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ON THE EFFECTIVE BEHAVIOUR OF A NONLINEAR DISPERSION OF SPHERES

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Константин Марков, Красимир Цвятков. ОБ ЭФФЕКТИВНОМ ПОВЕДЕНИИ НЕЛИНЕЙНОЙ СУСПЕНЗИИ СФЕР

Рассматриваеця эффективное поведение нелинейной суспензии сфер на примере распространения тепла; нелинейность понимаеця в том смысле, что коэффициент проводимости, разный для матрицы и для сфер, зависит также и от градиента температуры в каждой точке. Используеця вариационная процедура для оценки эффективных свойств суспензий; в случае малой концентрации сфер получены аналитические результаты.

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The effective behaviour of a nonlinear random dispersion of spheres is considered in the context of heat propagation; nonlinearity is understood in the sense that the coefficient of thermal conductivity, being different for the matrix and for the spheres, depends on the temperature gradient at each point. A variational technique is proposed which allows to estimate the effective properties of the dispersion. In the case of a dilute dispersion the estimates are explicitly evaluated.

1. INTRODUCTION

Consider a two-phase random medium which is statistically homogeneous and isotropic. For the sake of definiteness only, we shall call one of the constituents matrix and the other filler, though our considerations will for the moment hold for an arbitrary two-phase medium. All quantities connected with the matrix will be supplied with the subscript "m", and those connected with the filler — with "f". To describe the random constitution of the medium we introduce the random field $I_f(\mathbf{x})$ which equals 1 if $\mathbf{x}$ lies in the filler and vanishes otherwise. The set of all multipoint moments (or correlation functions) for $I_f(\mathbf{x})$ defines adequately the random medium [1, 2].

Consider the stationary heat propagation in the medium which we suppose to be governed by the nonlinear version of the Fourier law:

$$\begin{aligned} \nabla \cdot \mathbf{q}(\mathbf{x}) &= 0, \qquad \mathbf{q}(\mathbf{x}) = K(\mathbf{x}; T(\mathbf{x}))\nabla\theta(\mathbf{x}), \\ K(\mathbf{x}; T) &= K_m(T) + [K_f(T) - K_m(T)]I_f(\mathbf{x}), \end{aligned} \tag{1.1a}$$

$T = T(\mathbf{x}) = |\nabla\theta(\mathbf{x})|^2$, where $\mathbf{q}(\mathbf{x})$ is the opposite heat flux vector, $\theta(\mathbf{x})$ is the temperature field; this means that each of the constituents has a coefficient of thermal conductivity that depends on the magnitude of the local temperature gradient. We suppose that

$$0 < k_1 < K(\mathbf{x}; T) < k_2 < \infty, \qquad \forall \mathbf{x},\ \forall T. \tag{1.2}$$

Let the medium be subject to constant macroscopic temperature gradient, i.˜e.

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

where $\langle \cdot \rangle$ denotes ansemble averaging. The condition (1.1b) plays the role of a boundary one for the nonlinear random equation (1.1a). Under certain assumptions about the functions $K_m(T)$ and $K_f(T)$, see §2, the problem (1.1) possesses a solution $\theta(\mathbf{x})$ which is unique. By means of the latter we can evaluate the average heat flux, $\mathbf{Q}$, in the medium

$$\mathbf{Q} = \langle \mathbf{q}(\mathbf{x}) \rangle = \langle K(\mathbf{x}; T(\mathbf{x}))\nabla\theta(\mathbf{x}) \rangle = K^*(g)\mathbf{G}; \tag{1.3}$$

here $g = \mathbf{G} \cdot \mathbf{G}$ and K^* is the effective thermal conductivity of the medium which, similarly to that of the constituents, represents a certain nonlinear function of the average temperature gradient acting on the medium, $K^* = K^*(g)$.

Thus the given random geometry of the medium under study defines a nonlinear operator $\mathbf{K}$ that transforms the functions $K_m(\tau)$ and $K_f(\tau)$ into the function $K^*(g)$ which describes the effective behaviour of the medium;

$$K^*(\cdot) = \mathbf{K}[K_m(\cdot), K_f(\cdot)]. \tag{1.4}$$

The well-known homogenization problem consists in evaluation of the operator $\mathbf{K}$. Note that the problem of estimating the operator $\mathbf{K}$ for given statistics of the

medium is considered by J. Willis [3] making use of a variational principle of the type of the Hashin-Shtrikman's, see also [4].

Certain remarks concerning the operator $\mathbf{K}$ are now warranted.

First, in the case of linear behaviour of the constituents we have $K_m(T) \equiv \kappa_m$, $K_f(T) \equiv \kappa_f, K^*(g) \equiv \kappa^*$, where κ^* is the effective conductivity of the medium, so that the operator $\mathbf{K}$ reduces to a scalar function of two scalar arguments

$$\kappa^* = \mathbf{K}(\kappa_m, \kappa_f). \tag{1.5}$$

Even in this simplest case there is no general algorithm how to calculate κ^* for an arbitrary random geometry; the difficulty of such a calculation is well acknowledged in the literature, see, e. g., [2, 5], and it stems from the fact the basic random problem (1.1) is statistically nonlinear even when the behaviour of the constituents is linear, i. e., when they obey the classical Fourier law. In the case under consideration, the problem (1.1) is both statistically and physically nonlinear. The latter makes the analysis of nonlinear random media much more complicated. The interest in such media however, has increased in the recent years. One of the basic reasons is that the nonlinear effects become important in certain polarizable materials when the electric field (the exact counterpart of $|\nabla\theta(\mathbf{x})|$ here) is large, e. g., when a laser beam is directed into the material, cf. [6, 7] for details and references.

Second, the operator $\mathbf{K}$ is homogeneous in the sense that

$$\mathbf{K}[\lambda K_m(\cdot), \lambda K_f(\cdot)] = \lambda \mathbf{K}[K_m(\cdot), K_f(\cdot)], \tag{1.6}$$

for any $\lambda \in$R. In the linear case this yields that κ^* depends on the ratio κ_f/κ_m, but in the nonlinear case the latter statement cannot be generalized, i. e., $K^*(\cdot)$ does not depend on the ratio $K_f(\cdot)/K_m(\cdot)$ only, except when $K_m(\cdot) \equiv$ const.

Our aim here is to get certain results for the effective behaviour of the nonlinear composite, i. e. for the function $K^*(g)$, in the particular case of a random dispersion of spheres, making use of the variational procedure introduced and briefly discussed in [8].

2. THE VARIATIONAL PRINCIPLE

We replace the random problem (1.1) by the variational principle which states that the true temperature field in the medium minimizes the functional

$$\begin{aligned} W_A[\theta(\cdot)] &= \langle \Phi(\mathbf{x}; |\nabla\theta(\mathbf{x})|^2) \rangle \longrightarrow \min, \\ \Phi(\mathbf{x}; T) &= \tfrac{1}{2} \int_0^T K(\mathbf{x}; \tau) d\tau; \qquad T = T(\mathbf{x}) = |\nabla\theta(\mathbf{x})|^2, \end{aligned} \tag{2.1}$$

over the class of random fields $\theta(\mathbf{x})$ which satisfy (1.1b). Moreover,

$$\min W_A[\theta(\cdot)] = \frac{1}{2} K^*(g) g, \qquad g = \mathbf{G} \cdot \mathbf{G}. \tag{2.2}$$

This is a straightforward generalization of the classical variational principle in the linear case [1].

It is noteworthy that the variational principle (2.1) allows to sketch a simple proof of an existence and uniqueness theorem for the basic random problem (1.1).

Indeed, let us consider the class of random fields $\theta(\mathbf{x})$ whose gradients are statistically homogeneous. On using a statement of ergodic type, we replace ensemble averages by spatial ones

$$\langle\cdot\rangle = \frac{1}{V}\int\limits_V \cdot d^3x,$$

where V is a certain macro-region. We next introduce the standard Hilbert space $H_1(V)$ and consider the set $G \subset H_1(V)$ of functions that satisfy the boundary condition

$$\theta(\mathbf{x})\,|_{\partial V} = \mathbf{G}\cdot\mathbf{x}.$$

This is obviously a convex closed subset in $H_1(V)$. Moreover, the Gauss theorem easily yields that the functions $\theta \in G$ comply with the condition (1.1b). The functional W_A in (2.1) is strictly convex on $H_1(V)$ if the function Φ is such with respect to T. The latter holds in turn if the function $K(\mathbf{x};\tau)$ is strictly monotonically increasing with respect to τ. Due to (1.1a), this will be true if both functions $K_f(\tau)$ and $K_m(\tau)$ have the same monotonicity property:

$$\frac{d}{d\tau}K_f(\tau) > 0, \qquad \frac{d}{d\tau}K_m(\tau) > 0 \tag{2.3}$$

— something which will be assumed hereafter. The conditions (2.3) also yield that the functional W_A is coercitive, i. e.,

$$W_A[\theta(\cdot)] \longrightarrow \infty \ \text{ if } \ ||\theta|| \longrightarrow \infty. \tag{2.4}$$

According to a classical result of the convex analysis, see, e. g., [9, Ch. II, prop. 1.2], the property (2.4) suffices to claim that the functional W_A, strictly convex under the assumptions (2.3), possesses a unique minimizing element $\theta^*(\mathbf{x})$ over the convex set G; this element is just the true temperature field in the medium, i. e., the solution of the basic problem (1.1).

In what follows a special attention will be paid to the particular case of nonlinear constitutive relation [7] for which

$$K_f(\tau) = \kappa_f^0 + \kappa_f^1\tau, \quad K_m(\tau) = \kappa_m^0 + \kappa_m^1\tau. \tag{2.5}$$

The functional (2.1) in this case becomes

$$W_A[\theta(\cdot)] = \frac{1}{2}\langle\kappa^0(\mathbf{x})|\nabla\theta(\mathbf{x})|^2\rangle + \frac{1}{4}\langle\kappa^1(\mathbf{x})|\nabla\theta(\mathbf{x})|^4\rangle, \tag{2.6}$$

where

$$\kappa^i(\mathbf{x}) = \kappa_m^i + [\kappa^i]I_f(\mathbf{x}), \tag{2.7}$$

$[\kappa^i] = \kappa_f^i - \kappa_m^i$, $i = 0, 1$. The condition of strict convexity for the functional (2.6) reads

$$\kappa_f^i,\ \kappa_m^i > 0, \qquad i = 0, 1, \tag{2.8}$$

cf. (2.3), which will be assumed hereafter in order to ensure the existence and uniqueness for the solution of the random problem (1.1) in the particular case of constitutive relations (2.5).

The variational principle, dual to (2.1), can be easily derived if we reformulate the random problem (1.1) with respect to the heat flux $\mathbf{q}(\mathbf{x})$. From (1.1) and (2.5) we get

$$\nabla\theta(\mathbf{x}) = L(\mathbf{x}; \mathbf{q}(\mathbf{x}))\mathbf{q}(\mathbf{x}), \tag{2.9}$$

where L denotes the compliance field

$$\begin{aligned} L(\mathbf{x}; \mathbf{q}(\mathbf{x})) &= 1/K(\mathbf{x}; T(\mathbf{x})) \\ &= L^0(\mathbf{x}) - L^1(\mathbf{x})|\mathbf{q}(\mathbf{x})|^2 + L^2(\mathbf{x})|\mathbf{q}(\mathbf{x})|^4 - \cdots; \\ L^0(\mathbf{x}) &= 1/\kappa^0(\mathbf{x}), \\ L^1(\mathbf{x}) &= L^O(\mathbf{x})\kappa^1(\mathbf{x})/(\kappa^0(\mathbf{x}))^3, \qquad \text{etc.} \end{aligned} \tag{2.10}$$

Obviously the compliance field does not have the form (2.5) of the conductivity field. This form holds only if the nondimensional temperature gradient g_m, defined in (4.5) below, is small, $g_m \ll 1$. In the latter case, within the accuracy $o(g_m)$, the series (2.10) can be truncated after the second term and we thus will obtain the counterpart of (2.5), namely,

$$\nabla\theta(\mathbf{x}) = \{L^0(\mathbf{x}) - L^1(\mathbf{x})|\mathbf{q}(\mathbf{x})|^2\}\mathbf{q}(\mathbf{x}). \tag{2.11}$$

To the same order of accuracy we can reformulate the random problem (1.1) as follows

$$\mathbf{q}(\mathbf{x}) = \nabla \times \Psi(\mathbf{x}), \tag{2.12a}$$

$$\nabla\theta(\mathbf{x}) = \{L^0(\mathbf{x}) - L^1(\mathbf{x})|\mathbf{q}(\mathbf{x})|^2\}\mathbf{q}(\mathbf{x}), \tag{2.12b}$$

$$\langle \nabla \times \Psi(\mathbf{x}) \rangle = \mathbf{Q}, \tag{2.12c}$$

where $\mathbf{Q}$ is the prescribed macroscopic heat flux. The problem (2.12) is equivalent to the variational principle

$$W_B[\Psi(\cdot)] = \frac{1}{2}\langle L^0(\mathbf{x})|\nabla \times \Psi(\mathbf{x})|^2\rangle - \frac{1}{4}\langle L^1(\mathbf{x})|\nabla \times \Psi(\mathbf{x})|^4\rangle \longrightarrow \min, \tag{2.13}$$

where the functional W_B is considered over the class of vector potential fields $\Psi(\mathbf{x})$ that satisfy (2.12c). Moreover,

$$\min W_B = \frac{1}{2}L^*(q)q, \qquad q = \mathbf{Q} \cdot \mathbf{Q}, \tag{2.14}$$

where $L^*(q)$ is the effective compliance of the composite defined as follows

$$\mathbf{G} = L^*(q)\mathbf{Q}, \tag{2.15}$$

the counterpart of (1.3). The relation between $K^*(g)$ and $L^*(q)$ is a consequence of the identities

$$q = K^*(g)g, \qquad g = L^*(q)q, \tag{2.16}$$

cf. (1.3) and (2.15).

Finally, let us recall once more that all relations (2.11) to (2.16) are correct only to the order $O(g_m)$, i. e. for small values of the temperature gradient impressed on the medium.

As is well known, the variational principles allow to obtain estimates on the effective properties of random media [10, 11]. Here we shall use the principles (2.1) and (2.13) in order to derive such estimates on the function $K^*(\cdot)$ that defines the effective behaviour of a nonlinear dilute dispersion of spheres whose constituents obey (2.5).

3. THE VARIATIONAL PROCEDURE FOR A DISPERSION OF SPHERES

Let the medium be a random dispersion of equisized and nonoverlapping spheres of radius a and let $\mathbf{x}$ be the set of random points that serve as centers of spheres. After [12] we introduce the field

$$\psi(\mathbf{x}) = \sum_j \delta(\mathbf{x} - \mathbf{x}_j), \tag{3.1}$$

called the random density field for the dispersion. Obviously, $\langle\psi(\mathbf{x})\rangle = n$, where n is the number density of the spheres, $n = c/V_a$ and c is the volume fraction of the spheres. The field $\psi(\mathbf{x})$ provides an exhaustive description of the random set $\mathbf{x}_j$ and thus of the dispersion as well. Its application is very convenient here because the random fields in (2.7) have simple integral representations by means of $\psi(\mathbf{x})$, namely,

$$\kappa^i(\mathbf{x}) = \kappa^i_m + [\kappa^i]\int h(\mathbf{x} - \mathbf{y})\psi(\mathbf{y})d^3\mathbf{y}, \qquad i = 0, 1, \tag{3.2}$$

where $h(\mathbf{x})$ is the indicator function for a single sphere of radius a located at the origin. (Hereafter if the integration domain is not explicitly indicated, the integrals are taken over the whole R^3.)

Consider after [11] the class of trial fields

$$\theta(\mathbf{x}) = \mathbf{G}\cdot\mathbf{x} + \int P(\mathbf{x} - \mathbf{y})\psi'(\mathbf{y})d^3\mathbf{y}, \tag{3.3}$$

where $\psi'(\mathbf{y}) = \psi(\mathbf{y}) - n$ is the fluctuating part of $\psi(\mathbf{y})$ and $P(\mathbf{x})$ is a nonrandom kernel. The fields (3.3) are obviously admissible since they satisfy (1.1b). On introducing (3.2) and (3.3) into the functional (2.6) we make the latter an usual functional of the kernel $P(\mathbf{x})$ whose minimization yields the respective Euler-Lagrange equation for the minimizing $P(\mathbf{x})$. If this equation could be solved we would be able to obtain an upper bound of the function $K^*(g)$. Making use of the arguments of [11, p. 1], it can be shown that this estimate would be the best possible which employs the statistical information given by the l-point correlation functions for the dispersion up to $l = 5$. The said Euler-Lagrange equation is however very complicated; even in the dilute case, $c \ll 1$, when its form is much simpler:

$$\begin{aligned} &\langle\kappa^0\rangle\Delta P(\mathbf{x}) + \nabla\cdot\{[\kappa^0]h(\mathbf{x})[\mathbf{G} + \nabla P(\mathbf{x})] \\ &+[\langle\kappa^1\rangle + [\kappa^1]h(\mathbf{x})]|\mathbf{G} + \nabla P(\mathbf{x})|^2(\mathbf{G} + \nabla P(\mathbf{x}))\} = 0, \end{aligned} \tag{3.4}$$

it seems very diffficult to be handled analytically. That is why we shall employ a certain variational procedure of Ritz type, similar to that proposed by Beran [13] and discussed by the authors in [14] in the linear case.

Consider the perturbation solution of the problem (1.1) in the case of weakly inhomogeneous medium governed by the constitutive relations (2.5):

$$\theta(\mathbf{x}) = \mathbf{G}\cdot\mathbf{x} + \theta^{(1,0)}(\mathbf{x}) + \theta^{(0,1)}(\mathbf{x}) + \dots, \tag{3.5}$$

where $\theta^{(p,q)}(\mathbf{x})$ are centered, $\langle\theta^{(p,q)}(\mathbf{x})\rangle = 0$, and have the order of magnitude $(\delta\kappa^0)^p(\delta\kappa^1)^q$, p, $q = 0, 1, \dots$,

$$\delta\kappa^i = \max_x\left|\frac{\kappa^{i'}(\mathbf{x})}{\langle\kappa^i\rangle}\right|, \qquad i = 0, 1;$$

we suppose that both $\delta\kappa^i \ll 1$, but $\delta\kappa^0$ and $\delta\kappa^1$ could be small of different order of magnitudes. (The prime denotes the fluctuating part of the respective random variable.)

Simple arguments show that the functions $\theta^{(1,0)}(\mathbf{x})$ and $\theta^{(0,1)}(\mathbf{x})$ comply with the equations

$$[\langle\kappa^0\rangle + g\langle\kappa^1\rangle]\Delta\theta^{(1,0)}(\mathbf{x}) + 2\langle\kappa^1\rangle\mathbf{GG} : \nabla\nabla\theta^{(1,0)}(\mathbf{x}) + \nabla\cdot\{\kappa^{0'}(\mathbf{x})\mathbf{G}\} = 0, \tag{3.6a}$$

$$[\langle\kappa^0\rangle + g\langle\kappa^1\rangle]\Delta\theta^{(0,1)}(\mathbf{x}) + 2\langle\kappa^1\rangle\mathbf{GG} : \nabla\nabla\theta^{(0,1)}(\mathbf{x}) + \nabla\cdot\{\kappa^{1'}(\mathbf{x})\mathbf{G}\} = 0; \tag{3.6b}$$

the colon means contraction with respect to two pairs of indexes.

We next introduce the new variable $\mathbf{z}$ instead of $\mathbf{x}$ as follows: $\mathbf{z} = \mathbf{x} + \mu\mathbf{GG}\cdot\mathbf{x}$,with the scalar μ to be specified below. Obviously, $\nabla_x = (\mathbf{I} + \mu\mathbf{GG})\cdot\nabla_z$,which allows, after simple manipulations, to reduce eqns (3.6) to the Poisson equations

$$\begin{aligned} &\Delta_z\theta^{(1,0)}(\mathbf{z}) + \nabla_x\cdot\{\kappa^{0'}(\mathbf{x})\mathbf{G}\} = 0, \\ &\delta_z\theta^{(0,1)}(\mathbf{z}) + \nabla_x\cdot\{\kappa^{1'}(\mathbf{x})\mathbf{G}\} = 0, \end{aligned} \tag{3.7}$$

provided μ is chosen as one of the roots, μ_1 or μ_2, of the square equation

$$
\begin{aligned}
&A + 2B\mu + C\mu^2 = 0; \quad A = 2\langle\kappa^1\rangle, \\
&B = 3g\langle\kappa^1\rangle + \langle\kappa^0\rangle, \qquad C = 3g^2\langle\kappa^1\rangle + g\langle\kappa^0\rangle,
\end{aligned} \tag{3.8}
$$

whose discriminant

$$\Delta = [3g\langle\kappa^1\rangle + \langle\kappa^0\rangle][g\langle\kappa^1\rangle + \langle\kappa^0\rangle]$$

is obviously positive so that both roots μ_1 and μ_2 are real. In (3.7), $\Delta_z = \nabla_z \cdot \nabla_z$ is the Laplace operator with respect to $\mathbf{z}$.

In virtue of (3.7), the bounded everywhere solutions of eqns (3.6) have the form

$$
\begin{aligned}
\theta^{(1,0)}(\mathbf{z}) &= \mathbf{G} \cdot \int \frac{1}{4\pi|\mathbf{z}-\mathbf{y}|} \nabla_y \kappa^{0'}(\mathbf{y}) d^3\mathbf{y}, \\
\theta^{(0,1)}(\mathbf{z}) &= \mathbf{G} \cdot \int \frac{1}{4\pi|\mathbf{z}-\mathbf{y}|} \nabla_y \kappa^{1'}(\mathbf{y}) d^3\mathbf{y},
\end{aligned} \tag{3.9}
$$

where $\mathbf{z} = \mathbf{x} + \mu\mathbf{G}\mathbf{G} \cdot \mathbf{x}$ with μ being one of the said roots, μ_1 or μ_2. Note that it does not matter which one of the roots, μ_1 or μ_2, will be chosen becouse the expressions, say, for $\theta^{(1,0)}(\mathbf{x})$ thus obtained will both satisfy the nonhomogeneous linear equation (3.6a); their difference will then solve the homogeneous equation (3.6a) and since we deal with bounded everywhere solutions this difference will vanish.

Following the general idea of Beran [13], we introduce, instead of (3.3), the class of trial fields

$$\theta(\mathbf{x}) = \mathbf{G} \cdot \mathbf{x} + \lambda_1 \theta^{(1,0)}(\mathbf{x}) + \lambda_2 \theta^{(0,1)}(\mathbf{x}), \tag{3.10}$$

where λ_1, λ_2 are two adjustable scalar parameters. For two-phase media we, however, have

$$\kappa^{i'}(\mathbf{x}) = [\kappa^i] I'_f(\mathbf{x}), \qquad i = 0, 1,$$

see (2.7), where $I'_f(\mathbf{x})$ is the fluctuating part of the indicator function $I_f(\mathbf{x})$ for the filler phase. That is why the two functions $\theta^{(1,0)}(\mathbf{x})$ and $\theta^{(0,1)}(\mathbf{x})$ in (3.9) are proportional, so that (3.10) can be replaced by the class

$$\theta(\mathbf{x}) = \mathbf{G} \cdot \mathbf{x} + \lambda \theta^{(1,0)}(\mathbf{x}), \tag{3.11}$$

with a single adjustable parameter $\lambda \in R$. On introducing (3.11) into the functional (2.6) we make the latter a scalar function of λ whose minimization brings forth a certain upper Ritz' type bound on the effective function $K^*(\cdot)$ defined in (1.3). This bound takes into account statistical information given by the l-point correlation functions for the medium up to $l = 5$. Unlike the bound which could be obtained through the solution of the equation of the type (3.4), i. e., when considering the class (3.3) of trial functions, the said Ritz' type bound will not be optimal in general.

4. THE LOWER AND UPPER BOUNDS FOR DILUTE DISPERSIONS

To illustrate the performance of the above considered scheme and to get certain tangible results at the same time, we shall deal hereafter with the case of a dilute dispersion for which $c \ll 1$. The needed moments of the random density field $\psi(\mathbf{x})$ in this case are very simple, namely,

$$\langle \psi(\mathbf{y}_1) \dots \psi(\mathbf{y}_\rho) \rangle = n\delta(\mathbf{y}_\rho - \mathbf{y}_1) \dots \delta(\mathbf{y}_\rho - \mathbf{y}_{\rho-1}) + o(n), \tag{4.1}$$

$\rho = 1, 2, \dots$, where n, let us recall, is the number density of the spheres, $n = c/V_a$. Another simplification to be adopted consists in replacing the field $\theta^{(1,0)}(\mathbf{x})$ from (2.9) by the respective field from the linear case

$$\widetilde{\theta}(\mathbf{x}) = \mathbf{G} \cdot \int \frac{1}{4\pi|\mathbf{x} - \mathbf{y}|} \nabla_y \kappa^{0'}(\mathbf{y}) d^3\mathbf{y}. \tag{4.2a}$$

Making use of (3.2), it is easily seen that

$$\widetilde{\theta}(\mathbf{x}) = \int \widetilde{T}(\mathbf{x} - \mathbf{y}) \psi'(\mathbf{y}) d^3\mathbf{y}, \tag{4.2b}$$

where $\widetilde{T}(\mathbf{x}) = \mathbf{G} \cdot \nabla\phi(\mathbf{x})$, with $\phi(\mathbf{x})$ denoting the harmonic (Newtonian) potential of a single sphere located at the origin. As is well-known the kernel $\widetilde{T}(\mathbf{x})$ is proportional to the disturbances to the temperature field in a linear unbounded matrix, introduced by a single spherical inhomogeneities, when the temperature gradient at infinity equals $\mathbf{G}$.

We thus restrict the functional (2.6) over the class of trial fields

$$\theta(\mathbf{x}) = \mathbf{G} \cdot \mathbf{x} + \lambda\widetilde{\theta}(\mathbf{x}), \lambda \in R, \tag{4.3}$$

instead of the class (3.11), under the assumption (4.1).

Note that the choise of $\widetilde{\theta}(\mathbf{x})$ is reasonable because in what follows we shall be able to calculate explicitly the needed bounds only for small, in a certain sense to be explained below, magnitudes of the macrogradient $\mathbf{G}$ for which the difference between $\theta^{(1,0)}(\mathbf{x})$ and $\widetilde{\theta}(\mathbf{x})$ is negligible.

The functional W_A from (2.6), when restricted over the class (4.3), becomes a polynomial of fourth degree of λ:

$$W_A[\theta(\cdot)] = w_A(\lambda) = \frac{1}{2}\kappa_m^0 g\{A + B\lambda + C\lambda^2 + D\lambda^3 + E\lambda^4\}, \tag{4.4a}$$

and simple algebra gives the following expressions for the coefficients of this polynomial:

$$\begin{aligned} A &= 1 + (\alpha^0 - 1)c + \tfrac{1}{2}[1 + (\alpha^1 - 1)c]g_m, \\ B &= 2(\alpha^0 - 1)c + 2(\alpha^1 - 1)cg_m, \\ C &= (\alpha^0 + 2)c + (3\alpha^1 + 3.6)cg_m, \\ D &= 2(\alpha^1 - 0.4)cg_m, \ E = \tfrac{1}{2}(\alpha^1 + 1.6)cg_m. \end{aligned} \tag{4.4b}$$

Here $\alpha^i = \kappa_f^i/\kappa_m^i$, $i = 0, 1$, and

$$g_m = g\kappa_m^1/\kappa_m^0, \quad g_f = g\kappa_f^1/\kappa_f^0 \tag{4.5}$$

are nondimensional temperature gradients with respect to the matrix and filler constituent respectively, $g = \mathbf{G} \cdot \mathbf{G}$. The minimization of the function $w_A(\lambda)$, defined in (4.4a), with respect to λ leads to the cubic equation

$$B + 2C\lambda + 3D\lambda^2 + 4E\lambda^3 = 0. \tag{4.6}$$

This equation, under the assumption (2.8), has a unique real root

$$\lambda_0 = \lambda_0(\alpha^0, \alpha^1, g_m, c). \tag{4.7}$$

On inserting (4.7) into (4.4) and making use of the variational principle (2.1) and (2.2), we get an upper bound on the effective function $K^*(\cdot)$ for the dilute dispersion:

$$\begin{aligned} &K^*(g) \leq K^u, \\ &K^u = K^u(\alpha^0, \alpha^1, g_m) = \kappa_m^0\{A + B\lambda_0 + C\lambda_0^2 + D\lambda_0^3 + E\lambda_0^4\}. \end{aligned} \tag{4.8}$$

Obviously, only numerical results for the bound K^u can be obtained in general. It is important to note, however, that K^u, unlike the form (2.5), will not be already a quadratic function of $\mathbf{G}$ which indicates that in the constitutive law (1.3) for the dispersion all degrees of g will be present. The quadratic form (2.5) will hold for the dispersion only for small values of the gradient g_m, i.e., within the accuracy $o(g_m)$.

To get a lower bound for the effective function $K^*(\cdot)$, we shall employ the variational principle (2.13), dual to (2.1), and the class of trial fields

$$\Psi(\mathbf{x}) = \frac{1}{2}\mathbf{Q} \times \mathbf{x} + \eta \int \widetilde{\Psi}(\mathbf{x} - \mathbf{y})\psi'(\mathbf{y})d^3\mathbf{y}, \qquad \eta \in \mathbf{R}, \tag{4.9a}$$

where the kernel $\widetilde{\Psi}(\mathbf{x})$ is chosen, similarly to the kernel $\widetilde{T}(\mathbf{x})$ in (4.2b), as the vector potential of the heat flux disturbance, introdused in an unbounded linear matrix, by a single spherical inhomogeneity, when the heat flux at infinity is kept constant, $\mathbf{Q}$, i.e.,

$$\widetilde{\Psi}(\mathbf{x}) = \begin{cases} \mathbf{Q} \times \mathbf{x}, & \text{if } |\mathbf{x}| \leq a \\ -a^3\mathbf{Q} \times \nabla\dfrac{1}{|\mathbf{x}|}, & \text{if } |\mathbf{x}| > a. \end{cases} \tag{4.9b}$$

The functional W_B, defined in (2.13), when restricted over the class (4.9), becomes a polynomial of fourth degree of η

$$W_B[\Phi(\cdot)] = w_B(\eta), \tag{4.10}$$

whose minimization leads to a cubic equation, similar to (4.6). The real root of this equation,

$$\eta_0 = \eta_0(\alpha^0, \alpha^1, q, c), \tag{4.11}$$

is unique under the assumption (2.8). On introducing (4.11) into (4.10) we get an upper bound on the effective compliance $L^*(q)$ and thus, according to (2.16),a lower bound on $K^*(g)$ which corresponds to the upper bound of Ritz' type (4.8).

The evaluation of the above introduced bounds can be performed analytically to the order g_m, at $g_m \ll 1$; it shows that the upper and lower bounds coincide within this order of accuracy. In this way we find the following exact result for the effective conductivity function $K^*(\cdot)$ for the dilute dispersion of spheres:

$$K^*(g) = \kappa_0^* + \kappa_1^* g_m + \cdots, \tag{4.12a}$$

$$\kappa_0^* = \kappa_m^0(1 + 3\beta^0 c), \qquad \beta^0 = [\kappa^0]/(\kappa_f^0 + 2\kappa_m^0), \tag{4.12b}$$

$$\kappa_1^* = \kappa_m^0(1 + 3B_1 c), \tag{4.12c}$$

$$B_1 = \frac{1}{3}[\alpha^1(1 - \beta^{04}) + 1.6\beta^{04} + 1.6\beta^{03} + 7.2\beta^{02} + 4\beta^0 - 1].$$

As it should have been expected, (4.12b) coincides with the well-known Maxwell formula for the effective conductivity of a dilute dispersion of spheres, cf. [15], which is exact in the case of a linear behaviour of the constituents.

Consider the other limiting case when $g_m \longrightarrow \infty$ when we may neglect the first terms in the r. h. sides of (2.5) and think that both spheres and matrix obey the linear Fourier law with conductivities $\kappa_f g$ and $\kappa_m g$ respectively: the variance of g becomes negligible in this case. Making use of the above mentioned Maxwell formula for the dilute dispersion, see (4.12b), we get the following asymptotics for the function $K^*(\cdot)$:

$$\begin{aligned} &K^*(g) \underset{g_m \to \infty}{\longrightarrow} k_\infty^* g_m, \\ &k_\infty^* = \kappa_m^0(1 + 3\beta^1 c), \quad \beta^1 = [\kappa_m^1]/(2\kappa_f^1 + \kappa_m^1). \end{aligned} \tag{4.13}$$

The relations (4.12) and (4.13) can be combined within a certain Padé approximation of the type [2/1], cf. [16], for the effective function $K^*(\cdot)$ which may be employed for all values of g, namely:

$$K^*(g) = \frac{k_0^* + (k_0^* + k_1^*)g_m + k_\infty^* g_m^2}{1 + g_m}. \tag{4.14}$$

The formula (4.14) is the central result of the paper; it provides correct predictions for the function $K^*(\cdot)$ in both cases of small macrogradients, $g_m \ll 1$, to the order $o(g_m)$, and $g_m \longrightarrow \infty$, so that it could be expected to yield a reasonable approximation for the intermediate values of $g_m, 0 < g_m < \infty$ as well.

5. CONCLUDING REMARKS

To conclude the paper we shall show how the results of §4, valid for the particular case (2.5) of linear functions K_f and K_m, could be employed in order to give a certain approximation of Padé's type for the homogenization operator $\mathbf{K}$, defined in (1.4), in general case of arbitrary analytical functions $K_f(\cdot)$ and $K_m(\cdot)$ which comply with the monotonicity condition (2.3).

Let us expand the functions K_f and K_m, analytical on $[0,\infty]$ by assumption, as the Taylor series

$$K_m(g) = \kappa_m^0 + \kappa_m^1 g + \dots, \qquad K_f(g) = \kappa_f^0 + \kappa_m^1 g + \dots, \tag{5.1}$$

where

$$\begin{aligned} &\kappa_m^0 = K_m(0), \quad \kappa_f^0 = K_f(0), \\ &\kappa_m^1 = K_m'(0), \quad \kappa_f^1 = K_f'(0) \end{aligned} \tag{5.2}$$

are known constants. For small values of the nondimensional gradient $g_m = g\kappa_m^1/\kappa_m^0, g_m \ll 1$, we can replace the functions $K_f(g)$ and $K_m(g)$ by their linear approximations (5.1); such a replacement yields result also valid to the same order $o(g_m)$. That is why the formulas (4.12) are valid to the same order $o(g_m)$,

$$K^*(g) = k_0^* + k_1^* g_m + \dots, \tag{5.3}$$

where the coefficients k_0^* and k_1^* are given in (4.12b, c), provided

$$\alpha^0 = K_f(0)/K_m(0), \qquad \alpha^1 = K_f'(0)/K_m'(0),$$

cf. (5.2).

The limiting case $g_m \longrightarrow \infty$ is considered similarly to §4, though there are two possibilities here which should be dealt with separately, namely, (i) The function $K_m(\cdot)$ is bounded on $[0,\infty)$ and (ii). This function is unbounded on $[0,\infty)$, so that $K_m(\infty) = \infty$.

In the first case (i), when $K_m(\infty) < \infty$, we can think both constituents linear at $g_m \longrightarrow \infty$, so that the Maxwell formula (4.12b) yields

$$\begin{aligned} K^*(\infty) &= K_m(\infty)(1 + 3\beta^\infty c), \\ \beta^\infty &= (\alpha^\infty - 1)/(\alpha^\infty + 2), \qquad \alpha^\infty = K_m(\infty)/K_f(\infty). \end{aligned} \tag{5.4}$$

We can combine the formulas (5.3) and (5.4) in the following [1/1] Padé approximation:

$$K^*(g) = \frac{k_0^*[K^*(\infty) - k_0^*] + K^*(\infty)k_1^* g_m}{K^*(\infty) - k_0^* + k_1^* g_m}. \tag{5.5}$$

In the case (ii) we suppose for definiteness that $K_m(g)$ has the asymptotics

$$K_m(g) \sim \kappa_m^\infty g_m^\gamma \text{ at } \ g_m \longrightarrow \infty, \gamma \geq 1. \tag{5.6}$$

(The case $0 < \gamma < 1$ should be treated in a different manner.) Then the arguments, given in the end of §4, are applicable so that we may employ again the Maxwell formula (4.12b) which, together with (5.6), yields

$$
\begin{aligned}
&K^*(g) \sim k^*_\infty g_m^\gamma \text{ at } g_m \longrightarrow \infty, \quad k^*_\infty = \kappa_m^\infty(1 + 3\beta^\infty c),\\
&\beta^\infty = (\alpha^\infty - 1)/(\alpha^\infty + 2), \qquad \alpha^\infty = \lim_{g\to\infty} \tfrac{K_f(g)}{K_m(g)}.
\end{aligned} \tag{5.7}
$$

We then combine (5.3) and (5.7) into the Padé approximation, similar to (4.14):

$$
K^*(g) = \frac{k_0^* + (k_0^* + k_1^*)g_m + k^*_\infty g_m^\gamma}{1 + g_m}. \tag{5.8}
$$

The proposed formulas (4.14), (5.5) and (5.8), exact for both limiting cases of small and big magnitudes of the macrogradient $\mathbf{G}$ can be expected to predict reasonably enough the behaviour of the effective function $K^*(g)$ in the whole region $[0, \infty)$ for a dilute dispersion of spheres.

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R E F E R E N C E S

1. B e r a n, M. Statistical continuum theories. John Wiley, N. Y., 1968.
2. F r i s h, H. L. Statistics of random media. — Trans. Soc. Rheol., **9**, 1965, 293 — 312.
3. W i l l i s, J. R. Variational estimates for the overall response of an inhomogeneous nonlinear dielectric. — In: Proceedings of IMA workshop of homogenization and composites (October, 1984), Springer, Berlin — New York, 1986, 245–263.
4. T a l b o t, D. R. S., J. R. W i l l i s. A variational approach to the overall sink strength of a nonlinear lossy composite medium. — Proc. Roy. Soc., **A405**, 1986, 159–180.
5. M c C o y, J. J. Macroscopic response of continua with random microstructure. — In: Mechanics today (ed. by S. Nemat-Nasser), vol. **6**, Pergamon Press, N. Y., 1981, 1–40.
6. P a p a n i c o l a o u, G. C. Electromagnetic problems in composite materials in linear and nonlinear regimes. — In: Nonlinear electromagnetics (ed. by P. L. E. Uslenghi), Academic Press, N. Y., 1980, 253–261.
7. M i k s i s, M. I. Effective dielectric constant of a nonlinear composite material. — SIAM J. Appl. Math., **43**, 1983, 1140–1155.
8. M a r k o v, K. Z., K. D. Z v y a t k o v. On the nonlinear conductivity of a random suspension. — In: Proceedings 5th Bulg. Congress on Mechanics (Varna, 1985), Publ. House of the Bulg. Acad. Sci., vol. **2**, Sofia, 1985, 48–53.
9. E k e l a n d, I., R. T e m a m. Convex analysis and variational problems. North-Holland, Amsterdam, 1976.
10. W i l l i s, J. R. Variational and related methods for the overall properties of composites. — Adv. Appl. Mechanics, **21**, 1981, 1–78.

11. M a r k o v, K. Z. Application of Volterra-Wiener series for bounding the overall conductivity of heterogeneous media. I. General procedure, II. Suspensions of equi-sized spheres. — SIAM J. Appl. Math., **47**, 1987, 831–850, 851–870.
12. S t r a t o n o v i c h, R. L. Topics in theory of random noises, vol.1, Gordon and Breach, N. Y., 1967.
13. B e r a n, M. Use of a variational approach to determine bounds for the effective permittivity of a random medium. — Nuovo Cimento, **38**, 1965, 771–782.
14. M a r k o v, K. Z., K. D. Z v y a t k o v. On the bounds for the effective properties of random suspensions of spheres. — C. R. Acad. Bulg. Sci., **40**(3), 1987, 17–20.
15. M a x w e l l, J. C. A treatise on electricity and magnetism. Dover, N. Y., 1954. (Republication of 3rd ed. of 1891.)
16. B a k e r, G. A., P. G r a v e s-M o r i s. Padé approximants, Part I: Basic theory. Addison — Wesley, London, 1981.

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