NEW FORMULAS FOR THREE-POINT BOUNDS ON THE EFFECTIVE CONDUCTIVITY OF MULTI-PHASE SYMMETRIC CELL MATERIALS*

KRASIMIR D. TSVYATKOV

ABSTRACT: *New forms of trial fields for classical variational principles in the context of electrical conductivity are introduced. They generalize to an arbitrary random medium, not necessarily with discrete structure, of the trial fields of Phan-Thien and Milton (1982) introduced by them for an N-phase material with a periodic structure and leading to finding of three-point bounds on the effective conductivity of the composite. These new forms of their trial fields are used for symmetric cell materials and as a result simple formulas are obtained for the corresponding bounds on the effective conductivity of such materials.*

KEYWORDS: *Effective conductivity, Symmetric cell materials, Variational bounds*

1 Introduction

The problem of finding the effective physical properties of a heterogeneous medium requires knowledge of the complete statistical description of the medium [1]. In practice, however, we have only the information given by the first few correlation functions of the medium. Therefore, the only thing that can be rigourously done is to obtain bounds on these properties using this limited information. In the context of electrical conductivity, we will briefly consider finding variational bounds using only the first k-point moments up to $k = 3$ of the scalar random conductivity field $\sigma(\boldsymbol{x})$ of a d-dimensional medium at $d = 2, 3$. These bounds are referred to as three-point bounds.

We will assume that $\sigma(\boldsymbol{x})$ is a statistically homogeneous random field and relates the local current density field $\boldsymbol{J}(\boldsymbol{x})$ with the local electric field $\boldsymbol{E}(\boldsymbol{x})$ in the medium according to Ohm's law:

$$\boldsymbol{J}(\boldsymbol{x}) = \sigma(\boldsymbol{x})\boldsymbol{E}(\boldsymbol{x}), \tag{1}$$

We will also assume that the medium is macroscopically isotropic, i.e. its effective conductivity $\boldsymbol{\sigma}^*$, usually defined by the averaged Ohm's law:

$$\langle \boldsymbol{J}(\boldsymbol{x})\rangle = \boldsymbol{\sigma}^*\langle \boldsymbol{E}(\boldsymbol{x})\rangle, \tag{2}$$

is a scalar quantity; the brackets $\langle\ \rangle$, hereafter, denote ensemble averaging.

The electric field is irrotational: $\nabla \times \boldsymbol{E}(\boldsymbol{x}) = 0$, i.e. $\boldsymbol{E}(\boldsymbol{x}) = -\nabla\phi(\boldsymbol{x})$, where $\phi(\boldsymbol{x})$ is the electrical potential, and the current density field, in the absence of internal current sources, is solenoidal:

$$\nabla \cdot \boldsymbol{J}(\boldsymbol{x}) = 0. \tag{3}$$

The problem of finding the solution $\phi(\boldsymbol{x})$ of this equation under prescribed constant-average electric field, $\langle \boldsymbol{E}(\boldsymbol{x})\rangle = \boldsymbol{E}_0$, is equivalent to the classical energy variational principle, according to which among all the statistically homogeneous irrotational fields $\widehat{\boldsymbol{E}}(\boldsymbol{x})$ for which $\langle \widehat{\boldsymbol{E}}(\boldsymbol{x})\rangle = \boldsymbol{E}_0$, the functional

$$W\big[\widehat{\boldsymbol{E}}(\cdot)\big] = \frac{1}{2}\langle \sigma(\boldsymbol{x})\widehat{\boldsymbol{E}}(\boldsymbol{x}) \cdot \widehat{\boldsymbol{E}}(\boldsymbol{x})\rangle \tag{4}$$

*Partially supported by Scientific Research Grant RD-08-73/23.01.2020 of Shumen University.

is stationary and minimum for this solution, i.e. for the true electric field $\boldsymbol{E}(\boldsymbol{x})$ in the medium; so that $\min W = \dfrac{1}{2}\,\sigma^* E_0^2$, $E_0^2 = \boldsymbol{E}_0 \cdot \boldsymbol{E}_0$. The ergodic hypothesis for all statistically homogeneous fields is adopted.

The solution $\boldsymbol{E}(\boldsymbol{x})$ can be formally found in the form of a perturbation series for a weakly inhomogeneous medium. This series, truncated after its first integral term, forms the field

$$\widehat{\boldsymbol{E}}^{(b)}(\boldsymbol{x}) = \boldsymbol{E}_0 + \alpha\,\boldsymbol{E}_0 \cdot \int \nabla\nabla G(\boldsymbol{x}-\boldsymbol{y})\sigma'(\boldsymbol{y})\,\mathrm{d}\boldsymbol{y}, \tag{5}$$

where $G(\boldsymbol{x})$ is the Green function for the Laplace operator in $\mathbb{R}^d$, i.e. $G(\boldsymbol{x}) = 1/4\pi|\boldsymbol{x}|$ for $d = 3$ or $G(\boldsymbol{x}) = -\ln|\boldsymbol{x}|/2\pi$ for $d = 2$, and $\alpha = 1/\langle\sigma\rangle$; hereafter, $\Theta'(\boldsymbol{x}) = \Theta(\boldsymbol{x}) - \langle\Theta(\boldsymbol{x})\rangle$ is the fluctuation of a random field $\Theta(\boldsymbol{x})$ about its mean value $\langle\Theta(\boldsymbol{x})\rangle$.

In fact, Beran[2] adopted α as an adjustable parameter in (5) and then considered the classical variational principle on the fields $\widehat{\boldsymbol{E}}^{(b)}(\boldsymbol{x})$. The energy functional (4), when restricted over this class of trial fields, becomes a quadratic function of α, whose minimization brings forth an upper bound on σ^*:

$$\sigma^* \le \sigma_u^{(b)}, \quad \sigma_u^{(b)} = \langle\sigma\rangle \left\{ 1 - \frac{m_{2\sigma}^2/d^2}{m_{2\sigma}/d + A_\sigma^{(3)} m_{3\sigma}} \right\}, \tag{6}$$

where $m_{2\sigma} = \langle\sigma'^2\rangle/\langle\sigma\rangle^2$, $m_{3\sigma} = \langle\sigma'^3\rangle/\langle\sigma\rangle^3$, and

$$A_\sigma^{(3)} = \frac{1}{d\langle\sigma'^3\rangle} \iint M_3^\sigma(\boldsymbol{z}_1,\boldsymbol{z}_2)\nabla\nabla G(\boldsymbol{z}_1) : \nabla\nabla G(\boldsymbol{z}_2)\,\mathrm{d}\boldsymbol{z}_1\mathrm{d}\boldsymbol{z}_2 \tag{7}$$

is a dimensionless statistical parameter for the medium, depending on the three-point moment $M_3^\sigma(\boldsymbol{z}_1,\boldsymbol{z}_2) = \langle\sigma'(\boldsymbol{0})\sigma'(\boldsymbol{z}_1)\sigma'(\boldsymbol{z}_2)\rangle$ for the conductivity field $\sigma(\boldsymbol{x})$; the colon denotes contraction with respect to two pairs of indices.

We are now introducing the more general class of trial fields

$$\widehat{\boldsymbol{E}}(\boldsymbol{x}) = \boldsymbol{E}_0 + \boldsymbol{E}_0 \cdot \int \nabla\nabla G(\boldsymbol{x}-\boldsymbol{y})\lambda'(\sigma(\boldsymbol{y}))\,\mathrm{d}\boldsymbol{y}, \tag{8}$$

where $\lambda(\sigma)$ is a varying non-random function of the achieved conductivity values and the field $\lambda'(\sigma(\boldsymbol{x}))$ is the fluctuation of the random field $\lambda(\sigma(\boldsymbol{x}))$. Obviously, if $\lambda(\sigma) = \alpha\,\sigma$ with an adjustable constant α, the fields (8) coincide with the Beran's trial fields (5).

Let us assume only for a moment that the medium is an N-phase composite material. Thus each phase is characterized by its constant conductivity σ_i and its indicator function $\Omega_i(\boldsymbol{x})$, taking the value 1 if $\boldsymbol{x} \in i$-th phase, and 0 otherwise, where $i = 1,\ldots,N$. Then conductivity field $\sigma(\boldsymbol{x})$ is represented as a step function:

$$\sigma(\boldsymbol{x}) = \sigma_1\Omega_1(\boldsymbol{x}) + \cdots + \sigma_N\Omega_N(\boldsymbol{x}) \tag{9}$$

and, according to the definition of the function $\lambda(\sigma)$, the field $\lambda(\sigma(\boldsymbol{x}))$ has the same form:

$$\lambda(\sigma(\boldsymbol{x})) = \alpha_1\Omega_1(\boldsymbol{x}) + \cdots + \alpha_N\Omega_N(\boldsymbol{x}), \tag{10}$$

where $\alpha_i = \lambda(\sigma_i)$, $i = 1, \ldots, N$. Therefore, for N-phase medium, the general trial fields (8) can be represented in the form

$$\widehat{\boldsymbol{E}}(\boldsymbol{x}) = \boldsymbol{E}_0 + \boldsymbol{E}_0 \cdot \sum_{i=i}^{N} \alpha_i \int \nabla\nabla G(\boldsymbol{x} - \boldsymbol{y}) \Omega_i'(\boldsymbol{y}) \, \mathrm{d}\boldsymbol{y}, \tag{11}$$

where $\boldsymbol{\alpha} = (\alpha_1, \ldots, \alpha_N)$ is un adjustable constant vector. Keeping in mind that the Fourier transform $\mathcal{F}$ maps the integral terms in (11) into the tensors $\dfrac{\boldsymbol{k}\boldsymbol{k}}{k^2}\,\omega_i(\boldsymbol{k})$ in the case of three space dimensions, where $\omega_i(\boldsymbol{k}) = \mathcal{F}\left[\Omega_i'(\boldsymbol{x})\right]$, $i = 1, \ldots, N$, we note that the fields (11) are the same trial fields introduced through their Fourier series expansions by Phan-Thien and Milton [3] for an N-phase material with periodic internal structure, when the vector $\boldsymbol{E}_0$ is chosen to be the unit vector in the x-direction. Thus, it turned out that the trial fields (8), introduced here, are a generalization of the trial fields of Phan-Tien and Milton for an arbitrary statistically homogeneous medium, as well as for a two-dimensional medium. By minimizing the restriction of the energy functional (4) on their trial fields with respect to the vector $\boldsymbol{\alpha}$, they obtained an upper bound for σ^* in a form involving matrix multiplications, the thre-point statistics of the medium being represented by a three-dimensional matrix of $N(N-1)^2/2$ different geometrical parameters

$$A_{abc} = \iint \langle \Omega_a'(\boldsymbol{0}) \Omega_b'(\boldsymbol{z}_1) \Omega_c'(\boldsymbol{z}_2) \rangle \, \nabla\nabla G(\boldsymbol{z}_1) : \nabla\nabla G(\boldsymbol{z}_2) \, \mathrm{d}\boldsymbol{z}_1 \mathrm{d}\boldsymbol{z}_2, \qquad a, b, c = 1, \ldots, N. \tag{12}$$

It is worth noting that $\Omega_1(\boldsymbol{x}) + \cdots + \Omega_N(\boldsymbol{x}) = 1$, whence $\Omega_2'(\boldsymbol{x}) = -\Omega_1'(\boldsymbol{x})$ at $N = 2$. Taking into account also (9), we conclude that for a two-phase medium the trial fields (11) are reduced to the Beran's fields (5).

Referring to the form (11) of the trial fields of Phan-Thien and Milton [3], it can be easily shown that the trial fields of Pham [4] coincide with them. Therefore, the corresponding bounds obtained by Pham [4] again in a form involving matrix multiplications, but with other statistical geometrical parameters, coincide with the bounds of Phan-Thien and Milton. This conclusion is in line with the finding of Pham [4] that by using computer programs one can check that his bounds in three-dimensional case ($d = 3$) agree with those of Phan-Thien and Milton [3].

While minimizing the energy functional (4) on the class of trial fields (11) is reduced to solving a linear system with respect to the multipliers $\alpha_1, \ldots, \alpha_N$, the problem of minimizing it on the class of fields (8) for un arbitrary, not necessarily medium with a discrete structure, comes down to solving the corresponding Euler-Lagrange equation, which seems to be a complicated, albeit linear, integral equation with respect to the function $\lambda(\sigma)$. That is why here we will turn our attention to the restriction of the energy functional on this more general class of trial fields for symmetric cell materials, which seems to be the first meaningful class of materials for which the Beran bounds [2] are considered, starting with Miller's work [5].

2 Symmetric cell materials and Miller geometrical parameter

The symmetric cell material is constructed as follows. The space is divided into an infinite number of cells in such a way that the resulting geometric structure is statistically homogeneous. In this division, the shapes and sizes of the cells may be different. Then each cell is randomly assigned a constant conductivity σ with some probability density $P(\sigma)$, the value of which does not depend on either the geometry of the cell or the assigned conductivities of the surrounding cells. A classic example of such material is the random (two- or three-dimensional) chessboard.

Another example is the medium obtained when space is divided only into spherical cells whose radii vary from a certain finite to the infinitesimally small value.

It follows from the assumed statistical independence of cell conductivities that [6]

$$\langle \sigma'(\boldsymbol{r}_1)\sigma'(\boldsymbol{r}_2)\sigma'(\boldsymbol{r}_3)\rangle = \langle \sigma'^3\rangle\, g_3(\boldsymbol{z}_1,\boldsymbol{z}_2,\boldsymbol{z}_2), \tag{13}$$

where $g_3(\boldsymbol{z}_1,\boldsymbol{z}_2,\boldsymbol{z}_2)$ is the probability that all three points $\boldsymbol{r}_1$, $\boldsymbol{r}_2$, and $\boldsymbol{r}_3$ fall into the same cell. Due to the special representation (13), the Beran statistical parameter $A_\sigma^{(3)}$ in (7) is equal to the geometrical parameter

$$\mathcal{G} = \frac{1}{d}\iint g_3(\boldsymbol{z},\boldsymbol{w})\nabla\nabla G(\boldsymbol{z}_1) : \nabla\nabla G(\boldsymbol{z}_2)\,\mathrm{d}\boldsymbol{z}_1\mathrm{d}\boldsymbol{z}_2, \tag{14}$$

introduced and denoted by G by Miller [5], who considered the Beran bounds for a two-phase medium. He showed that the parameter G does not depend on the sizes and relative positions of the cells, but only on their shapes. Hori [6] expressed the parameter G for a material with an ellipsoidal shape of the cells (with uniform random orientations) through their depolarization factors L_i, $i = 1,2,3$, and also introduced tensor $\boldsymbol{A}^{(3)}$ generalizing G for the case when the medium is not statistically isotropic. The values of the parameter G for some cell shapes, more complex constructions of cell materials, and other known results concerning them, the reader can find in the books of Milton [7, ch. 15] and Torquato [8, ch. 8, 22] and the references in them.

3 Three-point bounds for cell materials

We will now consider deriving un upper bound on the effective conductivity σ^* by using the classical energy principle on the class of trial fields (8) and a lower bound by using its dual classical variational principle on the class of corresponding to them trial fields.

3.1 Upper bound

According to the classical energy principle, the inequality

$$\sigma^*\boldsymbol{E}_0\cdot\boldsymbol{E}_0 \le \widetilde{W}\,[\lambda(\cdot)] \tag{15}$$

is hold, where $\widetilde{W}\,[\lambda(\cdot)]$ is the restriction of the energy functional (4) on the class of trial fields (8). The functional $\widetilde{W}$ can be written in the form

$$\widetilde{W}\,[\lambda(\cdot)] = \boldsymbol{E}_0\cdot\widetilde{\boldsymbol{U}}\,[\lambda(\cdot)]\cdot\boldsymbol{E}_0, \tag{16}$$

where

$$\begin{aligned}\widetilde{\boldsymbol{U}}\,[\lambda(\cdot)] = \langle\sigma\rangle\boldsymbol{I} &+ 2\int\nabla\nabla G(\boldsymbol{z})\langle\sigma'(\boldsymbol{0})\lambda'(\sigma(\boldsymbol{z}))\rangle\,\mathrm{d}\boldsymbol{z}\\ &+\langle\sigma\rangle\iint\langle\lambda'(\sigma(\boldsymbol{z}_1))\lambda'(\sigma(\boldsymbol{z}_2))\rangle\nabla\nabla G(\boldsymbol{z}_1)\cdot\nabla\nabla G(\boldsymbol{z}_2)\,\mathrm{d}\boldsymbol{z_1}\mathrm{d}\boldsymbol{z_2}\\ &+\iint\langle\sigma'(\boldsymbol{0})\lambda'(\sigma(\boldsymbol{z}_1))\lambda'(\sigma(\boldsymbol{z}_2))\rangle\nabla\nabla G(\boldsymbol{z}_1)\cdot\nabla\nabla G(\boldsymbol{z}_2)\,\mathrm{d}\boldsymbol{z_1}\mathrm{d}\boldsymbol{z_2},\end{aligned} \tag{17}$$

$\boldsymbol{I}$ being the unit tensor. For the second integral we find

$$\iint\langle\lambda'(\sigma(\boldsymbol{z}_1))\lambda'(\sigma(\boldsymbol{z}_2))\rangle\nabla\nabla G(\boldsymbol{z}_1)\cdot\nabla\nabla G(\boldsymbol{z}_2)\,\mathrm{d}\boldsymbol{z_1}\mathrm{d}\boldsymbol{z_2} = -\int\nabla\nabla G(\boldsymbol{z})\langle\lambda'(\sigma(\boldsymbol{0}))\lambda'(\sigma(\boldsymbol{z}))\rangle\,\mathrm{d}\boldsymbol{z} \tag{18}$$

after four integrations by parts. To arrive at this equation, we relied on the statistical homogeneity of the field $\lambda'(\sigma(\boldsymbol{x}))$, the basic equation $\triangle G(\boldsymbol{x}) = -\delta(\boldsymbol{x})$ for the Green function, $\delta(\boldsymbol{x})$ being the Dirac delta function, and the assumption of no long-range correlations, which is certainly fulfilled for symmetric cell materials.

By choosing $\boldsymbol{E}_0$ to be the units vectors along the coordinate axes, from (15)–(18) and the equation $\mathrm{tr}\,[\nabla\nabla G(\boldsymbol{x})] = \triangle G(\boldsymbol{x})$ we find

$$\sigma^* \le \sigma_u, \quad \sigma_u = U\,[\lambda(\cdot)]\,, \tag{19}$$

where $U = \mathrm{tr}\,\widetilde{\boldsymbol{U}}$, i.e.

$$U\,[\lambda(\cdot)] = \langle\sigma\rangle - \frac{2}{d}\langle\sigma'\lambda'(\sigma)\rangle + \frac{1}{d}\langle\sigma\rangle\langle\lambda'^2(\sigma)\rangle + A_\lambda^{(3)}\langle\sigma'^3\rangle, \tag{20}$$

where

$$A_\lambda^{(3)} = \frac{1}{d\langle\sigma'^3\rangle}\iint\langle\sigma'(\boldsymbol{0})\lambda'(\sigma(\boldsymbol{z}_1))\lambda'(\sigma(\boldsymbol{z}_2))\rangle\nabla\nabla G(\boldsymbol{z}_1) : \nabla\nabla G(\boldsymbol{z}_2)\,\mathrm{d}\boldsymbol{z_1}\mathrm{d}\boldsymbol{z}_2. \tag{21}$$

As already mentioned, the Euler-Lagrange equation for the functional (20) seems difficult to solve for arbitrary medium. If we replace $\lambda(\sigma)$ with $\alpha\sigma$ in (20) and minimize the obtained function of the multiplier α, we will immediately get the upper Beran bound (6).

Since the field $\lambda(\sigma(\boldsymbol{x}))$ has the same statistical properties as the conductivity field $\sigma(\boldsymbol{x})$, for symmetric cell material the third moment involved in the integral in (21) is expressed analogously to that in (13), i.e.

$$\langle\sigma'(\boldsymbol{0})\lambda'(\sigma(\boldsymbol{z}_1))\lambda'(\sigma(\boldsymbol{z}_2))\rangle = \langle\sigma'(\boldsymbol{0})\lambda'^2(\sigma(\boldsymbol{0}))\rangle\, g_3(\boldsymbol{0},\boldsymbol{z}_1,\boldsymbol{z}_2), \tag{22}$$

and therefore $A_\lambda^{(3)} = \langle\sigma'\lambda'^2(\sigma)\rangle G/\langle\sigma'^3\rangle$, where G is the geometric Miller parameter (14).

Let us remember that the energy functional must be considered for functions $\lambda(\sigma)$ subjected to the constraint

$$\langle\lambda'(\sigma(\boldsymbol{x}))\rangle = \lambda(\sigma(\boldsymbol{x})) - \langle\lambda(\sigma(\boldsymbol{x}))\rangle = 0. \tag{23}$$

It turns out to be more convenient to consider the functional (20) with an argument the functional

$$\lambda'(\sigma) = \lambda(\sigma) - \langle\lambda(\sigma)\rangle = \lambda(\sigma) - \int\lambda(\sigma)P(\sigma)\,\mathrm{d}\sigma, \tag{24}$$

instead of the function $\lambda(\sigma)$, and to introduce the Lagrangian functional

$$\begin{aligned}\mathcal{L}\,[\lambda'(\cdot),\Lambda] &= U\,[\lambda'(\cdot)] + \Lambda\langle\lambda'(\sigma(\boldsymbol{x}))\rangle \\ &= \langle\sigma\rangle - \frac{2}{d}\langle\sigma'\lambda'(\sigma)\rangle + \frac{1}{d}\langle\sigma\rangle\langle\lambda'^2(\sigma)\rangle + \langle\sigma'\lambda'^2(\sigma)\rangle G + \Lambda\,\langle\lambda'(\sigma(\boldsymbol{x}))\rangle\end{aligned} \tag{25}$$

with unknown Lagrange multiplier Λ. The Euler-Lagrange equation for this functional is

$$-\frac{1}{d}\sigma' + \frac{1}{d}\langle\sigma\rangle\lambda'(\sigma) + \sigma'\lambda'(\sigma)G + \frac{1}{2}\Lambda = 0, \tag{26}$$

whose averaging when using the constraint (23) gives $\Lambda = -2\,\langle\sigma'\lambda'(\sigma)\rangle G$.

Multiplying now the equation (26) by $\lambda'(\sigma)$ and then averaging it, we can express the quadratic part of the functional (20) with its linear one, and thus we get

$$\min U\,[\lambda(\cdot)] = \langle\sigma\rangle - \frac{1}{d}\langle\sigma'\lambda'(\sigma)\rangle, \tag{27}$$

where $\lambda'(\sigma)$ is the solution of the equation (26). Expressing now the functional $\lambda'(\sigma)$ from this equation, we find

$$\lambda'(\sigma) = \frac{1}{d}\,\frac{\sigma'}{\widetilde{\sigma}} + \frac{1}{\widetilde{\sigma}}\,\langle\sigma'\lambda'(\sigma)\rangle G, \tag{28}$$

where $\widetilde{\sigma} = \langle\sigma\rangle/d + G\sigma'$. After averaging now the equation (28) under the constraint (23), we get the linear term

$$\langle\sigma'\lambda'(\sigma)\rangle = \frac{1}{d}\,\frac{\langle\sigma'/\widetilde{\sigma}\rangle}{\langle 1/\widetilde{\sigma}\rangle G}. \tag{29}$$

Finally, taking into account (19), (27), and (29), for the best upper bound on σ^* we obtain the formula

$$\sigma_u = \langle\sigma\rangle\left\{1 + (1-\alpha_u(G))\;\frac{B_u}{1 - L_u(G)\,B_u}\right\}, \tag{30}$$

where

$$B_u = \left\langle \frac{\sigma(\boldsymbol{x}) - \langle\sigma\rangle}{\langle\sigma\rangle + L_u(G)\,(\sigma(\boldsymbol{x}) - \langle\sigma\rangle)} \right\rangle, \quad \alpha_u(G) = \frac{d^2G - 1}{d^2G}, \quad L_u(G) = dG. \tag{31}$$

3.2 Lower bound

To derive lower bounds on the effective conductivity σ^* we use the complementary energy principle, according to which among all the statistically homogeneous solenoidal trial fields $\widehat{\boldsymbol{J}}(\boldsymbol{x})$ with prescribed constant-average value, $\langle\widehat{\boldsymbol{J}}(\boldsymbol{x})\rangle = \boldsymbol{J}_0$, the functional

$$W\big[\widehat{\boldsymbol{J}}(\cdot)\big] = \frac{1}{2}\langle\rho(\boldsymbol{x})\widehat{\boldsymbol{J}}(\boldsymbol{x})\cdot\widehat{\boldsymbol{J}}(\boldsymbol{x})\rangle \tag{32}$$

is stationary and minimum for the true current density field $\boldsymbol{J}(\boldsymbol{x})$ in the medium, so that $\min W = \frac{1}{2}\,\rho^* J_0^2$, $J_0^2 = \boldsymbol{J}_0\cdot\boldsymbol{J}_0$, where $\rho(\boldsymbol{x}) = 1/\sigma(\boldsymbol{x})$ and $\rho^* = 1/\sigma^*$ are the local and effective resistivity of the medium, respectively.

For this variational principle, we introduce the trial fields

$$\begin{aligned}\widehat{\boldsymbol{J}}(\boldsymbol{x}) &= \boldsymbol{J}_0 + \int \nabla\times[\boldsymbol{J}_0\times\nabla G(\boldsymbol{x}-\boldsymbol{y})]\,\mu'(\sigma(\boldsymbol{y}))\,\mathrm{d}\boldsymbol{y} \\ &= \boldsymbol{J}_0 + \mu'(\sigma(\boldsymbol{x}))\boldsymbol{J}_0 + \boldsymbol{J}_0\cdot\int\nabla\nabla G(\boldsymbol{x}-\boldsymbol{y})\mu'(\sigma(\boldsymbol{y}))\,\mathrm{d}\boldsymbol{y}\end{aligned} \tag{33}$$

– the counterpart of the fields (8), where $\mu(\sigma)$ is a varying non-random function of the achieved conductivity values and the field $\mu'(\sigma(\boldsymbol{x}))$ is the fluctuation of the random field $\mu(\sigma(\boldsymbol{x}))$. The derivation of the best lower bound on σ^* is similar to that of the upper bound, so we will omit some details.

After substituting the trial field (33) into the functional (32), we will get an expression for an upper bound ρ_u on ρ^*:

$$\rho^* \le \rho_u, \quad \rho_u = U\left[\mu(\cdot)\right], \tag{34}$$

where

$$U\left[\mu(\cdot)\right] = \langle\rho\rangle + 2\,\frac{d-1}{d}\langle\rho\mu'(\sigma)\rangle + \frac{d-2}{d}\langle\rho\mu'^2(\sigma)\rangle + \frac{1}{d}\langle\rho\rangle\langle\mu'^2(\sigma)\rangle + A_\mu^{(3)}\langle\rho'^3\rangle \tag{35}$$

is a functional involving the statistical parameter

$$A_\mu^{(3)} = \frac{1}{d\langle\rho'^3\rangle}\iint\langle\rho'(\mathbf{0})\mu'(\sigma(\boldsymbol{z}_1))\mu'(\sigma(\boldsymbol{z}_2))\rangle\nabla\nabla G(\boldsymbol{z}_1) : \nabla\nabla G(\boldsymbol{z}_2)\,\mathrm{d}\boldsymbol{z}_1\mathrm{d}\boldsymbol{z}_2. \tag{36}$$

If we replace $\mu(\sigma)$ with $\alpha\sigma$ in (35) and minimize the obtained quadratic function of α, we will get the lower bound $\sigma_\ell = 1/\rho_u$ on σ^* of Beran [2].

For symmetric cell materials, $A_\mu^{(3)} = \langle\rho'\mu'^2(\sigma)\rangle G/\langle\rho'^3\rangle$, where G is again the geometric Miller parameter (14). The corresponding Lagrangian functional

$$\mathcal{L}\left[\mu'(\cdot),\Lambda\right] = U\left[\mu'(\cdot)\right] + \Lambda\langle\mu'(\sigma(\boldsymbol{x}))\rangle, \tag{37}$$

considered under the the constraint $\langle\mu'(\sigma(\boldsymbol{x}))\rangle = 0$, leads to finding the following equation for the functional $\mu'(\sigma)$:

$$\frac{d-1}{d}\rho' + \frac{d-1}{d}\langle\rho\rangle\mu'(\sigma) + \left(\frac{d-2}{d} + G\right)\left[\rho'\mu'(\sigma) - \langle\rho'\mu'(\sigma)\rangle\right] = 0. \tag{38}$$

For the best upper bound ρ_u on ρ^*, we find

$$\rho_u = \langle\rho\rangle + \frac{(d-1)^2}{d(dG+d-2)}\,\frac{\langle\rho'/\widetilde{\rho}\rangle}{\langle 1/\widetilde{\rho}\rangle}, \tag{39}$$

where $\widetilde{\rho} = (d-1)\langle\rho\rangle + (dG+d-2)\rho'$. Finally, for the best lower bound $\sigma_\ell = 1/\rho_u$ on σ^*, we obtain the formula

$$\sigma_\ell = \sigma_H\left\{1 + (1-\alpha_\ell(G))\;\frac{B_\ell}{1-(L_\ell(G)-\alpha_\ell(G))\;B_\ell}\right\}, \tag{40}$$

where

$$B_\ell = \left\langle\frac{\sigma(\boldsymbol{x})-\sigma_H}{\sigma_H + L_\ell(G)\,(\sigma(\boldsymbol{x})-\sigma_H)}\right\rangle, \quad \alpha_\ell(G) = \frac{d^2G-1}{d(dG+d-2)}, \quad L_\ell(G) = \frac{1-dG}{d-1}, \tag{41}$$

and $\sigma_H = 1/\langle\rho\rangle = \langle\sigma^{-1}\rangle^{-1}$ is the harmonic mean of the conductivity σ.

4 Concluding remarks

The parameters B_u and B_ℓ involved in formulas (30) and (40) for the bounds σ_u and σ_ℓ, respectively, are easy to calculate for a N-phase medium and some simple probability densities $P(\sigma)$ (e.g., for a uniform or triangular distribution of σ). As expected, for a three-phase cell material, the new formulas (30) and (40) are equivalent to the explicit formulas given in [3]. In the case of spherical cell material [5–8], $G = 1/d^2$, whence we find $\alpha_u = \alpha_\ell = 0$ and $L_u = L_\ell = 1/d$ – the depolarization factor of a sphere. Then the quantities B_u and B_ℓ represent the average polarizability of the spheres of the material with respect to the medium with conductivity the arithmetic mean $\sigma_A = \langle\sigma\rangle$ and the geometric mean σ_H, respectively. For cells with a non-spherical shape, polarizability is a tensor and therefore cannot be so clearly reflected in our formulas for macroscopic isotropic materials. Consideration of statistical anisotropic media will lead to a clearer presence of cell polarizability in the corresponding formulas for effective conductivity. The relevant study will be done elsewhere.

REFERENCES:

[1] Brown, W. F., Solid mixture permittivities, Journal of Chemical Physics **23** (1955), 1514 – 1517.

[2] Beran, M.J., Use of the variational approach to determine bounds for the effective permittivity in random media. Nuovo Cimento **38** (1965), 771–782.

[3] Phan-Thien, N., Milton, G.W., New bounds on the effective thermal conductivity of N-phase materials. Proc. R. Soc. Lond. A **380** (1982), 333–348.

[4] Pham, D.C., Bounds on the effective conductivity of statistically isotropic multicomponent materials and cell polycristals. J. Mech. Phys.Solids **59** (2011), 497–510.

[5] Miller, M. N., Bounds for effective electrical, thermal, and magnetic properties ofheterogeneous materials. J. Math. Phys. **10** (1969), 1988 – 2004.

[6] Hori, M., Statistical theory of effective electrical, thermal, and magnetic properties ofrandom heterogeneous materials. I. Perturbation expansions for the effective permittivity of cell materials. J. Math. Phys. **14** (1973), 514–523.

[7] Milton, G. W., The theory of composites, Cambridge University Press, Cambridge, England (2001).

[8] Torquato, S., Random Heterogeneous Materials: Microstructure and Macroscopic Properties. Springer-Verlag (2002).