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VARIATIONAL TREATMENT OF AN ABSORPTION PROBLEM IN RANDOM HETEROGENEOUS MEDIA

K. Z. MARKOV[1]* and M. K. KOLEV[2]

[1] Istanbul Technical University, Istanbul, Turkey

[2] University of Sofia, Sofia, Bulgaria

ABSTRACT—The paper is devoted to the steady-state problem of absorption of a diffusing species in a random heterogeneous medium. The variational principles of classic type, using ensemble averaging, are first discussed and then used for derivation of variational estimates on the effective absorption coefficient (sink strength) of the medium. The estimates are three-point, i.e., they employ statistical information, contained in the ℓ-point correlation functions for the medium up to $\ell = 3$, and could be viewed as counterparts of the well-known Beran's bounds in the scalar conductivity problem. Moreover, the bounds are third-order in the weakly-inhomogeneous case. Explicit results are obtained for Miller's cellular media which indicate that the bounds remain useful even when the absorption capabilities of the constituents differ one hundred times.

1. INTRODUCTION

The problem of predicting the macroscopic properties of heterogeneous solids of random internal constitution, making use of adequate microstructure information, is a classic one and the respective literature is enormous. We shall only point out the surveys [1,2] of the papers most relevant to the spirit of this study. As is well seen from these surveys, the basic attention is paid to transport properties (say, heat conduction, elasticity, etc.) in which the randomly fluctuating coefficients enter the divergence part of the respective differential operator. In the simplest scalar case this is the problem

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

in which $\theta(\mathbf{x})$ could be treated as the random temperature field; then $\kappa(\mathbf{x})$ is the given conductivity field and $\mathbf{G}$—the prescribed macroscopic gradient of the temperature. Hereafter $\langle\cdot\rangle$ denotes ensemble averaging. The solution of this, as well as of similar random problems, is understood in statistical sense. This means, let us recall [3], that one is to find all multipoint correlations of the random field $\theta(\mathbf{x})$, and all joint correlations of the

* On leave from Faculty of Mathematics and Informatics, University of Sofia, 5 blvd J. Bourchier, 1164 Sofia, Bulgaria

fields $\theta(\mathbf{x})$ and $\kappa(\mathbf{x})$, provided that all needed correlations of $\kappa(\mathbf{x})$ are known. In particular, one is to find the one-point correlation (the mean flux), which is proportional to the macrogradient $\mathbf{G}$, i.e., $\langle\kappa(\mathbf{x})\nabla\theta(\mathbf{x})\rangle = \kappa^*\mathbf{G}$. The constant κ^* is the effective conductivity of the medium, which describes its macroscopic behaviour.

Much smaller attention is paid in the literature to the case when the equation under study is of the type of Helmholtz and the randomly fluctuating coefficient multiplies the unknown function, namely

$$\Delta\varphi(\mathbf{x}) - k^2(\mathbf{x})\varphi(\mathbf{x}) + K = 0. \tag{1.1}$$

Under some simplifying assumptions (constant diffusion coefficient), this equation governs, e.g., the steady-state concentration $\varphi(\mathbf{x})$ of a diffusing species (say, irradiation defects) in a random absorbing (lossy) medium; the species is generated at the constant rate K (for instance, due to radiation) and it is absorbed throughout the medium with an absorption coefficient $k^2(\mathbf{x})$, see [4] for more details and references, and also [5] for the respective physical background. The absorption coefficient $k^2(\mathbf{x})$ is a known random field, assumed positive and statistically homogeneous and isotropic.

Let us average eqn (1.1)

$$\Delta\langle\varphi(\mathbf{x})\rangle - \langle k^2(\mathbf{x})\varphi(\mathbf{x})\rangle + K = 0. \tag{1.2}$$

Due to the statistical homogeneity $\langle\varphi(\mathbf{x})\rangle$ is a constant which equals the mean (macroscopic) value of the defect concentration resulting from the steady-state equilibrium of the two processes taking place in the medium, namely, generation and absorption of defects. Thus

$$\langle k^2(\mathbf{x})\varphi(\mathbf{x})\rangle = K. \tag{1.3}$$

We next define the effective absorption coefficient (sink strength), k^{*2}, of the medium by means of the relation

$$\langle k^2(\mathbf{x})\varphi(\mathbf{x})\rangle = k^{*2}\langle\varphi(\mathbf{x})\rangle, \tag{1.4}$$

which means that we perform a "homogenization" of the basic equation (1.1), i.e., we replace the given random medium with a homogeneous ("effective") one which, from a macroscopical point of view, absorbs defects at the same rate as the given micro-inhomogeneous one. From (1.3) and (1.4), we have

$$k^{*2} = \frac{K}{\langle\varphi(\mathbf{x})\rangle}. \tag{1.5}$$

Thus the evaluation of k^{*2} needs knowledge of the one-point joint moment of the random fields $k^2(\mathbf{x})$ and $\varphi(\mathbf{x})$ or of the mean value (the one-point moment) of the field $\varphi(\mathbf{x})$, see (1.4) and (1.5). These moments are part of the full statistical solution of the problem (1.1), in the sense described above. To the best of the authors' knowledge, there is no proof of the existence and uniqueness theorem for the random equation (1.1), unlike the classical problem, concerning stationary heat propagation or elastic straining of heterogeneous media. It is to be noted also that there is no additional condition for eqn (1.1) (of the type

of prescribed value of macroscopic gradient), but rather $\langle\varphi(\mathbf{x})\rangle$ is to be determined in the course of the solution, among the rest of the multipoint correlation functions of the latter.

The only detailed study of the random equation (1.1) from the point of view of its homogenization, as far as we know, is due to Talbot and Willis [4], who considered certain approximate schemes for evaluating k^{*2} and then derived bounds on this quantity upon introducing a variational principle of Hashin-Shtrikman's type. It is noted that in the literature much more attention is paid to the "degenerate" case when the medium is two-phase (in particular, a random dispersion) for which one phase is a perfect absorber ($k^2 = \infty$) while the other does not absorb at all ($k^2 = 0$). In this case a number of variational estimates have been proposed, see, e.g., [6] and the references therein.

The aim of the present study is a systematic use of variational principles of classical type in connection with the problem (1.1) and its homogenization. We first derive the primal and dual principles in which ensemble averaging is used so that there is no need to take care for the respective boundary conditions. The primal principle allows to sketch in passing a simple proof of uniqueness and existence theorem for the problem (1.1) (Section 2). In Section 3 we discuss the variational procedure that uses truncated functional series, generated by the random absorption coefficient field, in the simplest nontrivial case, when it yields optimal three-point bounds on k^{*2}. The procedure is similar to that introduced and elaborated by one of the authors [7] in the scalar conductivity case. In Section 4 we simplify the procedure, using the respective Green functions as kernels and get as a result the counterparts of the well-known Beran bounds [8]. The bounds are shown to be third-order for a two-phase medium, and this seems to be the central result of the paper; they are explicitly evaluated in Section 5 for Miller's cellular material. We notice there in passing that Beran's type bounds are not optimal even for this comparatively simple microstructure. The numerical results clearly show that the bounds remain tolerably close even when the absorption coefficients of the constituents differ one hundred times.

2. VARIATIONAL PRINCIPLES IN THE ABSORPTION PROBLEM

2.1. *The primal principle*

Consider the functional

$$I[\varphi(\cdot)] = \frac{1}{2}\Big\langle |\nabla\varphi(\mathbf{x})|^2 + k^2(\mathbf{x})\varphi^2(\mathbf{x}) - 2K\varphi(\mathbf{x})\Big\rangle, \tag{2.1}$$

defined over the class of all statistically homogeneous random fields $\varphi(\mathbf{x})$, $\mathbf{x} \in R^3$, for which $I[\varphi(\cdot)]$ is finite. Eqn (1.1) is the Euler-Lagrange equation for the variational problem

$$I \to \inf . \tag{2.2}$$

Moreover, the minimum value, $I_{\min}$, of I is

$$I_{\min} = -\frac{K^2}{2k^{*2}}, \tag{2.3}$$

and it is attained on the true field $\varphi(\mathbf{x})$, i.e., on the solution of eqn (1.1).

Indeed,* the first variation of the functional I is

$$\delta I[\delta\varphi(\cdot)] = \Big\langle \nabla\varphi(\mathbf{x}) \cdot \nabla\delta\varphi(\mathbf{x}) + k^2(\mathbf{x})\varphi(\mathbf{x})\delta\varphi(\mathbf{x}) - K\delta\varphi(\mathbf{x}) \Big\rangle.$$

Note next that

$$\langle \nabla\varphi \cdot \nabla\delta\varphi \rangle = \langle \nabla \cdot (\delta\varphi\nabla\varphi) \rangle - \langle \delta\varphi\Delta\varphi \rangle;$$

but $\langle \nabla \cdot (\delta\varphi\nabla\varphi) \rangle = 0$, since differentiation commutes with ensemble averaging and $\langle \delta\varphi\nabla\varphi \rangle$ = const due to the statistical homogeneity assumed. (A similar idea was employed by Beran [3, p. 128].) Thus

$$\delta I[\delta\varphi(\cdot)] = -\Big\langle \delta\varphi(\mathbf{x})[\Delta\varphi(\mathbf{x}) - k^2(\mathbf{x})\varphi(\mathbf{x}) + K] \Big\rangle = 0, \quad \forall\, \delta\varphi(\mathbf{x}),$$

which implies that eqn (1.1) is indeed the Euler-Lagrange equation for the functional I. A direct check shows that the second variation of I is strongly positive, since $k^2(\mathbf{x}) > 0$, and therefore I attains its minimum value on the true field $\varphi(\mathbf{x})$.

To calculate $I_{\min}$ we notice that for any statistically homogeneous field $\varphi(\mathbf{x})$ we have

$$\langle |\nabla\varphi|^2 \rangle = \langle \nabla\varphi \cdot \nabla\varphi \rangle = \nabla \cdot \langle \varphi\nabla\varphi \rangle - \langle \varphi\Delta\varphi \rangle = -\langle \varphi\Delta\varphi \rangle,$$

since $\nabla \cdot \langle \varphi\nabla\varphi \rangle = 0$, due to the assumed homogeneity. Thus

$$I[\varphi(\cdot)] = -\frac{1}{2}\Big\langle \varphi(\mathbf{x}) \left[\Delta\varphi(\mathbf{x}) - k^2(\mathbf{x})\varphi(\mathbf{x}) + 2K\right] \Big\rangle.$$

The minimum value is attained when $\varphi(\mathbf{x})$ solves eqn (1.1), and the expression in the square brackets then equals K:

$$I_{\min} = -\frac{1}{2}K\langle \varphi(\mathbf{x}) \rangle.$$

Now (2.3) immediately follows, if (1.5) is recalled.

2.2. *A consequence of the variational principle*

As a consequence of the principle (2.2) we shall sketch now a proof of the existence and uniqueness theorem for the problem (1.1).

Let $(\Omega, \mathcal{F}, P)$ be a probabilistic space. Assume that for every realization of the field $k^2(\mathbf{x})$ the following inequalities hold

$$0 < M_1 \le k^2(\mathbf{x}) \le M_2 < \infty. \tag{2.4}$$

* This statement is obvious, if the averaging used were the volume one; here ensemble averaging is employed so that the proof requires a bit more effort.

We introduce, after Golden and Papanicolaou [9], the Hilbert space $\mathcal{H}$ of all statistically homogeneous random fields with the scalar product

$$\varphi \cdot \psi = \langle \varphi(\mathbf{x})\psi(\mathbf{x}) \rangle + \langle \nabla\varphi(\mathbf{x}) \cdot \nabla\psi(\mathbf{x}) \rangle$$

and, consequently, with the norm

$$\|\varphi\| = \sqrt{\langle \varphi^2(\mathbf{x}) \rangle + \langle |\nabla\varphi(\mathbf{x})|^2 \rangle}. \tag{2.5}$$

The functional I, due to the positiveness of $k^2(\mathbf{x})$, is strongly convex.

Note the obvious inequality

$$\langle \varphi(\mathbf{x}) \rangle^2 \le \langle \varphi^2(\mathbf{x}) \rangle, \tag{2.6}$$

which follows immediately from the definition of the ensemble averaging as an integral with respect to the probability measure $P(d\omega)$:

$$\langle \varphi(\mathbf{x}) \rangle = \int_\Omega \varphi(\mathbf{x};\omega) P(d\omega).$$

The continuity of the functional (2.1) with respect to the norm (2.5) is now an obvious consequence of eqns (2.4) and (2.6). Another obvious consequence of these equations is the chain of inequalities

$$\begin{aligned} I[\varphi(\cdot)] &= \frac{1}{2}\Big\langle |\nabla\varphi(\mathbf{x})|^2 + k^2(\mathbf{x})\varphi^2(\mathbf{x}) - 2K\varphi(\mathbf{x}) \Big\rangle \\ &\ge \langle |\nabla\varphi(\mathbf{x})|^2 \rangle + M_1 \frac{1}{2}\Big(\langle \varphi^2(\mathbf{x}) \rangle - 2K\sqrt{\langle \varphi^2(\mathbf{x}) \rangle} \Big) \\ &\ge \frac{1}{2}\Big(m\|\varphi\|^2 - 2K\|\varphi\| \Big), \qquad m = \min(1, M_1) > 0, \end{aligned}$$

which implies that the functional I is coercitive, i.e.,

$$I[\varphi(\cdot)] \to \infty \quad \text{at} \quad \|\varphi\| \to \infty.$$

It remains now to notice that the functional I, due to the positiveness of $k^2(\mathbf{x})$, is strongly convex and to recall the well-known result from the convex analysis [10] which states that a continuous, strongly convex and coercitive functional, defined over a close and convex set (the whole space $\mathcal{H}$ in our case) possesses a minimizing element which is unique. This proves the uniqueness and existence theorem for the absorption problem (1.1) under study.

2.3. *The dual variational principle*

Let us rewrite the functional (2.1) in the form

$$I[\mathbf{e}(\cdot), \varphi(\cdot)] = \Big\langle W_A(\mathbf{e}(\mathbf{x}), \varphi(\mathbf{x})) \Big\rangle,$$

where

$$W_A(\mathbf{e}(\mathbf{x}), \varphi(\mathbf{x})) = \frac{1}{2}\left(\mathbf{e}^2(\mathbf{x}) + k^2(\mathbf{x})\varphi^2(\mathbf{x}) - 2K\varphi(\mathbf{x})\right).$$

The functional I in these notations is defined over the class of admissible pairs

$$\mathcal{A} = \Big\{(\mathbf{e}, \varphi) \;\Big|\; \mathbf{e} = \nabla\varphi\Big\}. \tag{2.7}$$

Following the well known scheme of the variational calculus [10,11], we introduce the Fenchel-Young transform of the function W_A

$$\begin{aligned} W_A^*(\mathbf{e}^*, \varphi^*) &= \sup_{(\mathbf{e},\varphi)\in\mathcal{A}} \Big\{\mathbf{e}\cdot\mathbf{e}^* + \varphi\varphi^* - W_A(\mathbf{e}, \varphi)\Big\} \\ &= \sup_{(\mathbf{e},\varphi)\in\mathcal{A}} \Big\{\mathbf{e}\cdot\mathbf{e}^* + \varphi\varphi^* - \frac{1}{2}\mathbf{e}(\mathbf{x})^2 - \frac{1}{2}k^2(\mathbf{x})\varphi^2(\mathbf{x}) + K\varphi(\mathbf{x})\Big\}. \end{aligned}$$

The supremum is attained at

$$\mathbf{e} = \mathbf{e}^*, \quad \varphi = \alpha^2(\mathbf{x})(\varphi^* + K), \tag{2.8}$$

so that

$$W_A^*(\mathbf{e}^*, \varphi^*) = \frac{1}{2}\left(\mathbf{e}^{*2} + \alpha^2(\mathbf{x})(\varphi^* + K)^2\right),$$

where $\alpha^2(\mathbf{x}) = 1/k^2(\mathbf{x})$ is the "compliance" field.

Due to (2.4) the function $W_A(\mathbf{e}, \varphi)$ is convex and therefore $W_A^{**} = W_A$ which means that

$$W_A(\mathbf{e}, \varphi) = \sup_{(\mathbf{e}^*,\varphi^*)} \left\{\mathbf{e}\cdot\mathbf{e}^* + \varphi\varphi^* - W_A^*(\mathbf{e}^*, \varphi^*)\right\}.$$

Thus the primal principle can be recast as

$$\begin{aligned} I_{\min} &= \inf \langle W_A(\mathbf{e}, \varphi)\rangle \\ &= \inf_{(\mathbf{e},\varphi)\in\mathcal{A}} \sup_{(\mathbf{e}^*,\varphi^*)} \Big\langle \mathbf{e}\cdot\mathbf{e}^* + \varphi\varphi^* - W_A^*(\mathbf{e}^*, \varphi^*)\Big\rangle \\ &= \sup_{(\mathbf{e}^*,\varphi^*)} J[\mathbf{e}^*(\cdot), \varphi^*(\cdot)], \end{aligned} \tag{2.9}$$

where

$$J[\mathbf{e}^*(\cdot), \varphi^*(\cdot)] = \inf_{(\mathbf{e},\varphi)\in\mathcal{A}} \Big\langle \mathbf{e}\cdot\mathbf{e}^* + \varphi\varphi^* - W_A^*(\mathbf{e}^*, \varphi^*)\Big\rangle.$$

We have not put until now any restrictions on the dual variables $(\mathbf{e}^*, \varphi^*)$, unlike those that enter the primal principle, see (2.7). Such restrictions should be imposed, however, in order to get a finite value of the functional J. We first note that

$$\langle \mathbf{e}\cdot\mathbf{e}^*\rangle = \langle \mathbf{e}^*\cdot\nabla\varphi\rangle = \langle \nabla\cdot(\varphi\mathbf{e}^*)\rangle - \langle \varphi\nabla\cdot\mathbf{e}^*\rangle = -\langle \varphi\nabla\cdot\mathbf{e}^*\rangle,$$

since $\langle \nabla \cdot (\varphi \mathbf{e}^*) \rangle = \nabla \cdot \langle (\varphi \mathbf{e}^*) \rangle = 0$ due to the statistical homogeneity. Thus

$$J[\mathbf{e}^*(\cdot), \varphi^*(\cdot)] = \inf_{\varphi} \left\langle \varphi[\varphi^* - \nabla \cdot \mathbf{e}^*] - W_A^*(\mathbf{e}^*, \varphi^*) \right\rangle$$

and a finite value of the functional J is obtained provided

$$\nabla \cdot \mathbf{e}^* = \varphi^*.$$

The equality (2.9) allows us to formulate the needed dual principle, namely

$$\begin{gathered} J \to \max, \\ J[\mathbf{e}^*(\cdot), \varphi^*(\cdot)] = -\left\langle W_B\big(\mathbf{e}^*(\mathbf{x}), \varphi^*(\mathbf{x})\big) \right\rangle, \\ W_B(\mathbf{e}^*(\mathbf{x}), \varphi^*(\mathbf{x}) = \frac{1}{2}\left(\mathbf{e}^{*2} + \alpha^2(\mathbf{x})(\varphi^* + K)^2\right). \end{gathered} \tag{2.10}$$

Moreover,

$$J_{\max} = \sup_{(\mathbf{e}^*, \varphi^*) \in \mathcal{A}^*} J[\mathbf{e}^*(\cdot), \varphi^*(\cdot)] = -\frac{K^2}{2k^{*2}}, \tag{2.11}$$

where

$$\mathcal{A}^* = \left\{ (\mathbf{e}^*, \varphi^*) \;\middle|\; \nabla \cdot \mathbf{e}^* = \varphi^* \right\}$$

and thus $I_{\min} = J_{\max}$ as it should be. The proof of the latter fact needs of course a bit more attention, since it follows from the assumption $\inf \sup = \sup \inf$, tacitly made in (2.9). For the simple functionals under study it is, however, easily verified, so we skip the details.

2.4. *Variational principles using volume averaging*

Let us denote by $k^2(\mathbf{x}; \omega)$ one of the realizations of the random field $k^2(\mathbf{x})$ in the volume $V = V(\omega)$. Hereafter we assume the field $k^2(\mathbf{x})$, as well as the rest of the random fields that appear, ergodic, i.e., the ensemble and volume averages (for a given realization) coincide.

Consider the basic equation (1.1) in the volume V. Since the latter is finite, and the defects created should be absorbed inside the volume, it is necessary that the no-flux condition

$$\frac{\partial \varphi}{\partial n}\Big|_S = 0 \tag{2.12}$$

be imposed on the boundary $S = \partial V$ with outward unit vector $\mathbf{n}$. The variational principles of Sections 2.1 and 2.3 are formulated in the same manner using volume averaging. The only difference is that in the primal principle the admissible fields should satisfy the boundary condition (2.12), and those in the dual one the condition

$$\mathbf{e}^* \cdot \mathbf{n}\Big|_S = 0. \tag{2.13}$$

In these forms the primal and dual principles were formulated by Talbot and Willis [4, p.II] (with the purely formal difference that they used $s^* = \varphi^* + K$ as a dual variable). The necessity to take care for the boundary conditions (2.12) or (2.13), when choosing appropriate trial fields, well explains our efforts to reformulate the variational principles in the absorption problem in forms, containing ensemble averaging merely and no boundary conditions involved as a consequence.

3. THE VARIATIONAL PROCEDURE

3.1. *The simplest bounds*

To obtain estimates on the effective absorption coefficient we shall introduce, as usual, appropriate trial fields in the variational principles. The simplest choice is to use a constant such field $\varphi(\mathbf{x}) = \varphi_0$ in the functional (2.1):

$$I[\varphi(\cdot)] = I(\varphi_0) = \frac{1}{2}\left(\left\langle k^2(\mathbf{x})\right\rangle \varphi_0^2 - 2K\varphi_0\right) \to \min\,.$$

Minimization with respect to φ_0 yields

$$\varphi_0 = \frac{K}{\langle k^2\rangle} \quad \text{and} \quad \min_{\varphi_0} I(\varphi_0) = -\frac{K^2}{2\langle k^2\rangle} \tag{3.1}$$

which, upon comparing with (2.3), gives the estimate of the Voigt type

$$k^{*2} \le k_V^2, \quad k_V^2 = \langle k^2(\mathbf{x})\rangle. \tag{3.2}$$

To get a similar lower bound we should also choose $\varphi^* = \text{const}$ in the dual principle. But $\nabla \cdot \mathbf{e}^* = \varphi^*$ and since $\mathbf{e}^*$ should be statistically homogeneous, then $\mathbf{e}^* = 0$, $\varphi^* = 0$ is the simplest choice. From (2.10) and (2.11) it then follows:

$$-\frac{1}{2}\langle\alpha^2\rangle K^2 \le -\frac{K^2}{2k^{*2}},$$

i.e.,

$$k_R^2 \le k^{*2}, \quad k_R^2 = \frac{1}{\langle\alpha^2\rangle} \tag{3.3}$$

which is the Reuss type bound estimate.

3.2. *The optimal three-point upper bound*

To get tighter bounds than the simplest Voigt and Reuss type ones, it is necessary to introduce ampler classes of trial fields. A convenient source for such classes are truncated functional series, similar to those that appear in the scalar conductivity case [7]. In the simplest nontrivial case they have here the form

$$\varphi(\mathbf{x}) = \Phi_0 + \int \Phi_1(\mathbf{x}-\mathbf{y})\delta k^2(\mathbf{y})\, d^3\mathbf{y}, \tag{3.4}$$

where the constant Φ_0 and the function $\Phi_1(\mathbf{x})$ are nonrandom and adjustable quantities and $\delta k^2(\mathbf{y}) = k^2(\mathbf{y}) - \langle k^2\rangle$ is the fluctuating part of the field $k^2(\mathbf{y})$. (Hereafter, the integrals are taken over the whole R^3, if the integration domain is not explicitly indicated.)

Let us introduce (3.4) into (2.1)

$$I = I[\varphi(\cdot)] = I[\Phi_0, \Phi_1(\cdot)] = \frac{1}{2}\Big\langle \Big| \int \nabla\Phi_1(\mathbf{x}-\mathbf{y})\delta k^2(\mathbf{y})\, d^3\mathbf{y} \Big|^2 + k^2(\mathbf{x}) \Big| \Phi_0 + \int \Phi_1(\mathbf{x}-\mathbf{y})\delta k^2(\mathbf{y})\, d^3\mathbf{y} \Big|^2 - 2K\Phi_0 \Big\rangle.$$

Upon varying this functional with respect to Φ_0, we get

$$\left\langle k^2(\mathbf{x}) \left[\Phi_0 + \int \Phi_1(\mathbf{x}-\mathbf{y})\delta k^2(\mathbf{y})\, d^3\mathbf{y} \right] \right\rangle = K,$$

which means that the trial fields (3.4) should satisfy the same condition $\langle k^2(\mathbf{x})\varphi(\mathbf{x})\rangle = K$ as the true fields $\varphi(\mathbf{x})$, see (1.3). Consequently

$$\Phi_0 = \frac{1}{k_V^2}\left(K - \int \Phi_1(\mathbf{y}) M_2^k(\mathbf{y})\, d^3\mathbf{y} \right), \tag{3.5}$$

so that the kernel $\Phi_1(\mathbf{x})$ should be only varied; here $M_2^k(\mathbf{y}) = \langle \delta k^2(\mathbf{0})\delta k^2(\mathbf{y})\rangle$ is the two-point correlation function for the absorption coefficient $k^2(\mathbf{x})$.

Let us introduce again the fields (3.4) in the functional (2.1), taking now (3.5) into account. This makes I a functional of $\Phi_1(\mathbf{x})$ only, whose Euler-Lagrange equation has the form

$$\int \Delta\Phi_1(\mathbf{x}-\mathbf{y}) M_2^k(\mathbf{y})\, d^3\mathbf{y} - \int \Phi_1(\mathbf{x}-\mathbf{y}) \left[k_V^2 M_2^k(\mathbf{y}) + M_3^k(\mathbf{x},\mathbf{y}) \right] d^3\mathbf{y} + \frac{1}{k_V^2} M_2^k(\mathbf{x}) \left[\int \Phi_1(\mathbf{y}) M_2^k(\mathbf{y})\, d^3\mathbf{y} - K \right] = 0, \tag{3.6}$$

where $M_3^k(\mathbf{y}_1,\mathbf{y}_2) = \langle \delta k^2(\mathbf{0})\delta k^2(\mathbf{y}_1)\delta k^2(\mathbf{y}_2)\rangle$ is the three-point correlation function for the field $k^2(\mathbf{x})$.

Obviously, the class of trial fields (3.4) (with or without the constraint (3.5)) forms a convex set in the space $\mathcal{H}$ of statistically homogeneous random fields. Since the functional

I is strongly convex, its restriction on this class has a minimizing element which is unique. This means that the Euler-Lagrange equation (3.6) for the kernel $\Phi_1(\mathbf{x})$ possesses a solution which is unique; we denote this solution as $\widetilde{\Phi}_1(\mathbf{x})$. Upon inserting the latter into the functional (2.1) and taking (3.5) and (3.6) into account, we reach the following upper bound on the effective absorption coefficient:

$$k^{*2} \le k_u^2, \quad k_u^2 = \frac{K k_V^2}{K - \int \widetilde{\Phi}_1(\mathbf{y}) M_2^k(\mathbf{y})\, d^3\mathbf{y}}. \tag{3.7}$$

It should be emphasized that the bound k_u^2 is three-point, i.e., its evaluation needs knowledge of the ℓ-point correlation functions for the medium up to $\ell = 3$. Moreover, repeating the arguments from the scalar conductivity case [7], we can claim that k_u^2 is the optimal three-point bound in the sense that it is the tightest one which *only* employs the above mentioned statistical information.

The solution of eqn (3.6) for a given random constitution, i.e., for given functions M_2^k and M_3^k, is very difficult. That is why we shall consider its solution in the simplest possible case of a weakly inhomogeneous medium. The obtained solution will reappear later in a Ritz's type variational procedures for bounding the effective absorption coefficient k^{*2} (Section 4).

3.3. *The optimal upper bound for a weakly inhomogeneous medium*

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

$$\delta\kappa = \max_x \frac{\mid \delta k^2(\mathbf{x} \mid}{\langle k^2 \rangle} \ll 1.$$

We look for the solution of eqn (3.6) as the perturbation series

$$\widetilde{\Phi}_1(\mathbf{x}) = \Phi_1^{(0)}(\mathbf{x}) + \Phi_1^{(1)}(\mathbf{x}) + \cdots, \tag{3.8}$$

assuming that $\Phi_1^{(p)}(\mathbf{x}) \sim (\delta\kappa)^p$, $p = 0, 1, \ldots$. The introduction of (3.8) into eqn (3.6) leads in a standard way to the chain of equations

$$\int \Big[\Delta\Phi_1^{(0)}(\mathbf{x}-\mathbf{y}) - \langle k^2\rangle \Phi_1^{(0)}(\mathbf{x}-\mathbf{y})\Big]\, M_2^k(\mathbf{y})\, d^3\mathbf{y} - \frac{K}{\langle k^2\rangle} M_2^k(\mathbf{x}) = 0, \tag{3.9}$$

$$\begin{aligned} \int \Big[\Delta\Phi_1^{(1)}(\mathbf{x}-\mathbf{y}) - k_V^2 \Phi_1^{(1)}(\mathbf{x}-\mathbf{y})\Big] M_2^k(\mathbf{y})\, d^3\mathbf{y} \\ - \int \Phi_1^{(0)}(\mathbf{x}-\mathbf{y}) M_3^k(\mathbf{y})\, d^3\mathbf{y} = 0, \quad \text{etc.} \end{aligned} \tag{3.10}$$

The solution of eqn (3.9) is obvious

$$\Phi_1^{(0)}(\mathbf{x}) = -\frac{K}{\langle k^2\rangle} G_V(\mathbf{x}) \tag{3.11}$$

—it is proportional to the Green function $G_V(\mathbf{x}) = \dfrac{1}{4\pi|\mathbf{x}|}\exp(-k_V|\mathbf{x}|)$ of the operator $\Delta - k_V^2$, i.e.

$$\Delta G_V(\mathbf{x}) - k_V^2 G_V(\mathbf{x}) + \delta(\mathbf{x}) = 0.$$

Making use of (3.7) and (3.11), we get

$$\frac{k_u^2}{k_V^2} = 1 - \frac{M_k^2(\mathbf{0})}{k_V^4} I_2^k + o((\delta\kappa)^2), \tag{3.12}$$

where

$$I_2^k = \frac{k_V^2}{M_2^k(\mathbf{0})} \int G_V(\mathbf{y}) M_2^k(\mathbf{y})\, d^3\mathbf{y} \tag{3.13}$$

is a dimensionless statistical parameter for the medium, depending on the two-point correlation function $M_2^k(\mathbf{y})$ solely. It could be easily shown that for dispersions of nonoverlapping spheres I_2^k is simply connected with the parameter I that appeared in Talbot and Willis' approximate scheme for evaluating k^{*2}, see [4, p.I, eqn (3.25)], and in their bounds of Hashin-Shtrikman's type, derived in [4, p.II]. Moreover, these authors were able to evaluate I in the case when the two-point statistics of the dispersion is governed by the well-known Percus-Yevick approximation.

Unlike (3.9), eqn (3.10) is not, however, an equation of convolution type. When calculating the bound k_u^2 to the order $o((\delta\kappa)^3)$ we do not need the exact solution $\Phi_1^{(1)}(\mathbf{x})$, but only the integral

$$F_1(\mathbf{x}) = \int \Phi_1^{(1)}(\mathbf{x} - \mathbf{y}) M_k^2(\mathbf{y})\, d^3\mathbf{y}$$

at $\mathbf{x} = 0$. From (3.10) we get the differential equation for the function $F_1(\mathbf{x})$:

$$\Delta F_1(\mathbf{x}) - k_V^2 F_1(\mathbf{x}) + \int \Phi_1^{(0)}(\mathbf{x} - \mathbf{y}) M_3^k(\mathbf{x}, \mathbf{y})\, d^3\mathbf{y} = 0$$

which immediately gives

$$F_1(\mathbf{0}) = \frac{K}{k_V^2} \iint G_V(\mathbf{y}_1) G_V(\mathbf{y}_2) M_3^k(\mathbf{y}_1, \mathbf{y}_2)\, d^3\mathbf{y}_1 d^3\mathbf{y}_2,$$

taking (3.11) into account. Thus

$$\frac{k_u^2}{k_V^2} = 1 - \frac{M_k^2(\mathbf{0})}{k_V^4} I_2^k + \frac{M_k^3(\mathbf{0}, \mathbf{0})}{k_V^6} I_3^k + o((\delta\kappa)^3), \tag{3.14}$$

where we have introduced, similarly to (3.13), another dimensionless statistical parameter

$$I_3^k = \frac{k_V^4}{M_3^k(\mathbf{0}, \mathbf{0})} \iint G_V(\mathbf{y}_1) G_V(\mathbf{y}_2) M_3^k(\mathbf{y}_1, \mathbf{y}_2)\, d^3\mathbf{y}_1 d^3\mathbf{y}_2, \tag{3.15}$$

depending already on the three-point correlation $M_3^k(\mathbf{y}_1, \mathbf{y}_2)$.

3.4. *Coincidence of k^{*2} and k_u^2 to the order $o((\delta\kappa)^3)$*

A perturbation solution of the type of (3.8) could be obviously found for the basic problem (1.1):

$$\varphi(\mathbf{x}) = \varphi^{(0)}(\mathbf{x}) + \varphi^{(1)}(\mathbf{x}) + \cdots, \tag{3.16}$$

where $\varphi^{(p)}(\mathbf{x}) \sim (\delta\kappa)^p$ at $\delta\kappa \to 0$, $p = 0, 1, \ldots$. Together with (1.1), the decomposition (3.16) immediately yields the chain of equations

$$-k_V^2\varphi^{(0)} + K = 0,$$

$$\Delta\varphi^{(p)} - k_V^2\varphi^{(p)} - \delta k^2\varphi^{(p-1)} = 0,$$

$p = 1, 2, \ldots$, i.e.,

$$\varphi^{(0)} = \frac{K}{k_V^2},$$

$$\varphi^{(1)} = -\frac{K}{k_V^2}\int G_V(\mathbf{x} - \mathbf{y})\delta k^2(\mathbf{y})\, d^3\mathbf{y},$$

etc., so that $\varphi^{(p)}$ will be represented as a p-tuple integral containing the product of p Green functions G_V. It is easy to check, in turn, that

$$\frac{k^{*2}}{k_V^2} = 1 - \frac{M_2^k(\mathbf{0})}{k_V^4}I_2^k + \frac{M_3^k(\mathbf{0},\mathbf{0})}{k_V^6}I_3^k + o((\delta\kappa)^3). \tag{3.17}$$

The comparison between (3.14) and (3.17) shows that indeed the optimal bound k_u^2 coincides with the effective absorption coefficient k^{*2} to the order $o((\delta\kappa)^3)$ for a weakly inhomogeneous medium. A similar result has been derived in [7] in the scalar conductivity case.

3.5. *Optimal three-point lower bound*

Consider the functional J in the dual principle

$$J[\mathbf{e}^*(\cdot)] = -\frac{1}{2}\Big\langle \mathbf{e}^{*2}(\mathbf{x}) + \alpha^2(\mathbf{x})(\nabla\cdot\mathbf{e}^*(\mathbf{x}) + K)^2\Big\rangle \to \max, \tag{3.18}$$

$\alpha^2(\mathbf{x}) = 1/k^2(\mathbf{x})$; we used the fact that $\nabla\cdot\mathbf{e}^* = \varphi^*$ in order to view J as a functional of the field $\mathbf{e}^*(\mathbf{x})$ only.

Similarly to (3.4), we consider the class of trial fields

$$\mathbf{e}^*(\mathbf{x}) = \nabla\int\Psi(\mathbf{x} - \mathbf{y})\delta\alpha^2(\mathbf{y})\, d^3\mathbf{y}, \tag{3.19}$$

where $\delta\alpha^2(\mathbf{y}) = \alpha^2(\mathbf{y}) - \langle\alpha^2\rangle$ is the fluctuation of the "compliance" field $\alpha^2(\mathbf{y})$ and $\Psi(\mathbf{x})$ is a nonrandom kernel.

The class of trial fields (3.19) is the obvious counterpart of the class (3.4), having recalled that $\nabla \cdot \mathbf{e}^* = \varphi^*$. The only difference is that instead of $\delta k^2(\mathbf{y})$ the fluctuation $\delta\alpha^2(\mathbf{y})$ appears in (3.19). Note, however, that the random fields $\delta k^2(\mathbf{y})$ and $\delta\alpha^2(\mathbf{y})$ are simply interrelated only under certain additional assumptions, e.g., if the medium is two-phase, see Section 4.4 below.

The introduction of (3.19) into (3.18) turns J into a functional of the kernel Ψ, namely

$$\begin{aligned} J[\mathbf{e}^*(\cdot)] = J[\Psi(\cdot)] = -\frac{1}{2}\Big\{ & \int\!\!\int \nabla\Psi(\mathbf{y}_1)\cdot\nabla\Psi(\mathbf{y}_2) M_2^\alpha(\mathbf{y}_1-\mathbf{y}_2)\, d^3\mathbf{y}_1 d^3\mathbf{y}_2 \\ & + \int\!\!\int \Delta\Psi(\mathbf{y}_1)\Delta\Psi(\mathbf{y}_2)\Big[\langle\alpha^2\rangle M_2^\alpha(\mathbf{y}_1-\mathbf{y}_2) + M_3^\alpha(\mathbf{y}_1,\mathbf{y}_2)\Big]\, d^3\mathbf{y}_1 d^3\mathbf{y}_2 \\ & + 2K\int \Delta\Psi(\mathbf{y}) M_2^\alpha(\mathbf{y})\, d^3\mathbf{y} + K^2\langle\alpha^2\rangle\Big\}, \end{aligned} \tag{3.20}$$

where, similarly to the absorption field $k^2(\mathbf{x})$,

$$M_2^\alpha(\mathbf{y}) = \langle \delta\alpha^2(\mathbf{0})\delta\alpha^2(\mathbf{y})\rangle, \quad M_3^\alpha(\mathbf{y}_1,\mathbf{y}_2) = \langle \delta\alpha^2(\mathbf{0})\delta\alpha^2(\mathbf{y}_1)\delta\alpha^2(\mathbf{y}_2)\rangle$$

are the two- and three-point correlation functions for the compliance field $\alpha^2(\mathbf{x})$. The Euler-Lagrange equation for the functional (3.20) is obvious

$$\begin{aligned} \int\Big[\langle\alpha^2\rangle\Delta\Psi(\mathbf{x}-\mathbf{y}) - \Psi(\mathbf{x}-\mathbf{y})\Big]\Delta M_2^\alpha(\mathbf{y})\, d^3\mathbf{y} & \\ + \int \Delta\Psi(\mathbf{x}-\mathbf{y})\Delta_x M_3^\alpha(\mathbf{x},\mathbf{y})\, d^3\mathbf{y} + K\Delta M_2^\alpha(\mathbf{x}) = 0. & \end{aligned} \tag{3.21}$$

Eqn (3.21) is the counterpart of the Euler-Lagrange equation (3.6); repeating the respective arguments, concerning the latter, we can claim that the solution, $\widetilde{\Psi}(\mathbf{x})$, exists and it is unique. Introducing this solution into the functional $J[\Psi(\cdot)]$, see (3.20), and using eqn (3.21), leads us to a lower bound on the effective conductivity k^{*2}, namely

$$k_\ell^2 \le k^{*2}, \quad k_\ell^2 = \frac{K}{K\langle\alpha^2\rangle + \int \Delta\widetilde{\Psi}(\mathbf{y}) M_2^\alpha(\mathbf{y})\, d^3\mathbf{y}}, \tag{3.22}$$

which corresponds to the upper bound k_u^2, given in (3.7). Note that the bound (3.22) could be immediately obtained upon averaging eqn (2.8):

$$\frac{K}{k_\ell^2} = \langle\varphi\rangle = \langle\alpha^2(\mathbf{x})(\varphi^*(\mathbf{x}) + K)\rangle = \langle\alpha^2(\mathbf{x})\nabla\cdot\mathbf{e}^*(\mathbf{x})\rangle + K\langle\alpha^2\rangle.$$

This is obviously a three-point bound, since its evaluation needs knowledge of the ℓ-point correlation functions M_2^α and M_3^α for the compliance field up to $\ell = 3$. Repeating the arguments of [7] we can again state that the bound k_ℓ^2 is the optimal three-point lower

bound, i.e., the best one which employs *solely* the statistical information incorporated in these correlation functions.

3.6. *The optimal lower bound for a weakly inhomogeneous medium*

Let the medium be again weakly inhomogeneous, but in the sense that

$$\delta\alpha = \max_x \frac{\mid \delta\alpha^2(\mathbf{x}) \mid}{\langle \alpha^2 \rangle} \ll 1.$$

For a two-phase medium this assumption is equivalent to that that $\delta\kappa \ll 1$, adopted in Section 3.3. Similarly to the analysis, performed there, we represent the solution of eqn (3.21) as the perturbation series

$$\widetilde{\Psi}(\mathbf{x}) = \Psi^{(0)}(\mathbf{x}) + \Psi^{(1)}(\mathbf{x}) + \cdots, \tag{3.23}$$

assuming that $\Psi^{(p)}(\mathbf{x}) \sim (\delta\alpha)^p$, $p = 0, 1, \ldots$. The introduction of (3.23) into eqn (3.21) leads in a standard way to the chain of equations

$$\int \Big[\Delta\Psi^{(0)}(\mathbf{x}-\mathbf{y}) - k_R^2\Psi^{(0)}(\mathbf{x}-\mathbf{y})\Big]\Delta M_2^\alpha(\mathbf{y})\, d^3\mathbf{y} + Kk_R^2\Delta M_2^\alpha(\mathbf{x}) = 0, \tag{3.24}$$

$$\begin{aligned}\int \Big[\Delta\Psi^{(1)}(\mathbf{x}-\mathbf{y}) - k_R^2\Psi^{(1)}(\mathbf{x}-\mathbf{y})\Big]\Delta M_2^\alpha(\mathbf{y})\, d^3\mathbf{y} \\ + k_R^2 \int \Psi^{(0)}(\mathbf{x}-\mathbf{y})\Delta_x M_3^\alpha(\mathbf{x},\mathbf{y})\, d^3\mathbf{y} = 0, \quad \ldots .\end{aligned} \tag{3.25}$$

The solution of eqn (3.24) is obvious

$$\Psi^{(0)}(\mathbf{x}) = Kk_R^2 G_R(\mathbf{x}),$$

where $G_R(\mathbf{x}) = \dfrac{1}{4\pi|\mathbf{x}|}\exp(-k_R|\mathbf{x}|)$ is the Green function for the operator $\Delta - k_R^2$. For the integral, appearing in (3.22), we find

$$\int \Delta\Psi^{(1)}(\mathbf{y}) M_2^\alpha(\mathbf{y})\, d^3\mathbf{y} = Kk_R^4 \int\int \Delta G_R(\mathbf{y}_1)\Delta G_R(\mathbf{y}_2) M_3^\alpha(\mathbf{y}_1,\mathbf{y}_2)\, d^3\mathbf{y}_1 d^3\mathbf{y}_2,$$

repeating the respective arguments of Section 3.3. The eventual result for the optimal three-point lower bound (3.22) reads

$$\frac{k_\ell^2}{k_R^2} = 1 + k_R^4 M_2^\alpha(\mathbf{0}) I_2^\alpha - k_R^6 M_3^\alpha(\mathbf{0},\mathbf{0}) I_3^\alpha + o((\delta\alpha)^3), \tag{3.26}$$

where

$$\begin{aligned} I_2^\alpha &= -\frac{1}{M_2^\alpha(\mathbf{0})}\int \Delta G_R(\mathbf{y}) M_2^\alpha(\mathbf{y})\, d^3\mathbf{y}, \\ I_3^\alpha &= \frac{1}{M_3^\alpha(\mathbf{0},\mathbf{0})}\int\int \Delta G_R(\mathbf{y}_1)\Delta G_R(\mathbf{y}_2) M_3^\alpha(\mathbf{y}_1,\mathbf{y}_2)\, d^3\mathbf{y}_1 d^3\mathbf{y}_2 \end{aligned} \tag{3.27}$$

are the dimensionless statistical parameters, corresponding to those, introduced in (3.13) and (3.15) for the absorption field $k^2(\mathbf{x})$.

3.7. *Coincidence of k^{*2} and k_ℓ^2 to the order $o((\delta\alpha)^3)$*

To show this, we rewrite eqn (1.1) in the form

$$\alpha^2(\mathbf{x})\Delta\varphi(\mathbf{x}) - \varphi(\mathbf{x}) + \alpha^2(\mathbf{x})K = 0. \tag{3.28}$$

Its perturbation solution in this case has the form (3.16), with the only difference that $\varphi^{(p)}(\mathbf{x}) \sim (\delta\alpha)^p$ at $\delta\alpha \to 0$, $p = 0, 1, \ldots$. The introduction of (3.16) into (3.28) yields the respective chain of equations, similar to that in Section 3.4, so that

$$\begin{aligned} \varphi^{(0)} &= \frac{K}{k_R^2}, \quad \varphi^{(1)}(\mathbf{x}) = Kk_R^2 \int G_R(\mathbf{x}-\mathbf{y})\delta\alpha^2(\mathbf{y})\, d^3\mathbf{y}, \\ \varphi^{(p)}(\mathbf{x}) &= k_R^2 \int G_R(\mathbf{x}-\mathbf{y})\Delta\varphi^{(p-1)}(\mathbf{y})\delta\alpha^2(\mathbf{y})\, d^3\mathbf{y}, \end{aligned} \tag{3.29}$$

$p = 2, 3, \ldots$. A direct check shows that

$$\int \Delta G_R(\mathbf{x})\, d^3\mathbf{x} = 0$$

which implies that

$$\int \Delta\varphi^{(p)}(\mathbf{x})\, d^3\mathbf{x} = 0, \quad p = 0, 1, , \ldots . \tag{3.30}$$

To calculate k^{*2} to the needed order, we use the definition (1.5) and eqn (3.28):

$$\frac{K}{k^{*2}} = \langle\varphi(\mathbf{x})\rangle = \langle\alpha^2(\mathbf{x})(\Delta\varphi(\mathbf{x}) + K)\rangle.$$

Upon inserting (3.29) here and making use of (3.30), we get after simple algebra the expression of k^{*2} to the needed order $o((\delta\alpha)^3)$, which indeed coincides with that for k_ℓ^2, as given in (3.26).

4. BERAN'S TYPE BOUNDS

4.1. *Ritz's procedure*

Due to the difficulties connected with the solution of eqns (3.6) and (3.21) for the optimal kernels, we shall resort here to a simpler procedure of Ritz's type in which we replace, e.g., the class (3.4) of trial fields with the narrower one

$$\varphi(\mathbf{x}) = \Phi_0 + \lambda \int \overline{\Phi}(\mathbf{x}-\mathbf{y})\delta k^2(\mathbf{y})\, d^3\mathbf{y}, \tag{4.1}$$

where Φ_0 and λ are adjustable scalars and $\overline{\Phi}(\mathbf{x})$ is a fixed kernel. The constants Φ_0 and λ are subject to the constraint

$$\Phi_0 k_V^2 + \lambda \int \overline{\Phi}(\mathbf{y}) M_2^k(\mathbf{y})\, d^3\mathbf{y} = K, \tag{4.2}$$

as it follows from (3.5). The procedure is now straightforward. We insert (4.1) into the functional I, see (2.1), and take into account (4.2). This makes I a quadratic function of λ whose minimization yields an upper bound on the effective absorption coefficient k^{*2}. This bound will depend on the choice of the kernel $\overline{\Phi}(\mathbf{x})$ in the class (4.1). Similarly to the scalar conductivity case [12,13], we call the kernel $\overline{\Phi}(\mathbf{x})$ optimal, if the respective bound coincides with the optimal one, k_u^2. The problem of determining the optimal, in this sense, kernels is equivalent to solving eqn (3.6): since the solution of this equation is unique, all such kernels are proportional to it.

A natural and convenient choice of the kernel $\overline{\Phi}(\mathbf{x})$ is the Green function $G_V(\mathbf{x})$. It is natural because it already appeared in the perturbation solution of eqn (3.6), and of the basic eqn (1.1). The convenience is due to simplification of the needed calculations of the respective bounds. A similar choice of the kernel in the scalar conductivity problem was proposed by Beran [8]; that is why the bounds on k^{*2}, obtained hereafter, will be called Beran's. The problem of their optimality will be addressed in Section 5.

4.2. *Upper Beran's type bound*

In accordance with the foregoing, let us minimize the functional I in (2.1) over the class of trial fields

$$\varphi(\mathbf{x}) = \Phi_0 + \lambda \int G_V(\mathbf{x}-\mathbf{y}) \delta k^2(\mathbf{y})\, d^3\mathbf{y}, \tag{4.3}$$

under the constraint

$$\Phi_0 k_V^2 + \lambda \frac{M_2^k(\mathbf{0})}{k_V^2} I_2^k = K,$$

see (4.2) and the definition (3.13) of the dimensionless statistical parameter I_2^k. The minimization with respect to λ is straightforward and the final result is the upper Beran's type bound:

$$\frac{k^{*2}}{k_V^2} \le 1 - \frac{M_2^k(\mathbf{0})}{k_V^4} \frac{(I_2^k)^2}{I_2^k + \dfrac{M_3^k(\mathbf{0},\mathbf{0})}{k_V^2 M_2^k(\mathbf{0})} I_3^k}, \tag{4.4}$$

with I_3^k defined in (3.15).

Obviously, the upper bound (4.4) coincides to the order $o((\delta\kappa)^3)$ with both the optimal upper bound k_u^2, see (3.14), and with the exact value of the absorption coefficient k^{*2}, given in (3.17).

4.3. *Lower Beran's type bound*

The same arguments can be repeated in connection with the dual principle (2.10). In this case (3.19) is replaced by the narrower class

$$\mathbf{e}^*(\mathbf{x}) = \lambda \nabla \int \overline{\Psi}(\mathbf{x}-\mathbf{y}) \delta\alpha^2(\mathbf{y})\, d^3\mathbf{y},$$

where $\overline{\Psi}(\mathbf{x})$ is a fixed kernel and the constant λ is adjustable. If the bound so obtained coincides with k_l^2—the optimal three-point lower bound, the kernel $\overline{\Psi}$ is called optimal. The optimal kernels have again a simple description here—they are proportional to the solution $\widetilde{\Psi}(\mathbf{x})$ of eqn (3.21).

Similarly to Section 4.1, we may argue that a natural and convenient choice of the kernel $\overline{\Psi}(\mathbf{x})$ is the Green function $G_R(\mathbf{x})$. That is why we maximize the functional (2.10) over the class of trial fields

$$\mathbf{e}^*(\mathbf{x}) = \lambda\nabla\int G_R(\mathbf{x}-\mathbf{y})\delta\alpha^2(\mathbf{y})\,d^3\mathbf{y},$$

with respect to λ. The final result is the above mentioned lower bound of Beran's type

$$\left\{1 - k_R^4 M_2^\alpha(\mathbf{0})\frac{(I_2^\alpha)^2}{I_2^\alpha + k_R^2\dfrac{M_3^\alpha(\mathbf{0},\mathbf{0})}{M_2^\alpha(\mathbf{0})}I_3^\alpha}\right\}^{-1} \leq \frac{k^{*2}}{k_R^2}, \tag{4.5}$$

where I_2^α and I_3^α are the statistical parameters (3.27).

Obviously, the lower bound (4.5) coincides to the order $o((\delta\alpha)^3)$ with both the optimal lower bound k_ℓ^2, see (3.26), and with the exact value of the absorption coefficient k^{*2}, given in (3.17).

4.4. *Beran's type bounds are third-order*

Let us recall that Beran's bounds in the scalar conductivity case depend on two statistical parameters [8]. For a two-phase medium these two parameters are interrelated, so that both may be expressed by a single one, following, e.g., Milton [14]. In the absorption problem under study the needed parameters are four: I_2^k, I_3^k, I_2^α and I_3^α. We derived, however, two perturbation expansions for the effective absorption coefficient k^{*2}—the first is (3.17), in powers of $\delta\kappa$, the second is (3.26), in powers of $\delta\alpha$. It sounds appealing at a first glance to compare these expansions and get as a result two relations between the above mentioned four statistical parameters. This is, unfortunately, not possible. The basic reason is that for an arbitrary random medium $\delta\kappa$ and $\delta\alpha$ are not connected, so that no comparison between (3.17) and (3.26) can be done. The latter is possible only for a two-phase medium for which

$$k^2(\mathbf{x}) = k_m^2 + [k^2]I_f(\mathbf{x}), \quad \alpha^2(\mathbf{x}) = \alpha_m^2 + [\alpha^2]I_f(\mathbf{x}), \tag{4.6}$$

where $I_f(\mathbf{x})$ is the characteristic function of the volume, occupied by one of the constituents, whose characteristics are supplied hereafter with the subscript 'f': its absorption coefficient and "compliance" are thus k_f^2 and $\alpha_f^2 = 1/k_f^2$, the volume fraction is $c = c_f$. For the other constituents we use the subscript 'm', so that its parameters are, respectively, k_m^2, $\alpha_m^2 = 1/k_m^2$ and $c_m = 1 - c$. As usual, $[k^2] = k_f^2 - k_m^2$, etc. Then

$$\delta k^2(\mathbf{x}) = [k^2]I'_f(\mathbf{x}), \quad \delta\alpha^2(\mathbf{x}) = [\alpha^2]I'_f(\mathbf{x}), \tag{4.7}$$

where $I'_f(\mathbf{x}) = I_f(\mathbf{x}) - c$ is the fluctuating part of $I_f(\mathbf{x})$; besides

$$M_2^k(\mathbf{0}) = c(1-c)[k^2]^2, \quad M_3^k(\mathbf{0},\mathbf{0}) = c(1-c)(1-2c)[k^2]^3, \tag{4.8}$$

and similarly for M_2^α and M_3^α.

Making use of (4.8), we rewrite the bounds (4.4) and (4.5) for a two-phase medium in a bit simplified form

$$\begin{aligned} &k_R^2 \left\{ 1 - k_R^4 [\alpha^2]^2 \frac{c(1-c)(I_2^\alpha)^2}{I_2^\alpha + k_R^2[\alpha^2](1-2c)I_3^\alpha} \right\}^{-1} \\ &\leq k^{*2} \leq k_V^2 \left(1 - \frac{[k^2]^2}{k_V^4} \frac{c(1-c)(I_2^k)^2}{I_2^k + \dfrac{[k^2]}{k_V^2}(1-2c)I_3^k} \right), \end{aligned} \tag{4.9}$$

keeping in mind that now $k_V^2 = ck_f^2 + (1-c)k_m^2$ and $k_R^2 = (c/k_f^2 + (1-c)/k_m^2)^{-1}$. Due to (4.7), the four statistical parameters I_2^k, I_3^k, I_2^α and I_3^α depend only on the two- and three-point correlation functions of the field $I'_f(\mathbf{x})$. These correlation functions are, however, multiplied in the respective integrals by different functions, namely, by $G_V(\mathbf{x})$ or $G_R(\mathbf{x})$—a situation that does not appear in the scalar conductivity or elastic cases (one and the same Green function is used there in both upper and lower Beran's bounds). It is noted also that one of the bounds (4.9) degenerates at $k_f/k_m \to 0$ (the lower) or $k_f/k_m \to \infty$ (the upper).

For a two-phase medium, the perturbation expansions (3.17) and (3.26) may already be compared, because they both can be recast as expansions in powers of, say, $\Delta\kappa = [k^2]/k_m^2$. It is then readily checked that

$$\frac{k_R^2}{k_V^2} = 1 + O((\Delta\kappa)^2).$$

That is why, in the definitions (3.27) of the parameters I_2^α and I_3^α, we may replace k_R^2 by k_V^2, making an error of the order $O((\Delta\kappa)^2)$. These parameters multiply $(\Delta\kappa)^2$ and $(\Delta\kappa)^3$ in (3.26) (having made an obvious transition from $\delta\alpha$ to $\Delta\kappa$ there), so that, to the order $o((\Delta\kappa)^3)$ the expansion (3.26) will not be affected by the replacement of k_R^2 by k_V^2. A simple check will show, in turn, that both expansions (3.14) and (3.26) coincide to the same order $o((\Delta\kappa)^3)$.

Two conclusions can be drawn upon the foregoing reasoning.

The first is that the existence of two perturbation expansions (3.14) and (3.26) for the effective absorption coefficient k^{*2} does not lead to any relations between the four statistical parameters in the Beran type bounds (4.4) and (4.5) even for a two-phase medium. Thus they are all independent and should be somehow evaluated for a given random constitution.

The second is the fact that Beran's type bounds (4.9) for a two-phase medium are third-order, i.e., they coincide to the asymptotic order $o((\Delta\kappa)^3)$ in the weakly inhomogeneous case. As is well-known, the same fact holds in the scalar conductivity and elastic cases.

5. BOUNDS FOR CELLULAR MEDIA

To illustrate the performance of the Beran's type bounds (4.9) we consider a simple and quite realistic, for a wide class of two-phase media, model of a cellular medium, proposed by Miller [15]. It is obtained by dividing the space into closed regions, called cells. Afterwards the cells are randomly filled up with one of the two constituents possessing (in our context) absorption coefficients k_f^2 or k_m^2. The volume fractions of the latter are respectively c_f or c_m, see [15], [16] for details, and [17] for the most recent discussion and development.

Under the assumption of statistical isotropy and spherical shape of the cells, the two- and three-point correlation functions for such a cellular medium have, after Hori [16], the form:

$$\begin{aligned} M_2^k(\mathbf{y}) &= M_2^k(\mathbf{0})\frac{1}{V_a}\int h(\mathbf{z})h(\mathbf{y}-\mathbf{z})\,d^3\mathbf{z}, \\ M_3^k(\mathbf{y}_1,\mathbf{y}_2) &= M_3^k(\mathbf{0},\mathbf{0})\,\frac{1}{V_a}\int h(\mathbf{z})h(\mathbf{y}_1-\mathbf{z})h(\mathbf{y}_2-\mathbf{z})\,d^3\mathbf{z}, \end{aligned} \tag{5.1}$$

where $h(\mathbf{x})$ denotes the characteristic function of the mean cell—a ball of radius a, $V_a = \frac{4}{3}\pi a^3$ and $M_2^k(\mathbf{0})$ and $M_3^k(\mathbf{0},\mathbf{0})$ are given in (4.8). Similar expressions hold for the "compliance" field $\alpha^2(\mathbf{x}) = 1/k^2(\mathbf{x})$.

On introducing (5.1) into the definitions (3.13) and (3.15) of the statistical parameters I_2^k and I_3^k respectively, one gets integrals involving the Helmholtz potential $\chi_V = h*G_V(\mathbf{x})$ for a single sphere of radius a, located at the origin. Using the simple analytical form of the latter, the following expressions for these parameters are eventually obtained:

$$I_2^k = 1 - F_2(a_V), \quad I_3^k = 1 - 2F_2(a_V) + F_3(a_V), \tag{5.2a}$$

where a_V is the dimensionless parameter $a_V = ak_V$ and

$$\begin{aligned} F_2(x) &= 3\frac{1+x}{x^3}e^{-x}(x\cosh x - \sinh x), \\ F_3(x) &= \frac{3}{2}\frac{(1+x)^2}{x^3}e^{-2x}(\sinh x\cosh x - x)1. \end{aligned} \tag{5.3}$$

Similar calculations for the statistical parameters I_2^α and I_3^α, that enter the lower bound (4.5), involve the Helmholtz potential $\chi_R = h * G_R(\mathbf{x})$ for a sphere of radius a, located at the origin. It appears finally that

$$I_2^\alpha = F_2(a_R), \quad I_3^\alpha = F_3(a_R), \tag{5.2b}$$

where $a_R = ak_R$ and the functions F_2 and F_3 are defined in (5.3).

Note that $F_2(x)$ and $F_3(x)$ are monotonically decreasing functions of x and $F_2(0) = F_3(0) = 1$, $F_2(\infty) = F_3(\infty) = 0$. That is why the four statistical parameters I_2^k, I_3^k, I_2^α and I_3^α for a cellular medium always lie in the interval $[0,1]$.

Tables 1 and 2 illustrate the performance of the bounds (4.9) for a cellular medium at $a_m = 1$ and $a_m = 10$, respectively, where $a_m = ak_m$. It is well seen that the bounds remain tolerably close and supply useful information about k^{*2} even when the absorption ability of one of the constituents is one hundred times greater than that of the other ($k_f^2/k_m^2 = 100$ or 0.01).

The cellular model helps us also to demonstrate that Beran's type bounds (4.9) are not optimal even for this simple microgeometry. Indeed, their optimality, in the sense explained in Section 3, would mean here that eqn (3.6) has a solution $\Phi_1(\mathbf{x}) = \lambda G_V(\mathbf{x})$ for a certain scalar λ, provided the correlation functions are given in (5.1). It is easy to see that the latter would imply proportionality of the convolutions $h * h$ and $\chi_V * h$, which could be true only if the Helmholtz potential of a sphere, $\chi_V(\mathbf{x})$, is proportional to the characteristic function, $h(\mathbf{x})$, of the same sphere. But this is obviously wrong. Thus Beran's type upper bounds are not optimal three-point bounds even for cellular media; more restrictive three-point bounds could be obtained if the kernel $\Phi_1(\mathbf{x})$ is chosen more skillfully.

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