isotropic and homogeneous manner. The random conductivity field $\kappa(\mathbf{x})$ of the medium takes then the values κ_m or κ_f, depending on whether $\mathbf{x}$ lies in the matrix or in a particle, respectively. If $\mathbf{G}$ denotes the prescribed macroscopic temperature gradient imposed upon the medium, the governing equations of the problem, at the absence of body sources, read

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

under the condition

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

which plays the role of a "boundary" one. In equations (1.1a) $\mathbf{q}(\mathbf{x})$ is the flux vector and $\theta(\mathbf{x})$ is the random temperature field. Hereafter $\langle\cdot\rangle$ denotes ensemble averaging.

The random problem (1.1) possesses a solution, in a statistical sense, which is unique under the natural condition $0 < k_1 \le \kappa(\mathbf{x}) \le k_2 < \infty$, see [11]. This means, let us recall [1], that all multipoint moments of the temperature field $\theta(\mathbf{x})$, and the joint moments of $\theta(\mathbf{x})$ and $\kappa(\mathbf{x})$, can be specified by means of the known moments of the conductivity field. In particular, among the joint moments, the simplest one-point moment defines the well-known effective conductivity κ^* of the medium through the relation

$$\mathbf{Q} = \langle \mathbf{q}(\mathbf{x})\rangle = \langle \kappa(\mathbf{x})\nabla\theta(\mathbf{x})\rangle = \kappa^*\mathbf{G} \tag{1.2}$$

(having assumed statistical homogeneity and isotropy). Note that the definition (1.2) of the effective conductivity κ^* reflects the "homogenization" of the problem under study, in the sense that from a macroscopic point of view, when only the macroscopic values of the flux and temperature gradient are of interest, the medium behaves as if it were homogeneous with a certain macroscopic conductivity κ^*. This interpretation explains why κ^* and its counterparts, say, the effective elastic moduli, have been extensively studied in the literature on homogenization. There one can find a number of rigorous or approximate schemes of their evaluation, especially, in the context of mechanics of heterogeneous and composite media, see, e.g. [9, 21, 14] *et al.* However, κ^* is only a tiny part of the full statistical solution of the random problem (1.1). Moreover, its evaluation *cannot* be torn away from the full statistical solution of (1.1), i.e. of specifying *all* needed multipoint moments, as pointed out for the first time by Brown [8]. (The latter fact explains the failure of all schemes that try to determine solely the effective property κ^* without trying to solve the whole stochastic problem (1.1).) Besides, there are plenty of reasons why one should pay much more attention to other statistical characteristics of random fields like $\theta(\mathbf{x})$ in (1.1), that appear in problems in random heterogeneous media. For instance, in the context of waves in random media or turbulence phenomena, one of the most important quantities is the variance of local fields, connected with the square of its fluctuation, see [1].

The (undimensional) variances, which we shall discuss hereafter, are defined as

$$\sigma^2_{\nabla\theta} = \frac{\langle|\nabla\theta'(\mathbf{x})|^2\rangle}{G^2}, \quad \sigma^2_q = \frac{\langle|q'(\mathbf{x})|^2\rangle}{Q^2}; \tag{1.3}$$

the primes denote in what follows the fluctuating parts of the respective random fields, so that, in particular, $\nabla\theta'(\mathbf{x}) = \nabla\theta(\mathbf{x}) - \mathbf{G}$, and hence $\langle\nabla\theta'(\mathbf{x})\rangle = 0$.

It is noted that for any two-point medium the variances $\sigma^2_{\nabla\theta}$ and σ^2_q are simply interconnected. Indeed, since the conductivity field $\kappa(\mathbf{x})$ takes only two values, κ_f and κ_m, we have

$$\kappa^2(\mathbf{x}) = (\kappa_f + \kappa_m)\kappa(\mathbf{x}) - \kappa_f\kappa_m$$

and hence

$$\langle \mathbf{q}^2(\mathbf{x})\rangle = \langle \kappa^2(\mathbf{x})|\nabla\theta(\mathbf{x})|^2\rangle = (\kappa_f + \kappa_m)\kappa^* G^2 - \kappa_f\kappa_m \langle |\nabla\theta(\mathbf{x})|^2\rangle,$$

having used (1.2). A simple algebra yields eventually

$$\sigma^2_q = -\frac{\kappa_f\kappa_m}{\kappa^{*2}}\sigma^2_{\nabla\theta} - \frac{(\kappa^* - \kappa_f)(\kappa^* - \kappa_m)}{\kappa^{*2}}. \tag{1.4}$$

Let us point out immediately that the study of variances in particular, and of the multipoint moments in general, is much more complicated than that of the effective properties due to the fact that, as a matter of fact, no variational principles for the former have been proposed and applied in the literature. (Though, see the book [5, p. 143], where an extremely concise exposition and some ideas along this line are indicated.)

To the best of the authors' knowledge an investigation of the variances, in addition to the effective properties in the scalar conductivity context, was initiated by Beran *et al.* [2, 4, 3]. In particular, Beran [2] obtained bounds on the variances through the effective properties, investigated in great detail in the literature. The Beran's estimates are quite crude and this is inevitable since they are applicable to *any* statistically homogeneous and isotropic medium.

More restrictive bounds can be obtained only if additional information about the medium constitution is available and the needed random fields are specified at least to a certain extent. This is the case with random dispersion of spheres which we shall study in more detail later on.

The above mentioned results of Beran indicated that there may exist more intimate connection between variances and effective properties. Indeed, as shown independently by several authors [6, 7, 15], the variance is simply connected to the derivatives of the effective conductivity $\kappa^* = \kappa^*(\kappa_f, \kappa_m)$, treated as a function of the material properties κ_f, κ_m of the two constituents in a binary medium. This is an interesting and important result, but its practical application is limited by the fact that very rarely rigorous analytical formulae for $\kappa^*(\kappa_f, \kappa_m)$ are known for realistic random constitution. Rigorous bounds on $\kappa^*(\kappa_f, \kappa_m)$ are well-known, of course, but they obviously cannot supply any estimates for the appropriate derivatives.

In the present note we shall employ another method for studying variation in random dispersions. Namely, we shall use the fact that for the latter the full statistical solution of the problem (1.1) can be conveniently constructed by means of the functional series approach, see [10, 16, 17]. Moreover, the first two kernels of the series can be explicitly found, which results, in particular, in a formula for the needed variances, which is *exact* to the order c^2, where c is the volume fraction of the spheres. Then the observation that some of the terms in the appropriate formulae are sign-definite produces a bound on the variances which, as it turns out, can be simply expressed by means of the c^2-coefficient of the effective conductivity. The latter, as it is well-known, represents a quantity extensively studied in the literature.

2. C^2–SOLUTION OF THE BASIC PROBLEM (1.1) FOR DISPERSIONS OF SPHERES

To get certain rigorous results for the variance, one should somewhat narrow the class of two-phase random media. To this end, consider in more detail here a random dispersion of spheres as a typical representative of the wide and important class of particulate microinhomogeneous media, extensively studied in the literature.

Let us recall first the so-called virial (or density) expansion of κ^* in powers of the volume fraction c of the spheres:

$$\frac{\kappa^*}{\kappa_m} = 1 + a_1 c + a_2 c^2 + \cdots \tag{2.1}$$

Note that hereafter we shall try to cover simultaneously both 3-D case (dispersion of spheres) and its 2-D counterpart — a matrix containing an array of circular and aligned fibers subjected to a macroscopic gradient perpendicular to fiber axes. Depending on dimension, a will denote either the sphere radius (3-D) or the radius of the cylinder cross-section (2-D). For the volume fraction c of the spheres we have $c = nV_a$, $V_a = \frac{4}{3}\pi a^3$ in the 3-D case, or $c = nS_a$, $S_a = \pi a^2$ in 2-D, n is the number density of the spheres or of the fibers.

As it is well-known, the coefficient a_1 in (2.1) is the only thing rigorously calculated by Maxwell [20] in his classical theory of macroscopic conductivity of a random dispersion. The Maxwell result reads

$$a_1 = d\beta_d, \quad \beta_d = \frac{[\kappa]}{\kappa_f + (d-1)\kappa_m}, \quad [\kappa] = \kappa_f - \kappa_m; \tag{2.2}$$

hereafter $d = 3$ in the 3-D case and $d = 2$ in the 2-D-case.

For higher sphere fraction, the Maxwell theory [20] yields the well-known approximate relation

$$\frac{\kappa^*}{\kappa_m} = 1 + \frac{d\beta_d c}{1 - \beta_d c} \tag{2.3}$$

— the so-called Maxwell (or Clausius-Mossotti) formula [14]. The latter produces in turn the following approximation for the c^2-coefficient, namely:

$$a_2 = d\beta_d^2, \quad d = 2, 3. \tag{2.4}$$

The rigorous evaluation of a_2 has attracted the attention of many authors, because this is the simplest case in which the multiparticle interaction shows up in a nontrivial way. We refer here to the papers [22, 13, 12, 16] *et al.*, where a_2 has been expressed in a closed form, making use of the zero-density limit $g_0(r)$ of the so-called radial distribution function for the spheres, and of the one- and two-inclusion fields for the conductivity problem under study. (Recall that the radial distribution function $g(r) = f_2(r)/n^2$, where $r = |\mathbf{y}_1 - \mathbf{y}_2|$, so that $g(r) = g_0(r) + o(n)$ in the dilute limit $n \to 0$; $f_2(r) = f_2(\mathbf{y}_1 - \mathbf{y}_2)$ denotes the two-point probability density for the set of sphere centers.) In the 2-D case (fiber-reinforced material), the coefficient a_2 has been evaluated analytically by the authors [19], making use of the earlier reasoning of Peterson and Hermans [22].

As already mentioned, the full statistical solution of the problem (1.1) for a random dispersion can be conveniently constructed by means of the functional series approach, see [10, 16, 17] for details. In particular, as shown by one of the authors [16], the temperature gradient fluctuation in the dispersion of spheres, correct to the order c^2, has the form of the truncated functional series:

$$\begin{aligned}\nabla\theta'(\mathbf{x}) &= \int \nabla_x T_1(\mathbf{x}-\mathbf{y}) D_\psi^{(1)}(\mathbf{y})\, d\mathbf{y} \\ &+ \iint \nabla_x T_2(\mathbf{x}-\mathbf{y}_1, \mathbf{x}-\mathbf{y}_2) D_\psi^{(2)}(\mathbf{y}_1, \mathbf{y}_2)\, d\mathbf{y}_1 d\mathbf{y}_2,\end{aligned} \tag{2.5}$$

where

$$D_\psi^{(1)} = \psi'(\mathbf{x}) = \psi(\mathbf{x}) - n, \tag{2.6}$$

$$\begin{aligned}D_\psi^{(2)}(\mathbf{y}_1, \mathbf{y}_2) &= \psi(\mathbf{y}_1)[\psi(\mathbf{y}_2) - \delta(\mathbf{y}_1 - \mathbf{y}_2)] \\ &- n g_0(\mathbf{y}_1 - \mathbf{y}_2)[\psi'(\mathbf{y}_1) + \psi'\mathbf{y}_2)] - n^2 g_0(\mathbf{y}_1 - \mathbf{y}_2),\end{aligned} \tag{2.7}$$

and the kernel $T_2(\mathbf{y}_1, \mathbf{y}_2)$ is a symmetric function of its arguments $\mathbf{y}_1$ and $\mathbf{y}_2$. Recall that n is the number density of the spheres, so that their volume fraction is $c = n\,\frac{4}{3}\pi a^3$ in 3-D and $c = n\,\pi a^2$ in 2-D. The integrals hereafter are over the entire space $\mathbb{R}^d$ if the integration domain is not explicitly indicated. In (2.5) to (2.7)

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

is the random density field of Stratonovich [23], generated by the random set $\{\mathbf{x}_\alpha\}$ of sphere centers. The fields $D_\psi^{(1)}$, $D_\psi^{(2)}$ and the constant field $D_\psi^{(0)} = 1$ constitute a c^2-orthogonal family, i.e.

$$\left\langle D_\psi^{(1)} \right\rangle = \left\langle D_\psi^{(2)} \right\rangle = \left\langle D_\psi^{(1)} D_\psi^{(2)} \right\rangle = o(c^2), \tag{2.8}$$

which means that in the c^2-analysis performed below the averaged values in (2.8) can be neglected. We have also

$$\begin{aligned}&\left\langle D_\psi^{(1)}(\mathbf{y}_1) D_\psi^{(1)}(\mathbf{y}_2) \right\rangle = n\delta_{12} - n^2 R_0(\mathbf{y}_1 - \mathbf{y}_2), \quad R_0(\mathbf{y}) = 1 - g_0(\mathbf{y}), \\ &\left\langle D_\psi^{(2)}(\mathbf{y}_1, \mathbf{y}_2) D_\psi^{(2)}(\mathbf{y}_3, \mathbf{y}_4) \right\rangle = n^2 g_0(\mathbf{y}_1 - \mathbf{y}_2)\big(\delta_{13}\delta_{24} + \delta_{14}\delta_{23}\big),\end{aligned} \tag{2.9}$$

where $\delta_{ij} = \delta(\mathbf{y}_i - \mathbf{y}_j)$, and g_0 is the above mentioned low-density limit of the radial distribution function for the set of sphere centers, which is the only statistical characteristics of the dispersion needed in the c^2-statistical solution for the temperature gradient. The relations (2.9), as well as *all* formulae in the sequel, are correct to the order c^2 only.

The kernel T_1 that enters (2.5) has the form

$$T_1(\mathbf{x}) = T_{10}(\mathbf{x}) + nT_{11}(\mathbf{x}). \tag{2.10}$$

In (2.10) $T_{10}(\mathbf{x})$ is the "one-sphere" solution, i.e. the disturbance field superimposed by a single spherical inclusion of radius a (located at the origin) on a temperature field in the matrix with constant gradient $\mathbf{G}$ at infinity. Recall that $T_{10}(\mathbf{x})$ solves the equation

$$\kappa_m \Delta T_{10}(\mathbf{x}) + [\kappa]\nabla \cdot \{h(\mathbf{x})[\mathbf{G} + \nabla T_{10}]\} = 0 \tag{2.11a}$$

and hence

$$T_{10}(\mathbf{x}) = d\beta_d \mathbf{G} \cdot \nabla\varphi(\mathbf{x}), \tag{2.11b}$$

where $\varphi(\mathbf{x})$ is the Newtonian potential for a sphere (in 3-D) or for a circle (in 2-D) of radius a; $h(\mathbf{x})$ denotes the characteristic function of a single sphere (or disk in 2-D) centered at the origin, and β_d was defined in (2.2). As it is well known, the potential $\varphi(\mathbf{x})$ solves the equation

$$\Delta\varphi(\mathbf{x}) + h(\mathbf{x}) = 0, \tag{2.12}$$

which implies, in particular, that

$$h(\mathbf{x})\nabla T_{10}(\mathbf{x}) = -d\beta_d h(\mathbf{x})\mathbf{G}, \quad \Delta T_{10}(\mathbf{x}) = -d\beta_d \nabla \cdot \big(h(\mathbf{x})\mathbf{G}\big). \tag{2.13}$$

To specify $T_{11}(\mathbf{x})$, we should first note that to the order c^2 the kernel T_2 in (2.5) equals T_{20}. The latter solves the equation

$$\begin{aligned} 2\kappa_m \Delta T_{20}(\mathbf{x}, \mathbf{x} - \mathbf{z}) + [\kappa]\,\nabla \cdot \{2\big[h(\mathbf{x}) + h(\mathbf{x} - \mathbf{z})\big]\nabla T_{20}(\mathbf{x}, \mathbf{x} - \mathbf{z}) \\ + h(\mathbf{x})\nabla T_{10}(\mathbf{x} - \mathbf{z}) + h(\mathbf{x} - \mathbf{z})\nabla T_{10}(\mathbf{x})\} = 0. \end{aligned} \tag{2.14}$$

The differentiation hereafter is with respect to $\mathbf{x}$, and $\mathbf{z}$ plays the role of a parameter. Hence

$$2T_{20}(\mathbf{x} - \mathbf{z}; \mathbf{x}) = T^{(2)}(\mathbf{x}; \mathbf{z}) - T_{10}(\mathbf{x}) - T_{10}(\mathbf{x} - \mathbf{z}) \tag{2.15}$$

with $T^{(2)}(\mathbf{x}; \mathbf{z})$ denoting the "two-sphere" solution, i.e. 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 $\mathbf{z}$, $|\mathbf{z}| > 2a$, when the temperature gradient at infinity equals $\mathbf{G}$. Thus

$$\kappa_m \Delta T^{(2)}(\mathbf{x}; \mathbf{z}) + [\kappa]\,\nabla \cdot \Big\{[h(\mathbf{x}) + h(\mathbf{x} - \mathbf{z})]\,[\mathbf{G} + \nabla T^{(2)}(\mathbf{x}; \mathbf{z})]\Big\} = 0, \tag{2.16}$$

which is the counterpart of the "single-sphere" equation (2.11a).

The coefficient $T_{11}(\mathbf{x})$ can be represented as

$$T_{11}(\mathbf{x}) = \beta_d V_a T_{10}(\mathbf{x}) + 2L_{20}(\mathbf{x}), \quad L_{20}(\mathbf{x}) = \int_{|\mathbf{z}|>2a} g_0(\mathbf{z}) T_{20}(\mathbf{x}-\mathbf{z};\mathbf{x})\, d\mathbf{z}. \quad (2.17)$$

To calculate the effective conductivity κ^* through the kernels T_1 and T_2, note that the conductivity field $\kappa(\mathbf{x})$ of the dispersion has a form, similar to (2.5), namely,

$$\kappa(\mathbf{x}) = \langle\kappa\rangle + \kappa'(\mathbf{x}), \quad \kappa'(\mathbf{x}) = [\kappa]\int h(\mathbf{x}-\mathbf{y}) D_\psi^{(1)}(\mathbf{y})\, d\mathbf{y}. \quad (2.18)$$

That is why, inserting (2.5) and (2.17) into (1.2) and using the orthogonality of the fields $D_\psi^{(1)}$ and $D_\psi^{(2)}$, see (2.8), give

$$\begin{aligned} \kappa^*\mathbf{G} &= \langle\kappa(\mathbf{x})\nabla\theta(\mathbf{x})\rangle = \langle\kappa\rangle\,\mathbf{G} + \langle\kappa'(\mathbf{x})\nabla\theta'(\mathbf{x})\rangle \\ &= \langle\kappa\rangle\,\mathbf{G} + n[\kappa]\int h(\mathbf{x})\nabla S(\mathbf{x})\, d\mathbf{x} \end{aligned} \quad (2.19)$$

with the function

$$S(\mathbf{x}) = T_1(\mathbf{x}) - n\int T_1(\mathbf{x}-\mathbf{y}) R_0(\mathbf{y})\, d\mathbf{y} = S_0(\mathbf{x}) + nS_1(\mathbf{x}), \quad (2.20)$$

so that, due to (2.10),

$$S_0(\mathbf{x}) = T_{10}(\mathbf{x}), \quad S_1(\mathbf{x}) = T_{11}(\mathbf{x}) - \int T_{10}(\mathbf{x}-\mathbf{y}) R_0(\mathbf{y})\, d\mathbf{y}. \quad (2.21)$$

Inserting (2.10) and (2.21) into (2.9) and comparing the result with (2.16) give for the virial coefficients a_1 and a_2:

$$a_1 = \left(1-\beta_d\right)\frac{[\kappa]}{\kappa_m} = d\beta_d, \quad (2.22)$$

which indeed coincides with the exact value, given in (2.17), and

$$a_2 = d\beta_d^2 + a_2', \quad a_2'\mathbf{G} = 2\,\frac{[\kappa]}{\kappa_m}\frac{1}{V_a^2}\int h(\mathbf{x})\nabla L_{20}(\mathbf{x})\, d\mathbf{x}. \quad (2.23)$$

Note that the integrals in (2.17) and (2.23) are conditionally convergent, the mode of integration being extracted in the course of the statistical solution of the problem (1.1), see [16, 18] for details and discussion. Namely, one should integrate first with respect to the angular coordinates at fixed $r = R$ and only then with respect to the radial coordinate R. This mode of integration will be tacitly used hereafter to avoid convergent difficulties for some of the integrals in Section 4.

Let us point out finally that though the formula for a_2' in (2.23) is written for a 3-D dispersion, it holds as well in the 2-D case, with the only change that the volume V_a of the inclusions is replaced by their area S_a and the integrals are two-tuple. The same will hold true in all formulae in the sequel. Moreover, a closed form analytic formula for a_2' in the 2-D case was derived, let us recall, by the authors in [19].

3. C^2–FORMULA FOR THE VARIANCES $\sigma^2_{\nabla\theta}$ AND σ^2_Q

Insert the representation (2.5) into the definition (1.3) of the variance. Due to the orthogonality of the fields $D_\psi^{(1)}$ and $D_\psi^{(2)}$, see (2.8), we get

$$\sigma^2_{\nabla\theta} = \frac{1}{G^2}\left(\mathcal{A}_1 + \mathcal{A}_2\right), \tag{3.1}$$

where

$$\mathcal{A}_1 = \iint \nabla_x T_1(\mathbf{x}-\mathbf{y}_1)\cdot\nabla_x T_1(\mathbf{x}-\mathbf{y}_2)\left\langle D_\psi^{(1)}(\mathbf{y}_1)D_\psi^{(1)}(\mathbf{y}_2)\right\rangle d\mathbf{y}_1 d\mathbf{y}_2, \tag{3.2}$$

$$\begin{aligned}\mathcal{A}_2 &= \iiiint \nabla_x T_2(\mathbf{x}-\mathbf{y}_1,\mathbf{x}-\mathbf{y}_2)\cdot\nabla_x T_2(\mathbf{x}-\mathbf{y}_3,\mathbf{x}-\mathbf{y}_4)\\ &\quad\times\left\langle D_\psi^{(2)}(\mathbf{y}_1,\mathbf{y}_2)D_\psi^{(2)}(\mathbf{y}_3,\mathbf{y}_4)\right\rangle d\mathbf{y}_1\, d\mathbf{y}_2\, d\mathbf{y}_3\, d\mathbf{y}_4.\end{aligned} \tag{3.3}$$

Note that

$$\mathcal{A}_2 = \left\langle \left| \iint \nabla_x T_2(\mathbf{x}-\mathbf{y}_1,\mathbf{x}-\mathbf{y}_2) D_\psi^{(2)}(\mathbf{y}_1,\mathbf{y}_2)\, d\mathbf{y}_1\, d\mathbf{y}_2 \right|^2 \right\rangle,$$

which implies immediately that $\mathcal{A}_2 \geq 0$, and hence

$$\sigma^2_{\nabla\theta} \geq \frac{1}{G^2}\mathcal{A}_1. \tag{3.4}$$

An evaluation of the term $\mathcal{A}_1$ yields thus the lower estimate (3.4) of the variance.

Note that the evaluation of $\mathcal{A}_1$ is much easier than that of $\mathcal{A}_2$. The reason, as we shall see below (Section 4), is that to evaluate $\mathcal{A}_1$ only the single sphere solution T_{10} is needed together with the values, assumed known, of the c^2-term a_2 in (2.16). At the same time $\mathcal{A}_2$ involves already the double-sphere field $T^{(2)}$ in a nontrivial way, which essentially complicates the investigation. Note also that the term $\mathcal{A}_2$ has the order $O(c^2)$ (see (2.9)), so that the lower estimate (3.4) gives correct to the order $O(c)$ results in the dilute case $c \ll 1$. Hence from (3.4) the exact value of the c-coefficient A_1, see (3.12), and a lower bound on the c^2-coefficient A_2 in the virial expansion, see (4.5) below, of the variance $\sigma^2_{\nabla\theta}$ will follow in particular.

4. EVALUATION OF THE TERM $\mathcal{A}_1$

Using (2.9) into (3.2), we get

$$\mathcal{A}_1 = n\int \nabla T_1(\mathbf{x})\cdot\nabla S(\mathbf{x})\, d\mathbf{x} \tag{4.1}$$

with the function $S(\mathbf{x})$ defined in (2.20), and hence due to (2.10) and (2.21)

$$\mathcal{A}_1 = n\left(\mathcal{A}_{11} + n\mathcal{A}_{12}\right), \tag{4.2}$$

$$\mathcal{A}_{11} = \int \nabla T_{10}(\mathbf{x}) \cdot \nabla T_{10}(\mathbf{x})\, d\mathbf{x}, \tag{4.3}$$

$$\mathcal{A}_{12} = 2\int \nabla T_{10}(\mathbf{x}) \cdot \nabla T_{11}(\mathbf{x})\, d\mathbf{x} - \int \nabla T_{10}(\mathbf{x}) \cdot \int \nabla_x T_{10}(\mathbf{x}-\mathbf{y}) R_0(\mathbf{y})\, d\mathbf{y} d\mathbf{x}. \tag{4.4}$$

Let

$$\sigma^2_{\nabla\theta} = A_1 c + A_2 c^2 + \cdots, \quad \sigma^2_q = B_1 c + B_2 c^2 + \cdots \tag{4.5}$$

be the virial expansions of the variances, similar to the classical expansion (2.1) for the effective conductivity. The leading terms A_1 and B_1 can be easily found, since this requires an evaluation of the integral $\mathcal{A}_{11}$ from (4.3). To this end integrate by parts in (4.3), use (2.13), and once again integrate by parts

$$\mathcal{A}_{11} = -\int T_{10}(\mathbf{x}) \cdot \Delta T_{10}(\mathbf{x})\, d\mathbf{x} = d\beta_d \mathbf{G} \cdot \int h(\mathbf{x}) \nabla T_{10}(\mathbf{x})\, d\mathbf{x},$$

so that using (2.13) once more gives

$$\mathcal{A}_{11} = d\beta_d^2 V_a^2 G^2.$$

Together with (4.2) this gives the leading terms of the virial expansions (4.6) of the variances, namely,

$$A_1 = \beta_d a_1 = d\beta_d^2, \quad B_1 = d(d-1)\beta_d^2 = (d-1)A_1. \tag{4.6}$$

Turning to the evaluation of $\mathcal{A}_{12}$, we start with the first integral in (4.4). Integrating by parts and using (2.13) together with the formula (2.15) for T_{11}, we have

$$\int \nabla T_{10}(\mathbf{x}) \cdot \nabla T_{11}(\mathbf{x})\, d\mathbf{x} = \int \Delta T_{10}(\mathbf{x}) T_{11}(\mathbf{x})\, d\mathbf{x} = -d\beta_d \int \nabla \cdot (h(\mathbf{x})\mathbf{G}) T_{11}(\mathbf{x})\, d\mathbf{x}$$

$$= -d\beta_d \mathbf{G} \cdot \int h(\mathbf{x}) \nabla T_{11}(\mathbf{x})\, d\mathbf{x} = -d\beta_d \mathbf{G} \cdot \int h(\mathbf{x}) \left[\beta_d V_a \nabla T_{10}(\mathbf{x}) + 2\nabla L_{20}(\mathbf{x})\right] d\mathbf{x}$$

$$= d\beta_d^3 V_a^2 G^2 - 2d\beta_d \mathbf{G} \cdot \int h(\mathbf{x}) \nabla L_{20}(\mathbf{x})\, d\mathbf{x}.$$

The second term in the last formula is connected with the c^2-value a_2 of the effective conductivity, see (2.23), so that

$$\int \nabla T_{10}(\mathbf{x}) \cdot \nabla T_{11}(\mathbf{x})\, d\mathbf{x} = \left(\beta_d^3 - \frac{d\kappa_m}{\kappa_f + (d-1)\kappa_m} \left(a_2 - d\beta_d^2\right)\right) dV_a^2 G^2. \tag{4.7}$$

The second integral in (4.4) is similarly simplified through integration by parts, and applying (2.13):

$$\int \nabla T_{10}(\mathbf{x}) \cdot \int \nabla_x T_{10}(\mathbf{x}-\mathbf{y}) R_0(\mathbf{y})\, d\mathbf{y} d\mathbf{x} = -d\beta_d \mathbf{G} \cdot \int\int \nabla T_{10}(\mathbf{x}) h(\mathbf{x}-\mathbf{y}) R_0(\mathbf{y})\, d\mathbf{y} d\mathbf{x}$$

$$= -d^2\beta_d^2 \mathbf{G} \cdot \iint h(\mathbf{x}-\mathbf{y}) R_0(\mathbf{y}) \nabla\nabla\varphi(\mathbf{x})\, d\mathbf{y} d\mathbf{x} \cdot \mathbf{G}, \tag{4.8}$$

having used the representation (2.11b) of the single sphere solution $T_{10}(\mathbf{x})$ through the Newtonian potential of the sphere. But, due to the isotropy of the latter, the integral in the right-hand side of (4.8) represents a second rank isotropic tensor, so that

$$\iint h(\mathbf{x}-\mathbf{y}) R_0(\mathbf{y}) \nabla\nabla\varphi(\mathbf{x})\, d\mathbf{y} d\mathbf{x} = \gamma \mathbf{I} \tag{4.9}$$

and, upon contraction,

$$\gamma d = \iint h(\mathbf{x}-\mathbf{y}) R_0(\mathbf{y}) \Delta\varphi(\mathbf{x})\, d\mathbf{y} d\mathbf{x} = -\iint h(\mathbf{x}-\mathbf{y}) h(\mathbf{x}) R_0(\mathbf{y})\, d\mathbf{y} d\mathbf{x},$$

see (2.12). The product $h(\mathbf{x}-\mathbf{y})h(\mathbf{x})$ does not vanish however only in the sphere $|\mathbf{y}| \leq 2a$, where $R_0(\mathbf{y}) = 1 - g_0(\mathbf{y}) = 1$, due to the nonoverlapping assumption ($g_0(\mathbf{y}) = 0$ if $|\mathbf{y}| \leq 2a$, since the spheres are forbidden to intersect). Thus $\gamma d = -V_a^2$ and from (4.8) and (4.9) it follows that

$$\int \nabla T_{10}(\mathbf{x}) \cdot \int \nabla_x T_{10}(\mathbf{x}-\mathbf{y}) R_0(\mathbf{y})\, d\mathbf{y} d\mathbf{x} = -d\beta_d^2 V_a^2 G^2. \tag{4.10}$$

Combining (4.7) and (4.10) into (4.4) and using (3.1) and (2.5) give eventually

$$\mathcal{A}_{12} = \left(d\beta_d^2 - 2(1-\beta_d)a_2\right) V_a^2 G^2.$$

From (3.4), (4.4) and the last formula, as it was already discussed, immediately follows the lower bound

$$A_{12} \leq A_2, \quad A_{12} = d\beta_d^2 - 2(1-\beta_d)a_2, \tag{4.11}$$

for the c^2-coefficient of the variance $\sigma^2_{\nabla\theta}$. Using (1.4) gives in turn the following upper bound for the respective coefficient of the flux variance σ_q^2, namely,

$$B_2 \leq B_{12}, \quad B_{12} = d\beta_d^2 \left[d(1-2\alpha+2\alpha\beta_d) - \alpha \right] + \left[2(1-\beta_d)\alpha + \alpha - 1 \right] a_2. \tag{4.12}$$

5. CONCLUDING REMARKS

The estimates (4.11) and (4.12) for the c^2-coefficients in the virial expansions (4.5) of the variances represent the central result of the present note. They have been obtained without using variational arguments — instead the full statistical solution of the problem (1.1) has been appropriately exploited. The estimates account for the statistics of the dispersion through the well-known and extensively studied in the literature c^2-coefficient a_2 of the effective conductivity. Moreover, they remain finite for high-contrast media, when the ratio $\alpha = \kappa_f/\kappa_m$ goes to 0 or ∞. A more detailed investigation of the estimates (4.11), (4.12) and of their implications will be performed elsewhere.

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