ГОДИШНИК НА СОФИЙСКИЯ УНИВЕРСИТЕТ "СВ. КЛИМЕНТ ОХРИДСКИ" ФАКУЛТЕТ ПО МАТЕМАТИКА И ИНФОРМАТИКА Том 95

ANNUAIRE DE L'UNIVERSITE DE SOFIA "ST. KLIMENT OHRIDSKI"

FACULTE DE MATHEMATIQUES ET INFORMATIQUE

Tome 95

A GENERALIZATION OF THE VOIGT-REUSS BOUNDS FOR A BINARY MEDIUM

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In the present note a certain generalization of the well-known Voigt-Reuss bounds on the effective conductivity κ^* of a binary medium is proposed. For a fixed binary constitution the scalar function $f(\alpha)$ that gives the undimensional effective conductivity as a function of the ratio α of the constituents conductivities is considered. Certain inequalities for the derivative $f'(\alpha)$ of this function, which include α , $f(\alpha)$ and the volume fractions of the constituents, are derived. The inequalities are sharp if these fractions are solely known. More precisely, they turn into equalities for the familiar laminate media loaded along and across the layers. The Voigt and Reuss bounds on κ^* follow from the proposed inequalities, but the latter are stronger than the former bounds, since estimates are put here on the rate at which the effective conductivity changes when the constituents properties are varied at a fixed binary constitution of the medium. It is in this sense, namely, when it is claimed that our inequalities generalize the Voigt-Reuss bounds.

Keywords: random two-phase media, variances, effective properties, bounds on the effective conductivity

2000 MSC: 60G60, 60H15

1. INTRODUCTION

The aim of this note is to report some inequalities, concerning the rate of variation of the effective properties of a two-phase medium when, at a fixed random constitution, the properties of the constituents are varied. The heat conduction context is chosen for the sake of simplicity. The random constitution is assumed statistically homogeneous and isotropic.

Let κ_1 and κ_2 be the conductivities of the constituents, $\kappa_1, \kappa_2 > 0$. The random conductivity field $\kappa(x)$ of the medium takes then the values κ_1 or κ_2 , depending on whether x lies in the phase '1' or '2', respectively. If E denotes the prescribed macroscopic temperature gradient imposed upon the medium, the governing equations of the problem, at the absence of body sources, read

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

where $E(x) = \nabla \theta(x)$. Eq. (1.1a) is supplied with the condition

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

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

Having solved somehow the random problem (1.1), one can evaluate all multipoint moments of the temperature field $\theta(x)$ and the joint moments of $\theta(x)$ and $\kappa(x)$ by means of the known moments of the conductivity field, see [1, 9]. In particular, among the joint moments, the simplest one-point moment of the flux q(x) defines the well-known effective conductivity κ^* of the medium through the relation

$$Q = \langle q(x) \rangle = \langle \kappa(x)E(x) \rangle = \kappa^* E \tag{1.2}$$

(assuming statistical homogeneity and isotropy).

Note that the definition (1.2) of the effective conductivity κ^* reflects the "homogenization" of the problem under study in the sense that from a macroscopic point of view, when only the macroscopic values of the flux and temperature gradient are of interest, the medium behaves as if it were homogeneous with a certain macroscopic conductivity κ^* . This interpretation explains why κ^* and its counterparts, say, the effective elastic moduli, have been extensively studied in the literature on homogenization, see, e.g. [8, 14] et al., as well as the recent survey [10]. A well-known fact is to be only recalled, namely that κ^* can be defined also "energetically":

$$\kappa^* E^2 = \langle \kappa(\mathbf{x}) | \nabla \theta(\mathbf{x}) |^2 \rangle . \tag{1.3}$$

Besides κ^* , other statistical characteristics of the fields E(x) and q(x) deserve attention and, above all, the (undimensional) variances of these fields, defined as follows:

$$\sigma_e^2 = \frac{\left\langle E'(x)^2 \right\rangle}{E^2} \,, \quad \sigma_q^2 = \frac{\left\langle |q'(x)|^2 \right\rangle}{Q^2} \,. \tag{1.4}$$

The primes denote in what follows the fluctuating parts of the respective random fields, so that, in particular, E'(x)f = E(x) - E, and hence $\langle E'(x) \rangle = 0$.

It is to be noted immediately that for any two-point medium the variances σ_e^2 and σ_q^2 are simply interconnected:

$$\sigma_q^2 = -\frac{\kappa_1 \kappa_2}{\kappa^{*2}} \sigma_e^2 - \frac{(\kappa^* - \kappa_1)(\kappa^* - \kappa_2)}{\kappa^{*2}}.$$
 (1.5)

This formula, derived in [11], is a straightforward consequence of the fact that the medium under study is binary and hence the field $\kappa(x)$ takes the values κ_1 or κ_2 solely.

The variances (1.4) provide us with useful information about the deviation of the random fields under studies from their mean values. Also, they are connected to the mean energy of the appropriate fields, accumulated within the phases. That is why they have attracted some attention in the literature on heterogeneous media.

To the best of the authors' knowledge, an investigation of the variances, in addition to the effective properties in the scalar conductivity context, has been initiated by Beran et al., [2, 4, 3]. In particular, Beran [2] has obtained bounds on the variances through the effective properties. The Beran estimates are quite crude and this is inevitable since they are applicable to any statistically homogeneous and isotropic medium. More restrictive bounds are derived in [11], but only for dispersions of spheres, correct to the order "square of concentration."

Note that an application of such variances, concerned with the deviation from the Hooke law in heterogeneous materials, can be found in the recent authors' paper [12].

The above mentioned results of Beran indicate that there may exist more intimate connection between variances and effective properties. Indeed, as shown first by Bergman [5], see also [7, 13], the variance is simply connected to the derivatives of the effective conductivity $\kappa^* = \kappa^*(\kappa_1, \kappa_1)$, treated as a function of the material properties κ_1 and κ_2 of the constituents in a binary medium, at a fixed random constitution. This is an interesting and important result, but its practical application is limited by the fact that very rarely rigorous analytical formulae for $\kappa^*(\kappa_1, \kappa_1)$ are known for realistic random constitution. Rigorous bounds on $\kappa^*(\kappa_1, \kappa_1)$ are well-known, of course, but they obviously cannot supply any estimates for the above-mentioned derivatives.

It turns out that the variances (1.4) can be simply represented by means of κ^* and its derivatives $\partial \kappa^*/\partial \kappa_1$ and $\partial \kappa^*/\partial \kappa_2$ with respect to the constituents properties, having fixed the random constitution. The appropriate formulae are direct consequences of the Bergman formula [5], which will be rederived in Section 2. In turn, this formula will yield certain inequalities between the effective conductivities κ^* and its derivatives $\partial \kappa^*/\partial \kappa_1$ and $\partial \kappa^*/\partial \kappa_2$ (Section 3). These inequalities, when transformed into dimensionless form, have as a consequence the Voigt and Reuss bounds (Section 4). Both these bounds are, to say the least, well-known. The important point, however, is that the inequalities derived here bound not only the effective properties, but also the rate of their change when the constituents properties are varied. The proposed inequalities are closely connected as well to the convexity of the function $f(\alpha)$, discussed in Section 5. The latter easily follows from the spectral representation of $f(\alpha)$, due again to Bergman [5]. A certain appealing geometrical interpretation of the Voigt bound is proposed as a consequence of the convexity of the function $f(\alpha)$, namely, that this function should lie below each of its tangents and, in particular, below its tangent, drawn at the point $\alpha = 1$.

2. THE BERGMAN FORMULA

For the sake of completeness, we shall provide here a derivation of the Bergman formula. It is a bit more rigorous than the original one due to Bergman [5], since ensemble (instead of volume) averaging will be utilized. In the papers [7, 13], where the same formula has been rederived later on, volume averaging is used, similarly to the original Bergman reasoning.

The starting point is the energy definition (1.3) of the effective conductivity κ^* . Let us change the conductivity field of the medium, $\kappa(x)$, by the infinitesimal quantity

$$\delta(\mathbf{x}) = \chi_1(\mathbf{x})\delta\kappa_1 + \chi_2(\mathbf{x})\delta\kappa_2, \tag{2.1}$$

where $\chi_1(\mathbf{x})$ and $\chi_2(\mathbf{x})$ are the characteristic functions of the regions, occupied by the constituents '1' and '2', respectively. Then, at fixed $\mathbf{E} = \nabla \theta(\mathbf{x})$, the field $\theta(\mathbf{x})$ will change by $\delta \theta(\mathbf{x})$ and the effective conductivity – by $\delta \kappa^*$. According to (1.3), we have

$$(\kappa^* + \delta\kappa^*) E^2 = \langle (\kappa(x) + \delta\kappa(x)) | \nabla \theta(x) + \nabla \delta \theta(x) |^2 \rangle$$

= $\langle \kappa(x) | \nabla \theta(x) |^2 \rangle + \langle \delta\kappa(x) | \nabla \theta(x) |^2 \rangle + 2 \langle \kappa(x) \nabla \theta(x) \cdot \nabla \delta \theta(x) \rangle$, (2.2)

having neglected terms of order $(\delta \kappa)^2$. The first term in the right-hand side of (3.2) equals $\kappa^* E^2$, see (1.3), and the underlined term there vanishes, since

$$\nabla \cdot \langle \kappa(\boldsymbol{x}) \delta \theta(\boldsymbol{x}) \nabla \theta(\boldsymbol{x}) \rangle = \langle \delta \theta(\boldsymbol{x}) \nabla \cdot (\kappa(\boldsymbol{x}) \nabla \theta(\boldsymbol{x})) \rangle$$
$$+ \langle \kappa(\boldsymbol{x}) \nabla \theta(\boldsymbol{x}) \cdot \nabla \delta \theta(\boldsymbol{x}) \rangle = 0,$$

having taken (1.1a) into account. The reason is that the field $\kappa(x)\delta\theta(x)\nabla\theta(x)$ is statistically homogeneous and therefore its mean value is constant.

Hence

$$\delta \kappa^* E^2 = \langle \delta \kappa(\mathbf{x}) | \nabla \theta(\mathbf{x}) |^2 \rangle$$

= $\delta \kappa_1 \langle \chi_1(\mathbf{x}) | \nabla \theta(\mathbf{x}) |^2 \rangle + \delta \kappa_2 \langle \chi_2(\mathbf{x}) | \nabla \theta(\mathbf{x}) |^2 \rangle$.

The latter implies

$$\frac{\partial \kappa^*}{\partial \kappa_i} = \frac{1}{E^2} \left\langle \chi_i(\boldsymbol{x}) |\nabla \theta(\boldsymbol{x})|^2 \right\rangle, \quad i = 1, 2, \tag{2.3}$$

which is exactly the Bergman formula [5]. It obviously means that the mean value of the temperature gradient square within the constituent 'i' is proportional to the derivative $\partial \kappa^*/\partial \kappa_i$, i = 1, 2.

3. THE INEQUALITIES FOR THE DERIVATIVES $\partial \kappa^*/\partial \kappa_i$

Note first that

$$\begin{split} \frac{1}{E^2} \left\langle |\boldsymbol{E}(\boldsymbol{x})|^2 \right\rangle &= \frac{1}{E^2} \left\langle \chi_1(\boldsymbol{x}) |\boldsymbol{E}(\boldsymbol{x})|^2 \right\rangle \\ &+ \frac{1}{E^2} \left\langle \chi_2(\boldsymbol{x}) |\boldsymbol{E}(\boldsymbol{x})|^2 \right\rangle = \frac{\partial \kappa^*}{\partial \kappa_1} + \frac{\partial \kappa^*}{\partial \kappa_2} \end{split}$$

and hence

$$\sigma_e^2 = \frac{\left\langle |E'(x)|^2 \right\rangle}{E^2} = \frac{1}{E^2} \left[\left\langle |E(x)|^2 \right\rangle - 2E \cdot \left\langle E(x) \right\rangle + E^2 \right]$$

$$= \frac{1}{E^2} \left\langle |E(x)|^2 \right\rangle - 1 = \frac{\partial \kappa^*}{\partial \kappa_1} + \frac{\partial \kappa^*}{\partial \kappa_2} - 1.$$
(3.1)

Formula (3.1) provides us with the interconnection between the variance of the temperature gradient and the partial derivatives of the effective conductivity considered, at a fixed two-phase geometry, as a function of the constituents conductivities.

To recast (3.1) into dimensionless form, recall the obvious fact that $\kappa^* = \kappa^*(\kappa_1, \kappa_2)$ is a homogeneous function of first order, i.e.

$$\kappa^*(\lambda \kappa_1, \lambda \kappa_2) = \lambda \kappa^*(\kappa_1, \kappa_2), \quad \forall \ \lambda > 0.$$

This fact allows us to apply the Euler formula

$$\kappa_1 \frac{\partial \kappa^*}{\partial \kappa_1} + \kappa_2 \frac{\partial \kappa^*}{\partial \kappa_2} = \kappa^*,$$

i.e.

$$\frac{\partial \kappa^*}{\partial \kappa_1} = \frac{\kappa^*}{\kappa_1} - \frac{\kappa_2}{\kappa_1} \frac{\partial \kappa^*}{\partial \kappa_2}.$$
 (3.2)

Let us now fix the conductivity κ_1 of the first of the constituents and introduce the dimensionless variables

$$\alpha = \frac{\kappa_2}{\kappa_1}, \quad \alpha \in (0, \infty),$$

$$f(\alpha) = \frac{\kappa^*}{\kappa_1}, \quad f(\alpha) \ge 0.$$
(3.3)

Here $f(\alpha)$, for the fixed two-phase geometry under discussion, depends on the dimensionless ratio α solely. Using (3.2) and (3.3) into (3.1) gives

$$\sigma_e^2 = \frac{\partial \kappa^*}{\partial \kappa_1} + \frac{\partial \kappa^*}{\partial \kappa_2} - 1$$

$$= \frac{\kappa^*}{\kappa_1} - \left(\frac{\kappa_2}{\kappa_1} - 1\right) \frac{\partial (\kappa^*/\kappa_1)}{\partial (\kappa_2/\kappa_1)} - 1,$$
(3.4)

$$\sigma_{\epsilon}^2 = f(\alpha) - 1 - f'(\alpha)(\alpha - 1). \tag{3.5}$$

Hence any theory that predicts $f(\alpha)$, i.e. the effective conductivity as a function of α automatically predicts the variance σ_e^2 , since (3.4), as a consequence of the Bergman formula (2.3), is an *exact* relation.

The situation with the variance σ_q^2 of the heat flux is fully similar. In this case we should combine (1.5) and (3.6). The final result reads

$$\sigma_q^2 = \frac{\alpha(\alpha - 1)f'(\alpha) - f(\alpha)(f(\alpha) - 1)}{f^2(\alpha)}.$$
 (3.6)

It remains now to note that both variances σ_e^2 and σ_q^2 are nonnegative, as it follows from their definitions (1.3). Together with (3.5) and (3.6), this obvious fact yields the inequalities

$$\frac{f(\alpha)\left(f(\alpha)-1\right)}{\alpha} \le f'(\alpha)(\alpha-1) \le f(\alpha)-1. \tag{3.7}$$

Formula (3.7) is our generalization of the Voigt-Reuss bounds for a two-phase heterogeneous medium. The reason to call it generalization will become clear in the next section, where two basic consequences of (3.7) will be derived, namely, both the Voigt and Reuss bounds on the effective conductivity.

4. SOME CONSEQUENCES OF INEQUALITY (3.7)

Recall first the well-known perturbation expansion of the effective conductivity

$$\frac{\kappa}{\langle \kappa \rangle} = 1 - a_2 \left(\frac{\kappa}{\langle \kappa \rangle}\right)^2 + \cdots, \tag{4.1}$$

due to Brown [6]. Here $\langle \kappa \rangle = c_1 \kappa_1 + c_2 \kappa_2$, $[\kappa] = \kappa_2 - \kappa_1$ and c_i is the volume fraction of the constituent 'i', i = 1, 2, so that $c_1 + c_2 = 1$. In (4.1) $a_2 = \frac{1}{3}c_1c_2$, but this fact will not be needed here, since it affects only the $(\alpha - 1)^2$ -term in the Taylor expansion (4.2) below.

In the dimensionless variables (3.3), Eq. (4.1) is recast as

$$f(\alpha) = \frac{\kappa^*}{\kappa_1} = 1 + c_2(\alpha - 1) + o(|\alpha - 1|), \quad |\alpha - 1| \ll 1.$$
 (4.2)

Assume now that $\alpha > 1$. Rewrite the right-hand side inequality in (3.7) in the form

$$\frac{\mathrm{d}f(\alpha)}{f(\alpha)-1} \le \frac{\mathrm{d}\alpha}{\alpha-1}, \quad \alpha > 1,$$

and integrate the latter from $1 + \varepsilon$ to α , $\varepsilon > 0$. This gives

$$\ln \frac{f(\alpha) - 1}{f(1 + \varepsilon) - 1} \le \ln \frac{\alpha - 1}{\varepsilon}$$

and hence

$$f(\alpha) - 1 \le \left[f(1+\varepsilon) - 1 \right] \frac{\alpha}{\varepsilon}$$
.

Choosing now $\varepsilon \to 1 + 0$ in the latter inequality yields

$$f(\alpha) \le 1 + c_2(\alpha - 1), \tag{4.3}$$

having taken into account (4.2) as well. Repeating the above reasoning for α < 1 produces the same result (4.3). Hence (4.3) holds for arbitrary values of the constituents conductivities κ_1 and κ_2 . Using the definition (3.3) of α and $f(\alpha)$ allows us to recast (4.3) as

$$\kappa^* \le \kappa^{\mathsf{v}}, \quad \kappa^{\mathsf{v}} = c_1 \kappa_1 + c_2 \kappa_2, \tag{4.4}$$

and this is the familiar Voigt estimate upon the effective conductivity of the medium.

The treatment of the left-hand side inequality in (3.7) is fully similar. In this case, at $\alpha > 1$, we have

$$\frac{\mathrm{d}\alpha}{\alpha(\alpha-1)} \le \frac{\mathrm{d}f(\alpha)}{f(\alpha)(f(\alpha)-1)} \,. \tag{4.5}$$

An elementary integration of both sides of (4.5) over the interval $(1 + \varepsilon, \alpha)$ gives

$$\ln \frac{\alpha - 1}{\alpha} - \ln \frac{\varepsilon}{1 + \varepsilon} \le \ln \frac{f(\alpha) - 1}{f(\alpha)} - \ln \frac{f(1 + \varepsilon) - 1}{f(1 + \varepsilon)},$$

which simplifies as

$$\frac{c_2(\alpha-1)}{\alpha} \le \frac{f(\alpha)-1}{f(\alpha)},$$

having again taken (4.2) into account. Hence

$$\frac{\alpha}{\alpha - c_2(\alpha - 1)} \le f(\alpha).$$

Recalling the definition (3.3), the latter can be recast as

$$\kappa^{\mathrm{r}} \le \kappa^{*}, \quad \kappa^{\mathrm{r}} = \left(\frac{c_{1}}{\kappa_{1}} + \frac{c_{2}}{\kappa_{2}}\right)^{-1}$$
(4.6)

and this is just the familiar Reuss bound on the effective conductivity.

It is important to point out that the inequalities (3.7) are *sharp*, i.e. they cannot be improved provided only the volume fractions c_1 , c_2 are known.

Indeed, if the temperature gradient is along the layers of a laminate medium, the Voigt approximation (4.4) provides the exact value of the effective conductivity and hence

$$f(\alpha) = 1 + c_2(\alpha - 1).$$

The latter function turns the right-hand side of (3.7) into equality.

Similarly, if the temperature gradient is across the layers of a laminate medium, the Reuss value (4.6) represents exactly the effective conductivity. Then

$$f(\alpha) = \frac{\alpha}{\alpha - c_2(\alpha - 1)}$$

and this function assures equality sign in the left-hand side of (3.7).

5. DISCUSSION

Let us point out first that, since f(1) = 1,

$$f(\alpha) - 1 = f'(\xi)(\alpha - 1), \tag{5.1}$$

where $\xi \in (1, \alpha)$ or $\xi \in (\alpha, 1)$, depending on whether $\alpha > 1$ or $\alpha < 1$, respectively. (This is the well-known Lagrange theorem from the elementary calculus.) Together with the right-hand side of (3.7), Eq. (5.1) implies that for each α there exists an "intermediate" $\xi \in (1, \alpha)$ or $\xi \in (\alpha, 1)$, depending again on whether $\alpha > 1$ or $\alpha < 1$, such that

$$f'(\alpha) \le f'(\xi), \quad \xi < \alpha.$$
 (5.2)

From (5.2) it follows that f''(1) < 0 and hence the function $f(\alpha)$ is convex in a certain vicinity of $\alpha = 1$. (This is indeed so, because the coefficient a_2 , proportional to f''(1), in the Taylor expansion of $f(\alpha)$ about $\alpha = 1$ is negative, see the beginning of Section 4.) We do not know, however, whether (5.2) suffices to claim that the function $f(\alpha)$ is convex globally, i.e. $f''(\alpha) \leq 0$ on the whole semiaxis $\alpha \in (0, \infty)$.

However, the convexity of $f(\alpha)$ easily follows from the well-known spectral (pole) representation¹

$$f(\alpha) = 1 - F(s), \quad F(s) = \sum_{n} \frac{B_n}{s - s_n},$$

 $s = \frac{1}{1 - \alpha}, \quad B_n \ge 0,$ (5.3)

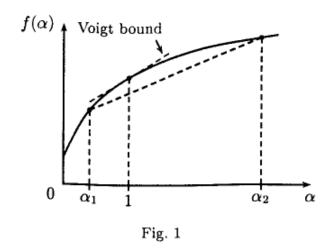
due again to Bergman [5] (see also [9]). Indeed, a straightforward differentiation of (5.3) shows that $f''(\alpha) \leq 0$, $\forall \alpha \in (0, \infty)$ (recall that $B_n \geq 0$).

The convexity of $f(\alpha)$ means geometrically that the function lies below each of its tangents, i.e.

$$f(\alpha) \le f'(\alpha_0) (\alpha - \alpha_0) + f(\alpha_0), \quad \forall \alpha, \alpha_0 \in (0, \infty).$$
 (5.4)

In particular, the Voigt bound can be interpreted geometrically as the obvious fact that the function $f(\alpha)$ falls below its tangent, drawn at the point $\alpha = 1$, see Fig. 1.

¹The authors thank D. Bergman for this observation (in a private communication).



Obviously, the inequality (5.4) reduces to the right-hand side of (3.7) if $\alpha_0 = 1$. It is clear, however, that (5.4) is of little practical use, because $f(\alpha_0)$ and $f'(\alpha_0)$ are unknown, in general, unless $\alpha_0 = 1$.

Recall also that the convexity of $f(\alpha)$ can be alternatively defined by the requirement that its graph in the interval (α_1, α_2) lies higher than the chord between the points $(\alpha_1, f(\alpha_1))$ and $(\alpha_2, f(\alpha_2))$, see Fig. 1. In other words, the following inequality holds:

$$\frac{f(\alpha_1) - f(\alpha_2)}{\alpha_1 - \alpha_2} \alpha + \frac{\alpha_1 f(\alpha_2) - \alpha_2 f(\alpha_1)}{\alpha_1 - \alpha_2} \le f(\alpha), \tag{5.5}$$

 $\forall \alpha \in (\alpha_1, \alpha_2), \alpha_1, \alpha_2 \in (0, \infty)$. Hence (5.5) provides a certain lower bound on the effective conductivity provided we have somehow measured the values of the latter for two given values α_1 , α_2 of the ratio of the constituents conductivities. Observe, however, that (5.5) is a lower bound only in the interval $\alpha \in (\alpha_1, \alpha_2)$. Outside this interval (5.5) becomes an upper bound on $f(\alpha)$.

6. CONCLUDING REMARKS

We have derived certain inequalities, cf. (3.7), for the rate of change $f'(\alpha)$ of the dimensionless effective conductivity $f(\alpha)$ of a binary medium when the constituents properties are varied at fixed random constitution. The inequalities are of first order, in the sense that they include, besides $f(\alpha)$ and the dimensionless ratio α of the constituents conductivities, only the volume fractions of the constituents. They indicate that the above-mentioned rate of change $f'(\alpha)$ cannot be arbitrary for a realistic binary constitution. It is rather "guided" by the value $f(\alpha)$ of the effective conductivity at any given α . Presumably, higher-order counterparts of the inequalities (3.7) exist as well. They should provide tighter estimates for $f'(\alpha)$ at the expense of including the appropriate higher-order statistical information for the medium.

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Received December 10, 2001 Revised January 7, 2002

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