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ON THE OPTIMAL THIRD-ORDER BOUNDS ON THE EFFECTIVE ELASTIC MODULI OF RANDOM DISPERSIONS OF SPHERES

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Константин Марков, Красимир Цвятков. ОБ ОПТИМАЛЬНЫХ ГРАНИЦ ТРЕТЬЕГО ПОРЯДКА ДЛЯ ЭФФЕКТИВНЫХ УПРУГИХ МОДУЛЕЙ СЛУЧАЙНЫХ СУСПЕНЗИЙ СФЕР.

Исследуется вопрос оптимальности вариационных границ Берана-Молине, Маккоя и др. для эффективных модулей упругости двухфазных сред. Оптимальность понимается в смысле получения наиболее узких границ при учете только статистической информации, необходимой для подсчета этих границ, именно двух- и трехточечных корреляционных функций. На примере случайной суспензии сфер показано, что аналогично скалярному случаю, эти границы в общем случае неоптимальны. Оптимальность имеет место лишь до порядка c^2, где c объемная концентрация сфер. Для суспензий границы Берана-Молине и Маккоя подсчитаны явно до порядка c^2 и полученные результаты использованы для исследования применимости некоторых эвристических методов механики композитных материалов.

Konstantin Markov, Krassimir Zvyatkov. ON THE OPTIMAL THIRD-ORDER BOUNDS ON THE EFFECTIVE ELASTIC MODULI OF RANDOM DISPERSIONS OF SPHERES.

The problem of optimality of the variational bounds, due to Beran-Molyneux, McCoy, et al., on the effective elastic moduli of two-phase random media is considered. Optimality is understood in the sense that bounds should be the tightest ones that use the statistical information needed for their evaluation; for the said bounds these are the two- and three-point correlation functions for the medium. For random dispersion of spheres it is shown that the bounds are optimal to the order c^2 only, where c is the volume fraction of the spheres. The Beran-Molyneux and McCoy bounds are then explicitly calculated to the order c^2 for the dispersions and used for a study of applicability of some known schemes of mechanics of composite media.

INTRODUCTION

The paper is devoted to the problem of variational bounding of the effective elastic moduli of two-phase random media. Generalizing the scalar conductivity arguments of [1] we first rederive the Beran-Molyneux [2] and the McCoy [3] bounds on the effective bulk and shear moduli of the media, respectively, as simple Ritz-type approximation within the frame of the general variational procedure given in [4]. Then we pose the central for the paper problem of the optimality of the said bounds. Optimality is understood here in the sense that they should be the tightest ones that use the statistical information needed for their evaluation. For the said bounds these are the two- and three- point correlation functions for the medium. Similarly to the scalar conductivity case [1], it appears that the Beran-Molyneux and the McCoy bounds are not optimal in general. For random dispersions of spheres, however, they are optimal to the order c^2, where c denotes the volume fraction of the spheres. We next calculate explicitly the said bounds to the order c^2. The so-obtained c^2-bounds represent, in particular, a rigorous basis for a comparison with the predictions of some heuristic models in mechanics of composite materials. In this way certain conclusions (mostly negative), concerning the applicability of some known formulas in elasticity of random dispersions, are finally reached.

THE BOUNDING PROCEDURE IN THE ELASTIC CASE

Consider a two-phase elastic random medium, which is statistically homogeneous and isotropic. For definiteness in this moment only we shall call constituents filler and matrix. We assume the constituents isotropic, so that the fourth-rank tensor of elastic moduli of the medium, $\boldsymbol{L}(\boldsymbol{x})$, is a random field of the form

$$\boldsymbol{L}(\boldsymbol{x}) = 3k(\boldsymbol{x})\boldsymbol{J}' + 2\mu(\boldsymbol{x})\boldsymbol{J}'', \tag{2.1a}$$

where $\boldsymbol{J}'$ and $\boldsymbol{J}''$ are the basic isotropic fourth-rank tensors with the Cartesian components

$$J'_{ijkl} = \frac{1}{3}\delta_{ij}\delta_{kl}, \qquad J''_{ijkl} = \frac{1}{2}\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \frac{2}{3}\delta_{ij}\delta_{kl}\right),$$

and

$$k(\boldsymbol{x}) = \langle k \rangle + k'(\boldsymbol{x}), \qquad k'(\boldsymbol{x}) = [k] I'(\boldsymbol{x}), \tag{2.1b}$$

$$\mu(\boldsymbol{x}) = \langle \mu \rangle + \mu'(\boldsymbol{x}), \qquad \mu'(\boldsymbol{x}) = [\mu] I'(\boldsymbol{x}),$$

$[k] = k_f - k_m$, $[\mu] = \mu_f - \mu_m$, k and μ stand everywhere for the bulk and shear modulus, respectively. Hereafter, all quantities, pertaining to the filler, are supplied with the subscript "f" and those for the matrix — with "m", the volume fraction of the filler and matrix are respectively c and $1 - c$. In (2.1b) $I'(\boldsymbol{x}) \doteq I(\boldsymbol{x}) - c$ is the fluctuating part of the indicator function $I(\boldsymbol{x})$ for the region, occupied by the filler constituent, i.e.

$$I(\boldsymbol{x}) = I_f(\boldsymbol{x}) = \begin{cases} 1, & \text{if } \boldsymbol{x} \in \text{filler}, \\ 0, & \text{if } \boldsymbol{x} \in \text{matrix}, \end{cases} \tag{2.2}$$

The Lame equations for the medium, at the absense of body forces, read

$$\nabla \cdot \sigma(\boldsymbol{x}) = 0, \qquad \sigma(\boldsymbol{x}) = \boldsymbol{L}(\boldsymbol{x}) : \varepsilon(\boldsymbol{x}), \tag{2.3a}$$

where σ denotes the stress tensor, $\varepsilon = \frac{1}{2}(\nabla \boldsymbol{u} + \boldsymbol{u}\nabla)$ is the small strain tensor generated by the displacement field $\boldsymbol{u}(\boldsymbol{x})$, the colon denotes contraction with respect to two pairs of indices. In the isotropic case under consideration we have

$$\sigma(\boldsymbol{x}) = k(\boldsymbol{x})\theta(\boldsymbol{x})\boldsymbol{I} + 2\mu(\boldsymbol{x})\boldsymbol{d}(\boldsymbol{x}), \tag{2.4}$$

$$\varepsilon(\boldsymbol{x}) = \frac{1}{3}\theta(\boldsymbol{x})\boldsymbol{I} + \boldsymbol{d}(\boldsymbol{x}), \qquad \theta(\boldsymbol{x}) = \operatorname{tr} \varepsilon(\boldsymbol{x}), \tag{2.5}$$

(cf. (2.1)) so that (2.5) is the decomposition of the strain tensor as a sum of its spherical and deviatoric parts, $\boldsymbol{I}$ stands here for the unit second-rank tensor.

We prescribe also the average strain tensor $\boldsymbol{E}$, imposed on the medium

$$\langle \varepsilon(\boldsymbol{x}) \rangle = \boldsymbol{E}, \tag{2.3b}$$

where $\boldsymbol{E}$ is a given symmetrical second-rank tensor, the brackets $\langle \cdot \rangle$ hereafter denote ensemble averaging. Eqns (2.3) represent the basic random problem (with respect to displacements) in elasticity of composite media. This is the elastic counterpart of the scalar problem, considered in [1].

The random problem (2.3) is equivalent to the variational problem

$$W_A[\boldsymbol{u}(\cdot)] = \langle \varepsilon(\boldsymbol{x}) : \boldsymbol{L}(\boldsymbol{x}) : \varepsilon(\boldsymbol{x}) \rangle \longrightarrow \min. \tag{2.6}$$

The functional W_A is considered over the class of random fields $\boldsymbol{u}(\boldsymbol{x})$, which generate strain fields $\varepsilon(\boldsymbol{x})$ satisfying (2.3b). Moreover, $\min W_A = \boldsymbol{E} : \boldsymbol{L}^* : \boldsymbol{E}$, where $\boldsymbol{L}^*$ is the tensor of effective elastic moduli for the medium. In the statistically isotropic case under consideration $\boldsymbol{L}^* = 3k^*\boldsymbol{J}' + 2\mu^*\boldsymbol{J}''$, where k^* and μ^* are the effective bulk and shear modulus of the random medium respectively.

In order to obtain bounds on the effective properties of a random medium it was proposed in [4] to employ certain truncated functional series as classes of trial fields for the respective variational principles. For an elastic medium the class of such trial fields, in the simplest nontrivial case of interest, is

$$K^{(1)} = \{\boldsymbol{u}(\boldsymbol{x}) | \boldsymbol{u}(\boldsymbol{x}) = \boldsymbol{E}\cdot\boldsymbol{x} + \int \boldsymbol{T}(\boldsymbol{x}-\boldsymbol{y}) I'(\boldsymbol{y}) d^3\boldsymbol{y}\}. \tag{2.7}$$

Hereafter the integrals are over the whole R^3, if the integration domain is not explicity indicated.

The energy functional (2.6), when restricted over the class (2.7), becomes an usual functional of the nonrandom kernel $\boldsymbol{T}(\boldsymbol{x})$, namely

$$W_A[\boldsymbol{T}(\cdot)] = \langle\lambda\rangle \mathrm{tr}^2\boldsymbol{E} + 2\langle\mu\rangle\boldsymbol{E}:\boldsymbol{E}$$

$$+2\int\{[\lambda]\nabla\cdot\boldsymbol{T}(\boldsymbol{y}) + 2[\mu]\boldsymbol{E}:\mathrm{def}\ \boldsymbol{T}(\boldsymbol{y})\}M_2(\boldsymbol{y})d^3\boldsymbol{y}$$

$$+2\int\int\nabla\cdot\boldsymbol{T}(\boldsymbol{y}_1)\nabla\cdot\boldsymbol{T}(\boldsymbol{y}_2)\{\langle\lambda\rangle M_2(\boldsymbol{y}_1-\boldsymbol{y}_2) + [\lambda]M_3(\boldsymbol{y}_1,\ \boldsymbol{y}_2)\}d^3\boldsymbol{y}_1 d^3\boldsymbol{y}_2$$

$$+2\int\int\mathrm{def}\ \boldsymbol{T}(\boldsymbol{y}_1):\mathrm{def}\ \boldsymbol{T}(\boldsymbol{y}_2)\{\langle\mu\rangle M_2(\boldsymbol{y}_1-\boldsymbol{y}_2) + [\mu]M_3(\boldsymbol{y}_1,\ \boldsymbol{y}_2)\}d^3\boldsymbol{y}_1 d^3\boldsymbol{y}_2,$$

where $\mathrm{def}\ \boldsymbol{T}(\boldsymbol{x}) = \frac{1}{2}(\nabla\boldsymbol{T}(\boldsymbol{x}) + \boldsymbol{T}(\boldsymbol{x})\nabla)$,

$$M_2(\boldsymbol{x}) = \langle I'(0)I'(\boldsymbol{x})\rangle, \qquad M_3(\boldsymbol{x},\boldsymbol{y}) = \langle I'(0)I'(\boldsymbol{x})I'(\boldsymbol{y})\rangle$$

are the two- and three-point moments of the indicator field $I(\boldsymbol{x})$, defined in (2.2). Hereafter the differentiation is with respect to $\boldsymbol{x}$.

The Euler-Lagrange equation for the functional $W_A[\boldsymbol{T}(\cdot)]$ reads

$$\boldsymbol{E}:[\boldsymbol{L}]\cdot\nabla M_2(\boldsymbol{x}) + \int\nabla M_2(\boldsymbol{x}-\boldsymbol{y})\cdot\boldsymbol{L}_m : \mathrm{def}\ \boldsymbol{T}(\boldsymbol{y})d^3\boldsymbol{y} \tag{2.8}$$

$$+\int\nabla M_3(\boldsymbol{x},\ \boldsymbol{y})\cdot[\boldsymbol{L}]:\mathrm{def}\ \boldsymbol{T}(\boldsymbol{y})d^3\boldsymbol{y} = 0.$$

It could be shown, employing simple convexity arguments, that the solution of eqn (2.8) does exist and is unique. The solution, $\boldsymbol{T}(\boldsymbol{y})$, is to be inserted into the second equation of (2.3a) which, upon averaging, will bring forth certain upper bounds $k^{(3)}$ and $\mu^{(3)}$ on the effective bulk and shear moduli of the medium. The superscript "3" indicates that the evaluation of the bounds $k^{(3)}$ and $\mu^{(3)}$ requires knowledge of the r-point moments for the field $I(\boldsymbol{x})$ up to $r = 3$. In this sense these bounds are called third-order, similarly to the scalar conductivity case [1, 4]. More important, it could be shown, extending the scalar conductivity arguments of [4], that $k^{(3)}$ and $\mu^{(3)}$ are the optimal third-order bounds in the sense that they are the best ones which can be obtained, making use of the said statistical information, i.e. M_2 and M_3 only.

The explicit solution of the integro-differential equation (2.8) is very difficult in general. That is why we introduce, after [5], a simpler procedure. Let $\widetilde{\boldsymbol{T}}(\boldsymbol{x})$ be a fixed kernel. Consider the set of trial fields

$$\widetilde{K}^{(1)} = \{\boldsymbol{u}(\boldsymbol{x}) | \boldsymbol{u}(\boldsymbol{x}) = \boldsymbol{E} \cdot \boldsymbol{x} + \alpha \int \widetilde{\boldsymbol{T}}(\boldsymbol{x} - \boldsymbol{y}) I'(\boldsymbol{y}) d^3\boldsymbol{y}\} \subset K^{(1)}, \tag{2.9}$$

where $\alpha \in R^1$ is adjustable parameter. The functional W_A, when restricted on $\widetilde{K}^{(1)}$, becomes a quadratic function of α, whose monimization brings forth certain third-order bounds $\widetilde{k}^{(3)}$ and $\widetilde{\mu}^{(3)}$ on the effective bulk and shear moduli. Such bounds, due to obvious reasons, are called in [5] Ritz-type ones. Though not optimal in general, the bounds $\widetilde{k}^{(3)}$ and $\widetilde{\mu}^{(3)}$ could be explicitly evaluated, if the kernel $\widetilde{\boldsymbol{T}}(\boldsymbol{x})$ is skillfully chosen. As a matter of fact, Beran and Molineux [2] and McCoy [3] have pointed out that such a choice of the kernel is supplied by the first-order terms in the perturbation solution of the basic elastic problem (2.3). (A similar observation in the scalar conductivity case is due again to Beran ([6].) In our terminology the above mentioned authors have calculated the bounds $\widetilde{k}^{(3)}$ and $\widetilde{\mu}^{(3)}$ for the said choice of the kernel $\widetilde{\boldsymbol{T}}(\boldsymbol{x})$. Their derivations will be repeated below in the frame of our scheme and then the problem of optimality of the respective bounds for random dispersions of spheres will be addressed. But before this it is necessary that the perturbation solution of the elastic problem (2.3) should be considered at some length.

PERTURBATION SOLUTION OF THE BASIC ELASTIC PROBLEM (8.3)

Let the medium be weakly inhomogeneous, i.e. the ratios

$$\delta k = \max_{\boldsymbol{x}} \frac{|k'(\boldsymbol{x})|}{\langle k \rangle}, \qquad \delta \mu = \max_{\boldsymbol{x}} \frac{|\mu'(\boldsymbol{x})|}{\langle \mu \rangle} \tag{3.1}$$

are small, δk, $\delta \mu \ll 1$, noting, however, that δk, $\delta \mu$ may be small of different orders of magnitude. Consider the perturbation series for the displacement field that solves the problem (2.3)

$$u(x) = u^{(0,0)}(x) + u^{(1,0)}(x) + u^{(0,1)}(x) + \sum_{p,q=1}^{\infty} u^{(p,q)}(x), \tag{3.2}$$

where $u^{(0,0)}(x) = E \cdot x$ and $u^{(p,q)}(x)$ has the order of magnitude $(\delta k)^p(\delta\mu)^q$, besides, $\langle u^{(p,q)}(x)\rangle = 0$, $p, q = 0, 1, \dots, p^2 + q^2 \neq 0$.

On introducing (3.2) into (2.3a), we get straightforwardly

$$u^{(1,0)}(x) = \frac{3\mathrm{tr}\, E}{3\langle k\rangle + 4\langle\mu\rangle} \int \nabla \frac{1}{4\pi|x-y|} k'(y) d^3y, \tag{3.3}$$

$$u^{(0,1)}(x) = \frac{1}{2\pi\langle\mu\rangle} E_d : \int \left\{ \nabla \frac{1}{|x-y|} \otimes I + \overline{\varkappa} \nabla\nabla\nabla|x-y| \right\} \mu'(y) d^3y, \tag{3.4}$$

where $E_d = E - \frac{1}{3} I \mathrm{tr}\, E$ is the deviatoric average strain, hereafter all gradients are with respect to x, $\nabla = \nabla_x$, and

$$\overline{\varkappa} = -\frac{1}{4(1-\overline{\nu})}, \qquad \overline{\nu} = \frac{3\langle k\rangle - 2\langle\mu\rangle}{6\langle k\rangle + 2\langle\mu\rangle}, \tag{3.5}$$

so that $\overline{\nu}$ is the Poisson ratio of a medium with elastic moduli $\langle k\rangle$ and $\langle\mu\rangle$. (Note that $\overline{\nu} \neq \langle\nu\rangle$.) The well-known Green tensor for the Lame equation in the isotropic case is used in an obvious manner, when deriving (3.3) and (3.4).

The reformulation of the problem (2.3) for the stress field is well-known (cf. [7]):

$$\sigma(x) = \nabla \times \Phi(x) \times \nabla; \tag{3.6a}$$

$$\nabla \times (M(x) : \sigma(x)) \times \nabla = 0. \tag{3.6b}$$

Here $M(x) = L^{-1}(x)$ is the fourth-rank compliance tensor field for the medium, and $\Phi(x)$ is the symmetrical second-rank "tensor potential" field for the stress — the stress function of Maxwell and Morrera, which assures that the equilibrium equation (2.3a) is identically satisfied. Similarly to (2.3b), we prescribe the mean value, Σ, for the stress tensor

$$\langle\sigma(x)\rangle = \Sigma. \tag{3.6c}$$

Eqns (3.6) represent the basic random elastic problem, with respect to stress, in elasticity of composite media. The variational formulation of this problem is the principle of minimum complementary energy. Consider the functional

$$W_B[\Phi(\cdot)] = \langle \sigma(\boldsymbol{x}) : \boldsymbol{M}(\boldsymbol{x}) : \sigma(\boldsymbol{x}) \rangle \longrightarrow \min, \tag{3.7}$$

where the field $\sigma(\boldsymbol{x})$ is the birotor of $\Phi(\boldsymbol{x})$, cf. (3.6a), such that (3.6c) holds. Then the solution $\Phi^*(\boldsymbol{x})$ of the problem (3.6) minimizes W_B, so that $\sigma(\boldsymbol{x}) = \nabla \times \Phi^*(\boldsymbol{x}) \times \nabla$ is the real stress field in the medium. Moreover,

$$\min W_B = \Sigma : \boldsymbol{M}^* : \Sigma, \tag{3.8}$$

where $\boldsymbol{M}^* = \boldsymbol{L}^{*-1}$ is the effective compliance tensor of the medium.

In the isotropic case under study we have

$$W_B[\Phi(\cdot)] = \frac{1}{9}\left\langle \frac{1}{k(\boldsymbol{x})} \mathrm{tr}^2 \sigma(\boldsymbol{x}) \right\rangle + \frac{1}{2}\left\langle \frac{1}{\mu(\boldsymbol{x})} \boldsymbol{s}(\boldsymbol{x}) : \boldsymbol{s}(\boldsymbol{x}) \right\rangle, \tag{3.9}$$

$$\min W_B[\Phi(\cdot)] = \frac{1}{9k^*}\mathrm{tr}^2\Sigma + \frac{1}{2\mu^*}\Sigma_d : \Sigma_d, \tag{3.10}$$

where $\boldsymbol{s}(\boldsymbol{x}) = \sigma(\boldsymbol{x}) - \frac{1}{3}\boldsymbol{I}\mathrm{tr}\, \sigma(\boldsymbol{x})$ is the stress deviator and Σ_d is the deviatoric part of the macrostress tensor Σ.

The construction of the Ritz-type lower bounds, similar to the upper ones of Beran and Molineux, needs the first-order perturbation terms in the solution of the random problem (3.6), i.e. the counterparts of the fields $\boldsymbol{u}^{(1,0)}(\boldsymbol{x})$ and $\boldsymbol{u}^{(0,1)}(\boldsymbol{x})$, given in (3.3) and (3.4) respectively . As noted by McCoy [3], the straightforward construction of these terms is however lengthy and tedioús. That is why we shall use another scheme of arguments, suggested and, as a matter of fact employed in the same paper [3]. The scheme consists in the following.

Let us insert the perturbation solution (3.2) into the Hooke law

$$\begin{aligned} \sigma(\boldsymbol{x}) &= \boldsymbol{L}(\boldsymbol{x}) : \varepsilon(\boldsymbol{x}) \\ &= \{\langle \boldsymbol{L} \rangle + \boldsymbol{L}'(\boldsymbol{x})\} : \{\boldsymbol{E} + \nabla \boldsymbol{u}_1(\boldsymbol{x}) + o(\delta \boldsymbol{L})\} \\ &= \langle \boldsymbol{L} \rangle : \boldsymbol{E} + \sigma_1(\boldsymbol{x}) + o(\delta \boldsymbol{L}), \end{aligned} \tag{3.11}$$

where

$$\begin{gathered} \sigma_1(\boldsymbol{x}) = \boldsymbol{L}'(\boldsymbol{x}) : \boldsymbol{E} + \langle \boldsymbol{L} \rangle : \nabla \boldsymbol{u}_1(\boldsymbol{x}), \\ \delta \boldsymbol{L} = \max_{\boldsymbol{x},i,j,k,l} |L'_{ijkl}(\boldsymbol{x})|/L, \quad \boldsymbol{L}^2 = \langle L_{\alpha\beta\gamma\delta} L_{\alpha\beta\gamma\delta} \rangle, \end{gathered} \tag{3.12a}$$

$L'(x) = L(x) - \langle L \rangle$ being the fluctuating part of the field $L(x)$. In the isotropic case under consideration we have $\delta L = \max(\delta k,\ \delta\mu)$ and

$$u_1(x) = u_1^{(1,0)}(x) + u_1^{(0,1)}(x). \tag{3.12b}$$

On averaging (3.11) we get

$$\Sigma = \langle L \rangle : E + o(\delta L), \tag{3.13}$$

so that the field $\sigma_1(x)$, to the order $o(\delta L)$, has the form

$$\sigma_1(x) = L'(x) : \langle L \rangle^{-1} : \Sigma + \langle L \rangle : \nabla u_1(x). \tag{3.14}$$

Since $\langle \sigma_1(x) \rangle = 0$, we have in virtue of (3.12) that

$$\sigma(x) = \Sigma + \sigma_1(x) + o(\delta L)$$

and thus $\sigma_1(x)$ is the needed first-order term in the perturbation expansion of the solution of the problem (3.6).

In the isotropic case

$$\sigma_1(x) = \sigma^{(1,0)}(x) + \sigma^{(0,1)}(x),$$

where $\sigma^{(1,0)}(x)$ and $\sigma^{(0,1)}(x)$ have the orders of magnitude δk and $\delta\mu$ respectively. Moreover $\sigma^{(1,0)}(x) = 0$ if $\mu'(x) = 0$, i.e. if the constituents have the same shear modulus, and $\sigma^{(0,1)}(x) = 0$ if $k'(x) = 0$, i.e. if the bulk modulus is the same. The analytic forms of $\sigma^{(1,0)}(x)$ and $\sigma^{(0,1)}(x)$ easily follow from (3.7) and (3.8):

$$\begin{aligned} \sigma^{(1,0)}(x) = {} & \frac{1}{3}\frac{k'(x)}{\langle k \rangle} I \operatorname{tr} \Sigma + \frac{1}{3}(3\langle k \rangle - 2\langle \mu \rangle) I \nabla \cdot u^{(1,0)} \\ & + \langle \mu \rangle (\nabla u^{(1,0)} + u^{(1,0)} \nabla), \end{aligned} \tag{3.15}$$

$$\begin{aligned} \sigma^{(0,1)}(x) = {} & \frac{\mu'(x)}{\langle \mu \rangle} \Sigma_d + \frac{1}{3}(3\langle k \rangle - 2\langle \mu \rangle) I \nabla \cdot u^{(0,1)} \\ & + \langle \mu \rangle (\nabla u^{(0,1)} + u^{(0,1)} \nabla). \end{aligned} \tag{3.16}$$

The eventual form of these fields would be obtained, if the expressions (3.3) and (3.4) for $u^{(1,0)}(x)$ and $u^{(0,1)}(x)$ are inserted into (3.15) and (3.16) respectively, and transition from E to Σ is made according to (3.13). Such explicit formulas are not needed in what follows, however, because we can use the respective expressions from the evaluation of the upper bounds, which involve contractions of tensors like $\nabla u^{(1,0)}$ and $\nabla u^{(0,1)}$. Therefore the evaluation of the lower Ritz-type bounds can be readily performed if the respective upper bounds are already calculated. In this way the difficulties that appear, due to the presence of birotors in (3.6), are avoided. That is why we shall give in the following the formulas for the lower bounds without any comments.

BERAN — MOLINEUX (BM) BOUNDS ON THE EFFECTIVE BULK MODULUS

We start with the construction of certain Ritz-type bounds on the effective elastic moduli of the two-phase material, making use of the above constructed first-order perturbation fields $\boldsymbol{u}^{(1,0)}$, $\boldsymbol{u}^{(0,1)}$, $\sigma^{(1,0)}$ and $\sigma^{(0,1)}$. The discussion of the problem of their optimality, in the above explained sense, will be postponed till sec. 6.

Let us consider, after Beran and Molineux [2], the class of trial displacement fields

$$\boldsymbol{u}(\boldsymbol{x}) = \boldsymbol{E}\cdot\boldsymbol{x} + \alpha\boldsymbol{u}^{(1,0)}(\boldsymbol{x}), \tag{4.1}$$

where $\boldsymbol{E}$ is spherical, $\boldsymbol{u}^{(1,0)}(\boldsymbol{x})$ is given in (3.3) and α is adjustable scalar parameter, cf. (2.9). On inserting (4.1) into the energy functional (2.6) and minimizing the result with respect to α, one gets the following upper bound on the effective bulk modulus k^*, obtained by the above authors:

$$k^* \leqq k^u_{BM}, \qquad k^u_{BM} = \langle k\rangle\{1 - \langle k'^2\rangle^2/K_u\}, \tag{4.2a}$$

where

$$K_u = \{[\langle\lambda\rangle + 2\langle\mu\rangle]\langle k'^2\rangle + \langle\lambda' k'^2\rangle + 2J\}\langle k\rangle, \tag{4.3}$$

$$J = \int\int\langle\mu'(0),\ k'(\boldsymbol{z})k'(\boldsymbol{w})\rangle\nabla\nabla\frac{1}{4\pi|\boldsymbol{z}|} : \nabla\nabla\frac{1}{4\pi|\boldsymbol{w}|}d^3\boldsymbol{z}d^3\boldsymbol{w}$$

is a certain statistical parameter and $\lambda = k - \dfrac{2}{3}\mu$ is the Lame constant. Hereafter the prime denotes the fluctuating part of the respective random fields. Simple analysis, based on the relations (2.2), shows that

$$J = [\mu][k]^2 A, \tag{4.4}$$

where A is the dimensionless statistical parameter, introduced as follows

(4.5)

$$A = \int\int i(\boldsymbol{z},\ \boldsymbol{w})\nabla\nabla\frac{1}{4\pi|\boldsymbol{z}|} : \nabla\nabla\frac{1}{4\pi|\boldsymbol{w}|}d^3\boldsymbol{z}d^3\boldsymbol{w}, \qquad i(\boldsymbol{z},\ \boldsymbol{w}) = \langle I'(0,\ I'(\boldsymbol{z})I'(\boldsymbol{w})\rangle.$$

The lower BM-bound is obtained when the functional (3.9) is minimized over the class of trial stress fields

$$\sigma(\boldsymbol{x}) = \Sigma + \alpha\sigma^{(1,0)}(\boldsymbol{x}), \qquad \alpha\in R^1,$$

with a spherical Σ and $\sigma^{(1,0)}(\boldsymbol{x})$, defined in (3.15). The final result, in the original Beran — Molyneux form, reads

$$k^l_{BM} \leqq k^*,\ (k^l_{BM})^{-1} = \left\langle \frac{1}{k} \right\rangle - \langle k'^2 \rangle^2 / K_l, \tag{4.2b}$$

$$K_l = \left\langle \frac{k'^2}{k} \right\rangle - \frac{3}{8} \left\langle \frac{k'^2}{\mu} \right\rangle + \frac{9}{8} J',$$

where J' is the statistical parameter

$$J' = \int\int \left\langle \frac{k'(\boldsymbol{z})k'(\boldsymbol{w})}{\mu(0)} \right\rangle \nabla\nabla \frac{1}{4\pi|\boldsymbol{z}|} : \nabla\nabla \frac{1}{4\pi|\boldsymbol{w}|} d^3\boldsymbol{z} d^3\boldsymbol{w}. \tag{4.6}$$

Since

$$\frac{1}{\mu(\boldsymbol{x})} = \left\langle \frac{1}{\mu} \right\rangle + \left[\frac{1}{\mu} \right] I'(\boldsymbol{x}), \tag{4.7a}$$

we have

$$J' = \left\langle \frac{1}{\mu} \right\rangle \langle k'^2 \rangle + \left[\frac{1}{\mu} \right] [k]^2 A, \tag{4.7b}$$

and thus the upper and lower BM-bounds (4.2) depend on the same statistical parameter A. This parameter appears also in the Beran bounds [6] on the effective conductivity, as it could be easily shown. In turn, BM-bounds may be expressed in a concise form [8] by means of the Milton parameters ξ_1 and ξ_2, defined as

$$\xi_2 = 1 - \xi_2 \tag{4.8}$$

$$= \frac{9}{c(1-c)} \int\int \frac{d^3\boldsymbol{z} d^3\boldsymbol{w}}{16\pi^2|\boldsymbol{z}|^3|\boldsymbol{w}|^3} \left\{ S_3(\boldsymbol{z},\ \boldsymbol{w}) - \frac{S_2(\boldsymbol{z})S_2(\boldsymbol{w})}{c} \right\} P_2(u),$$

where $u = \cos\varphi$, φ being the angle between the vectors $\boldsymbol{z}$ and $\boldsymbol{w}$, $P_2(u) = \frac{1}{2}(3u^2-1)$ is the Legendre polynomial of order two, and

$$S_2(\boldsymbol{x}) = \langle I(0)I(\boldsymbol{x}) \rangle, \qquad S_3(\boldsymbol{x},\ \boldsymbol{y}) = \langle I(0)I(\boldsymbol{x})I(\boldsymbol{y}) \rangle \tag{4.9}$$

are the so-called [9] two- and three-point functions respectively. Let us recall that the quantities $S_2(\boldsymbol{x})$ and $S_3(\boldsymbol{x},\ \boldsymbol{y})$ are, respectively, the probabilities of finding in the filler phase (phase "2" in our case) the end points O (the origin, chosen arbitrarily) and O' of the line segment $\overrightarrow{OO'} = \boldsymbol{x}$ and the vertices of the triangle $OO'O''$, where $\overrightarrow{OO'} = \boldsymbol{x}$, $\overrightarrow{OO''} = \boldsymbol{y}$.

Note that the relation between the parameters ξ_2 and A is readily deducible from their definitions, if one takes into account that

$$(4.10) \qquad \nabla\nabla\frac{1}{4\pi|\boldsymbol{z}|} : \nabla\nabla\frac{1}{4\pi|\boldsymbol{w}|} = \frac{6P_2(u)}{|\boldsymbol{z}|^3|\boldsymbol{w}|^3}, \qquad u = \frac{\boldsymbol{z}\cdot\boldsymbol{w}}{|\boldsymbol{z}|\,|\boldsymbol{w}|},$$

and it reads

$$(4.11) \qquad \xi_2 = 1 - \xi_1 = \frac{1}{2}\left(4c + \frac{3A}{c(1-c)} - 1\right).$$

Let the medium have constant shear modulus, i.e. $\mu_f = \mu_m$ and $\mu'(\boldsymbol{x}) = 0$, so that the bulk modulus only varies in position. In this case the lower and the upper BM-bounds coincide yielding the exact values of the effective bulk modulus, namely

$$(4.12) \qquad k^* = \langle k\rangle - \frac{\langle k'^2\rangle}{\langle k\rangle + \frac{4}{3}\langle\mu\rangle + [k](1-2c)}.$$

The same value of k^* can be obtained from the Hashin-Shtrikman bounds [10] on k^*, which also coincide if $\mu_f = \mu_m$. Note that the exact value (4.12) of k^* in the case under consideration was first pointed out by Hill [11].

McCOY (MC) BOUNDS ON THE EFFECTIVE SHEAR MODULUS

The reasoning of McCoy [3] is fully similar to that in sec. 4, namely we assume that $\operatorname{tr}\boldsymbol{E} = 0$, i.e. the macrostrain tensor is deviatoric, and then take the class of trial displacement fields

$$\boldsymbol{u}(\boldsymbol{x}) = \boldsymbol{E}\cdot\boldsymbol{x} + \alpha\boldsymbol{u}^{(0,1)}(\boldsymbol{x}), \qquad \alpha \in R^1,$$

for the energy functional (2.3). Minimization of the latter with respect to α yields the upper MC-bound on the effective shear modulus of the medium, which we write in the form

$$(5.1a) \qquad \mu^* \leqq \mu^u_{MC}, \qquad \mu^u_{MC} = \langle\mu\rangle - \frac{4(4-5\overline{\nu})^2\langle\mu'^2\rangle^2}{15M_u},$$

$$M_u = 2(1-\overline{\nu})(4-5\overline{\nu}\langle\mu\rangle\langle\mu'^2\rangle + \frac{1}{2}(1-2\overline{\nu})^2\langle k'\mu'^2\rangle I_{k\mu\mu}$$

$$+\langle\mu'^3\rangle\left\{I_\mu\frac{1}{3}(13\overline{\nu}^2 - 22\overline{\nu} + 10)\right\},$$

with the dimensionless statistical parameters, defined as follows

$$(5.2)\quad I_{k\mu\mu} = \frac{1}{\langle k'\mu'^2\rangle}\int\int \langle k'(0)\mu'(z)\mu'(w)\rangle \nabla\nabla\frac{1}{4\pi|z|} : \nabla\nabla\frac{1}{4\pi|w|}d^3zd^3w,$$

$$I_\mu = \frac{3}{4}I_\mu^{(2)} + (7\overline{\nu}^2 - 10\overline{\nu} + 1)I_\mu^{(1)},$$

$$I_\mu^{(1)} = \frac{1}{\langle \mu'^3\rangle}\int\int M_3^\mu(z,\ w)\nabla\nabla\frac{1}{4\pi|z|} : \nabla\nabla\frac{1}{4\pi|w|}d^3zd^3w,$$

$$I_\mu^{(2)} = \frac{1}{16\pi^2\langle\mu'^3\rangle}\int\int M_3^\mu(z,\ w)\nabla\nabla\nabla\nabla|z| \bullet \nabla\nabla\nabla\nabla|w|d^3zd^3w,$$

Where $M_3^\mu(z,\ w) = \langle\mu'(0)\mu'(z)\mu'(w)\rangle$ is the three-point correlation function for the random field $\mu(x)$, and ν is defined in (3.5). The bold-faced point in (5.2) denotes full contraction, i.e. contraction with respect to all four pairs of indices.

The parameters $I_{k\mu\mu}$ and $I_\mu^{(1)}$ obviously coincide, being proportional to the above introduced parameter A:

$$(5.3a)\quad I_\mu^{(1)} = I_{k\mu\mu} \doteq \frac{A}{c(1-c)(1-c)}.$$

For the parameter $I_\mu^{(2)}$ we have

$$(5.3b)\quad I_\mu^{(2)} = \frac{4A_1}{c(1-c)(1-2c)},$$

where

$$(5.4)\quad A_1 = \frac{1}{64\pi^2}\int\int i(z,\ w)\nabla\nabla\nabla\nabla|z| \bullet \nabla\nabla\nabla\nabla|w|d^3zd^3w$$

is another statistical parameter for the medium, independent of A, $i(z,\ w) = \langle I'(0)I'(z)I'w)\rangle$. The parameter A_1 is introduced by Milton and Phan-Thien [12], eqn (63). Milton [12, 13] has employed the statistical parameters η_1, η_2, similar to the ξ's, defined in (4.8):

$$(5.5)\quad \eta_2 = 1 - \eta_1 = \frac{5}{21}\xi_2$$

$$+\frac{150}{c(1-c)}\int\int\frac{d^3zd^3w}{16\pi^2|z|^3|w|^3}\left\{S_3(z,\ w) - \frac{S_2(z)S_2(w)}{c}\right\}P_4(u),$$

the same notations being used here as those in (4.8), where $P_4(u) = \frac{1}{8}(35u^4 - 20u^2 + 3)$ is the Legendre polynomial of order four.

The relation between the parameters $\eta_{1,2}$ and A_1 is given in [12], eqn (29). It reads

$$\eta_2 = 1 - \eta_1 = \frac{5}{6}\left(\frac{c + 4A_1 - 3A}{c(1-c)} + \frac{1-c}{5}\right) \tag{5.6}$$

and follows from the formula

$$\nabla\nabla\nabla\nabla|\boldsymbol{z}| \bullet \nabla\nabla\nabla\nabla|\boldsymbol{w}| = \frac{72P_4(u)}{|\boldsymbol{z}|^3|\boldsymbol{w}|^3}, \qquad u = \frac{\boldsymbol{z}\cdot\boldsymbol{w}}{|\boldsymbol{z}|\,|\boldsymbol{w}|}, \tag{5.7}$$

similar to (4.10).

It is important to point out that both statistical parameters ξ_1 and η_1 (and thus ξ_2 and η_2 as well) lie in the interval [0, 1]. Moreover, they satisfy the inequality

$$21\eta_2 - 5\xi_2 \geqq 0, \tag{5.8}$$

as it is shown by Milton and Phan-Thien [12], eqn (52). This inequality is a consequence of the fact that the upper MC-bound should be always greater than the lower one.

The lower MC-bound μ^l_{MC} on the shear modulus μ^* of the medium is obtained when minimizing the functional (3.9) over the class of trial stress fields

$$\sigma(\boldsymbol{x}) = \Sigma + \alpha\sigma^{(0,1)}(\boldsymbol{x}), \qquad \alpha \in R^1,$$

with a deviatoric Σ and $\sigma^{(0,1)}(\boldsymbol{x})$, defined in (3.16). The final result reads

$$\mu^l_{MC} \leqq \mu^*, \quad (\mu^l_{MC})^{-1} = \left\langle\frac{1}{\mu}\right\rangle - \frac{(7-5\overline{\nu})^2\langle\mu'/\mu\rangle^2}{5M_l}, \tag{5.1b}$$

$$M_l = \frac{2}{3}(1+\overline{\nu})^2\left\{3J_{kk\mu} - \langle\mu'^2/k\rangle\right\} + \frac{9}{4}J^{(2)}_{k\mu\mu}$$
$$+3(7\overline{\nu}^2 - 10\overline{\nu} + 1)J^{(1)}_\mu - (2\overline{\nu}^2 + 4\overline{\nu} - 7)\langle\mu'/\mu\rangle,$$

with the following statistical parameters

$$J_{k\mu\mu} = \int\int\left\langle\frac{\mu'(\boldsymbol{z})\mu'(\boldsymbol{w})}{k(0)}\right\rangle\nabla\nabla\frac{1}{4\pi|\boldsymbol{z}|} : \nabla\nabla\frac{1}{4\pi|\boldsymbol{w}|}d^3\boldsymbol{z}d^3\boldsymbol{w}, \tag{5.9}$$

$$J^{(1)}_\mu = \int\int\left\langle\frac{\mu'(\boldsymbol{z})\mu'\boldsymbol{w})}{\mu(0)}\right\rangle\nabla\nabla\frac{1}{4\pi|\boldsymbol{z}|} : \nabla\nabla\frac{1}{4\pi|\boldsymbol{w}|}d^3\boldsymbol{z}d^3\boldsymbol{w},$$

$$J^{(2)}_\mu = \frac{1}{16\pi^2}\int\int\left\langle\frac{\mu'(\boldsymbol{z})\mu'(\boldsymbol{w})}{\mu(0)}\right\rangle\nabla\nabla\nabla\nabla|\boldsymbol{z}| \bullet \nabla\nabla\nabla\nabla|\boldsymbol{w}|d^3\boldsymbol{z}d^3\boldsymbol{w},$$

similar to (5.2).

Simple analysis, based on the relations of the type (4.7), shows that the lower MC-bound (5.1) depends on the same statistical parameters as the upper one. They may be chosen either as A and A_1, defined in (4.5) and (5.4) respectively, or as the Milton parameters ξ_1 and η_1, defined in (4.8) and (5.6) (cf. [8]).

THE CLUSTER BOUNDS FOR DISPERSIONS OF SPHERES

Let the medium be a random dispersion of equisized nonoverlapping spheres of radius a and let $\boldsymbol{x}_j$ be the set of random points that serve as centers of the spheres. The random constitution of the dispersion is exhaustively described by the Stratonovich random density function [1, 4]

$$\psi(\boldsymbol{x}) = \sum \delta(\boldsymbol{x} - \boldsymbol{x}_j). \tag{6.1}$$

Then

$$k'(\boldsymbol{x}) = [k] \int h(\boldsymbol{x} - \boldsymbol{y})\psi'(\boldsymbol{x})d^3\boldsymbol{x}, \tag{6.2}$$

$$\mu'(\boldsymbol{x}) = [\mu] \int h(\boldsymbol{x} - \boldsymbol{y})\psi'(\boldsymbol{x})d^3\boldsymbol{x},$$

where $\psi'(\boldsymbol{x})$ is the fluctuating part of $\psi(\boldsymbol{x})$ and $h(\boldsymbol{x})$ is the characteristic function of a single sphere of radius a, located at the origin. On introducing (6.2) into (3.3) and (3.4), we make, similarly to that in [1, 5], a transition from the basic random field $I(\boldsymbol{x})$ (cf. (2.2)) to the random density field $\psi(\boldsymbol{x})$. The first-order perturbation fields $\boldsymbol{u}^{(1,0)}(\boldsymbol{x})$ and $\boldsymbol{u}^{(0,1)}(\boldsymbol{x})$ then become

$$\boldsymbol{u}^{(1,0)}(\boldsymbol{x}) = \frac{3[k]}{3\langle k\rangle + 4\langle\mu\rangle} \operatorname{tr} \boldsymbol{E} \int \Gamma_s(\boldsymbol{x} - \boldsymbol{y})\psi'(\boldsymbol{y})d^3\boldsymbol{y}, \tag{6.3}$$

$$\boldsymbol{u}^{(0,1)}(\boldsymbol{x}) = 2\frac{[\mu]}{\langle\mu\rangle}\boldsymbol{E}_d : \int \Gamma_d(\boldsymbol{x} - \boldsymbol{y})\psi'(\boldsymbol{y})d^3\boldsymbol{y}, \tag{6.4}$$

where

$$\Gamma_s(\boldsymbol{x}) = \nabla\varphi(\boldsymbol{x}), \quad \Gamma_d(\boldsymbol{x}) = \nabla\varphi(\boldsymbol{x}) \otimes \boldsymbol{I} + \overline{\varkappa}\nabla\nabla\nabla\chi(\boldsymbol{x}), \quad \overline{\varkappa} = -\frac{1}{4(1-\overline{\nu})}, \tag{6.5}$$

and $\varphi = h * \frac{1}{4\pi|\boldsymbol{x}|}$, $\chi = h * \frac{1}{4\pi}|\boldsymbol{x}|$ are respectively the harmonic (Newtonian) and the biharmonic potentials for a single sphere of radius a, located at the origin.

Obviously, the kernel $\Gamma_s(\boldsymbol{x})$ in (6.3) is proportional to the disturbance of the displacement field in an unbounded elastic matrix (of moduli k_m and μ_m), introduced by a single spherical inhomogeneity (of moduli k_f and μ_f), when the strain tensor at infinity is spherical (cf. [14]). This means that in the latter case the class of trial fields (4.1) is just the superposition of such disturbances, multiplied by an adjustable scalar parameter, over the set of spheres in the dispersion. Therefore the BM-bound (4.2a) on the bulk modulus k^*, which corresponds to the class (4.1), coincides with the first-order cluster bound in the sense of Torquato [15] — a conclusion fully similar to that, already reached in [5] for the scalar conductivity case.

The situation with the displacement field $\boldsymbol{u}^{(0,1)}(\boldsymbol{x})$ is a bit more involved. Recalling again the Eshelby result [14], one can easily notice that $\boldsymbol{u}^{(0,1)}(\boldsymbol{x})$ is proportional to the disturbance of the displacement field in an unbounded matrix with shear modulus μ_m and the Poisson ratio $\overline{\nu}$, introduced by a single spherical inhomogeneity with elastic moduli k_m and μ_m, when deviatoric strain is applied at infinity. Thus the MC-bound (5.1a) represents a first-order cluster bound in the sense that the field (3.4) is proportional to the disturbance, generated by a single spherical inhomogeneity. Strictly speaking, however, it is not a cluster bound in the sense of Torquato [15], because the field $\boldsymbol{u}^{(0,1)}(\boldsymbol{x})$ is not the single-sphere disturbance, generated in the matrix material with the moduli k_m and μ_m, i.e. with the Poisson ratio ν_m. The reason is that $\overline{\nu} \neq \nu_m$ and thus $\overline{\varkappa} \neq \varkappa_m$ as well. It could be easily seen, however, that $\overline{\nu} - \nu_m = O(c)$ and thus $\overline{\varkappa} - \varkappa_m = O(c)$ as well. That is why the kernel $\boldsymbol{E}_d : \Gamma_d(\boldsymbol{x})$ in (6.4) is proportional, to the order $O(c)$, to the single-sphere disturbance in the matrix material. This fact, as we shall see in the following sec. 8, suffices to claim that the MC-bounds together with the BM-ones are optimal to the order c^2 for the random dispersions under study.

A GENERALIZATION OF THE McCOY BOUNDS

The very form (3.4) of the field $\boldsymbol{u}^{(0,1)}(\boldsymbol{x})$ hints the following idea. Consider the class of trial displacements

$$(7.1) \qquad \boldsymbol{u}(\boldsymbol{x}) = \boldsymbol{E}_d \cdot \boldsymbol{x} + \boldsymbol{E}_d : \int \left\{ \alpha_1 \nabla \frac{1}{4\pi|\boldsymbol{x}-\boldsymbol{y}|} \otimes \boldsymbol{I} + \alpha_2 \nabla\nabla\nabla \frac{1}{4\pi}|\boldsymbol{x}-\boldsymbol{y}| \right\} I'(\boldsymbol{y}) d^3\boldsymbol{y},$$

where $\boldsymbol{E}_d$ is deviatoric and α_1, α_2 are two adjustable scalar parameters. The minimization of the energy functional (2.6) with respect to α_1 and α_2 brings forth a certain upper bound $\widetilde{\mu}_{MC}$ on the effective shear modulus μ^* of the random medium.

This bound could be called generalized MC-bound. Obviously, the latter coincides with the upper MC-bound, $\widetilde{\mu}_{MC} = \mu_{MC}$, if

$$A_{\min} = \frac{\alpha_2^{\min}}{\alpha_1^{\min}} = \overline{\varkappa}, \tag{7.2}$$

where $\alpha_1^{\min}$, $\alpha_2^{\min}$ are respectively the values of the parameters α_1 and α_2 that minimize the functional (2.6) in the class (7.1), $\overline{\varkappa}$ is defined in (3.5).

As a matter of fact, the class of trial fields (7.1) has been introduced by Milton and Phan-Thien [12], sec. 5a, who considered two-phase random media of periodic internal constitution and employed the Fourier transform of the fields from the class (7.1).

Let the medium be a random dispersion of spheres. On making transition to the random density field $\psi(\boldsymbol{x})$, cf. (6.1), we recast the trial fields (7.1) as

$$\boldsymbol{u}(\boldsymbol{x}) = \boldsymbol{E}_d \cdot \boldsymbol{x} + \boldsymbol{E}_d : \int \{\alpha_1 \nabla\varphi(\boldsymbol{x}-\boldsymbol{y}) \otimes \boldsymbol{I} + \alpha_2 \nabla\nabla\nabla\chi(\boldsymbol{x}-\boldsymbol{y})\}\psi'(\boldsymbol{y})d^3\boldsymbol{y}. \tag{7.3}$$

Using once more the arguments from sec. 6, we note that $\widetilde{\mu}_{MC}$ resembles again the cluster bound of Torquato, because the best kernel in the integral of (7.3) is proportional to the field

$$\boldsymbol{E}_d : \{\nabla\varphi(\boldsymbol{x}-\boldsymbol{y}) \otimes \boldsymbol{I} + A_{\min} \nabla\nabla\nabla\chi(\boldsymbol{x}-\boldsymbol{y})\}, \tag{7.4}$$

with $A_{\min}$, defined in (7.2). In turn, the field (7.4) is proportional to the single-sphere disturbance with the deviatoric strain $\boldsymbol{E}_d$, acting at infinity. However, this disturbance could exist in an elastic matrix material only if $-0.5 \leqq A_{\min} \leqq -0.25$, because the Poisson ratio $\nu \in (0, 0.5)$.

A detailed study with many examples and figures, concerning the Beran-Molyneux, McCoy, generalized McCoy and other new and more restrictive bounds (of fourth-order) on the effective moduli of random elastic media is performed in the above mentioned paper [12], to which we refer the reader for further information. We shall turn now to the problem of optimality of the aforementioned bounds for random dispersions of spheres and their explicit evaluation to the order c^2.

THE OPTIMAL THIRD-ORDER BOUNDS ON THE ELASTIC MODULI

At mentioned in sec. 2, the optimal third-order bounds on the effective elastic moduli could be obtained by solving the Euler-Lagrange equation (2.8). The foregoing Ritz-type bounds will be optimal if the respective kernels satisfy eqn (2.8). The scalar conductivity arguments, presented in [1], can be easily extended to the elastic case as well, so that we could claim that the Beran-Molyneux, the McCoy and the generalized McCoy bounds are not the optimal third-order bounds.

To show however that the said bounds are optimal to the order c^2 for a dispersion of spheres, we shall use again the scheme of arguments of [1]. The arguments for the moment hold for anisotropic constituents with tensors of elastic moduli $\boldsymbol{L}_M$ (for the matrix) and $\boldsymbol{L}_f$ (for the filler particles). Let

$$\boldsymbol{T}(\boldsymbol{x}) = \boldsymbol{T}(\boldsymbol{x}; n) = \boldsymbol{T}_0(\boldsymbol{x}) + n\boldsymbol{T}_1(\boldsymbol{x}) + n^2\boldsymbol{T}_2(\boldsymbol{x}) + \cdots \tag{8.1}$$

be the virial expansion of the optimal kernel $\boldsymbol{T}(\boldsymbol{x})$. We have to underline that it depends on the number density of the spheres n.

Let us insert (8.1) into the functional (2.6), restricted over the class of trial fields (2.7), and expand the result in powers of n:

$$W[\boldsymbol{T}(\cdot)] = \boldsymbol{E} : \langle \boldsymbol{L} \rangle : \boldsymbol{E} + nW_1[\boldsymbol{T}_0(\cdot)] + n^2 W_2[\boldsymbol{T}_0(\cdot),\ \boldsymbol{T}_1(\cdot)] + o(n^2). \tag{8.2}$$

The functionals W_1 and W_2 depend on the indicated virial coefficients as follows:

$$W_1(\boldsymbol{T}_0(\cdot)] = \int \varepsilon_0(\boldsymbol{x}) : \boldsymbol{L}_m : \varepsilon_0(\boldsymbol{x}) d^3\boldsymbol{x} + \int h(\boldsymbol{x})\{\varepsilon_0(\boldsymbol{x}) + 2\boldsymbol{E}\} : [\boldsymbol{L}] : \varepsilon_0(\boldsymbol{x}) d^3\boldsymbol{x}; \tag{8.3}$$

$$W_2[\boldsymbol{T}_0(\cdot),\ \boldsymbol{T}_1(\cdot)] = \overline{W}_2[\boldsymbol{T}_0(\cdot)] + 2\int \{\varepsilon_0(\boldsymbol{x}) : \boldsymbol{L}_m + h(\boldsymbol{x})[\varepsilon_0(\boldsymbol{x}) + \boldsymbol{E} : [\boldsymbol{L}]\} : \varepsilon_1(\boldsymbol{x}) d^3\boldsymbol{x}; \tag{8.4}$$

$$\begin{aligned}\overline{W}_2[\boldsymbol{T}_0(\cdot)] = {} & V_a \int \varepsilon_0(\boldsymbol{x}) : [\boldsymbol{L}] : \varepsilon_0(\boldsymbol{x}) d^3\boldsymbol{x} \\ & - \int\int \varepsilon_0(\boldsymbol{x} - \boldsymbol{y}_1) : \boldsymbol{L}_m : \varepsilon_0(\boldsymbol{x} - \boldsymbol{y}_2) R_0(\boldsymbol{y}_1 - \boldsymbol{y}_2) d^3\boldsymbol{y}_1 d^3\boldsymbol{y}_2 \\ & - \int\int h(\boldsymbol{x} - \boldsymbol{y}_1) R_0(\boldsymbol{y}_1 - \boldsymbol{y}_2)[2\boldsymbol{E} + 2\varepsilon_0(\boldsymbol{x} - \boldsymbol{y}_1) + \varepsilon_0(\boldsymbol{x} - \boldsymbol{y}_2)] \\ & : [\boldsymbol{L}] : \varepsilon_0(\boldsymbol{x} - \boldsymbol{y}_2) d^3\boldsymbol{y}_1 d^3\boldsymbol{y}_2.\end{aligned} \tag{8.5}$$

When deriving (8.3) — (8.5), the well-known formulas for the moments of the random density field $\psi(\boldsymbol{x})$, correct to the order n^2, are used, namely

$$\langle\psi(\boldsymbol{y})\rangle = n, \qquad \langle\psi(\boldsymbol{y}_1)\psi(\boldsymbol{y}_2)\rangle = n\delta(\boldsymbol{y}_2-\boldsymbol{y}_1)+n^2 g_0(\boldsymbol{y}_1-\boldsymbol{y}_2)+o(n^2),$$

$$\langle\psi(\boldsymbol{y}_1)\psi(\boldsymbol{y}_2)\psi(\boldsymbol{y}_3)\rangle = n\delta(\boldsymbol{y}_2-\boldsymbol{y}_1)\delta(\boldsymbol{y}_3-\boldsymbol{y}_1) +3n^2\{\delta(\boldsymbol{y}_1-\boldsymbol{y}_2)g_0(\boldsymbol{y}_2-\boldsymbol{y}_3)\}_s+o(n^2). \tag{8.6}$$

Here g_0 is the zero-density limit of the two-point probability density function for the random set $\boldsymbol{x}_j$ of sphere centers, $\{\cdot\}_s$ denotes symmetrization with respect to all different combination of the indices in the braces, $R_0(\boldsymbol{y}) = 1 - g_0(\boldsymbol{y})$, cf. [4, 16], also $\varepsilon_i(\boldsymbol{x}) = \frac{1}{2}(\nabla \boldsymbol{T}_i + \boldsymbol{T}_i\nabla)$ for $i = 0, 1$, $[\boldsymbol{L}] = \boldsymbol{L}_f - \boldsymbol{L}_m$.

The minimizing kernel satisfies the equation $\delta W = 0$, so that we have in particular $\delta W_1[\boldsymbol{T}_0(\cdot)] = 0$, $\delta W_2[\boldsymbol{T}_0(\cdot), \boldsymbol{T}_1(\cdot)] = 0$. The first of these equations yields straightforwardly

$$\nabla\cdot\{[\boldsymbol{L}_m+[\boldsymbol{L}]h(\boldsymbol{x})][\boldsymbol{E}+\varepsilon_0(\boldsymbol{x})]\} = 0, \tag{8.7}$$

which is just the equation for the disturbance in the displacement field in an unbounded matrix of moduli $\boldsymbol{L}_m$, introduced by a single spherical inhomogeneity of moduli $\boldsymbol{L}_f$, when the strain at infinity is $\boldsymbol{E}$. The functional W_2 is then independent of $\boldsymbol{T}_1(\boldsymbol{x})$ (see (8.5), (8.7)):

$$W_2[\boldsymbol{T}_0(\cdot), \boldsymbol{T}_1(\cdot)] = \overline{W}_2[\boldsymbol{T}_0(\cdot)].$$

This means that for the bounds to be optimal to the order c^2 it suffices the zero-order coefficient $\boldsymbol{T}_0(\boldsymbol{x})$ in (8.1) to be proportional to the single-sphere disturbance field — the solution of eqn (8.7). (Obviously, this conclusion holds also for dispersions of identical and identically oriented inclusions, randomly and nonoverlappingly distributed in a matrix.) Since the first-order perturbation kernels in (6.3) and (6.4) in the isotropic case either coincide ($\Gamma_s(\boldsymbol{x})$) or coincide to the order $O(c)$ ($\Gamma_d(\boldsymbol{x})$), with the spherical and deviatoric parts, respectively, of the single-sphere disturbance field in the matrix material, we can claim that the BM- and MC-bounds are c^2-optimal. This fact implies, in turn, that the generalized MC-bound $\widetilde{\mu}^u_{MC}$ (cf. sec. 6) coincides to the order c^2 with the MC-bound μ^u_{MC}, given in (5.1a):

$$\widetilde{\mu}^u_{MC} = \mu^u_{MC} + o(c^2). \tag{8.7}$$

The reason is that when evaluating the bound $\widetilde{\mu}_{MC}$ we employ statistical information, given by the two- and three-point moments and thus it cannot be better than the optimal third-order bound. The latter, however, coincides to the order c^2 with the MC-bound $\widetilde{\mu}^u_{MC}$.

EVALUATION OF THE BERAN-MOLYNEUX AND THE McCOY BOUNDS TO ORDER c^2

In order to obtain the explicit forms of the BM- and the MC-bounds for a dispersion of spheres to the order c^2, it suffices to calculate the statistical parameters A and A_1 to the same order of accuracy.

To the order n^2, i.e. c^2, the three-point correlation function $i(\boldsymbol{z}, \boldsymbol{w})$ for the field $I(\boldsymbol{x})$, see (2.2), has the form

(9.1)
$$i(\boldsymbol{z}, \boldsymbol{w}) = n\int h(\boldsymbol{x})h(\boldsymbol{z}-\boldsymbol{x})h(\boldsymbol{w}-\boldsymbol{x})d^3\boldsymbol{x}$$
$$-n^2\int\int h(\boldsymbol{z}-\boldsymbol{x}_1)[h(\boldsymbol{w}-\boldsymbol{x}_1)h(\boldsymbol{x}_2)+h(\boldsymbol{w}-\boldsymbol{x}_2)h(\boldsymbol{x}_2)$$
$$+h(\boldsymbol{w}-\boldsymbol{x}_2)h(\boldsymbol{x}_1)]R_0(\boldsymbol{x}_1-\boldsymbol{x}_2)d^3\boldsymbol{x}_1d^3\boldsymbol{x}_2+o(n^2),$$

because $I'(\boldsymbol{x}) = \int h(\boldsymbol{x}-\boldsymbol{y})\psi'(\boldsymbol{x})d^3\boldsymbol{y}$. On inserting (9.1) into (4.5), we get

(9.2a)
$$A = (a_0 - a_1c)c + o(o^2),$$

where

(9.2b)
$$a_0 = \frac{1}{V_a}\int h(\boldsymbol{x})\nabla\nabla\varphi(\boldsymbol{x}) : \nabla\nabla\varphi(\boldsymbol{x})d^3\boldsymbol{x} = \frac{1}{3},$$

(9.2c)
$$a_1 = \frac{1}{V_a^2}\int F_0(\boldsymbol{x})\nabla\nabla\varphi(\boldsymbol{x}) : \nabla\nabla\varphi(\boldsymbol{x})d^3\boldsymbol{x}$$
$$+\frac{2}{V_a^2}\int R_0(\boldsymbol{x}_1-\boldsymbol{x}_2)h(\boldsymbol{x}_1)\nabla\nabla\varphi(\boldsymbol{x}_1) : \nabla\nabla\varphi(\boldsymbol{x}_2)d^3\boldsymbol{x}_1d^3\boldsymbol{x}_2 = \frac{5}{3} - m_2,$$

making use of the well-known properties of the Newtonian potential $\varphi(\boldsymbol{x})$ for a sphere. In (9.2c)

$$F_0(\boldsymbol{x}) = \int h(\boldsymbol{x}-\boldsymbol{y})R_0(\boldsymbol{y})d^3\boldsymbol{y},$$

and

(9.3)
$$m_2 = 2\int_2^\infty \frac{\lambda^2}{(\lambda^2-1)^3}g_0(\lambda a)d\lambda, \qquad \lambda = r/a,$$

is the statistical parameter for the dispersion, which appears in the Beran bounds on the effective conductivity of the dispersion.

The relations (9.2) can be summarized as follows

$$A = \frac{1}{3}[1-(5-3m_2)c]c + o(c^2), \tag{9.4}$$

and thus the first two coefficients in the virial expansion of the statistical parameter A, defined in (4.5), are calculated for the dispersion under study.

It is worth mentioning that if we insert (9.4) into (4.11), we shall obtain

$$\xi_2 = \frac{3}{2}m_2 c + o(c), \tag{9.5}$$

so that the statistical parameter (9.3) appears to be proportional to the coefficient of the leading c-term in the virial expansion of the Milton parameter (4.8) for the dispersion.

The c^2-evaluation of the statistical parameter A_1, defined in (5.4), is similar. We have

$$A_1 = (b_0 - b_1 c)c + o(c^2). \tag{9.6a}$$

On introducing (9.1) into (5.4), we get

$$b_0 = \frac{1}{4V_a}\int h(\boldsymbol{x})\nabla\nabla\nabla\chi(\boldsymbol{x}) \bullet \nabla\nabla\nabla\chi(\boldsymbol{x})d^3\boldsymbol{x} = \frac{1}{5}, \tag{9.6b}$$

availing of the well-known properties of the biharmonic potential $\lambda(\boldsymbol{x})$ for a sphere. In turn, after simple algebra, we find

$$b_1 = \frac{1}{4V_a^2}\left\{\int F_0(\boldsymbol{x})\nabla\nabla\nabla\chi(\boldsymbol{x}) \bullet \nabla\nabla\nabla\chi(\boldsymbol{x})d^3\boldsymbol{x}\right.$$
$$\left. + \frac{1}{2\pi}\int F_0(\boldsymbol{x}_1-\boldsymbol{x}_2)h(\boldsymbol{x}_1)\nabla\nabla\nabla\nabla|\boldsymbol{x}_2| \bullet \nabla\nabla\nabla\chi(\boldsymbol{x}_1)d^3\boldsymbol{x}_1 d^3\boldsymbol{x}_2\right\} = \frac{7}{5} - m_2', \tag{9.6c}$$

where

$$m_2' = \frac{9}{2\pi}\int_{2a}^{\infty}\frac{F_0'(r)}{r^3}\left\{\frac{a^4}{5r^4} - \frac{2a^2}{5r^2} + \frac{1}{3}\right\}dr \tag{9.7}$$

is a new statistical parameter for the dispersion, similar to m_2.

Since the function $F_0'(\boldsymbol{x})$ depends linearly on $g_0(r)$, the parameter m_2' will be also a linear functional of $g_0(r)$. Its explicit form could be derived by using the method of [4, sec. 11], by means of which the relation (9.3) was reached. The final result reads

$$(9.8) \quad m_2' = \frac{6}{5}\int_2^\infty \frac{\lambda^2(5\lambda^8 - 30\lambda^6 + 51\lambda^4 - 4\lambda^2 + 2)}{(\lambda^2-1)^7} g_0(\lambda a)d\lambda, \qquad \lambda = r/a.$$

Note that in the so-called well-stirred case, for which $g_0(r) = g(r) = 1$ if $r \geqq 2a$, and vanishes otherwise, we have

$$(9.9) \qquad m_2 = \frac{5}{18} - \frac{1}{8}\ln 3 \approx 0.14045, \qquad m_2' \approx 0.25016.$$

The relations (9.6) can be now summarized as follows

$$(9.10) \qquad A_1 = \frac{1}{5}[1-(7-5m_2')c]c + o(c^2),$$

which is the counterpart of (5.3). In turn, for the second Milton parameter η_2, defined in (5.5), we obtain

$$(9.11) \qquad \eta_2 = \frac{6}{5}M_2 c + o(c), \qquad M_2 = 4m_2' - 3m_2,$$

as a consequence of (5.6) and (9.10).

Note that the inequality (5.8) together with (9.5) and (9.11) yields

$$(9.12) \qquad m_2' \geqq \frac{6}{7}m_2.$$

A simple inspection of the kernels in the integral representations (9.3) and (9.11) for the statistical parameters m_2 and m_2' shows that the stronger inequality

$$(9.13) \qquad m_2' \geqq \frac{6}{5}m_2$$

holds for dispersions of nonoverlapping spheres, since $g_0(r)$ is nonnegative. Moreover, the inequality (9.13) is the best in the sense that the constant 6/5 cannot be made bigger. This fact implies that the equality in (9.12) is never realizable for dispersions, so that the equality in the Phan-Thien-Milton inequality (5.8) is never attainable whatever be the random distribution of the spheres.

Let

$$\frac{k^*}{k_m} = 1 + a_{1k}c + a_{2k}c^2 + \cdots \tag{9.14}$$

be the virial expansion for the effective bulk modulus of the dispersion. Making use of eqns (4.4), (4.5) and (9.3), we get as a consequence of the BM-bounds (4.2)

$$a_{1k} = \frac{[k]}{k_m + \alpha_m[k]}, \qquad \alpha_m = \frac{3k_m}{3k_m + 4\mu_m}, \tag{9.15}$$

so that the upper and lower BM-bounds coincide to the order c and for the c^2-coefficient the following inequalities hold

$$a_{2k}^l \leqq a_{2k} \leqq a_{2k}^u, \tag{9.16a}$$

$$a_{2k}^l = \alpha_m a_{1k}^2 \left\{ 1 + 2\alpha_m \frac{\mu_m[\mu]}{\mu_f k_m} m_2 \right\}, \tag{9.16b}$$

$$a_{2k}^u = \alpha_m a_{1k}^2 \left\{ 1 + 2\alpha_m \frac{[\mu]}{k_m} m_2 \right\},$$

where m_2 is the statistical parameter (9.3).

Let

$$\frac{\mu^*}{\mu_m} = 1 + a_{1\mu}c + a_{2\mu}c^2 + \cdots \tag{9.17}$$

be the virial expansion for the effective shear modulus of the dispersion. Making use of eqns (5.2), (5.3) and (9.10), we get as a consequence of the MC-bounds (5.1)

$$a_{1\mu} = \frac{[\mu]}{\mu_m + \beta_m[\mu]}, \qquad \beta_m = \frac{6(k_m + 2\mu_m)}{5(3k_m + 4\mu_m)}, \tag{9.18}$$

so that the upper and lower MC-bounds, similarly to the BM-ones, coincide to the order c. For the c^2-coefficient $a_{2\mu}$ we get the inequalities

$$a_{2\mu}^l \leqq a_{2\mu} \leqq a_{2\mu}^u, \tag{9.19a}$$

where

$$a_{2\mu}^l = \beta_m a_{1\mu}^2 \left\{ 1 + f(\nu_m) \frac{k_m}{k_f} \frac{[k]}{\mu_m} m_2 + \chi \frac{[\mu]}{\mu_f} \right\}, \tag{9.19b}$$

$$a_{2\mu}^l = \alpha_m a_{1k}^2 \left\{ 1 + f(\nu_m) \frac{[k]}{\mu_m} m_2 + \chi \frac{[\mu]}{\mu_m} \right\}$$

with the notations

$$f(\nu_m) = \frac{3(1-2\nu_m)^2}{4(1-\nu_m)(4-5\nu_m)}, \tag{9.19c}$$

$$\chi = \chi(\nu_m,\ m_2,\ m_2') = \frac{3m_2' + (7\nu^2 - 10\nu_m + 1)m_2}{4(1-\nu_m)(4-5\nu_m)},$$

where ν_m is the Poisson ratio of the matrix.

The c^2-bounds (9.16) and (9.19) on the effective elastic moduli of a random dispersion of spheres have been reported in [17], using slightly different notations.

In the case of an incompressible matrix, $\nu_m = 0.5$, the bounds (9.19) are significantly simplified

$$a_{2\mu}^l = \frac{2}{5}\left(\frac{5[\mu]}{3\mu_m + 2\mu_f}\right)^2 \left(1 + \frac{[\mu]}{2\mu_f}M_2\right), \tag{9.20}$$

$$a_{2\mu}^u = \frac{2}{5}\left(\frac{5[\mu]}{3\mu_m + 2\mu_f}\right)^2 \left(1 + \frac{[\mu]}{2\mu_m}M_2\right).$$

The c^2-bounds on μ^* in this case depend on a single statistical parameter $M_2 = 4m_2' - 3m_2$ — the same that appeared in the c-term of the virial expansion (9.11) of the second Milton parameter η_2. The situation is thus fully similar to that for the c^2-bounds on the effective conductivity, considered in detail in [4], with the only difference that a new statistical parameter appears.

Let us assume that the spheres are rigid, so that $\mu_f = \infty$ as well. The upper bound (9.19) then degenerates since $M_2 > 0$, cf. (9.13), and thus

$$\frac{5}{2}\left(1 + \frac{1}{2}M_2\right) \leqq a_{2\mu} < \infty. \tag{9.21}$$

We can conclude from (9.21) that the value 2.5 for $a_{2\mu}$ is never attainable for dispersions with incompressible constituents. For a well-stirred dispersion we get, moreover, that $3.2241 \leqq a_{2\mu} < \infty$, in virtue of (9.9).

SOME IMPLICATIONS OF THE c^2-BOUNDS

The foregoing c^2-bounds (9.16) and (9.19) are third-order also in the sense that they coincide for a weakly inhomogeneous dispersion to the order $([k]/k_m)^p([\mu]/\mu_m)^q$, $p + q = 3$. For instance, for the bounds (9.16) on the bulk modulus we have

$$a_{2k} = \alpha_m a_{1k}^2 \left\{1 + 2\alpha_m \frac{[\mu]}{k_m} m_2\right\} + o\left(\left(\frac{[k]}{k_m}\right)^2 \frac{[\mu]}{\mu_m}\right). \tag{10.1}$$

In turn, the bounds (9.19) coincide to the order $([\mu]/\mu_m)^3$. These facts allow to check on the applicability of the known theories in mechanics of composite media for the case of random dispersions of spheres, making use of the method, proposed in [4, p. II], when studying effective scalar conductivity. The basic idea of the method is to consider the formulae for the effective properties, predicted by some of these theories, to the order c^2 and in the limiting case of a weakly inhomogeneous medium, and to compare the results with the relations of the type of (10.1). In this way the values of the statistical parameters m_2 and m_2', which correspond to the theory under examination, could be obtained. To illustrate the method we shall consider here only two examples: the well-known self-consistent theory of elastic composites, due to Hill [18] and Budiansky [19], and the approximate c^2-theory of random elastic dispersion, due to Willis and Acton [20].

10.1. The self-consistent theory of random dispersions is based on the assumption that each sphere is embedded in an unbounded matrix material that possesses the unknown effective moduli k^*, μ^*, see for more details [19, 20]. This assumption eventually yields the following system for the moduli k^* and μ^*:

$$k^* = k_m + \frac{[k]k^*c}{k^* + \alpha^*(k_f - k^*)}, \tag{10.2a}$$

$$\mu^* = \mu_m + \frac{[\mu]\mu^*c}{\mu^* + \beta^*(\mu_f - \mu^*)},$$

where

$$\alpha^* = \frac{3k^*}{3k^* + 4\mu^*}, \qquad \beta^* = \frac{6(k^* + 2\mu^*)}{5(3k^* + 4\mu^*)}. \tag{10.2b}$$

Let

$$\frac{k^*}{k_m} = 1 + a_{1k}c + a_{2k}c^2 + \cdots, \tag{10.3}$$

$$\frac{\mu^*}{\mu_m} = 1 + a_{1\mu}c + a_{2\mu}c^2 + \cdots$$

be the virial expansions of the solution $k^* = k^*(c)$, $\mu^* = \mu^*(c)$ of the system (10.2) at $c \ll 1$. It is easily seen that the c-coefficients a_{1k} and μ_{1k} in (10.3) coincide with those, given in (9.14) and (9.17) respectively. In the case of incompressible spheres, $k_f = \infty$, we get the following expression for a_{2k}:

$$a_{2k} = \alpha_m a_{1k}^2 \left\{ 1 + \frac{4\mu_m[\mu]}{3\alpha_m k_m(\mu_m + \beta_m[\mu])} \right\} \tag{10.4}$$

which meets the bounds (9.16) only if

$$(10.5) \qquad m_2 = m_2^{sc} = \frac{2}{3}.$$

This value of m_2 may be also obtained by comparing (10.1) and (10.4) in the weakly inhomogeneous case $[\mu]/\mu_m \ll 1$. It is noteworthy that the same value (10.5) for m_2 has been found in [4] when analyzing the applicability of the self-consistent theory of effective scalar conductivity for random dispersions.

Suppose that the matrix is also incompressible, $k_m = \infty$, so that $k^* = \infty$ as well. The second equation (10.2a) then simplifies and one easily obtains

$$(10.6) \qquad a_{2\mu} = \frac{2}{5}a_{1\mu}^2\left(1 + \frac{3[\mu]}{3\mu_m + 2\mu_f}\right),$$
$$= \frac{2}{5}a_{1\mu}^2\left(1 + \frac{3[\mu]}{5\mu_m}\right) + o(([\mu]/\mu_m)^3).$$

Having compared (10.6) and (9.18), and taking into account (10.5), we get the value of the second statistical parameter m_2', corresponding to the self-consistent theory, to be

$$(10.7) \qquad m_2' = \frac{4}{5}.$$

However, the values (10.5) and (10.7) of the parameters m_2 and m_2' respectively do not satisfy the inequality (9.13), which should hold for any dispersion of nonoverlapping spheres. We therefore conclude that the predictions of the self-consistent theory, eqns (10.2), are not applicable in general to such dispersions even to the order c^2, whatever be the random distribution of the spheres.

10.2. The approximate c^2-theory of Willis and Acton. In the theory of Willis and Acton [20] the effective elastic moduli of the dispersion are expressed in terms of the solution of an integral equation for the so-called p o l a r i z a t i o n f i e l d. It is proposed that the equation be solved by iterations and the first two such iterations are analytically found, yielding approximate formulas for the c^2-coefficients a_{1k} and $a_{1\mu}$ of the virial expansions (9.14) and (9.17) of the effective moduli k^* and μ^* respectively.

The formula for a_{2k} of the said authors, in our notations, reads (cf. [20], eqn (5.20)):

$$(10.8) \qquad a_{2k} = \alpha_m a_{1k}^2\left\{1 + \frac{4}{5}\Lambda\alpha_m\frac{[\mu]}{k_m}\frac{\mu_m}{\mu_m + \beta_m[\mu]}\right\},$$

where

(10.9)
$$\Lambda = 3\int_2^{\infty} \frac{g_0(\lambda a)}{\lambda^4} d\lambda$$

is the statistical parameter, introduced in [20, eqn (5.18)] and denoted there by λ. (In the well-stirred case $\Lambda = 1/8$.) The expression (10.8) meets the bounds (9.16) only if

(10.10)
$$m_2 = \frac{2}{3}\Lambda.$$

A simple inspection of the kernels in the integral representations (9.3) and (10.9) for the parameters m_2 and Λ, respectively, shows that

(10.11)
$$m_2 > \frac{2}{3}\Lambda,$$

since $g_0(r) > 0$. This means that the c^2- approximation (10.8) for a_{2k} violates the bounds (9.16) whatever be the function $g_0(r)$.

For incompressible constituents, $k_m = k_f = \infty$, the approximate formula of Willis and Acton for a_{2k} is (cf. [20], eqn (6.1)):

(10.12)
$$a_{2k} = \frac{2}{5}a_{1\mu}^2\left\{1 + 15\Lambda\frac{[\mu]}{3\mu_m + 2\mu_f}\right\}, \qquad a_{1\mu} = \frac{5[\mu]}{3\mu_m + 2\mu_f}.$$

When compared to (9.20), eqn (10.12) yields

(10.13)
$$6\Lambda = 4m_2' - 3m_2,$$

which is violated for the well-stirred dispersion, cf. (9.9). Unlike the case of bulk modulus there exist, however, random constitutions, i.e. functions $g_0(r)$, for which (10.13) holds.

CONCLUDING REMARKS

The method of truncated functional series [4] has been systematically applied in this paper, in order to investigate certain third-order bounds on the effective elastic properties of two-phase random media, i.e. bounds that employ statistical information, given by the two- and three-point correlation functions. In this way we were, first, able to unify the existing bounding procedures, due to Beran and Molyneux and McCoy, as certain Ritz-type procedures, corresponding to the choice of the respective perturbation kernels in the one-tuple term of the truncated functional series. Second, and more important, we were led to the problem of optimality of the bounds, due to the mentioned authors, in the sense whether the bounds are the most respective ones under the statistical information, used in their evaluation. The answer appears negative even for the classical example of a random dispersion of nonoverlapping spheres. However, the bounds in the latter case are optimal to the order c^2, similarly to the scalar conductivity case. The explicit evaluation of the said bounds to the order c^2 leads to the appearance of two statistical parameters, which linearly depend on the zero-density limit of the radial distribution function for the random set of sphere centers. The parameters are closely related to the coefficients of the leading c-terms of the Milton parameters ζ_2 and η_2 for the dispersion. This fact indicates once more the importance of the Milton parameters in the theory of two-phase random media. Similarly to [4], the obtained c^2-bounds allow to check on the applicability of certain heuristic theories in elasticity of composite materials for random dispersions of spheres. The most curious result of such a check here is that the well-known self-consistent theory, due to Hill and Budiansky, is not applicable to random dispersion even to the order c^2, because its predictions violate the respective bounds whatever be the random distribution of the spheres.

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