

Estimates for two-phase nonlinear conductors via iterated homogenization

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Research



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Estimates for the overall response of two-phase nonlinear conductors are derived via an iterated homogenization approach. The approach consists in the construction of sequentially laminated microgeometries for which the overall response can be determined exactly. The estimate depends on the one- and two-point microstructural correlations through the volume fractions of each constituent phase and the H-measures of the microstructure. Sample results for power-law random conductors are provided and compared with earlier predictions. The work concludes with a discussion on the possible extremal character of sequential laminates.

1. Introduction

The purpose of this work is to develop estimates for the overall electrical response of *nonlinear* composite conductors in terms of the electrical responses of its constituent phases and their geometrical arrangement. The focus is on composites consisting of inclusions of one phase dispersed in a continuous matrix of a second phase.

Several strategies have been proposed by now to *estimate* the overall response of random composite materials. One particular strategy often used when the conductors exhibit a *linear* response consists in

identifying special classes of microgeometries that reproduce the essential geometrical features of the actual composite microstructure, while at the same time allow the exact computation of the overall response via homogenization theory. These the so-called *solvable microgeometries* include, for instance, laminates [1] and composite-sphere assemblages [2]. The works of Maxwell [1] and Bruggeman [3] have shown that the set of solvable microgeometries could be enlarged by following iterative procedures whereby the constituent phases in a solvable microgeometry are themselves identified with solvable microgeometries at a lower length scale, thus producing hierarchical microgeometries of increasing complexity whose overall response can be determined via iterated homogenization. For instance, Maxwell [1] constructed particulate composites with an isotropic response by iterating anisotropic laminated microgeometries; the resulting microgeometries are known as sequential laminates. Bruggeman [3], in turn, constructed particulate composites with arbitrary volume fractions of particles by iterating microgeometries with dilute volume fractions of particles; this approach is known as the differential scheme, and has been generalized by Norris *et al.* [4] and others. Sequential laminates and differential schemes have proved very useful in estimating the overall response of two-phase *linear* conductors and linear composites more generally—see, for instance, the monograph by Milton [5]. By contrast, their use in the context of *nonlinear* conductors has remained limited, mainly because iteration is encumbered by the fact that the overall response in nonlinear composites does not exhibit, in general, the same functional dependence on the electric field as the local responses. The few available results [6–9] make use of special lamination sequences and assume isotropic local responses. The first objective of the present contribution is to construct a fairly general class of sequential laminates that can represent two-phase composites with any local response—isotropic as well as anisotropic—and particulate microstructure, including isotropic and (ellipsoidal) anisotropic particle dispersions. This is achieved by following an iterative scheme recently pursued by Idiart [10] in the mathematically related context of viscoplasticity, which hinges upon the simultaneous use of a nonlinear differential scheme and sequential laminations (see also [11] in the context of finite elasticity). Because the resulting estimates are realizable, by construction, their predictions are guaranteed to exhibit the required convexity properties, and to agree exactly to second order with the small-contrast expansion of Blumenfeld & Bergman [12,13].

Along with estimating the overall response of composites with special types of microstructures, a common alternative strategy consists in *bounding* the entire range of possible responses for composite conductors with general classes of microstructures. Bounds are useful for two additional reasons: they provide benchmarks to test estimates and they can be used as estimates themselves. A particularly fruitful and fairly general approach to generate bounds was advanced for linear composites by Hashin & Shtrikman [2], generalized and further developed for nonlinear composites by Willis [14,15] and Talbot & Willis [16]. In this approach, a variational formulation is used to bound the overall response of a composite conductor in terms of ‘polarization’ fields relative to a suitably chosen *homogeneous reference medium*. A more general variational approach making use of a *linear comparison composite* was proposed by Ponte Castañeda [6,17]. When used in combination with the Hashin–Shtrikman bounds for linear composites, it recovers the earlier bounds of Talbot & Willis [16]; however, this method can also be used with other bounds and estimates for linear composites to generate corresponding bounds and estimates for nonlinear composites. For instance, nonlinear generalizations of the three-point bounds of Beran [18] and Milton [19] were given by Ponte Castañeda [6]. This approach was initially developed for locally isotropic composites, and a generalization for anisotropic materials was later proposed by Idiart & Ponte Castañeda [20]. While these are the best general bounds for nonlinear composites available to date, they are generally exact only to first order in the heterogeneity contrast, even when the comparison bound is exact to second order. This implies that, in general, the bounds are not optimal, and should, therefore, be amenable to improvement. In a recent work on viscoplastic composites, Ponte Castañeda [21] found that improved bounds could be obtained by iterating the earlier linear comparison bounds of the Hashin–Shtrikman type.

The link between these two complementary homogenization strategies is provided by the fact that some of these solvable microgeometries have been used to show the attainability—and, therefore, optimality—of the bounds, at least for linear composites. Thus, for example, the Hashin composite-sphere assemblage was used to demonstrate the optimality of the Hashin–Shtrikman (H–S) bound for isotropic composites consisting of two isotropic phases in linear conductivity [2]. But, more importantly for our purposes, is the fact that sequential laminates can also be used to saturate the H–S bounds not only for composites with isotropic microstructures [22], but also for two-phase composites with more general anisotropic behaviour for the ‘well-ordered’ phases and anisotropic microstructures [23]. In fact, it turns out that, again in the linear case, the sequential laminates have been found to depend on certain H-measures [24,25] of the two-point correlations of the phase distributions, which are precisely the same microstructural quantities defining the class of microstructures for which the H–S bounds hold. A second objective of this work is to explore corresponding ideas in the context of nonlinear composites by first obtaining estimates for sequential laminates, and then by comparing them to available bounds.

We begin in §2 with some preliminaries on nonlinear homogenization, its iterated version and H-measures. In §3, we obtain an exact expression for the effective energy-density function of finite-rank, sequentially laminated composites with nonlinear phases, and remark that—unlike the corresponding energies for their linear counterparts—they actually depend on additional microstructural information beyond the H-measures. However, consistent with earlier observations by deBotton [11] and Idiart [10], it is found that, in the limit of dilute concentrations of the ‘matrix’ phase, the expression for the energies of the nonlinear laminates depends only on the H-measures of the microstructures. As a consequence, we are able to iterate this result in the volume fraction of the matrix phase—again as proposed by deBotton [11] and Idiart [10]—to obtain an exact expression for the energies of these sequentially iterated composites consisting of inclusions of the dilute sequentially layered materials that are in turn repeatedly coated with infinitesimal layers of the matrix material until the desired final volume fraction of the matrix is reached. Note that although this second iteration on the volume fraction is essentially a differential scheme, it is not of the standard type, because it starts with 100 per cent of the inclusion phase and it is the matrix phase that is added incrementally (instead of starting with the matrix phase and incrementing the volume fraction of the inclusion phase, as in the usual differential self-consistent scheme). In §4, the expressions of the prior section for the iterated laminated structures are specialized to power-law behaviour for the constituents, and the results are compared with available bounds. Finally, in §5, we demonstrate explicitly that the effective behaviour of the finite-concentration laminates with the same finite concentrations of the phases and (transversely) isotropic H-measures is not only anisotropic, but in fact depends on the order of the lamination sequence. As a consequence of the anisotropy of the finite-concentration laminates, we are able to demonstrate explicitly that the (dilutely) iterated laminate construction does not provide a bound for the class of nonlinear composite conductors with prescribed H-measures, contradicting a conjecture made by Idiart [10] to this effect. We conclude the paper with some remarks concerning the possible extremal character of the estimates.

2. Preliminaries

We consider material systems made up of a continuous matrix containing a uniform dispersion of inclusions. The matrix phase will be identified with the index $r = 1$, whereas the inclusions will be collectively identified with the index $r = 2$.

The nonlinear response of each constituent phase is characterized by a *convex* potential $w^{(r)}$, such that the current density \mathbf{J} and the electric field \mathbf{E} are related by

$$\mathbf{J} = \frac{\partial w}{\partial \mathbf{E}}(\mathbf{x}, \mathbf{E}), \quad w(\mathbf{x}, \mathbf{E}) = \sum_{r=1}^2 \theta^{(r)}(\mathbf{x}) w^{(r)}(\mathbf{E}), \quad (2.1)$$

where the characteristic functions $\theta^{(r)}$ serve to describe the microstructure, being 1 if the position vector \mathbf{x} is in phase r , and 0 otherwise, such that

$$\theta^{(1)}(\mathbf{x}) + \theta^{(2)}(\mathbf{x}) = 1. \quad (2.2)$$

This constitutive framework can be used to model weakly nonlinear as well as strongly nonlinear behaviours. It can also be used to model non-smooth behaviours, such as dielectric breakdown and current-induced superconductivity breakdown, provided the derivative with respect to \mathbf{E} is understood as the subdifferential of convex analysis.

(a) Homogenization

The focus is on material systems where the size of the characteristic particle size is much smaller than the size of the specimen. In that case, the functions $\theta^{(r)}$ exhibit rapid oscillations and homogenization theory states that the *overall response* of the composite is given by the relation between the average current density and the average electric field over a ‘representative volume element’ Ω . Then, letting $\langle \cdot \rangle$ denote the volume average over Ω , and letting $\bar{\mathbf{J}} = \langle \mathbf{J} \rangle$ and $\bar{\mathbf{E}} = \langle \mathbf{E} \rangle$, the overall response can be characterized by the *effective potential* \tilde{w} , such that [14,15]

$$\bar{\mathbf{J}} = \frac{\partial \tilde{w}}{\partial \bar{\mathbf{E}}}(\bar{\mathbf{E}}), \quad \tilde{w}(\bar{\mathbf{E}}) = \min_{\mathbf{E} \in \mathcal{K}(\bar{\mathbf{E}})} \langle w(\mathbf{x}, \mathbf{E}) \rangle, \quad (2.3)$$

where $\mathcal{K}(\bar{\mathbf{E}})$ is the set of admissible fields \mathbf{E} , such that there is a continuous scalar field φ satisfying $\mathbf{E} = \nabla \varphi$ in Ω and $\varphi = \bar{\mathbf{E}} \cdot \mathbf{x}$ on $\partial \Omega$.

In general, the effective potential \tilde{w} depends on the microstructural variables $\theta^{(r)}$ in an extremely intricate manner. In this work we will construct a particular—yet fairly general—class of microgeometries that allows \tilde{w} to be computed. It turns out that for these microgeometries, the effective potential does not depend on all microstructural details but only on the one- and two-point correlation functions as defined by $c^{(r)} = \langle \theta^{(r)}(\mathbf{x}) \rangle$ and $c^{(rs)}(\mathbf{z}) = \langle \theta^{(r)}(\mathbf{z} + \mathbf{x}) \theta^{(s)}(\mathbf{x}) \rangle$ ($r, s = 1, 2$). Note that the one-point correlation functions $c^{(r)}$ correspond to the volume fractions of the phases in Ω , and are such that $c^{(1)} + c^{(2)} = 1$.

The dependence on the two-point correlation functions $c^{(rs)}$ will actually enter through certain geometrical objects known as the H-measures, introduced by Gérard [25] and Tartar [24]. These are measures which quantify in phase space the lack of compactness of weakly converging sequences of characteristic functions $[\theta^{(r)}(\mathbf{x}) - c^{(r)}]$ —see Allaire & Maillot [26], Francfort [27]—and provide a partial characterization of microstructural oscillations along different directions in physical space. For periodic microstructures, the H-measures can be written as—see, Kohn [28] and Milton [5]

$$\mu^{(rs)}(\mathbf{n}) = \text{Re} \sum_{\mathbf{k} \neq 0} \mathcal{F} \theta^{(r)}(\mathbf{k}) \overline{\mathcal{F} \theta^{(s)}(\mathbf{k})} \delta(\mathbf{n} - \mathbf{k}/|\mathbf{k}|), \quad (2.4)$$

where $\mathcal{F} \theta^{(r)}$ denotes the Fourier coefficient of $\theta^{(r)}$, $\delta(\cdot)$ denotes the vector-valued delta function, the overbar denotes the complex conjugate and the argument \mathbf{n} is a unit vector. On the other hand, for random microstructures that are statistically uniform and ergodic the H-measures can be written as—see, for instance, Smyshlyaev & Willis [29]

$$\mu^{(rs)}(\mathbf{n}) = -\frac{1}{8\pi^2} \int_{\mathbb{R}^3} \delta''(\mathbf{n} \cdot \mathbf{x}) (c^{(rs)}(\mathbf{x}) - c^{(r)} c^{(s)}) \, \mathrm{d}\mathbf{x}, \quad (2.5)$$

where δ'' denotes the second derivative of the Dirac delta function.

In view of the relation (2.2), the H-measures satisfy the following properties—see Kohn [28], Smyshlyaev & Willis [30]

$$\left. \begin{aligned} \mu^{(rs)}(\mathbf{n}) &= \mu^{(sr)}(\mathbf{n}), \quad \mu^{(rs)}(\mathbf{n}) = \mu^{(rs)}(-\mathbf{n}), \quad \sum_{s=1}^2 \mu^{(rs)}(\mathbf{n}) = 0, \\ \int_S \mu^{(rs)}(\mathbf{n}) \, dS(\mathbf{n}) &= \delta_{rs} c^{(r)} - c^{(r)} c^{(s)} \\ \text{and} \quad \sum_{r,s=1}^2 \int_S \mu^{(rs)}(\mathbf{n}) \phi^{(r)}(\mathbf{n}) \phi^{(s)}(\mathbf{n}) \, dS(\mathbf{n}) &\geq 0, \end{aligned} \right\} \quad (2.6)$$

for any continuous test functions $\phi^{(r)}$ defined over the unit sphere S . (Here, δ_{rs} denotes the Kronecker delta.) Therefore, we can express them as

$$\mu^{(rs)}(\mathbf{n}) = c^{(r)} (\delta_{rs} - c^{(s)}) \nu(\mathbf{n}), \quad (2.7)$$

where the function ν is a ‘reduced’ H-measure such that

$$\nu(\mathbf{n}) \geq 0 \quad \text{and} \quad \int_S \nu(\mathbf{n}) \, dS(\mathbf{n}) = 1. \quad (2.8)$$

This reduced H-measure will play a central role in our developments.

(b) Iterated homogenization

When the microstructural properties oscillate along multiple well-separated length scales, the computation of \tilde{w} can be carried out sequentially by successively homogenizing over the different scales, from the smallest to the largest one. In effect, this amounts to expressing the effective potential of the multi-scale composite in terms of the effective potentials of each of the smaller-scale composite structures. This follows rigorously from the so-called iterated homogenization theorem. A proof of this theorem for the case of nonlinear convex materials—such as those considered here—can be found in Braides & Lukkassen [31].

The microstructural correlations of the multi-scale composite can also be expressed in terms of the correlations of each of the smaller-scale composite structures. To fix ideas, consider a two-phase particulate composite with a microstructure oscillating along two well-separated length scales. The characteristic functions of each phase can be written as

$$\theta^{(r)}(\mathbf{x}) = \hat{\theta}^{(1)}(\mathbf{x}) \hat{\theta}^{(1,r)}(\mathbf{x}) + \hat{\theta}^{(2)}(\mathbf{x}) \hat{\theta}^{(2,r)}(\mathbf{x}), \quad (2.9)$$

where the $\hat{\theta}^{(\alpha)}$ ($\alpha = 1, 2$) are the characteristic functions of the domains $\hat{\Omega}^{(\alpha)}$ —such that $\hat{\Omega}^{(1)} \cup \hat{\Omega}^{(2)} = \Omega$ —at the larger scale, and the $\hat{\theta}^{(\alpha,r)}$ are the characteristic functions of the subdomains $\hat{\Omega}^{(\alpha,r)}$ —such that $\hat{\Omega}^{(\alpha,1)} \cup \hat{\Omega}^{(\alpha,2)} = \hat{\Omega}^{(\alpha)}$ —at the smaller scale; this is shown schematically in figure 1.¹

Let $\hat{c}^{(\alpha)}$ denote the volume fraction of the domain Ω occupied by $\hat{\Omega}^{(\alpha)}$, and let $\hat{c}^{(\alpha,r)}$ denote the volume fraction of the domain $\hat{\Omega}^{(\alpha)}$ occupied by $\hat{\Omega}^{(\alpha,r)}$. The total volume fraction of each phase in the two-scale composite is then given by

$$c^{(r)} = \hat{c}^{(1)} \hat{c}^{(1,r)} + \hat{c}^{(2)} \hat{c}^{(2,r)}. \quad (2.10)$$

Similarly, the multi-point correlation functions of the two-scale composite can be expressed in terms of the multi-point correlation functions of each length scale. In the limit as the ratio ϵ of the characteristic length scales over which the smaller and larger microgeometries oscillate tends to zero, the H-measures $\mu^{(rs)}$ associated with the two-scale microgeometry $\theta^{(r)}$ can be expressed in terms of the H-measures $\hat{\mu}^{(rs)}$ of the larger-scale microgeometry $\hat{\theta}^{(r)}$ and the H-measures

¹The two-phase composite is particulate provided the microgeometries at the smaller length scale are particulate; the microgeometry at the larger scale need not be particulate.

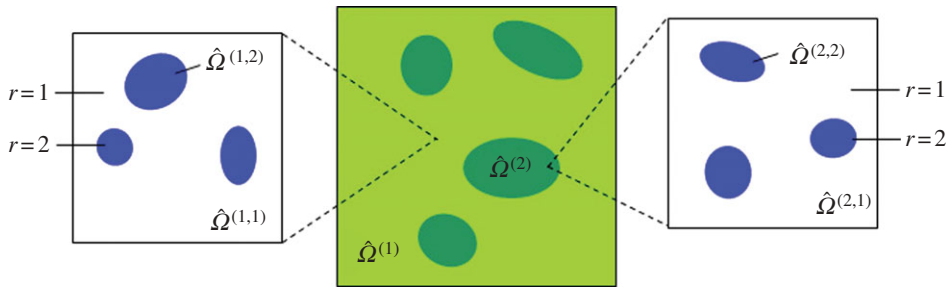


Figure 1. A two-phase particulate composite with a two-scale microgeometry. (Online version in colour.)

$\hat{\mu}^{(\alpha,rs)}$ of the smaller-scale microgeometries $\hat{\theta}^{(\alpha,r)}$ by means of the so-called ‘mixing formula’ for H-measures—see Smyshlyaev & Willis [30] and Kohn [28]

$$\mu^{(rs)} = \hat{c}^{(1)}\hat{\mu}^{(1,rs)} + \hat{c}^{(2)}\hat{\mu}^{(2,rs)} + (\hat{c}^{(1,r)} - \hat{c}^{(2,r)})(\hat{c}^{(1,s)} - \hat{c}^{(2,s)})\hat{\mu}^{(rs)}. \quad (2.11)$$

Because the microgeometries at both length scales are composed of two distinct phases, the measures $\hat{\mu}^{(\alpha,rs)}$ and $\hat{\mu}^{(rs)}$ satisfy the relations

$$\hat{\mu}^{(\alpha,rs)}(\mathbf{n}) = \hat{c}^{(\alpha,r)}(\delta_{rs} - \hat{c}^{(\alpha,s)})\hat{v}^{(\alpha)}(\mathbf{n}) \quad \text{and} \quad \hat{\mu}^{(rs)}(\mathbf{n}) = \hat{c}^{(r)}(\delta_{rs} - \hat{c}^{(s)})\hat{v}(\mathbf{n}), \quad (2.12)$$

where the reduced measures $\hat{v}^{(\alpha)}(\mathbf{n})$ and $\hat{v}(\mathbf{n})$ are positive and have an average value of 1.

Central to our approach is the observation that if the reduced measures $\hat{v}^{(\alpha)}$ and \hat{v} are both equal to some measure v_0 , the resulting reduced measure v of the two-scale composite is also equal to v_0 ; this is easily shown by making use of expressions (2.10)–(2.12). Thus, by combining two particulate microgeometries with certain volume fractions of inclusions and reduced measure v , a new (two-scale) particulate microgeometry with a *different* volume fraction of inclusions but with the *same* reduced measure v is generated. This observation holds more generally for multi-scale composites with an arbitrary number of length scales.

3. Estimates via sequential laminations

In this section, a specific class of two-phase composites—known as sequential laminates—is constructed whose effective potential can be determined exactly in terms of the volume fractions and reduced H-measure of the microgeometry, and the resulting potentials are then proposed as estimates for two-phase particulate composites conforming to the assumed microstructural correlations.

(a) Finite-rank sequential laminates

A sequential laminate is an iterative construction obtained by layering laminated materials (which in turn have been obtained from lower-order lamination procedures) with other laminated materials, or directly with the homogeneous phases that make up the composite (see, [5]). The *rank* of the laminate refers to the number of layering operations required to reach the final sequential laminate. Throughout this section a subscript k is used to denote quantities corresponding to the rank- k laminate.

Our interest is to construct two-phase microgeometries of particulate type. To that end, we follow a lamination sequence used by Idiart [10] in the context of viscoplasticity. The sequence is formed by layering at every step a laminate with the matrix material, here identified with $r = 1$. Thus, a rank-1 laminate corresponds to a simple laminate with a given layering direction \mathbf{n}_1 , with phases 1 and 2 in proportions $(1 - \hat{f}_1)$ and \hat{f}_1 , as shown in figure 2a. The characteristic functions describing these microstructures depend on \mathbf{x} only through the combination $\mathbf{x} \cdot \mathbf{n}_1$. It then follows that the exact electric field in a simple laminate, subjected to affine boundary conditions $\varphi = \bar{\mathbf{E}} \cdot \mathbf{x}$,

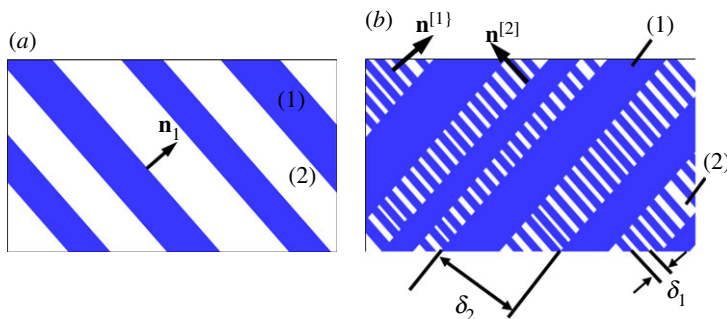


Figure 2. Two-phase laminates: (a) simple or rank 1 laminate, (b) rank 2 laminate ($\delta_2 \gg \delta_1$). (Online version in colour.)

is uniform per phase. The effective potential \tilde{w}_1 of the rank-1 laminate has been given by Ponte Castañeda [6] and can be written as

$$\tilde{w}_1(\bar{\mathbf{E}}) = \min_{a_1} \{ \hat{f}_1 w^{(2)}(\bar{\mathbf{E}} - (1 - \hat{f}_1) a_1 \mathbf{n}_1) + (1 - \hat{f}_1) w^{(1)}(\bar{\mathbf{E}} + \hat{f}_1 a_1 \mathbf{n}_1) \}. \quad (3.1)$$

In this expression, the arguments of the potentials $w^{(r)}$ represent the local electric fields in each phase r , and the minimization with respect to the scalar a_1 enforces continuity of current density across the internal material interfaces.

A rank-2 laminate is constructed by layering the rank-1 laminate with phase 1, along a layering direction \mathbf{n}_2 , in proportions \hat{f}_2 and $1 - \hat{f}_2$, as shown schematically in figure 2b. The key assumption in this construction process is that the length scale of the *embedded* laminate is taken to be much smaller than the length scale of the *embedding* laminate, i.e. $\delta_1 \ll \delta_2$ in figure 2b. According to the iterated homogenization theorem, the effective potential \tilde{w}_2 of the rank-2 laminate is then given by the formula (3.1) for a simple laminate, with the potential $w^{(2)}$ replaced by \tilde{w}_1 . Note that the total concentration of material $r = 2$ in the two-scale rank-2 laminate is $\hat{f}_1 \hat{f}_2$.

A rank- M laminate is obtained by repeating this process M times, always laminating a rank- m laminate with material $r = 1$, in proportions \hat{f}_m and $(1 - \hat{f}_m)$, respectively, along a layering direction \mathbf{n}_m . Making repeated use of the formula (3.1) for simple laminates, it can be shown—see Idiart [10] for a derivation in the context of viscoplasticity—that the effective potential \tilde{w}_M of the rank- M laminate is given by

$$\tilde{w}_M(\bar{\mathbf{E}}) = \min_{\alpha_i} \left\{ f^{(2)} w^{(2)}(\bar{\mathbf{E}}^{(2)}) + f^{(1)} \sum_{i=1}^M \alpha_i w^{(1)}(\bar{\mathbf{E}}_i^{(1)}) \right\}, \quad (3.2)$$

where

$$f^{(2)} = 1 - f^{(1)} = \prod_{i=1}^M \hat{f}_i \quad \text{and} \quad \alpha_i = \frac{(1 - \hat{f}_i)}{\hat{f}_i} \frac{\prod_{j=i}^M \hat{f}_j}{1 - \prod_{j=1}^M \hat{f}_j} \quad (3.3)$$

are microstructural variables representing, respectively, the total volume fractions of each material r —such that $f^{(1)} + f^{(2)} = 1$ —and the fraction of matrix material added at the i th lamination—such that $\alpha_i > 0$ and $\sum_{i=1}^M \alpha_i = 1$ —and the vectors $\bar{\mathbf{E}}_i^{(1)}$ and $\bar{\mathbf{E}}^{(2)}$ are local fields given by

$$\bar{\mathbf{E}}_i^{(1)} = \bar{\mathbf{E}} + a_i \mathbf{n}_i - \sum_{j=i}^M (1 - \hat{f}_j) a_j \mathbf{n}_j, \quad i = 1, \dots, M \quad (3.4)$$

and

$$\bar{\mathbf{E}}^{(2)} = \bar{\mathbf{E}} - \sum_{j=1}^M (1 - \hat{f}_j) a_j \mathbf{n}_j. \quad (3.5)$$

Thus, computing the effective potential (3.2) requires the solution of a convex optimization problem with respect to M scalars a_i . Note that the potential $w^{(2)}$ is evaluated at a single value of the electric field, which means that the fields in phase $r = 2$ are uniform. For two-dimensional systems, expression (3.2) is equivalent to the recursive formula derived by Hariton & deBotton [9].

In the above sequentially laminated constructions the discontinuous inclusion phase is made up of material $r = 2$, whereas the continuous matrix phase is made up of material $r = 1$, see figure 2*b*. Their effective potential \tilde{w}_M depends on the total volume fractions $f^{(r)}$ of each phase r and on higher-order microstructural correlations through the set of quantities $\{(\hat{f}_i, \mathbf{n}_i), i = 1, \dots, M\}$. Thus, in order to use these constructions as model composites, the quantities $(\hat{f}_i, \mathbf{n}_i)$ must be expressed in terms of the multi-point correlations of the microgeometries. In general, this is not feasible. One strategy consists then in identifying subclasses of sequentially laminated constructions for which the dependence of the effective potential on higher-order correlations can be made explicit [10].

To that end, it proves convenient to derive an alternative formula for \tilde{w}_M involving H-measures of the sequentially laminated microgeometries. At each step in the iterative process described above, the reduced H-measure of the two-phase single-scale laminate is given by—see Kohn [28]

$$\hat{v}_i(\mathbf{n}) = \delta(\mathbf{n} - \mathbf{n}_i). \quad (3.6)$$

The reduced H-measure of the multi-scale rank- M laminate can be obtained by making repeated use of the mixing formula (2.12) with (3.6); the result is

$$\nu_M(\mathbf{n}) = \sum_{i=1}^M \check{v}_i \delta(\mathbf{n} - \mathbf{n}_i), \quad \text{with } \check{v}_i = \frac{1}{f^{(1)}} \frac{1 - \hat{f}_i}{\hat{f}_i} \prod_{j=1}^i \hat{f}_j, \quad (3.7)$$

where the quantities \check{v}_i are such that

$$\check{v}_i \geq 0 \quad \text{and} \quad \sum_{i=1}^M \check{v}_i = 1. \quad (3.8)$$

Relations (3.7)₂ can be inverted to express the \hat{f}_i in terms of the \check{v}_i as

$$\hat{f}_i = \frac{1 - f^{(1)}(1 + \check{v}_i - \sum_{j=i}^M \check{v}_j)}{1 - f^{(1)}(1 - \sum_{j=i}^M \check{v}_j)}. \quad (3.9)$$

Upon replacing the \hat{f}_i in (3.2)–(3.5) by (3.9), we obtain an alternative expression for the effective potential \tilde{w}_M that depends on the underlying microgeometry through the total volume fractions $f^{(r)}$ of each material r and the set $\{(\check{v}_i, \mathbf{n}_i)\}$ subject to the constraints (3.8). Note that the reduced H-measure $\nu_M(\mathbf{n})$ is fully determined by the set, but the set is not fully determined by the reduced H-measure ν_M . This is due to the facts that there is no one-to-one correspondence between the function $\nu_M(\mathbf{n})$ and the \check{v}_i when lamination sequences involve repeated lamination directions, and that the function ν_M is insensitive to the order of the elements in the set. The effective potential \tilde{w}_M , therefore, depends, in general, on microstructural information *beyond* the volume fractions and the H-measure.

(b) Dilute sequential laminates

We now consider rank- M laminates with prescribed $\{(\check{v}_i, \mathbf{n}_i)\}$ and decreasing volume fraction $f^{(1)}$ of *matrix* material. Expanding relations (3.9) to first order as $f^{(1)} \rightarrow 0$ we obtain

$$\hat{f}_i = 1 - \check{v}_i f^{(1)} + O(f^{(1)2}). \quad (3.10)$$

In turn, assuming that the expansion of the optimal a_i for small $f^{(1)}$ is regular, expanding terms inside the curly brackets in (3.2) to first order in $f^{(1)}$ and making use of (3.3)–(3.5), we obtain the

following expression for the effective potential

$$\tilde{w}_M(\bar{\mathbf{E}}) = w^{(2)}(\bar{\mathbf{E}}) - \left[w^{(2)}(\bar{\mathbf{E}}) + \max_{a_i} \sum_{i=1}^M \dot{v}_i \left(a_i \frac{\partial w^{(2)}}{\partial \mathbf{E}}(\bar{\mathbf{E}}) \cdot \mathbf{n}_i - w^{(1)}(\bar{\mathbf{E}} + a_i \mathbf{n}_i) \right) \right] f^{(1)} + O(f^{(1)^2}). \quad (3.11)$$

In this expression, each variable a_i appears only in one term of the sum. Thus, the optimality conditions for the variables a_i decouple to first order in $f^{(1)}$.² Then, introducing the weighted orientational average

$$\langle \cdot \rangle_{v_M} \equiv \int_S (\cdot) v_M(\mathbf{n}) \, dS(\mathbf{n}), \quad (3.12)$$

with $v_M(\mathbf{n})$ given by (3.7)₁, expression (3.11) can be written as

$$\tilde{w}_M(\bar{\mathbf{E}}) = w^{(2)}(\bar{\mathbf{E}}) - \left[w^{(2)}(\bar{\mathbf{E}}) + \max_{a(\mathbf{n})} \left\langle a \frac{\partial w^{(2)}}{\partial \mathbf{E}}(\bar{\mathbf{E}}) \cdot \mathbf{n} - w^{(1)}(\bar{\mathbf{E}} + a\mathbf{n}) \right\rangle_{v_M} \right] f^{(1)} + O(f^{(1)^2}), \quad (3.13)$$

where the optimization is now carried out with respect to vector-valued functions $a(\mathbf{n})$ defined over the set of unit vectors \mathbf{n} . Expression (3.13) reveals that the effective potential of these dilute sequential laminates depends on the microstructure *only* through the volume fractions $f^{(r)}$ of each phase r and the reduced H-measure v_M .

(c) Estimates based on differential laminates

The above dilute sequential laminates are now used as seed microgeometries in an iterative differential scheme to generate microgeometries of *finite* volume fractions and prescribed H-measures. The scheme presented here follows the works of Idiart [10] and Lopez-Pamies [32]. Throughout this subsection, a subscript $[k]$ is used to denote quantities associated with the k th iteration in the differential scheme.

The starting point is to consider a certain class of particulate composites with matrix and inclusion phases characterized by $w^{(1)}$ and $w^{(2)}$. Let $\hat{f}_{[1]}$ denote the volume fraction of the matrix phase in each member of the class. Now, consider a sequence of members with decreasing $\hat{f}_{[1]}$, and assume that the effective potential $\tilde{w}_{[1]}$ associated with those members exhibits a regular asymptotic behaviour as $\hat{f}_{[1]} \rightarrow 0$. We can thus write

$$\tilde{w}_{[1]}(\bar{\mathbf{E}}) = w^{(2)}(\bar{\mathbf{E}}) - \mathcal{H}[\bar{\mathbf{E}}, w^{(1)}, w^{(2)}] \hat{f}_{[1]} + O(\hat{f}_{[1]}^2), \quad (3.14)$$

where \mathcal{H} is a functional that depends on the limiting values of the multi-point correlations associated with the sequence of microgeometries.

Next, consider composites with the *same* two-phase microgeometries as those of the above composites, where the matrix phase is again made up of material $r = 1$, but the inclusion phase is made up of the limiting composite of the previous step with its microstructure oscillating at a much smaller length scale. Denote the volume fraction of the matrix domain in this second iteration by $\hat{f}_{[2]}$, and, again, consider a sequence of members with decreasing $\hat{f}_{[2]}$. According to the iterated homogenization theorem, the effective potential $\tilde{w}_{[2]}$ of these two-scale composites is given to first order in $\hat{f}_{[2]}$ by the formula (3.14), with the same functional \mathcal{H} , but with $w^{(2)}$ replaced by $\tilde{w}_{[1]}$. Note that the resulting two-scale microgeometries can be described by characteristic functions of the form (2.9) with the matrix domain being uniform. The *total* volume fraction of material $r = 1$ is then $\hat{f}_{[2]} + \hat{f}_{[1]}(1 - \hat{f}_{[2]})$. Thus, an infinitesimal amount $\hat{f}_{[2]}(1 - \hat{f}_{[1]})$ of phase 1 has been added to the two-phase composite in this second iteration.

In order to construct a microgeometry with the desired finite volume fraction $c^{(1)}$ of material $r = 1$, the iterative process must be repeated an infinite number of times. The equation for the

²This was first noted by deBotton [11] in the context of certain hyper-elastic sequential laminates.

resulting effective potential can be obtained by considering an arbitrary iteration k and taking the limit $k \rightarrow \infty$. At the k iteration, the effective potential is given by

$$\tilde{w}_{[k]}(\bar{\mathbf{E}}) = \tilde{w}_{[k-1]}(\bar{\mathbf{E}}) - \mathcal{H}[\bar{\mathbf{E}}, w^{(1)}, \tilde{w}_{[k-1]}] \hat{f}_{[k]}, \quad (3.15)$$

which contains a *total* volume fraction of phase 1 given by

$$f_k = 1 - \prod_{j=1}^k (1 - \hat{f}_{[j]}). \quad (3.16)$$

The *increment* in total concentration of the phase $r = 1$ in this iteration—i.e. in passing from $k - 1$ to k —is given by

$$f_k - f_{k-1} = \prod_{j=1}^{k-1} (1 - \hat{f}_{[j]}) - \prod_{j=1}^k (1 - \hat{f}_{[j]}) = \hat{f}_{[k]} (1 - f_{k-1}), \quad (3.17)$$

and therefore,

$$\hat{f}_{[k]} = \frac{f_k - f_{k-1}}{1 - f_{k-1}}. \quad (3.18)$$

Substituting (3.18) in expression (3.15) yields the difference equation

$$(1 - f_{k-1}) \frac{\tilde{w}_{[k]}(\bar{\mathbf{E}}) - \tilde{w}_{[k-1]}(\bar{\mathbf{E}})}{f_k - f_{k-1}} + \mathcal{H}[\bar{\mathbf{E}}, w^{(1)}, \tilde{w}_{[k-1]}] = 0, \quad \tilde{w}_{[0]}(\bar{\mathbf{E}}) = w^{(2)}(\bar{\mathbf{E}}). \quad (3.19)$$

Letting $t_k \doteq -\ln(1 - f_k)$ and $k \rightarrow \infty$, and noting that the increment $f_k - f_{k-1}$ at each iteration is infinitesimally small, we can recast the difference equation (3.19) as a differential equation for the function $\tilde{w}(\bar{\mathbf{E}}, t)$ given by

$$\frac{\partial \tilde{w}}{\partial t}(\bar{\mathbf{E}}, t) + \mathcal{H}[\bar{\mathbf{E}}, w^{(1)}, \tilde{w}] = 0, \quad \tilde{w}(\bar{\mathbf{E}}, 0) = w^{(2)}(\bar{\mathbf{E}}), \quad (3.20)$$

which must be integrated with respect to the time-like concentration variable t over the interval $[0, -\ln c^{(2)}]$, $c^{(2)} = 1 - c^{(1)}$ being the total concentration of material $r = 2$ in the final multi-scale composite.

Thus, starting from the effective potential of a given *dilute* microgeometry, as determined by the functional \mathcal{H} , the above differential scheme yields the *exact* effective potential of a material with *finite* values of phase concentrations. If the dilute microgeometry is of particulate type, the above scheme generates non-dilute microgeometries that are also particulate. The multi-point correlations in the resulting multi-scale composite are, in general, extremely complicated functions. However, as a consequence of the scale-invariance of the H-measures described in the previous section, the reduced H-measure of the multi-scale composite is the same as the reduced measure of the seed microgeometries considered in the first step of the process. Furthermore, if the functional \mathcal{H} depends on the microgeometry only through the H-measures, so does the effective potential satisfying equation (3.20). This is precisely the case of the dilute sequential laminates of the previous subsections. In that case, the functional \mathcal{H} is given by the square brackets in (3.13). The use of those dilute sequential laminates as seed microgeometries in the differential scheme thus generates non-dilute particulate microgeometries, referred to as *differential laminates*, whose effective potential is the solution to the Hamilton–Jacobi equation

$$\frac{\partial \tilde{w}}{\partial t}(\bar{\mathbf{E}}, t) + H\left(\bar{\mathbf{E}}, \tilde{w}(\bar{\mathbf{E}}, t), \frac{\partial \tilde{w}}{\partial \bar{\mathbf{E}}}(\bar{\mathbf{E}}, t)\right) = 0, \quad \tilde{w}(\bar{\mathbf{E}}, 0) = w^{(2)}(\bar{\mathbf{E}}), \quad (3.21)$$

with $t \in [0, -\ln c^{(2)}]$ and Hamiltonian

$$H(\bar{\mathbf{E}}, \tilde{w}, \bar{\mathbf{J}}) = \tilde{w} + \max_{a(\mathbf{n})} \langle a\mathbf{n} \cdot \bar{\mathbf{J}} - w^{(1)}(\bar{\mathbf{E}} + a\mathbf{n}) \rangle_\nu, \quad (3.22)$$

where $c^{(2)}$ and ν are the volume fraction of inclusions and reduced H-measure, respectively, and where subindices have been dropped to ease the notation. The resulting potential can be regarded as an estimate for two-phase particulate composites with prescribed volume fractions

$c^{(r)}$ and reduced H-measure ν . This estimate is general enough to allow for any matrix and particle potentials and particle distributions. Moreover, by construction, this estimate is *realizable*, in the sense that it is exact for a given class of microgeometries. Consequently, it is guaranteed to be theoretically sound and to give physically sensible predictions. In particular, it is guaranteed to be convex, to satisfy all pertinent bounds, and to agree exactly with the weak-contrast expansion of Blumenfeld & Bergman [12,13] to second order.

4. Results for power-law random composites

(a) Formulae

The estimate presented above is used in this section to explore the effects of material nonlinearity on the electrical response of two-phase composite conductors. Specific results are computed for material systems with *isotropic* phases characterized by power-law potentials of the form

$$w^{(r)}(\mathbf{E}) = \frac{\chi^{(r)}}{1+m} |\mathbf{E}|^{1+m}, \quad (4.1)$$

where the $\chi^{(r)}$ are nonlinear conductivity constants, and m is a nonlinearity index such that $0 < m < \infty$. The quantity $(\chi^{(r)})^{-1/m}$ is often referred to as the nonlinear resistivity. An index $m = 1$ corresponds to a linear behaviour, while $m \rightarrow 0$ and $m \rightarrow \infty$ correspond to strongly nonlinear threshold-type behaviours. This type of response is observed, for instance, in composite materials used as varistors [33–35], and it is particularly suitable to study the effects of nonlinearity on material response.

We restrict attention to composites with *statistically isotropic, random* microstructures. In this case, the reduced H-measure is the same in all directions, and in view of the property (2.8)₂ must be given by

$$\nu(\mathbf{n}) = \frac{1}{4\pi}. \quad (4.2)$$

Furthermore, the overall response is isotropic, and so the effective energy density must be of the power-law form—see [6]

$$\tilde{w}(\bar{\mathbf{E}}) = \frac{\tilde{\chi}}{1+m} |\bar{\mathbf{E}}|^{1+m}, \quad (4.3)$$

where the effective conductivity constant $\tilde{\chi}$ depends on the local constants $\chi^{(r)}$, on the phase concentrations $c^{(r)}$, and on the nonlinearity index m . Thus, the effective behaviour exhibits the same nonlinearity index as the constituent phases and is fully characterized by the effective conductivity.

The estimate (3.21) for \tilde{w} translates into an estimate for $\tilde{\chi}$ that solves the ordinary differential equation

$$\frac{d\tilde{g}}{dt} + H_{\text{pl}}(\tilde{g}) = 0, \quad \tilde{g}(0) = \frac{\chi^{(2)}}{\chi^{(1)}}, \quad (4.4)$$

where $\tilde{g}(t) \doteq \tilde{\chi}(t)/\chi^{(1)}$ and

$$H_{\text{pl}}(\tilde{g}) = \tilde{g} + \max_{\alpha(\varphi)} \frac{1}{2} \int_0^\pi ((1+m)\tilde{g}\alpha(\varphi) \cos \varphi - [1 + 2\alpha(\varphi) \cos \varphi + \alpha(\varphi)^2]^{(1+m)/2}) \sin \varphi \, d\varphi. \quad (4.5)$$

The differential equation must be integrated over the interval $[0, -\ln c^{(2)}]$. Upon integration, the concentration of phase 2 can be written explicitly in terms of $\tilde{\chi}$ as

$$\ln c^{(2)} = \int_{\chi^{(2)}/\chi^{(1)}}^{\tilde{\chi}/\chi^{(1)}} \frac{d\tilde{g}}{H_{\text{pl}}(\tilde{g})}. \quad (4.6)$$

This integral must be evaluated numerically, in general.

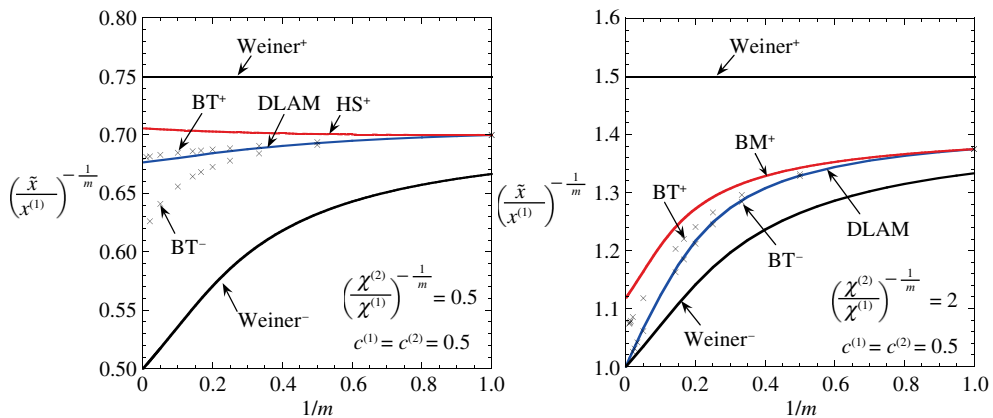


Figure 3. Effective resistivity $\tilde{\chi}^{-1/m}$ of isotropic power-law composites, as a function of the nonlinearity index $m \geq 1$. Bounds and estimates for volume fractions $c^{(1)} = c^{(2)} = 0.5$, and two values of the resistivity contrast: (a) $(\chi^{(2)}/\chi^{(1)})^{-1/m} = 0.5$ and (b) $(\chi^{(2)}/\chi^{(1)})^{-1/m} = 2$. (Online version in colour.)

The new estimates based on differential laminates (DLAM) are compared next with the elementary bounds of Weiner [36], the nonlinear Hashin–Shtrikman bounds (HS) of Talbot & Willis [16] and Ponte Castañeda [6], and the three-point nonlinear Beran–Milton (BM) of Ponte Castañeda [6]. Also included are the full-field numerical simulations by Barrett & Talbot [37] for composite spheres subject to an affine electric potential or to a uniform current density on the external boundary. These simulations provide, respectively, upper and lower bounds (BT) for the overall conductivity of a particular class of composites known as composite-sphere assemblages; when the phases are linear, the simulations provide the exact result.

Note that upper bounds on the effective conductivity $\tilde{\chi}$ correspond to lower bounds on the effective resistivity $\tilde{\chi}^{-1/m}$, and vice versa.

(b) Results for $m \geq 1$

Figure 3 shows various predictions for the effective resistivity $(\tilde{\chi})^{-1/m}$, normalized by the resistivity of the matrix phase $(\chi^{(1)})^{-1/m}$, as a function of the nonlinearity index $m \geq 1$, for the choice $c^{(1)} = c^{(2)} = 0.5$ and two values of the resistivity contrast. Recall that in this range of nonlinearity index m , the HS results provide rigorous upper bounds for the resistivity of all composites with statistically isotropic microstructures, whereas the BM results provide upper bounds for the resistivity of all composites with isotropic microstructures with extreme third-order Milton parameter. The differential laminates constructed in §3 may be interpreted as belonging to these classes—see sections 23.3 and 26.2 in Milton [5].

We begin by noting that for linear materials ($m = 1$) the nonlinear HS and BM bounds agree exactly with the linear bounds of Hashin & Shtrikman [2], Beran [18] and Milton [19] on which they are based. These linear bounds are known to be attained by composites with isotropic composite-sphere assemblage and sequentially laminated microstructures [5], and that is why the BT and DLAM results also agree with the bounds in this case. For nonlinear materials ($m > 1$), the various methods give somewhat different predictions, but they all predict a decreasing resistivity with increasing nonlinearity. In particular, the DLAM estimates are seen to lie well below the linear-comparison bounds and above the Weiner lower bound for all m , as expected from their realizability.

In the extremely nonlinear limit $m \rightarrow \infty$, the local potentials lose strict convexity and the current density field may localize along one-dimensional lines running throughout the composite avoiding the particles when these are more resistive than the matrix or going through the particles when these are more conductive—see, Duxbury *et al.* [38]. For composites with more resistive particles, the DLAM estimates and BT⁻ bound predict no effect on the effective resistivity owing

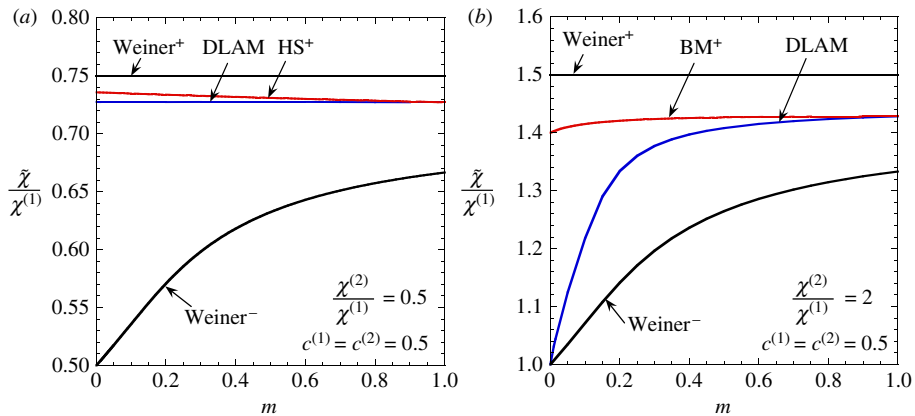


Figure 4. Effective resistivity $\tilde{\chi}$ of isotropic power-law composites, as a function of the nonlinearity index $m \leq 1$. Bounds and estimates for volume fractions $c^{(1)} = c^{(2)} = 0.5$ and two values of the resistivity contrast: (a) $\chi^{(2)}/\chi^{(1)} = 0.5$, (b) $\chi^{(2)}/\chi^{(1)} = 2$. (Online version in colour.)

to the addition of particles and are, therefore, consistent with current densities localized along straight lines running through the matrix, whereas the HS bound and BT^+ bound do predict an effect and are, therefore, consistent with current densities that are either diffuse or localized along curved rather than straight lines. The fact that the BT^+ bound is obtained by prescribing uniform current densities on the boundary of a composite sphere prevents it from capturing the presence of localized current fields. For composites with less resistive particles, the DLAM estimates lie very close to the BT^+ bound and not far from the HS bound, whereas the BT^- bound lies somewhat below. This is because the DLAM estimates are based on differential laminates where the current density field is uniform in the inclusion phase, and are therefore unable to capture localized current densities going through the particles.

The above trends have been found to hold regardless of particle volume fraction.

(c) Results for $m \leq 1$

Figure 4 shows various predictions for the effective conductivity $\tilde{\chi}$, normalized by the conductivity of the matrix phase $\chi^{(1)}$, as a function of the nonlinearity index $m \leq 1$, for the choice $c^{(1)} = c^{(2)} = 0.5$ and two values of the conductivity contrast. Recall that in this range of nonlinearity index m , the HS results provide rigorous upper bounds for the conductivity of all composites with statistically isotropic microstructures, whereas the BM results provide upper bounds for the conductivity of all composites with isotropic microstructures with extreme third-order Milton parameter. Recall also that the differential laminates belong to these classes.

The results show that the bounds and estimates for $m < 1$ exhibit the same qualitative trends as those already observed for $m > 1$. In particular, the DLAM estimates are seen to satisfy all bounds, as expected. In the limit $m \rightarrow 0$ the local potentials lose strict convexity and the electric field may localize on two-dimensional surfaces running through the composite. These surfaces tend to avoid the particles when these are more conductive than the matrix, and tend to seek the particles when these are more resistive—see, Duxbury *et al.* [38]. For composites with more conductive particles, the DLAM estimates predict no effect on the effective conductivity owing to the addition of particles and are, therefore, consistent with electric fields localized on flat surfaces running through the matrix, whereas the BM bound does predict an effect and is, therefore, consistent with electric fields that are either diffuse or localized along curved rather than flat surfaces. For composites with less conductive particles, the DLAM estimates lie close to the HS bound, which is a consequence of the fact that they are based on differential laminates where the electric field is uniform in the inclusion phase, and are, therefore, unable to capture localized electric fields going through the particles.

Once again, the above trends have been found to hold regardless of particle volume fraction.

5. On the possible extremal character of differential laminates

Idiart [10] conjectured that the differential laminates of §3c could exhibit extremal properties within the class of two-phase well-ordered composites with prescribed phase concentrations and H-measures.³ This conjecture was based on the fact that those laminates attain the Hashin–Shtrikman bounds for that class of composites whenever the matrix phase is linear—see also Idiart [39]. It is shown here via a counterexample that the conjecture does *not* hold in general, but may still hold in a restricted sense.

To that end, we focus on materials with power-law phase potentials (4.1) and *cylindrical* microstructural symmetry, subject to *in-plane* electric fields $\bar{\mathbf{E}}$. In this case, the reduced H-measure $\nu(\mathbf{n})$ takes non-zero values on the unit circle in the transverse plane perpendicular to the axis of microstructural symmetry—see, Idiart [10]. In turn, the effective potential \tilde{w} can be written in the form (4.3) with an effective conductivity $\tilde{\chi}$ depending on the angle $\bar{\beta}$ formed by the applied field $\bar{\mathbf{E}}$ and a reference in-plane direction $\hat{\mathbf{e}}_1$. Within this class of material systems, we consider three different microgeometries.

First, we consider differential laminates with transversely isotropic H-measures. In this case, the reduced H-measure is supported on the unit circle with uniform line density $(2\pi)^{-1}$ in view of the constraint (2.8)₂, and the effective conductivity $\tilde{\chi}$ is the solution to the Hamilton–Jacobi equation (4.4) for $\tilde{g} = \tilde{\chi}/\chi^{(1)}$ with Hamiltonian

$$H_{\text{pl}}(\tilde{g}) = \tilde{g} + \max_{\alpha(\varphi)} \frac{1}{2\pi} \int_0^{2\pi} ((1+m)\tilde{g}\alpha(\varphi)\cos\varphi - [1 + 2\alpha(\varphi)\cos\varphi + \alpha(\varphi)^2]^{(1+m)/2}) d\varphi. \quad (5.1)$$

Note that the effective response of these differential laminates is transversely isotropic so that the effective conductivity is independent of the angle $\bar{\beta}$.

Secondly, we consider rank- M sequential laminates with $\check{\nu}_i = 1/M$ and two different sets of (in-plane) lamination directions $\mathbf{n}_i = \cos\theta_i\hat{\mathbf{e}}_1 + \sin\theta_i\hat{\mathbf{e}}_2$ where

$$\text{LAM-1: } \{\theta_i\} = \bigcup_{i=1}^M \left\{ 2\pi \frac{i-1}{M} \right\} \quad (5.2)$$

and

$$\text{LAM-2: } \{\theta_i\} = \bigcup_{i=1}^{M/4} \left\{ 2\pi \frac{i-1}{M}, \frac{\pi}{2} + 2\pi \frac{i-1}{M}, \pi + 2\pi \frac{i-1}{M}, \frac{3\pi}{2} + 2\pi \frac{i-1}{M} \right\}. \quad (5.3)$$

The rank M must be a multiple of 4; figure 5a shows LAM-1 and LAM-2 sequences represented as points of equal weight on the unit circle for $M=8$; LAM-1 is a counterclockwise incremental sequence of angles, whereas LAM-2 alternates between quadrants. In any event, the important point is that both lamination sequences are such that in the limit $M \rightarrow \infty$ their H-measures become transversely isotropic. The effective conductivity of these rank- M laminates can be computed by evaluating expression (3.2) numerically. The results to be given below have been obtained by setting $M=100$. However, increasing M beyond that value did not change significantly the results and so the results for $M=100$ can be taken as accurate approximations of the corresponding results for $M \rightarrow \infty$.

Figure 5b shows plots of the effective conductivity $\tilde{\chi}$ versus $\bar{\beta}$, normalized by the matrix conductivity $\chi^{(1)}$, of the above sequential (LAM-1, LAM-2) and differential (DLAM) laminates with transversely isotropic H-measures, for the choice $c^{(2)} = 0.2$, $m = 0.1$ and $\chi^{(2)}/\chi^{(1)} = 0.2$. Also included in the figure are the nonlinear Weiner and Hashin–Shtrikman (HS^+) upper bounds. In the linear case, all these materials exhibit the same transversely isotropic response and attain the Hashin–Shtrikman bound. In the nonlinear case considered in the figure, however, the

³The conjecture was made in the mathematically related context of viscoplasticity.

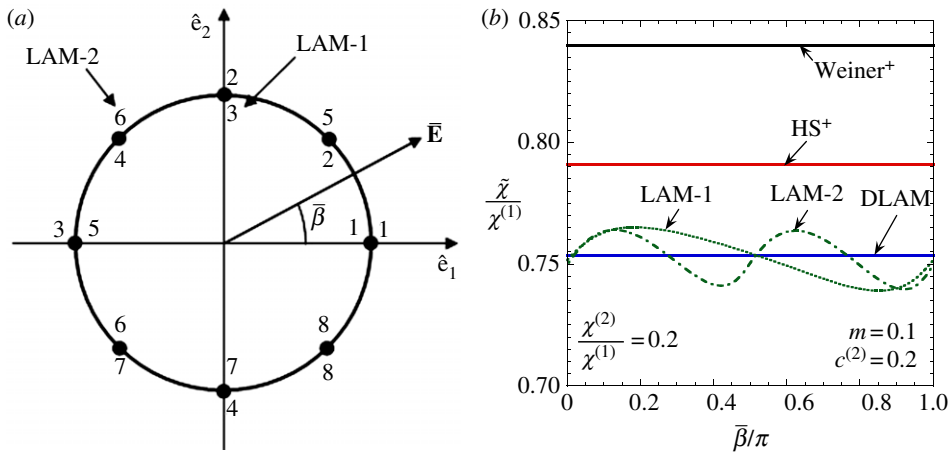


Figure 5. Power-law composites with cylindrical symmetry. (a) Lamination sequences of LAM-1 and LAM-2 microgeometries of rank $M = 8$; (b) effective in-plane conductivity $\tilde{\chi}$ of LAM-1 and LAM-2 laminates of rank $M = 100$, as a function of the applied field direction $\bar{\beta}$, compared with that of the differential laminates (DLAM) and the Hashin–Shtrikman upper bound (HS^+) for composites with transversely isotropic H-measures. The volume fraction of particles is $c^{(2)} = 0.2$ and the heterogeneity contrast is $\chi^{(2)} / \chi^{(1)} = 0.2$. (Online version in colour.)

various materials exhibit different responses. In particular, it is observed that the response of the sequential laminates becomes anisotropic and that the LAM-1 and LAM-2 sequences give rise to different responses, even though their underlying microgeometries have both transversely isotropic H-measures. This means that the effective potential of these materials depends on microstructural information beyond the H-measures, as already anticipated in §3a. The effective potential of the differential laminates, on the other hand, depends on the H-measure only, and consequently, remains transversely isotropic in the nonlinear case. In any case, all these materials are well-ordered such that $w^{(1)} \geq w^{(2)}$, and so according to the aforementioned conjecture the DLAM results should provide an upper bound in this case. However, the LAM-1 and LAM-2 results are seen to be larger than the DLAM results for certain ranges of $\bar{\beta}$ and, therefore, the conjecture of Idiart [10] is disproved. On the other hand, note that the HS^+ bound still holds for all three laminates with transversely isotropic H-measures, regardless of whether they exhibit overall transversely isotropic behaviour or not. This is because while linear-comparison bounds depend only on the volume fractions and H-measures, they do not exclude the possibility that the overall response of the nonlinear composite may be more general than that suggested by the symmetry of the H-measures. Even though the linear-comparison bound is (transversely) isotropic in the particular example considered in figure 5b, it includes composites with microstructures that do not have isotropic overall behaviour, and, therefore, it is not evident that it can be improved without further restrictions on the overall behaviour of the composites in the given class.

In conclusion, the differential laminates of §3c cannot be bounds for the class of two-phase, well-ordered, nonlinear composites with prescribed phase concentrations and H-measures. On the other hand, the question still remains open as to whether, or not, the linear comparison bounds of the HS-type can be significantly improved without further restrictions on the above-defined class of microstructures. In this connection, it should be remarked—as already noted by deBotton & Harriton [40] and Harriton & deBotton [9]—that the macroscopic response of the finite-concentration sequential laminates (LAM-1 and LAM-2) becomes transversely isotropic as the lamination sequences are repeated many times. In fact, we have shown that it tends to the response of the differential laminates (DLAM), which follows from the fact that the concentration of the matrix phase in the first lamination sequence for the finite-concentration sequential laminates must tend to zero as the number of lamination sequences increases. As

a consequence, it is conceivable that the differential laminates (DLAM) may be extremal for appropriate *subclasses* of microstructures with additional hypotheses on the overall macroscopic symmetry of the nonlinear composites. Additional work will be required to further elucidate this point.

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