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Abstract

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Three-point bounds and other estimates for strongly nonlinear composites

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A *variational* procedure due to Ponte Castañeda *et al.* [Phys. Rev. B **46**, 4387 (1992)] is used to determine three-point bounds and other types of estimates for the effective response of strongly nonlinear composites with random microstructures. The variational procedure makes use of estimates for the effective properties of “linear comparison composites” to generate corresponding estimates for nonlinear composites. Several equivalent forms of the variational procedure are derived. In particular, it is shown that the mean-field theory of Wan *et al.* [Phys. Rev. B **54**, 3946 (1996)], which also makes use of a linear comparison composite, together with a certain “decoupling approximation,” leads to results that are precisely identical to those that can be obtained from the earlier variational procedure. Finally, three-point bounds and other estimates are computed for power-law composites with cell-type microstructures, and the results are compared with random resistor network simulations available from the literature. [S0163-1829(98)03619-4]

I. INTRODUCTION

The computation of the effective properties of inhomogeneous media is a classical problem that has attracted the attention of numerous investigators in many different fields. However, most of the efforts to date have concentrated on the effective properties of *linear* systems. In particular, the problem of estimating the effective conductivity (or, analogously, the effective dielectric constant) of a linear composite conductor (or dielectric) has played a central role in this endeavor.^{1,2} By comparison, the study of *nonlinear* heterogeneous systems has not received nearly as much attention,³ in spite of its relative importance in the context of many different physical phenomena, including dielectric breakdown, burning out of fuses, and laser phenomena. Additional examples could be given in the realms of electric, magnetic, and other physical and mechanical properties of matter.

The aim of this paper is to make use of a *variational procedure*, originally developed by Ponte Castañeda and co-workers,^{5,4} to obtain three-point bounds and other estimates for strongly nonlinear composite conductors. The variational procedure allows the estimation of the effective energy-density function of a nonlinear composite with a given random microstructure in terms of the effective properties of a fictitious “linear comparison composite” with precisely the same microstructure as the nonlinear composite. This means that *any* estimate for the effective properties of the “linear comparison composite” can be used to generate a corresponding estimate for the nonlinear composite. In particular, explicit estimates of the Maxwell-Garnett⁶ (MGA) and effective medium⁷ (EMA) type have been given⁵ for power-law dielectrics with statistically isotropic distributions of perfectly conducting and insulating inclusions. Similarly, MGA-type estimates have also been given⁴ for two-phase power-law composites in terms of simple one-dimensional optimization problems (which are equivalent to the solution of one nonlinear algebraic equation). Because of a well-known connection between the linear MGA estimates and Hashin-Shtrikman⁸ bounds, the nonlinear MGA-type esti-

mate corresponding to the Hashin-Shtrikman lower bound for the effective conductivity could also be interpreted as a rigorous lower bound for the effective nonlinear conductivity of the class of all two-phase statistically isotropic power-law composites. In addition, it has been shown^{5,4} that the variational procedure is capable of reproducing exactly the estimates of Zeng, Bergman, Hui, and Stroud¹⁰ for *weakly nonlinear* composites, as well as the Hashin-Shtrikman bounds of Talbot and Willis⁹ for composites with arbitrary nonlinearity. However, the variational procedure^{5,4} is in agreement *only* to first order in contrast with the small-contrast perturbation results of Blumenfeld and Bergman¹¹ for strongly nonlinear composites. A general procedure that reproduces the small-contrast perturbation estimates of Blumenfeld and Bergman¹¹ exactly to second-order in the contrast has been proposed recently by Ponte Castañeda and co-workers.¹² Unfortunately, this “second-order” procedure cannot be used to obtain bounds.

In this paper, new three-point bounds of the Beran¹³ type are given for power-law composites. For this purpose, use is made of a simplified form of the Beran bounds, due to Milton.¹⁴ The results are compared with the random resistor network (RRN) simulations of Lee and Yu,¹⁵ by making use of the result of Miller¹⁶ to estimate the three-point Milton parameters for composites with *symmetric cell* microstructures, which are akin to the RRN microstructures. Comparisons with the EMA, MGA and other bounds and estimates are also given.

A secondary aim of this work is to make connections with the recent work of Hui and collaborators,¹⁷ who also developed estimates for strongly nonlinear composites making use of corresponding estimates for linear composites. Their approach involves a certain decoupling approximation,^{18,19} together with a well-known result^{2,3} to estimate the second moment of the electric field in the linear comparison composite. Thus, it is shown that the approach of Hui and collaborators^{17,18} in general leads to *precisely the same* estimates as the variational approach of Ponte Castañeda and co-workers^{5,4} (and not just to second-order in the contrast, as

noted by Hui and collaborators¹⁷). One distinct advantage of the variational approach of Ponte Castañeda and co-workers^{5,4} is that the resulting estimates are given in the form of bounds for the effective energy function of the composite, and thus implicitly contains more information than the corresponding approach of Hui and collaborators.^{17,18} In passing, it is also shown that the approach of Hui and collaborators^{17,18} is closely related (and therefore also equivalent) to a “modified secant” approach first proposed by Suquet²⁰ for nonlinear elastic systems. Other recent works of interest include the papers of Gao and Li,²¹ as well as the articles of Levy and Bergman,^{23,24} Sali and Bergman,²⁵ and Palenberg and Felderhof.²²

The nonlinear composite conductor occupies a region in space Ω , and its constitutive behavior is characterized by an energy-density function w , depending on the position vector \mathbf{x} and the electric field \mathbf{E} , such that the current field \mathbf{J} is given by

$$\mathbf{J}(\mathbf{x}) = \frac{\partial w}{\partial \mathbf{E}}(\mathbf{x}, \mathbf{E}). \quad (1)$$

It is assumed that the composite conductor is made up of N homogeneous, isotropic phases, so that

$$w(\mathbf{x}, \mathbf{E}) = \sum_{r=1}^N \theta^{(r)}(\mathbf{x}) \phi^{(r)}(E), \quad (2)$$

where $\theta^{(r)}$ and $\phi^{(r)}$ denote, respectively, the indicator function of phase r and its isotropic potential. The functions $\theta^{(r)}$ ($r=1, \dots, N$) are such that $\theta^{(r)}=1$ if \mathbf{x} is in phase r and 0 otherwise. The functions $\phi^{(r)}$ are taken to be convex in the magnitude of the electric field $E=|\mathbf{E}|$ and are assumed to be such that $\phi^{(r)}(E) \geq 0$ and that $\phi^{(r)}(0)=0$. A commonly used form for the phase potentials is

$$\phi^{(r)}(E) = \frac{1}{n+1} \chi^{(r)} |E|^{n+1}, \quad (3)$$

where $\chi^{(r)}$ is the nonlinear conductivity and the nonlinearity exponent n is taken to be between 0 and ∞ . The special case when $n=1$ corresponds to a linear conductor.

It is known³ that the *effective* constitutive behavior of the composite conductor may be expressed in terms of the averages of the fields, $\bar{\mathbf{J}} = \langle \mathbf{J} \rangle$ and $\bar{\mathbf{E}} = \langle \mathbf{E} \rangle$, where angular brackets are used to denote volume averages over Ω , via

$$\bar{\mathbf{J}} = \frac{\partial \bar{W}}{\partial \bar{\mathbf{E}}}(\bar{\mathbf{E}}). \quad (4)$$

In this relation, the effective energy-density function of the composite \bar{W} is most easily described in terms of the minimum energy principle

$$\bar{W}(\bar{\mathbf{E}}) = \min_{\mathbf{E} \in K} \langle w(\mathbf{x}, \mathbf{E}) \rangle = \min_{\mathbf{E} \in K} \left\{ \sum_{r=1}^N c^{(r)} \langle \phi^{(r)}(E) \rangle^{(r)} \right\}, \quad (5)$$

where K is the set of trial electric fields, defined by

$$K = \{ \mathbf{E} | \mathbf{E} = -\nabla \varphi(\mathbf{x}) \text{ in } \Omega, \text{ and } \varphi = -\bar{\mathbf{E}} \cdot \mathbf{x} \text{ on } \partial\Omega \}, \quad (6)$$

and where $c^{(r)} = \langle \theta^{(r)} \rangle$ is the volume fraction of phase r and the symbol $\langle \cdot \rangle^{(r)}$ is used to denote a volume average over phase r . This variational formulation of the conductivity problem for the composite is equivalent to the standard boundary-value-problem formulation in terms of the equations $\nabla \cdot \mathbf{J} = \mathbf{0}$ and $\nabla \times \mathbf{E} = \mathbf{0}$, together with the uniform boundary condition $\varphi = -\bar{\mathbf{E}} \cdot \mathbf{x}$ on $\partial\Omega$, where φ is the electrostatic potential.

The main advantage of the variational formulation is that the effective behavior of the nonlinear composite is then characterized in terms of only one scalar variable, namely, \bar{W} . The main difficulty associated with the computation of the effective energy function of the composite (11) lies in the fact that the exact fields are extremely difficult to determine in general. However, approximate methods have been developed to address this problem in the context of *linear* constitutive behavior for the composite. In the following, a variational principle that allows for the use of known estimates for linear composites to obtain corresponding estimates for nonlinear composites is recalled.

II. THE VARIATIONAL PRINCIPLE

Following an analogous development for nonlinearly elastic solids,²⁶ Ponte Castañeda^{5,4} proposed a variational method to estimate the effective energy functions of *nonlinear* composite conductors in terms of the energy functions of appropriately chosen *linear* composite conductors. Such “linear comparison composites” are defined by the quadratic energy-density function

$$w_0(\mathbf{x}, \mathbf{E}) = \frac{1}{2} \sigma_0(\mathbf{x}) E^2, \quad (7)$$

where $\sigma_0(\mathbf{x})$ is the conductivity of the fictitious linear composite conductor.

Under the hypothesis that the potentials $\phi^{(r)}$ of the nonlinear composite are convex on E^2 , it can be shown⁵ that

$$\phi^{(r)}(E) = \max_{\sigma_0 \geq 0} \left\{ \frac{1}{2} \sigma_0 E^2 - v^{(r)}(\sigma_0) \right\}, \quad (8)$$

where

$$v^{(r)}(\sigma_0) = \max_E \left\{ \frac{1}{2} \sigma_0 E^2 - \phi^{(r)}(E) \right\}. \quad (9)$$

This representation is based on Legendre duality for the function $f^{(r)}$ defined by $f^{(r)}(E^2) = \phi^{(r)}(E)$. In fact, $\phi^{(r)}(E) = (f^{(r)})^{**}(E^2)$ and $v^{(r)}(\sigma_0) = f^{(r)*}(\frac{1}{2}\sigma_0)$, where f^* denotes the Legendre transform of f . Note that if the $\phi^{(r)}$ are smooth, the maximum conditions in Eqs. (8) and (9) are attained at the stationarity conditions

$$\frac{1}{2} E^2 = \frac{\partial v^{(r)}}{\partial \sigma_0} \quad \text{and} \quad \sigma_0 = \frac{1}{E} \frac{\partial \phi^{(r)}}{\partial E}, \quad (10)$$

respectively, which are inverses of each other.

When relation (8) for $\phi^{(r)}$ is used in expression (5) for \bar{W} , and the order of the maximum over σ_0 is interchanged with the minimum over \mathbf{E} , it follows^{5,4} that

$$\tilde{W}(\bar{\mathbf{E}}) = \max_{\sigma_0(\mathbf{x}) \geq 0} \left\{ \tilde{W}_0(\bar{\mathbf{E}}) - \sum_{r=1}^N c^{(r)} \langle v^{(r)}(\sigma_0(\mathbf{x})) \rangle^{(r)} \right\}, \quad (11)$$

where \tilde{W}_0 denotes the effective energy function of the linear comparison composite, with local energy function (7), such that

$$\tilde{W}_0(\bar{\mathbf{E}}) = \min_{\mathbf{E} \in K} \langle w_0(\mathbf{x}, \mathbf{E}) \rangle. \quad (12)$$

Thus, expression (11), together with Eqs. (9) and (12), provide an alternative variational representation of the effective energy function of the *nonlinear* composite in terms of the effective energy function of a fictitious *linear* composite material, the choice of which is determined by Eq. (11). It is emphasized that the conductivity coefficient $\sigma_0(\mathbf{x})$ of the comparison composite in Eq. (11) is an arbitrary non-negative function of position. In addition, it is important to note that the minimum principle (11) is valid only under the hypothesis that the potentials $\phi^{(r)}$ are convex on E^2 . If, on the other hand, the $\phi^{(r)}$ are concave on E^2 [as would be the case if $0 \leq n < 1$ in Eq. (3)], an analogous result would hold, but with the maximum in Eq. (11) replaced by a minimum, and with the functions $v^{(r)}$ redefined such that the maximum in Eq. (9) is replaced by a minimum.

III. BOUNDS AND ESTIMATES FOR EFFECTIVE RESPONSE

A. Composites with generally nonlinear isotropic phases

Even if the properties of the nonlinear phases are assumed to be homogeneous, the solutions for the comparison conductivities $\sigma_0(\mathbf{x})$ in the variational principle (11) are not in general constant over the individual phases, unless the actual fields happen to be constant over the phases. However, a lower bound for \tilde{W} may be obtained by restricting the class of trial comparison conductivity fields to be constant within each phase, that is, by letting

$$\sigma_0(\mathbf{x}) = \sum_{r=1}^N \theta^{(r)}(\mathbf{x}) \sigma_0^{(r)}, \quad (13)$$

with $\sigma_0^{(r)}$ constant. This follows from the fact that the maximum over a set is, in general, larger than the maximum over any subset of the original set. Therefore, from Eqs. (11) and (12), it follows that

$$\tilde{W}(\bar{\mathbf{E}}) \geq \max_{\sigma_0^{(r)} > 0} \left\{ \tilde{W}_0(\bar{\mathbf{E}}) - \sum_{r=1}^N c^{(r)} v^{(r)}(\sigma_0^{(r)}) \right\}, \quad (14)$$

where now

$$\tilde{W}_0(\bar{\mathbf{E}}) = \frac{1}{2} \bar{\mathbf{E}} \cdot (\tilde{\sigma}_0 \bar{\mathbf{E}}) = \min_{\mathbf{E} \in K} \left\{ \frac{1}{2} \sum_{r=1}^N c^{(r)} \sigma_0^{(r)} \langle E^2 \rangle^{(r)} \right\}. \quad (15)$$

In this last relation, $\tilde{\sigma}_0$ is the effective conductivity tensor of a linear comparison composite with precisely the same microstructure as the original nonlinear composite. (Note that $\tilde{\sigma}_0$ is in general anisotropic.)

The expression (14) was first given by Ponte Castañeda²⁶ in the context of nonlinear elastic composites, and Ponte Castañeda and co-workers^{5,4} for nonlinear conductor composites. It is important to note that the above estimates for N -phase nonlinear composites are in the form of lower bounds for \tilde{W} . Thus, lower bounds for $\tilde{\sigma}_0$ may be used to generate corresponding lower bounds for \tilde{W} , but, on the other hand, upper bounds for $\tilde{\sigma}_0$ may not be used to generate upper bounds for \tilde{W} . In practice, however, one is usually only interested in obtaining estimates for the effective constitutive relations of a specific type of composite. Because bounds for \tilde{W} do not usually translate into bounds for the constitutive relations (4), one could ignore the inequality in relation (14), and reinterpret it as an *approximate* equality to obtain estimates for the effective potentials of specific types of composites. Thus, for example, Ponte Castañeda⁴ suggested the use of the Hashin-Shtrikman bounds (or MGA estimates) for $\tilde{\sigma}_0$ to generate estimates for the effective potentials \tilde{W} of nonlinear composites with *particulate* microstructures. Alternatively, an EMA estimate for $\tilde{\sigma}_0$ was used⁵ to estimate \tilde{W} for composite materials with *granular* microstructures.

Denoting by $\hat{\sigma}_0^{(s)}$ the optimal values of $\sigma_0^{(s)}$ from relations (14), it follows, from Eq. (4), that

$$\bar{\mathbf{J}} = \tilde{\sigma}_0(\hat{\sigma}_0^{(1)}, \dots, \hat{\sigma}_0^{(N)}) \bar{\mathbf{E}} + \sum_{r=1}^N \left\{ \frac{1}{2} \bar{\mathbf{E}} \cdot \left[\frac{\partial \tilde{\sigma}_0}{\partial \sigma_0^{(r)}}(\hat{\sigma}_0^{(1)}, \dots, \hat{\sigma}_0^{(N)}) \bar{\mathbf{E}} \right] - c^{(r)} \frac{\partial v^{(r)}}{\partial \sigma_0^{(r)}}(\hat{\sigma}_0^{(r)}) \right\} \frac{\partial \hat{\sigma}_0^{(r)}}{\partial \bar{\mathbf{E}}}, \quad (16)$$

so that, if the maximum in the expression (14) for the general bound is attained at the stationarity condition

$$\begin{aligned} \frac{1}{2} \bar{\mathbf{E}} \cdot \left[\frac{\partial \tilde{\sigma}_0}{\partial \sigma_0^{(s)}}(\hat{\sigma}_0^{(1)}, \dots, \hat{\sigma}_0^{(N)}) \bar{\mathbf{E}} \right] \\ = c^{(s)} \frac{\partial v^{(s)}}{\partial \sigma_0^{(s)}}(\hat{\sigma}_0^{(s)}) \quad (s = 1, \dots, N), \end{aligned} \quad (17)$$

then the effective constitutive relation of the nonlinear composite reduces to

$$\bar{\mathbf{J}} = \tilde{\sigma}_0(\hat{\sigma}_0^{(1)}, \dots, \hat{\sigma}_0^{(N)}) \bar{\mathbf{E}}. \quad (18)$$

Note that, in spite of its appearance, this effective constitutive relation is fully nonlinear because the variables $\sigma_0^{(s)}$ depend nonlinearly on the $\bar{\mathbf{E}}$.

Next, it is observed from its definition (15) that $\tilde{\sigma}_0$ is a *homogeneous function of degree one* in the conductivity constants $\sigma_0^{(s)}$ of the linear comparison composite, so that, by Euler's theorem,

$$\sum_{r=1}^N \sigma_0^{(r)} \frac{\partial \tilde{\sigma}_0}{\partial \sigma_0^{(r)}} = \tilde{\sigma}_0. \quad (19)$$

It then follows that the expression (14) for \tilde{W} can be rewritten in the form

$$\tilde{W}(\bar{\mathbf{E}}) \geq \max_{\sigma_0^{(r)} > 0} \sum_{r=1}^N c^{(r)} \left\{ \frac{1}{2} \frac{\sigma_0^{(r)}}{c^{(r)}} \bar{\mathbf{E}} \cdot \left[\frac{\partial \tilde{\sigma}_0}{\partial \sigma_0^{(r)}} \bar{\mathbf{E}} \right] - v^{(r)}(\sigma_0^{(r)}) \right\}, \quad (20)$$

and, because $\partial \tilde{\sigma}_0 / \partial \sigma_0^{(r)}$ is homogeneous of degree zero, it follows from relations (8) that

$$\tilde{W}(\bar{\mathbf{E}}) \geq \sum_{r=1}^N c^{(r)} \phi^{(r)}(\hat{E}^{(r)}), \quad (21)$$

where

$$\hat{E}^{(s)} = \left\{ \frac{1}{c^{(s)}} \bar{\mathbf{E}} \cdot \left[\frac{\partial \tilde{\sigma}_0}{\partial \sigma_0^{(s)}} (\hat{\sigma}_0^{(1)}, \dots, \hat{\sigma}_0^{(N)}) \bar{\mathbf{E}} \right] \right\}^{1/2} \quad (s=1, \dots, N). \quad (22)$$

It is noted that $\hat{E}^{(r)} = [\langle E^2 \rangle^{(r)}]^{1/2}$, where $\langle E^2 \rangle^{(r)}$ is the second moment of the electric field in the linear comparison com-

posite. [This relation is easily demonstrated^{2,3} by differentiation of Eq. (15) with respect to $\sigma_0^{(s)}$.] Also, it is noted that the stationary conditions (17) for the $\hat{\sigma}_0^{(s)}$ can be rewritten in the form

$$\frac{1}{2} [\hat{E}^{(s)}]^2 = \frac{\partial v^{(s)}}{\partial \sigma_0^{(s)}} (\hat{\sigma}_0^{(s)}) \quad (s=1, \dots, N), \quad (23)$$

which, by the equivalence of the relations (10), can be inverted to give

$$\hat{\sigma}_0^{(s)} = \frac{1}{\hat{E}^{(s)}} \frac{\partial \phi^{(s)}}{\partial E} (\hat{E}^{(s)}) \quad (s=1, \dots, N). \quad (24)$$

Using these expressions in Eq. (22), the following set of implicit relations is obtained for the variables $\hat{E}^{(s)}$:

$$c^{(s)} [\hat{E}^{(s)}]^2 = \bar{\mathbf{E}} \cdot \left[\frac{\partial \tilde{\sigma}_0}{\partial \sigma_0^{(s)}} \left(\frac{1}{\hat{E}^{(1)}} \frac{\partial \phi^{(1)}}{\partial E} (\hat{E}^{(1)}), \dots, \frac{1}{\hat{E}^{(N)}} \frac{\partial \phi^{(N)}}{\partial E} (\hat{E}^{(N)}) \right) \bar{\mathbf{E}} \right] \quad (s=1, \dots, N). \quad (25)$$

The form (21) for the effective potential \tilde{W} and its relation to the second moments $\langle E^2 \rangle^{(r)}$ of the electric field in the linear comparison composite were first given by Suquet²⁰ in the context of nonlinear elasticity.

Finally, the effective constitutive relation of the nonlinear composite conductor can be written in the form

$$\bar{\mathbf{J}} = \tilde{\sigma}_0 \left(\frac{1}{\hat{E}^{(1)}} \frac{\partial \phi^{(1)}}{\partial E} (\hat{E}^{(1)}), \dots, \frac{1}{\hat{E}^{(N)}} \frac{\partial \phi^{(N)}}{\partial E} (\hat{E}^{(N)}) \right) \bar{\mathbf{E}}, \quad (26)$$

where the variables $\hat{E}^{(r)}$ are obtained from relations (25) as functions of the applied field $\bar{\mathbf{E}}$, the nonlinear properties of the constituent phases of the composite conductor, and appropriate statistical information about the microstructure.

In conclusion, there are two completely *equivalent* ways of expressing the bounds for \tilde{W} and the estimates for the effective constitutive relation (4) that arise from the variational principle (11). The first is given in terms of the optimal comparison conductivities $\hat{\sigma}_0^{(s)}$, as determined by the relations (17), and it involves expression (14) for \tilde{W} and expression (18) for the effective constitutive relation. The second is given in terms of the second moment variables $\hat{E}^{(s)}$, defined by relations (25), and it involves expression (21) for \tilde{W} and expression (26) for the effective constitutive

relation. Note that the first gives a simpler form for the effective constitutive relation and the second gives a simpler form for the effective potential.

B. Composites with power-law isotropic phases

An important class of composite conductors is that defined by the form (3) for the phase potentials $\phi^{(r)}$. For this special class of nonlinear composites, it is possible to simplify further the two equivalent forms (14) and (21) for \tilde{W} . Thus, from Eqs. (19) and (22), note that

$$\sum_{r=1}^N c^{(r)} \sigma_0^{(r)} [\hat{E}^{(r)}]^2 = \bar{\mathbf{E}} \cdot (\tilde{\sigma}_0 \bar{\mathbf{E}}). \quad (27)$$

Also, for a power-law composite, it follows, from Eqs. (21) and (24), that

$$\tilde{W}(\bar{\mathbf{E}}) \geq \sum_{r=1}^N c^{(r)} \phi^{(r)}(\hat{E}^{(r)}) = \frac{1}{n+1} \sum_{r=1}^N c^{(r)} \sigma_0^{(r)} [\hat{E}^{(r)}]^2. \quad (28)$$

Putting these two results together, it is concluded that, for a power-law composite,

$$\tilde{W}(\bar{\mathbf{E}}) \geq \frac{1}{n+1} \bar{\mathbf{E}} \cdot \left[\tilde{\sigma}_0 \left(\frac{1}{\hat{E}^{(1)}} \frac{\partial \phi^{(1)}}{\partial E} (\hat{E}^{(1)}), \dots, \frac{1}{\hat{E}^{(N)}} \frac{\partial \phi^{(N)}}{\partial E} (\hat{E}^{(N)}) \right) \bar{\mathbf{E}} \right]. \quad (29)$$

When the microstructure is statistically isotropic, the effective potential of the power-law composite conductor takes the form

$$\tilde{W}(\bar{\mathbf{E}}) = \frac{1}{n+1} \bar{\chi} \bar{E}^{n+1}, \quad (30)$$

which, via Eq. (29), defines an effective nonlinear conductivity $\bar{\chi}$ such that

$$\bar{\chi} \geq \frac{1}{\bar{E}^{n-1}} \bar{\sigma}_0 \left(\frac{1}{\hat{E}^{(1)}} \frac{\partial \phi^{(1)}}{\partial E} (\hat{E}^{(1)}), \dots, \frac{1}{\hat{E}^{(N)}} \frac{\partial \phi^{(N)}}{\partial E} (\hat{E}^{(N)}) \right), \quad (31)$$

where $\bar{\sigma}_0$ is now the isotropic conductivity of the linear comparison composite. This is the relation first given by Hui and co-workers.¹⁷ Although these authors derived this result as an estimate (not a bound), by making use of a certain decoupling approximation,^{18,19} it follows from the above derivation that it is strictly a special case of the variational bound (14).

IV. RESULTS FOR TWO-PHASE SYSTEMS

As already noted, for two-phase systems, $\partial \bar{\sigma}_0 / \partial \sigma_0^{(s)}$ is homogeneous of degree zero in the variables $\hat{\sigma}_0^{(1)}$ and $\hat{\sigma}_0^{(2)}$, so that, from Eq. (22), the variables $\hat{E}^{(1)}$ and $\hat{E}^{(2)}$ can depend on $\hat{\sigma}_0^{(1)}$ and $\hat{\sigma}_0^{(2)}$ only through the ratio $\hat{\sigma}_0^{(1)} / \hat{\sigma}_0^{(2)}$. This means that a single nonlinear equation for $\hat{\sigma}_0^{(1)} / \hat{\sigma}_0^{(2)}$ can be generated from relations (24), by taking their ratio, so that

$$\frac{\hat{\sigma}_0^{(1)}}{\hat{\sigma}_0^{(2)}} = \frac{\hat{E}^{(2)} \partial \phi^{(1)} / \partial E (\hat{E}^{(1)})}{\hat{E}^{(1)} \partial \phi^{(2)} / \partial E (\hat{E}^{(2)})}. \quad (32)$$

Thus, given an estimate for the effective conductivity $\bar{\sigma}_0$ of the linear comparison composite, this equation can be solved numerically for the ratio $\hat{\sigma}_0^{(1)} / \hat{\sigma}_0^{(2)}$. Next, the variables $\hat{E}^{(1)}$ and $\hat{E}^{(2)}$ can be computed explicitly in terms of this ratio and the result can be used in relation (21) to obtain a bound for the effective potential \tilde{W} .

A. General ellipsoidal microstructures

In this section, the special case is considered of two-phase nonlinear composite conductors with *anisotropic* random microstructures exhibiting ‘‘ellipsoidal symmetry.’’ Ellipsoidal symmetry is a generalization of statistical isotropy, due to Willis,²⁷ which assumes that the two-point probability function for the distribution of the two phases in the composite is given by $P^{(rs)}(\mathbf{x} - \mathbf{x}') = P^{(rs)}(|\mathbf{Z}(\mathbf{x} - \mathbf{x}')|)$, for some symmetric tensor \mathbf{Z} . Note that the limiting case where \mathbf{Z} is equal to the identity tensor \mathbf{I} corresponds to statistically isotropic microstructures. In order to be able to make use of the results of Sec. III A to obtain estimates for the effective energy function \tilde{W} of composites with generally nonlinear phases, corresponding estimates for the effective conductivity tensor $\bar{\sigma}_0$ of the linear comparison conductor defined by relation (15) are required in terms of the phase conductivities $\sigma_0^{(r)} = \sigma_0^{(r)} \mathbf{I}$ and the microstructure.

A sufficiently general expression,²⁷ from which MGA and EMA estimates for composites with general ellipsoidal symmetry may be obtained, is given by

$$\bar{\sigma}_0 = \left\{ \sum_{r=1}^2 c^{(r)} \sigma_0^{(r)} [\mathbf{I} + \mathbf{T}^{(0)} (\sigma_0^{(r)} - \sigma_0^{(0)})]^{-1} \right\} \times \left\{ \sum_{s=1}^2 c^{(s)} [\mathbf{I} + \mathbf{T}^{(0)} (\sigma_0^{(s)} - \sigma_0^{(0)})]^{-1} \right\}^{-1}, \quad (33)$$

where $\sigma_0^{(0)}$ denotes the conductivity tensor of a reference material, and $\mathbf{T}^{(0)}$ is an associated tensor characterizing the microstructure of the random composite. For general ellipsoidal symmetry,

$$\mathbf{T}^{(0)} = \frac{1}{4\pi \det \mathbf{Z}} \int_{|\xi|=1} \frac{\xi \otimes \xi}{\xi \cdot (\sigma_0^{(0)} \xi)} |\mathbf{Z}^{-1} \xi|^{-3} d\omega, \quad (34)$$

and the two different types of MGA estimates are obtained by setting $\sigma_0^{(0)}$ equal to either $\sigma_0^{(1)}$ or $\sigma_0^{(2)}$. The corresponding EMA estimate is obtained by setting $\sigma_0^{(0)}$ equal to $\bar{\sigma}_0$ and solving the resulting implicit equation for $\bar{\sigma}_0$.

When one of the phases in the nonlinear composite, say phase 2, is taken to be either perfectly insulating [$\phi^{(r)}(E) = 0$], or perfectly conducting [$\phi^{(r)}(E) \rightarrow \infty$, unless $E = 0$, in which case $\phi^{(r)} = 0$], the expressions of Sec. III A can be shown to simplify further. Thus, the expression (33) for the effective conductivity of the linear comparison composite can be shown to take the form

$$\bar{\sigma}_0 = \sigma_0^{(1)} \Sigma, \quad (35)$$

where the tensor Σ depends on the type of estimate (MGA or EMA), but not on $\sigma^{(1)}$. It then follows, from Eq. (21), that the effective potential for this special class of composites can be written in the form

$$\tilde{W}(\bar{\mathbf{E}}) \geq c^{(1)} \phi^{(1)} \left(\frac{1}{c^{(1)}} \bar{\mathbf{E}} \cdot \Sigma \bar{\mathbf{E}} \right), \quad (36)$$

which depends on the type of estimate (MGA or EMA) via Σ . For example, the MGA expressions for the perfectly insulating and perfectly conducting cases are, respectively,

$$\Sigma = c^{(1)} [\mathbf{I} - \sigma_0^{(1)} \mathbf{T}^{(1)}] [\mathbf{I} - c^{(1)} \sigma_0^{(1)} \mathbf{T}^{(1)}]^{-1}$$

and

$$\Sigma = \mathbf{I} + \frac{c^{(2)}}{c^{(1)}} [\sigma_0^{(1)} \mathbf{T}^{(1)}]^{-1}.$$

The corresponding EMA expressions are more complicated, requiring numerical computation in general.

B. Statistically isotropic microstructures

For *statistically isotropic* nonlinear composites, it is justified to consider only isotropic linear comparison composites with $\tilde{W}_0(\bar{\mathbf{E}}) = \frac{1}{2} \bar{\sigma}_0 \bar{E}^2$, where $\bar{\sigma}_0$ is now a scalar function of the nonlinear conductivities $\sigma_0^{(r)}$, the volume fractions $c^{(r)}$, and the microstructure. For the special case of two-phase composites, there are several closely related bounds and estimates for linear composite materials, which can be all characterized in terms of the equation

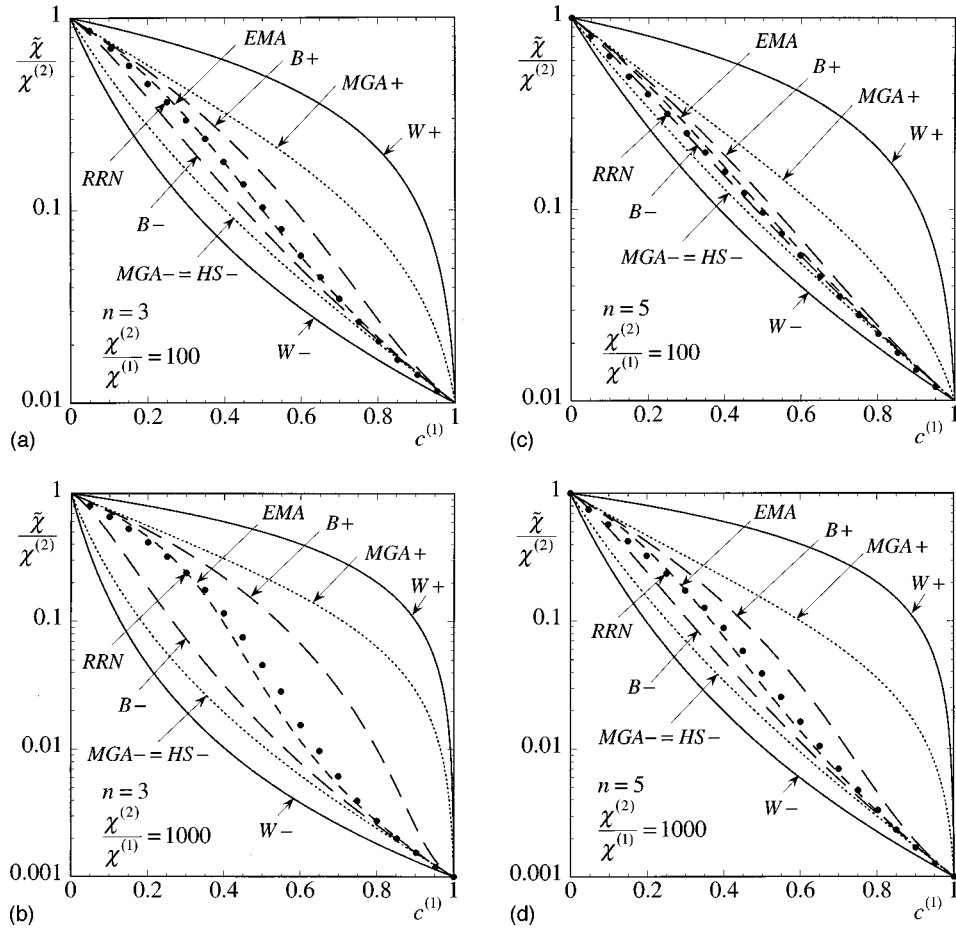


FIG. 1. Plots of various bounds and estimates for the effective nonlinear conductivity $\tilde{\chi}$ of two-dimensional ($d=2$), statistically isotropic, two-phase, power-law conductors with power exponents $n=3$ and 5 , and conductivity ratios $\chi^{(2)}/\chi^{(1)}=100$ and 1000 .

$$\tilde{\sigma}_0 = c^{(1)}\sigma_0^{(1)} + c^{(2)}\sigma_0^{(2)} - \frac{c^{(1)}c^{(2)}(\sigma_0^{(1)} - \sigma_0^{(2)})^2}{c^{(2)}\sigma_0^{(1)} + c^{(1)}\sigma_0^{(2)} + (d-1)\sigma_0}, \quad (38)$$

where d stands for the dimension of the underlying space, and σ_0 takes on different values for the different types of estimates, as described below. Assuming that $\sigma_0^{(1)} > \sigma_0^{(2)}$, the choices $\sigma_0 \rightarrow \infty$ and $\rightarrow 0$ correspond, respectively, to the Wiener²⁸ upper and lower bounds. The choices $\sigma_0 = \sigma_0^{(1)}$ and $\sigma_0^{(2)}$ yield the two MGA approximations for particulate microstructures with phases 1 and 2, respectively, in the matrix phase—they also lead to the Hashin-Shtrikman⁸ upper and lower bounds. The choices $\sigma_0 = \zeta^{(1)}\sigma_0^{(1)} + \zeta^{(2)}\sigma_0^{(2)}$ and $(\zeta^{(1)}/\sigma_0^{(1)} + \zeta^{(2)}/\sigma_0^{(2)})^{-1}$ give the upper and lower bounds of Beran,¹³ in terms of the three-point parameter $\zeta^{(1)} = 1 - \zeta^{(2)}$ of Milton.¹⁴ Finally, the choice $\sigma_0 = \tilde{\sigma}_0$ gives the EMA approximation.

As already noted at the beginning of this section, for the special case of two-phase composites, it is possible to obtain an expression for \tilde{W} involving only one nonlinear equation for the ratio $\hat{\sigma}_0^{(1)}/\hat{\sigma}_0^{(2)}$. Computing the variables $\hat{E}^{(1)}$ and $\hat{E}^{(2)}$ in terms of this ratio, the nonlinear conductivity $\tilde{\chi}$ for the power-law composite may then be obtained from Eq. (30). For illustrative purposes, results for $\tilde{\chi}$ are presented in Fig. 1 for two-dimensional ($d=2$), statistically isotropic, two-phase, power-law conductors with $n=3$ and 5 , and

$\chi^{(2)}/\chi^{(1)}=100$ and 1000 . The various curves are described below. The labels $W-$ and $W+$ correspond to the rigorous upper and lower Wiener bounds for composites with arbitrary microstructures. The labels $MGA-$ and $MGA+$ correspond to the Maxwell-Garnett estimates for particulate microstructures with fewer and more conducting materials occupying the matrix phase, respectively. Because the $MGA-$ estimate for the conductivity of a linear composite happens to coincide with the Hashin-Shtrikman lower bound for the set of all statistically isotropic composites, the $MGA-$ curve is identical to the rigorous nonlinear Hashin-Shtrikman lower bound, which is denoted $HS-$ and was first given by Ponte Castañeda *et al.*⁴ The label RRN corresponds to the random resistor network simulations of Wan *et al.*,¹⁸ following Lee and Yu.¹⁵ The label $B-$ corresponds to the rigorous Beran lower bound^{5,29} for statistically isotropic microstructures with the choice of the Milton three-point parameter $\zeta^{(1)} = c^{(1)}$. This choice is appropriate for “symmetric cell” microstructures,¹⁶ which are similar in character to the discrete RRN microstructures. The label $B+$ is used to denote the estimate (not a rigorous bound) that is obtained by making use of the Beran upper bound for the linear comparison composite. Finally, the label EMA is used to describe the effective medium approximation. These estimates are identical to those first given by Wan *et al.*¹⁸ for this case. The main observations in the context of this figure are as follows. First, as already noted¹⁷ the EMA estimates are in good

agreement with the RRN simulations, in spite of the fact that the EMA and RRN estimates correspond to continuum and discrete systems, respectively. Second, the Weiner, Hashin-Shtrikman, and Beran bound progressively narrow the range of possible behavior by introducing first-, second-, and third-order statistical information, respectively. Although the MGA and EMA are believed to be good approximations for *particulate* and *granular* type microstructures, respectively, the Beran-Milton nonlinear bounds given in this work provide a way of characterizing more general types of microstructures, for which the MGA and EMA estimates may not be appropriate. Implementation of these new bounds would, of course, require computation of the relevant three-point parameters, as in Torquato.³⁰

V. CONCLUDING REMARKS

In this work, three-point bounds and other estimates have been computed for the effective response of strongly nonlinear composites by means of the variational procedure of Ponte Castañeda and co-workers,^{5,4} making use of the notion of a linear comparison composite. The results were compared with random resistor network simulations available from the literature and found to be very accurate. One way to

explain the relatively good accuracy of the procedure is to note that the method is based on a variational approximation consisting in the use of appropriate trial fields in the context of an *exact* minimum principle for the effective energy-density function of the nonlinear composite. In particular, this means that the variational procedure of Ponte Castañeda and co-workers^{5,4} (and the equivalent method of Hui and co-workers¹⁷) provides the “best” possible approximation within the context of linear comparison composites with microstructures identical to those of the nonlinear composites. According to this variational interpretation, improved estimates could only be obtained by making use of more sophisticated linear comparison composites accounting for the distribution of the electric field within the various phases of the nonlinear composite. An example of such improved estimates is provided by the exact estimates given⁵ for sequentially laminated nonlinear composites, where the distribution of the electric field is piecewise constant within each phase.

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