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Bounds and Estimates for the Properties of Nonlinear Heterogeneous Systems

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Bounds and estimates for the properties of nonlinear heterogeneous systems

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Contents

A powerful and versatile variational principle, allowing the estimation of the effective properties of nonlinear heterogeneous systems, has been introduced recently by Ponte Castañeda (1992). The central idea is to express the effective energy-density function of a given nonlinear composite in terms of an optimization problem involving the effective energy-density functions of linear comparison composites with similar microstructure. This permits the computation of bounds and estimates for the effective properties of given classes of nonlinear heterogeneous systems directly from well-known bounds and estimates for the effective properties of corresponding classes of linear comparison composites. In this paper, we review the variational principle and apply it to determine bounds and estimates for the effective properties of certain classes of nonlinear composite dielectrics with homogeneous, isotropic phases. Thus, nonlinear bounds of the Hashin–Shtrikman and Beran types are obtained for composites with overall isotropy and prescribed volume fractions (of the phases). While nonlinear (second-order) bounds of the Hashin–Shtrikman type have been obtained previously, in different form, by other methods, the nonlinear

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(higher-order) Beran bounds are the first of their type. Finally, exact estimates are also obtained for nonlinear composites with 'sequentially layered' microstructures. These special composites, which have proved to be extremely useful in assessing the optimality of bounds for linear systems, are also useful, although to a lesser extent, in assessing the sharpness of the nonlinear bounds.

1. Introduction

This paper deals mainly with the development of methods for estimating and bounding the effective properties of nonlinear heterogeneous systems. Although the presentation of the analyses and results will be phrased in the context of nonlinear electrostatics, the emphasis is not on determining the nonlinear dielectric constant of any specific material system, but rather to develop methods that would be of general application in connection with several of the physical and mechanical properties of material systems. Thus, we could equally well have chosen to set our problem in the context of nonlinear conductivity, magnetostatics or diffusion. More rudimentary versions of the methods in this paper have also been applied by Ponte Castañeda (1991*a*, *b*) to more complex systems, such as nonlinearly viscous materials.

The study of the effective behaviour of heterogeneous systems is a classical problem that has attracted the attention of numerous investigators in many different fields. However, most of the efforts thus far have concentrated on the effective behaviour of linear systems. In particular, the problem of estimating the effective dielectric constant (or, analogously, the effective conductivity) of a linear dielectric (or conductor) has been a central one. On the other hand, the study of nonlinear heterogeneous systems had not received much attention until very recently, except in some highly specialized areas, such as nonlinear optics. Some examples of nonlinear material behaviour in the context of electrical-transport phenomena are provided by dielectric breakdown, burning out of fuses and laser phenomena. Many other examples could be given in the realm of electrical, and other physical and mechanical properties of matter. In the next few paragraphs, I briefly review some of the key relevant developments in the theory of linear heterogeneous systems, as well as some of the more recent developments in the emerging nonlinear theories.

As is by now well known, the effective properties of most heterogeneous systems are not characterized by a simple average of the properties of the constituent phases weighted by their respective volume fractions, and in general involve dependence on microstructural parameters other than the volume fractions. Thus a proper definition of effective properties is required to have a well-founded theory. One possibility that applies to a large number of systems, introduced by Hill (1963) and Hashin (1964) in the context of elasticity, is to define these properties in terms of the effective, or overall, energy of the heterogeneous system resulting from special classes of uniform boundary conditions. According to this definition, different boundary conditions will in general lead to different effective properties for the system. However, on physical grounds, it is assumed that, as the size of the typical heterogeneity becomes small compared to the size of the specimen under consideration, the heterogeneous material behaves like a homogeneous material with effective properties that are independent of the specific boundary conditions applied to the system.

This intuitive notion of effective properties has been made mathematically rigorous by the field of homogenization, which deals with the study of the existence and uniqueness of such effective properties in the limit of vanishingly small microstructure. Thus homogenization theory is concerned with the description of an idealized material that corresponds to the limit of a sequence of heterogeneous materials with two distinct length scales: one microscopic, l, corresponding to the size of the typical heterogeneity, and one macroscopic, L, corresponding to the size of the specimen of interest and the scale of variation of the boundary conditions and forcing functions applied to the system. The effective behaviour of the homogenized materials is then obtained by considering the limit of the behaviour of the sequence of heterogeneous materials as the ratio of scales, $\epsilon = l/L$, tends to zero. I also refer to this idealized limit material as a 'composite'. Two standard references in the context of periodic homogenization are provided by the monographs of Sanchez-Palencia (1980) and Bensoussan *et al.* (1978).

Many different methods have been proposed in the literature to predict the effective properties of linear heterogeneous systems, and they can generally be classified into three different categories. The first class of methods consists in identifying specific microstructures for which the effective properties can be computed exactly (e.g. periodic microstructures). An alternative approach, which is often more useful in practice (because the exact microstructure of most composite materials is usually not known precisely), is to define classes of composites for which the microstructure is only partially specified in terms of some known microstructural parameters. The goal of this approach is then to determine the range of possible behaviours for a given material class. Thus this approach seeks to characterize the effective behaviour of classes of material systems by specifying optimal bounds (i.e. bounds that are attainable by special members of the class) on the effective properties of the system. A third approach consists in the postulation of approximate models attempting to capture the essential features of the microstructure of a given system, or class of systems.

Examples of the first approach are provided by the periodic computations of McPhedran & McKenzie (1978), the composite-spheres assemblage models of Hashin & Shtrikman (1962) and the sequentially layered laminates, introduced by Bruggeman (1935), and used more recently by Schulgasser (1976) and Milton (1986), among others. Examples of the second approach are given by the bounds of Wiener (1912), Hashin & Shtrikman (1962) and Beran (1965). The Hashin–Shtrikman bounds have been extended and rederived in a number of fundamentally different ways by several authors. Walpole (1966, 1969), Willis (1977), and Kohn & Milton (1988) proposed extensions of the Hashin–Shtrikman variational principles to include, in particular, anisotropic effective behaviour. Bergman (1978) made use of analytic function theory to obtain an alternative derivation of the Hashin–Shtrikman bounds. Tartar (1985) proposed yet a different method, developed jointly with Murat (1978) (see also Murat & Tartar 1985), for bounding the effective properties of anisotropic composites making use of the notions of compensated compactness. Lurie & Cherkaev (1984, 1986) independently proposed closely related methods, which also were designed to deal with anisotropic composites. A detailed comparison between the different methods is given by Kohn & Milton (1986) and Milton (1990). The more stringent bounds of Beran have also been given a simpler form by Milton (1981) for the case of two-phase isotropic composites in terms of one geometric parameter (additional to the volume fractions) containing third-order statistical information. (The Hashin–Shtrikman bounds contain only up to second-order information.) Finally, examples of the third approach are provided by the self-consistent and other effective-medium theories, also initiated by Bruggeman (1935), and used more recently by numerous investigators (see Landauer 1978; Milton 1985).

For nonlinear heterogeneous systems, the number of investigations dealing with effective behaviour is comparatively small. Thus, in the context of the definition of effective properties, Willis (1986) has applied the approach of Hill (1963) to nonlinear dielectrics, and Marcellini (1978) has extended the results of homogenization theory to nonlinear media in the periodic context. On the other hand, the actual computation of effective properties in the nonlinear context has involved for the most part approximate methods that are generally problem-specific. For example, Miksis (1983) obtained results for the effective properties of periodic arrays, and random (in a self-consistent sense) distributions of nonlinear spherical inclusions in a linear matrix. Other approximate methods have been proposed recently by Stroud & Hui (1988) and Zeng *et al.* (1988) for weakly nonlinear materials. In terms of bounding methods, Willis (1986) and Talbot & Willis (1985) have proposed extensions of the Hashin–Shtrikman variational principles to nonlinear heterogeneous systems. Additionally, Talbot & Willis (1987) have used this method to compute bounds for the effective properties of nonlinear heterogeneous dielectrics, and compared them with the results of some self-consistent embedding procedures.

In this work, I apply variational principles (Ponte Castañeda 1992) to establish bounds and exact estimates for the effective properties of nonlinear heterogeneous systems with isotropic constituents. I begin, in the next section, by introducing the definition of effective properties in terms of the classical principles of minimum energy and minimum complementary energy. Additionally, I summarize the variational principles of Ponte Castañeda (1992) (see Theorems 2.1 and 2.2). These variational principles are designed to yield the effective properties of nonlinear heterogeneous systems in terms of the effective properties of suitably optimized linear heterogeneous systems. This procedure enables the translation of the large number of results available for linear composites directly into corresponding estimates for nonlinear composites. In §3, I use the variational principles (see Corollaries 3.1 and 3.2) to determine rigorous bounds for the effective properties of arbitrarily nonlinear heterogeneous dielectrics with prescribed volume fractions. Thus I show that the so-called Weiner bounds for arbitrarily anisotropic composites, which can be obtained directly from the classical minimum principles, can also be recovered from the new variational principles. More interestingly, I also develop sharper bounds for the class of isotropic nonlinear heterogeneous dielectrics by making use of the corresponding Hashin–Shtrikman bounds for linear dielectrics, and obtain even sharper bounds still for the class of two-phase, isotropic nonlinear systems by applying the Beran bounds for linear systems to the new variational principles. In §4, I introduce special microgeometries for which the effective nonlinear properties can be computed exactly. The new variational principles also play a central role in the derivation of these exact results (see Corollaries 4.1 and 4.2). The special microgeometries correspond to sequentially laminated materials, which have been found to yield the extremal properties of many classes of linear systems, both isotropic and anisotropic, and hence have been used in the linear theories to study the sharpness and optimality of bounds. Thus in §4, I also show that the nonlinear Weiner bounds are sharp. More importantly, I find in §§4 and 5 that, at least for two-phase systems, sequentially laminated nonlinear materials can be constructed closely approximating the nonlinear isotropic bounds of the Hashin–Shtrikman type obtained in §3. This suggests that, although the nonlinear bounds of

§3 may not be optimal in general, they are probably not too far from the optimal bounds (which, presumably, would be extremely difficult to determine precisely).

2. Effective properties

(*a*) *Definition*

I consider a heterogeneous dielectric occupying a region in space of unit volume Ω. The nonlinear constitutive behaviour of the material may be characterized in terms of an electric energy-density function, $w(\boldsymbol{x}, \boldsymbol{E})$, depending on the position vector $\boldsymbol{x}$ and the electric field $\boldsymbol{E}(\boldsymbol{x})$, such that the electric displacement field $\boldsymbol{D}(\boldsymbol{x})$ is expressed by

$$\boldsymbol{D}(\boldsymbol{x}) = \partial_{\boldsymbol{E}} w(\boldsymbol{x}, \boldsymbol{E}), \tag{2.1}$$

where $\partial_{\boldsymbol{E}}$ denotes differentiation with respect to $\boldsymbol{E}$, assuming differentiability of w. However, in the context of the present work, it will suffice to assume convexity and continuity of w on $\boldsymbol{E}$. In this event, $\partial_{\boldsymbol{E}}$ can still be given the interpretation of the subdifferential of convex analysis (Ekeland & Temam 1974; §I.5). As stated in §1, I further assume local isotropy, so that I can write

$$w(\boldsymbol{x}, \boldsymbol{E}) = \phi(\boldsymbol{x}, E), \tag{2.2}$$

where $\phi: \Omega \times R \rightarrow R$ is continuous, convex and coercive (in a sense to be specified later) in the magnitude of the electric field E. Here, R is the set of the extended real numbers. Additionally, I assume that ϕ satisfies the conditions

$$\phi(\boldsymbol{x}, E) \geqslant 0 \,\forall \boldsymbol{x}, E, \quad \phi(\boldsymbol{x}, 0) = 0 \,\forall \boldsymbol{x}. \tag{2.3}$$

It was shown by Hill (1963) (see also Hashin 1964; Willis 1986) that the effective constitutive behaviour of the heterogeneous dielectric may be defined by a relation analogous to (2.1),

$$\bar{\boldsymbol{D}} = \partial_{\bar{\boldsymbol{E}}} \tilde{W}(\bar{\boldsymbol{E}}), \tag{2.4}$$

relating the spatial averages of the fields, $\bar{\boldsymbol{D}}$ and $\bar{\boldsymbol{E}}$, through the effective energy-function of the nonlinear dielectric, $\tilde{W}(\bar{\boldsymbol{E}})$. I emphasize that the effective behaviour of the heterogeneous dielectric, as characterized by $\tilde{W}(\bar{\boldsymbol{E}})$, may in general be anisotropic, even though the phases themselves are assumed to be isotropic.

In principle, $\tilde{W}(\bar{\boldsymbol{E}})$ may be determined by solving the electrostatic problem on Ω, given by the restricted version of Faraday's law

$$\nabla \times \boldsymbol{E} = 0, \tag{2.5}$$

and Gauss's law for a vanishing distribution of free charge

$$\nabla \cdot \boldsymbol{D} = 0, \tag{2.6}$$

subject to a uniform boundary condition

$$\varphi = -\bar{\boldsymbol{E}} \cdot \boldsymbol{x} \text{ on } \partial\Omega, \tag{2.7}$$

where φ is the electrostatic potential and is such that $\boldsymbol{E} = -\nabla\varphi(\boldsymbol{x})$ in Ω (which is equivalent to Faraday's law). Note that boundary condition (2.7) ensures that the average of the electric field is in fact $\bar{\boldsymbol{E}}$, in the sense that

$$\bar{\boldsymbol{E}} = \int_{\Omega} \boldsymbol{E}(\boldsymbol{x})\, \mathrm{d}x. \tag{2.8}$$

On the other hand, the average displacement field is defined by the similar relation

$$\bar{\boldsymbol{D}} = \int_{\Omega} \boldsymbol{D}(\boldsymbol{x})\,\mathrm{d}x. \tag{2.9}$$

Thus the effective energy of the heterogeneous dielectric $\tilde{W}(\bar{\boldsymbol{E}})$ is computed by evaluating the pertinent energy functional for the heterogeneous dielectric

$$W(\boldsymbol{E}) = \int_{\Omega} w(\boldsymbol{x}, \boldsymbol{E}(\boldsymbol{x}))\,\mathrm{d}x,$$

at the actual electric field solving the electrostatic problem, as defined by (2.5) to (2.7), for a given microstructure. The effective constitutive behaviour of the heterogeneous material, in the form of a functional relation between $\bar{\boldsymbol{E}}$ and $\bar{\boldsymbol{D}}$, then follows from (2.4). However, due to the complexity of the microstructure of actual materials, it is often impractical to solve the electrostatic problem. For this reason, alternative variational formulations of the problem are helpful. There are two standard (dual) variational formulations of the electrostatic problem: the minimum energy and minimum complementary-energy principles. These will be used in the next subsection to provide alternative descriptions of the effective energy functions of the nonlinear heterogeneous dielectrics.

(*b*) *Classical minimum energy principles*

The first standard variational principle is the minimum energy principle. This variational principle, expressed in terms of the energy functional W, may be used to obtain the following expression for the effective energy of the heterogeneous dielectric, namely,

$$\tilde{W}(\bar{\boldsymbol{E}}) = \min_{\boldsymbol{E} \in K} W(\boldsymbol{E}), \tag{2.10}$$

where

$$K = \{\boldsymbol{E} \,|\, \boldsymbol{E} = -\nabla\varphi(\boldsymbol{x}) \text{ in } \Omega, \text{ and } \varphi = -\bar{\boldsymbol{E}}\cdot\boldsymbol{x} \text{ on } \partial\Omega\} \tag{2.11}$$

is the set of admissible electric fields.

Note that to guarantee the existence of a minimizer of (2.10) certain conditions on the growth of w (or ϕ) as $E \rightarrow \infty$ are needed. Thus I assume that w is coercive in the sense that $w \rightarrow \infty$ as $E \rightarrow \infty$. Additionally, I remark that strict convexity of w guarantees uniqueness of the solution, and that convexity of w ensures the convexity of $\tilde{W}$ (see, for example, the Appendix of Ponte Castañeda & Willis 1988). Finally, note that if the fields are smooth enough, statement (2.10) is equivalent to the electrostatics problem, as given by (2.5) to (2.7). More generally, however, (2.10) provides a weak statement of the electrostatics problem allowing the possibility of discontinuous fields, such as those that would arise in materials with discontinuous properties separated by sharp interfaces.

The second characterization of the effective constitutive behaviour of the heterogeneous dielectric is obtainable from the effective complementary-energy function for the dielectric, $\tilde{U}$. This energy function may be defined in terms of the principle of minimum complementary energy via

$$\tilde{U}(\bar{\boldsymbol{D}}) = \min_{\boldsymbol{D} \in S} U(\boldsymbol{D}), \tag{2.12}$$

where

$$U(\boldsymbol{D}) = \int_{\Omega} w^{*}(\boldsymbol{x}, \boldsymbol{D}(\boldsymbol{x}))\,\mathrm{d}x$$

is the complementary energy functional, expressed in terms of the Legendre–Fenchel dual (convex polar) of the energy function w,

$$w^*(\boldsymbol{x},\boldsymbol{D}) = \sup_{\boldsymbol{E}}\{\boldsymbol{E}\cdot\boldsymbol{D} - w(\boldsymbol{x},\boldsymbol{E})\}, \tag{2.13}$$

and where
$$S = \{\boldsymbol{D}\,|\,\nabla\cdot\boldsymbol{D} = 0 \text{ in } \Omega, \text{ and } \boldsymbol{D}\cdot\boldsymbol{n} = \bar{\boldsymbol{D}}\cdot\boldsymbol{n} \text{ on } \partial\Omega\} \tag{2.14}$$

is the set of admissible electric displacement fields. Note that the first condition in this set must be given a weak interpretation if the displacement field happens to be discontinuous. Then, given the conditions implicit in (2.14), relation (2.12) is a weak statement of (2.5). Further, if (2.8) is reinterpreted as a definition for the average electric field, we have the following relation analogous to (2.4), namely,

$$\bar{\boldsymbol{E}} = \partial_{\bar{\boldsymbol{D}}}\tilde{U}(\bar{\boldsymbol{D}}), \tag{2.15}$$

where it now follows from (2.14) that $\bar{\boldsymbol{D}}$ must satisfy the relation (2.9).

I emphasize, however, that the duality relation between the local energy functions w and w^* (recall that w is convex and continuous, and hence by Propositions I.3.1 and I.4.1 of Ekeland & Temam (1974))

$$w(\boldsymbol{x},\boldsymbol{E}) = \sup_{\boldsymbol{D}}\{\boldsymbol{E}\cdot\boldsymbol{D} - w^*(\boldsymbol{x},\boldsymbol{D})\}, \tag{2.16}$$

does not necessarily carry over to the effective energy functions $\tilde{W}$ and $\tilde{U}$ (which is also known to be convex). In fact, Willis (1989) has shown that, in general,

$$\tilde{W}(\bar{\boldsymbol{E}}) \geqslant \tilde{U}^*(\bar{\boldsymbol{E}}). \tag{2.17}$$

The reason for the inequality is related to the fact that definitions (2.10) for $\tilde{W}$ and (2.12) for $\tilde{U}$ correspond to different boundary conditions on the heterogeneous material (Dirichlet versus Neumann conditions), thus leading to generally distinct effective energies.

However, as I mentioned in §1, it follows from the results of homogenization theory that strict equality holds in the above relation for a composite (in the sense of a heterogeneous material that is homogeneous in a large enough scale). In particular, Marcellini (1978) has defined the effective properties of a nonlinear composite with periodic microstructure in terms of relations analogous to (2.4) with (2.10) (or, dually, (2.15) with (2.12)). In this context, the effective properties of the composite are defined by the solution of the electrostatics problem over a unit cell of the composite, with the uniform boundary conditions in (2.11) (or (2.14)) being replaced by periodic conditions over the unit cell. One advantage of this formulation is that equality then holds exactly in (2.17). Although either definition could be used in the present work, I prefer the earlier definition of effective properties (i.e. (2.4) with (2.10), or dually (2.15) with (2.12)), because it has fewer technical requirements (since the limit as the size of the typical heterogeneity vanishes is never evaluated explicitly).

Finally, I record here for later reference that (Ekeland & Temam 1974; §I.4.2), under the hypotheses given by (2.2) and (2.3),

$$w^*(\boldsymbol{x},\boldsymbol{D}) = \phi^*(\boldsymbol{x},D), \tag{2.18}$$

where ϕ^* is the convex polar function (Legendre transform) of ϕ, and D is the magnitude of $\boldsymbol{D}$.

(c) *New variational principles*

In this subsection, I discuss briefly the variational principles recently proposed by Ponte Castañeda (1992), which will be used in the next sections to determine bounds and estimates for the effective energy functions of nonlinear composites. These variational principles have been shown to be equivalent to the standard variational principles, discussed in the previous subsection, under appropriate hypothesis on the energy-density function. Also, the variational principles, just like the classical complementary-energy and Hashin–Shtrikman variational principles, are based on the Legendre transformation; however, the transformation is carried out on a suitably modified set of variables to achieve comparison with a linear heterogeneous comparison material with effective energy that can be estimated by using the linear theory. Depending on whether I start from the minimum energy or complementary-energy formulation, I obtain two versions of essentially the same result. I begin by considering the minimum energy formulation.

The new variational principle centres around a change of variables $r = h(E)$, with $h: R^+ \rightarrow R^+$ (R^+ is the set of non-negative reals) given by $h(E) = E^2$. By composition of ϕ with h^{-1}, we obtain a function $f\colon \Omega \times R^+ \rightarrow R^+$, such that

$$f(\boldsymbol{x}, r) = \phi(\boldsymbol{x}, E) = w(\boldsymbol{x}, \boldsymbol{E}). \tag{2.19}$$

We note that f has the same dependence on $\boldsymbol{x}$ as ϕ and w, and that it is continuous and coercive (but not necessarily convex) in r. Also, from (2.3), f is a non-negative function satisfying the condition that $f(\boldsymbol{x}, 0) = 0\, \forall \boldsymbol{x}$. Then, if I define the Legendre transform (convex polar) of f by

$$f^*(\boldsymbol{x}, p) = \sup_{r \geqslant 0} \{rp - f(\boldsymbol{x}, r)\}, \tag{2.20}$$

it follows that

$$f(\boldsymbol{x}, r) \geqslant \sup_{p \geqslant 0} \{rp - f^*(\boldsymbol{x}, p)\}. \tag{2.21}$$

Note that $\boldsymbol{x}$ is fixed in the above operations, and that the suprema are evaluated over the sets of non-negative r and p, respectively (this follows from the fact that f is non-negative and such that $f(\boldsymbol{x}, 0) = 0$; see the Appendix of Ponte Castañeda (1992)). Also, the right-hand side of inequality (2.21) is the bipolar of f, which has the geometric interpretation of the convex envelope of f, and hence the inequality. Propositions I.3.1 and I.4.1 of Ekeland & Temam (1974) ensure that equality is achieved in (2.21) if f is convex and continuous (in r). Therefore, assuming that the energy function w in (2.20) is such that f is convex (note that f convex implies that w is convex), I obtain from (2.21) the following representation for the local energy-density function of the nonlinear heterogeneous material, namely,

$$w(\boldsymbol{x}, \boldsymbol{E}) = \sup_{\epsilon_0 \geqslant 0} \{w_0(\boldsymbol{x}, \boldsymbol{E}) - v(\boldsymbol{x}, \epsilon_0)\}, \tag{2.22}$$

where (from (2.20))

$$v(\boldsymbol{x}, \epsilon_0) = \sup_{\boldsymbol{E}} \{w_0(\boldsymbol{x}, E) - w(\boldsymbol{x}, \boldsymbol{E})\}. \tag{2.23}$$

In the above relations, p has been identified with $\frac{1}{2}\epsilon_0$ and r with E^2, in such a fashion that $w_0(\boldsymbol{x}, \boldsymbol{E}) = \frac{1}{2}\epsilon_0(\boldsymbol{x}) E^2$ and $v(\boldsymbol{x}, \epsilon_0) = f^*(\boldsymbol{x}, \frac{1}{2}\epsilon_0)$. Thus w_0 corresponds to the local energy-density function of a linear, heterogeneous comparison material with arbitrary (not necessarily constant) non-negative dielectric coefficient $\epsilon_0(\boldsymbol{x})$.

The minimum energy formulation of the variational principle follows by making use of the representation (2.22) in the classical minimum energy principle (2.10), and interchanging the order of the infimum in (2.20) and the supremum in (2.22), which is allowed by an appropriate version of the saddle point theorem. The details of the proof are given in Ponte Castañeda (1992, §3.1), but the result is as follows.

Theorem 2.1. *Let the local energy-density function w of a given nonlinear heterogeneous material with isotropic phases satisfy condition (2.19) with f a non-negative, continuous, coercive and convex function of $r = E^2$, further satisfying $f(\boldsymbol{x}, 0) = 0\,\forall \boldsymbol{x}$. Then, the effective energy function of the nonlinear heterogeneous material $\tilde{W}$ is determined by the variational principle*

$$\tilde{W}(\bar{\boldsymbol{E}}) = \sup_{\epsilon_0(\boldsymbol{x}) \geqslant 0} \{\tilde{W}_0(\bar{\boldsymbol{E}}) - V(\epsilon_0)\}, \tag{2.24}$$

where
$$V(\epsilon_0) = \int_\Omega v(\boldsymbol{x}, \epsilon_0(\boldsymbol{x}))\,\mathrm{d}x, \tag{2.25}$$

is the functional generated by the function $v(\boldsymbol{x}, \epsilon_0)$, and where $\tilde{W}_0$ denotes the effective energy function of a linear heterogeneous comparison material with local energy function w_0, such that

$$\tilde{W}_0(\bar{\boldsymbol{E}}) = \min_{\boldsymbol{E} \in K} \int_\Omega w_0(\boldsymbol{x}, \boldsymbol{E})\,\mathrm{d}x. \tag{2.26}$$

I emphasize that the dielectric coefficient $\epsilon_0(\boldsymbol{x})$ of the comparison material in (2.24) is an arbitrary non-negative function in some appropriate functional space, so that (2.24) is indeed a variational statement. Later, however, I make use of piecewise constant approximations of $\epsilon_0(\boldsymbol{x})$, leading to finite-dimensional optimization problems for bounds and estimates on $\tilde{W}$.

The dual, or complementary-energy, formulation of the new variational principle follows in a similar fashion from the change of variables $s = h(D)$, where h is the same as before. This change of variables induces (by composition) a function $g\colon \Omega \times R^+ \rightarrow R^+$ such that

$$g(\boldsymbol{x}, s) = \phi^*(\boldsymbol{x}, D) = w^*(\boldsymbol{x}, \boldsymbol{D}). \tag{2.27}$$

Again, we note that g has the same dependence on $\boldsymbol{x}$ as ϕ^* and w^*, and that it is continuous and coercive in s. Similarly, it follows from (2.3) that ϕ^*, and therefore g, are non-negative functions satisfying the condition that $g(\boldsymbol{x}, 0) = \phi^*(\boldsymbol{x}, 0) = 0\,\forall \boldsymbol{x}$. Then, if we define the concave polar of g by (see Van Tiel 1974, §7.14)

$$g_*(\boldsymbol{x}, q) = \inf_{s \geqslant 0} \{sq - g(\boldsymbol{x}, s)\}, \tag{2.28}$$

it follows that
$$g(\boldsymbol{x}, s) \leqslant \inf_{q \geqslant 0} \{sq - g_*(\boldsymbol{x}, q)\}, \tag{2.29}$$

with equality if g is concave. Then, assuming that the complementary energy function w^* of the nonlinear heterogeneous material is such that g in (2.27) is concave, it follows from (2.29) that

$$w^*(\boldsymbol{x}, \boldsymbol{D}) = \inf_{\epsilon_0 \geqslant 0} \{w_0^*(\boldsymbol{x}, \boldsymbol{D}) + v(\boldsymbol{x}, \epsilon_0)\}, \tag{2.30}$$

where q has been identified with $(2\epsilon_0)^{-1}$ and s with D^2, such that $w_0^*(\boldsymbol{x}, \boldsymbol{D}) = (1/2\epsilon_0(\boldsymbol{x}))\,D^2$ is the complementary-energy function of the linear, heterogeneous comparison material with arbitrary non-negative dielectric coefficient $\epsilon_0(\boldsymbol{x})$. Here, $v(\boldsymbol{x}, \epsilon_0) = -g_*(\boldsymbol{x}, 1/2\epsilon_0)$, which can alternatively be expressed in the form

$$v(\boldsymbol{x}, \epsilon_0) = \sup_{\boldsymbol{D}} \{w^*(\boldsymbol{x}, \boldsymbol{D}) - w_0^*(\boldsymbol{x}, \boldsymbol{D})\}. \tag{2.31}$$

We note that this latter result is consistent with definition (2.23) of v, as can be seen by making use of (2.13) in the right-hand side of (2.31), and interchanging the resulting suprema.

The complementary-energy form of the variational principle then follows by letting w^* in (2.12) take the form given by (2.30), and interchanging the order of the infima. The final result may be stated (see Ponte Castañeda 1992, §3.2) in the following form.

Theorem 2.2. *Let the* (*convex*) *local complementary-energy function* w^* *of a given nonlinear heterogeneous material with isotropic phases satisfy condition* (2.27) *with* g *a non-negative, continuous, coercive and concave function of* $s = D^2$, *further satisfying* $g(\boldsymbol{x}, 0) = 0\,\forall \boldsymbol{x}$. *Then, the effective complementary-energy function of the nonlinear heterogeneous material* $\tilde{U}$ *is given by*

$$\tilde{U}(\bar{\boldsymbol{D}}) = \inf_{\epsilon_0(\boldsymbol{x}) \geqslant 0} \{\tilde{U}_0(\bar{\boldsymbol{D}}) + V(\epsilon_0)\}, \tag{2.32}$$

where

$$\tilde{U}_0(\bar{\boldsymbol{D}}) = \min_{\boldsymbol{D} \in S} \int_{\Omega} w_0^*(\boldsymbol{x}, \boldsymbol{D})\, \mathrm{d}x \tag{2.33}$$

stands for the effective complementary-energy function of the linear comparison composite, and the functional V *is as described by* (2.25).

In the above developments of the energy and complementary-energy formulations of the variational principle, the respective hypotheses of convexity of f and concavity of g were introduced independently of each other. It is natural to ask whether there is any relation between these two hypotheses. Further, it is important to emphasize that, without these hypotheses, equivalence between the classical minimum energy and new variational principles would not hold. Both of these issues are addressed in Ponte Castañeda (1992, §3.3), where it is proved that concavity of g implies convexity of f, and thus that concavity of g suffices to ensure the lack of a duality gap between the two versions of the variational principle (2.24) and (2.32), when applied to the same composite. Thus, in the developments to follow, I assume, unless stated otherwise, that w is such that g in (2.27) is concave.

I conclude this section by considering the implications of the hypothesis of concavity of g on the growth conditions on w. Recall that so far we have only assumed explicitly that w is convex and coercive ($w \rightarrow \infty$ as $E \rightarrow \infty$). Since concavity of g implies convexity of f, it implies in turn that $w \geqslant \alpha E^2$ (for some $\alpha > 0$) as $E \rightarrow \infty$. Thus a sensible growth condition may be that $w(\boldsymbol{x}, \boldsymbol{E}) \sim E^{1+n}$ ($n \geqslant 1$) as $E \rightarrow \infty$ (i.e. w is stronger than, or as strong as, quadratic). Then, f is stronger than, or as strong as, affine at infinity, which is consistent with convexity of f. On the other hand, the above assumption for w implies that $w^*(\boldsymbol{x}, \boldsymbol{D}) \sim D^{1+1/n}$ as $D \rightarrow \infty$, and therefore g is weaker than, or as weak as, affine at infinity, which is consistent with concavity of g. For a more detailed discussion of this point the reader is referred to Ponte Castañeda (1992, §3.3), but in the developments to follow the above growth conditions will be assumed implicitly. Other growth conditions are possible, but the bounds/estimates that follow may need reinterpretation, if these are different. For instance, we could let n in the above conditions be such that $0 < n \leqslant 1$. Then, the suprema and infima in the above relations would have to be replaced by infima and suprema, respectively. Further, the role of the bounds and estimates in the following discussion on bounds will also have to be appropriately adjusted.

3. Bounds

This section is concerned with the determination of bounds and estimates for the effective energy functions of classes of nonlinear heterogeneous dielectrics that are defined by the specification of appropriate statistical information on their microstructure, such as, for example, the volume fractions of their constituent phases. The idea is to make use of corresponding bounds and estimates for linear heterogeneous comparison materials in the context of the variational principles discussed in the previous section. Although the prescriptions determined by this approach can be applied to more general types of bounds and estimates, in this section, we limit our attention to bounds of the Weiner, Hashin–Shtrikman and Beran types.

In particular, we are interested in heterogeneous materials with n homogeneous phases, characterized by the isotropic energy functions $\phi^{(r)} (r = 1, \ldots, n)$, such that the local energy function of the heterogeneous material w, as defined by (2.2), takes the form

$$w(\boldsymbol{x}, \boldsymbol{E}) = \sum_{r=1}^{n} \chi^{(r)}(\boldsymbol{x})\, \phi^{(r)}(E), \tag{3.1}$$

where $\chi^{(r)}(\boldsymbol{x})$ is the characteristic function of phase r (this function vanishes, unless $\boldsymbol{x}$ is in phase r, in which case it equals unity). We further assume that the volume fractions $c^{(r)}$ of the constituent phases are fixed. These are given by the relations

$$c^{(r)} = \int_{\Omega} \chi^{(r)}(\boldsymbol{x})\, \mathrm{d}x, \text{ and are such that } \sum_{r=1}^{n} c^{(r)} = 1.$$

Additionally, we may, or may not, prescribe further information on the microstructure, such as overall isotropy. From hereon, I refer to the material characterized by (3.1) as a nonlinear composite (even though the relation may also correspond to a family of composites), and hence I assume that the scale of variation of the characteristic functions is small enough.

Because of the approximations that need to be made in the application of the variational principle (2.24) and its dual (2.32), most of the results of this section will be in the form of lower bounds for $\tilde{W}$. However, corresponding ‘upper estimates’, or rather estimates for the upper bounds, are also proposed. Thus §3*a* deals with the Weiner lower bound and is concerned with generally anisotropic nonlinear composites with isotropic phases in prescribed volume fractions. On the other hand, §§3*b* and *c*, dealing with Hashin–Shtrikman and Beran lower bounds, respectively, are concerned with nonlinear composites with isotropic phases in prescribed volume fractions, which are additionally distributed in such a way that the composite depicts overall isotropy. In addition, §3*d* addresses the difficulty in obtaining rigorous upper bounds for $\tilde{W}$. More specifically, it is shown that, although the well-known Weiner upper bounds are easily obtainable from the new variational principles, upper bounds of the Hashin–Shtrikman and Beran varieties are much harder to determine; instead, ‘upper estimates’ of these types are proposed and computed.

We remark that the above choices for classes of bounds are given on account of their simplicity, and to illustrate the general method. Other possibilities would include application to the linear bounds and self-consistent estimates of Willis (1977) for anisotropic composites with prescribed two-point correlation functions, and to the bounds of Murat & Tartar (1985) and Lurie & Cherkaev (1984, 1986) for generally anisotropic composites with prescribed volume fractions only (G-closures). This latter class of bounds on the effective conductivity tensor are sharper than the Weiner bounds, but would require, for implementation purposes, alternative expression in the form of energy bounds, analogous to those determined by Kohn & Lipton (1988) and Allaire & Kohn (1991) for linearly elastic systems. Before proceeding with the determination of the bounds and estimates, I state the following useful corollaries to Theorems 2.1 and 2.2.

Corollary 3.1. *Let* (3.1) *characterize the local energy-density function of an n-phase nonlinear composite, additionally satisfying the hypotheses of Theorem* 2.1. *Then, the effective energy function* $\tilde{W}$ *of the composite satisfies the inequality*

$$\tilde{W}(\bar{\boldsymbol{E}}) \geqslant \sup_{\epsilon_0^{(r)}>0} \left\{ \tilde{W}_0(\bar{\boldsymbol{E}}) - \sum_{r=1}^{n} c^{(r)} v^{(r)}(\epsilon_0^{(r)}) \right\}, \tag{3.2}$$

where $\tilde{W}_0$ *denotes the effective energy function of a linear comparison composite with n phases of dielectric constant* $\epsilon_0^{(r)}$ *in volume fraction* $c^{(r)}$, *such that the dielectric coefficient of the comparison composite is given by*

$$\epsilon_0(\boldsymbol{x}) = \sum_{r=1}^{n} \chi^{(r)}(\boldsymbol{x})\, \epsilon_0^{(r)},$$

and where the function $v^{(r)}$ *is obtained by specializing relation* (2.23) *to the rth phase.*

Note that the supremum in (3.2) is evaluated over the set of constants $\epsilon_0^{(r)} (r = 1, \ldots, n)$.

Proof. This result follows from expression (2.24) in Theorem 2.1 by restricting the set of (arbitrarily variable) comparison dielectric coefficients $\epsilon_0(\boldsymbol{x})$ to the set of piecewise constant fields, with a different dielectric constant over each nonlinear phase of the composite (namely, $\epsilon_0^{(r)}$ over phase r). The inequality is a consequence of the fact that the supremum over the set of piecewise constant fields is smaller than the supremum over the original (larger) set of fields. Note that the linear comparison composite has the same microstructure as the nonlinear composite, and that, in particular, it is constituted by n phases of dielectric constants $\epsilon_0^{(r)}$ in volume fractions $c^{(r)}$.

Corollary 3.2. *Let the appropriate dual version of* (3.1) *characterize the local complementary-energy function* w^* *of an n-phase nonlinear composite, additionally satisfying the hypotheses of Theorem* 2.2. *Then, the effective complementary-energy function* $\tilde{U}$ *satisfies*

$$\tilde{U}(\bar{\boldsymbol{D}}) \leqslant \inf_{\epsilon_0^{(r)}>0} \left\{ \tilde{U}_0(\bar{\boldsymbol{D}}) + \sum_{r=1}^{n} c^{(r)} v^{(r)}(\epsilon_0^{(r)}) \right\}, \tag{3.3}$$

where $\tilde{U}_0$ *denotes the effective complementary-energy function of a linear comparison composite with n phases of dielectric constant* $\epsilon_0^{(r)}$ *in volume fraction* $c^{(r)}$, *such that the dielectric coefficient of the comparison composite is given by*

$$\epsilon_0(\boldsymbol{x}) = \sum_{r=1}^{n} \chi^{(r)}(\boldsymbol{x})\, \epsilon_0^{(r)},$$

and where the function $v^{(r)}$ *is obtained by specializing relation* (2.31) (*or* (2.23)) *to the rth phase.*

Proof. The derivation of this result is analogous to the derivation of the previous result. In this case, we make use of expression (2.32) (Theorem 2.2) and of the fact that the infimum over the set of piecewise fields is larger than the infimum over the set of arbitrarily variable fields.

(*a*) *Weiner lower bound*

In this subsection, I make use of the lower bound of Weiner (1912) for linear, anisotropic composites with prescribed volume fractions to generate a corresponding bound for nonlinear, anisotropic composites. I refer to the resulting bound as the nonlinear Weiner bound. I emphasize that, although the nonlinear Weiner bounds may be obtained in exactly the same way as the corresponding linear bounds (directly from the classical variational principles), here I make use of the alternative variational principles of §2*c* for two reasons. First, the basic ideas in the derivation of the Weiner lower bound via the new variational principles are essentially the same as those to be used later in the derivation of the Hashin–Shtrikman and Beran bounds, and thus it is useful to illustrate the general method in the simplest possible case. Second, the resulting form of the Weiner lower bound for the case under consideration (isotropic phases) is new, and simpler than (although equivalent to) the alternative form resulting from direct application of the classical variational principle.

The Weiner lower bound may be specified as a bound on the effective energy functions of linear composites with dielectric constants $\epsilon_0^{(1)}, \epsilon_0^{(2)}, \ldots, \epsilon_0^{(n)}$ in prescribed volume fractions $c^{(1)}, c^{(2)}, \ldots, c^{(n)}$ via the relation

$$\tilde{W}_0(\bar{\boldsymbol{E}}) \geqslant \frac{1}{2}\left(\sum_{r=1}^{n} \frac{c^{(r)}}{\epsilon_0^{(r)}}\right)^{-1} \bar{E}^2, \tag{3.4}$$

where $\tilde{W}_0(\bar{\boldsymbol{E}}) = \frac{1}{2}(\tilde{\boldsymbol{\varepsilon}}_0\,\bar{\boldsymbol{E}})\cdot\bar{\boldsymbol{E}}$ is the effective energy function of linear composites with effective dialectric tensors $\tilde{\boldsymbol{\varepsilon}}_0$. Note that beyond the volume fractions of the constituent phases, nothing else is specified about the microstructure of the composite, and that, in particular, the composite may be anisotropic.

The nonlinear Weiner lower bound for the effective energy functions $\tilde{W}$ of the class of nonlinear composites with prescribed volume fractions is obtained by applying (3.2) of Corollary 3.1 to the set of nonlinear composites with prescribed volume fractions, and combining this result with the lower bound (3.4) for the class of linear comparison composites with prescribed volume fractions. Therefore,

$$\tilde{W}(\bar{\boldsymbol{E}}) \geqslant \sup_{\epsilon_0^{(r)}>0}\left\{\frac{1}{2}\left(\sum_{r=1}^{n} \frac{c^{(r)}}{\epsilon_0^{(r)}}\right)^{-1} \bar{E}^2 - \sum_{r=1}^{n} c^{(r)} v^{(r)}(\epsilon_0^{(r)})\right\}, \tag{3.5}$$

where, explicitly,

$$v^{(r)}(\epsilon_0^{(r)}) = \sup_{s>0}\{\tfrac{1}{2}\epsilon_0^{(r)} s^2 - \phi^{(r)}(s)\}. \tag{3.6}$$

Clearly, the optimizations implicit in (3.5) and (3.6) are $2n$ in number, but this number may be significantly reduced by means of the following identity, first utilized by deBotton & Ponte Castañeda (1992) and proved in the Appendix, namely,

$$\left(\sum_{r=1}^{n} \frac{c^{(r)}}{\epsilon_0^{(r)}}\right)^{-1} = \inf_{\substack{\omega^{(r)} \\ \bar{\omega}=0}}\left\{\sum_{r=1}^{n} c^{(r)}\epsilon^{(r)}(1-\omega^{(r)})^2\right\}, \tag{3.7}$$

where the infimum is over the set of variables $\omega^{(r)}$ $(r = 1, \ldots, n)$, which are subject to a zero-average constraint

$$\bar\omega = \sum_{r=1}^{n} c^{(r)}\omega^{(r)} = 0.$$

This identity, when applied to the nonlinear lower bound for $\tilde W$ in (3.5), yields the result

$$\tilde W(\bar{\boldsymbol E}) \geqslant \sup_{\epsilon_0^{(r)}>0}\left\{\inf_{\substack{\omega^{(r)}\\ \bar\omega=0}}\left\{\sum_{r=1}^{n} c^{(r)}[\tfrac12\epsilon^{(r)}(1-\omega^{(r)})^2\bar E^2 - v^{(r)}(\epsilon_0^{(r)})]\right\}\right\}, \tag{3.8}$$

which in turn leads to

$$\tilde W(\bar{\boldsymbol E}) \geqslant \inf_{\substack{\omega^{(r)}\\ \bar\omega=0}}\left\{\sum_{r=1}^{n} c^{(r)} \sup_{\epsilon_0^{(r)}>0}\{\tfrac12\epsilon^{(r)}(1-\omega^{(r)})^2\bar E^2 - v^{(r)}(\epsilon_0^{(r)})\}\right\}. \tag{3.9}$$

In this last step, I have made use of the fact that the argument of the nested supremum and infimum is concave in the $\epsilon_0^{(r)}$ (since the functions $v^{(r)}$ are convex in $\epsilon_0^{(r)}$) and convex in the $\omega^{(r)}$, and of the saddle point theorem to justify the interchange of the supremum and infimum operations.

Then, finally, application of relation (2.22), specialized to each phase in the form

$$\phi^{(r)}(s) = \sup_{\epsilon_0^{(r)}>0}\{\tfrac12\epsilon_0^{(r)}s^2 - v^{(r)}(\epsilon_0^{(r)})\}, \tag{3.10}$$

leads to the following result for the lower bound

$$\tilde W(\bar{\boldsymbol E}) \geqslant \inf_{\substack{\omega^{(r)}\\ \bar\omega=0}}\left\{\sum_{r=1}^{n} c^{(r)}\phi^{(r)}(|1-\omega^{(r)}|\,\bar E)\right\}. \tag{3.11}$$

The form of this bound is much simpler than that given by (3.5) and (3.6), and involves only a n-dimensional optimization, with one linear constraint. This constraint can easily be embedded in the optimization by suitable relabelling of the optimization variables. For instance, for the case of a two-phase composite, the above bound reduces to

$$\tilde W(\bar{\boldsymbol E}) \geqslant \inf_{\omega}\{c^{(1)}\phi^{(1)}(|1-c^{(2)}\omega|\bar E) + c^{(2)}\phi^{(2)}(|1+c^{(1)}\omega|\,\bar E)\}, \tag{3.12}$$

where the optimization variable ω is unconstrained.

Next, for completeness, note that an alternative expression for this lower bound may be obtained dually from expression (3.3) of Corollary 3.2. In this event, I obtain the following upper bound for the effective complementary-energy function $\tilde U$ of the nonlinear composite, namely,

$$\tilde U(\bar{\boldsymbol D}) \leqslant \inf_{\epsilon_0^{(r)}>0}\left\{\frac12\left(\sum_{r=1}^{n}\frac{c^{(r)}}{\epsilon_0^{(r)}}\right)\bar D^2 + \sum_{r=1}^{n} c^{(r)}v^{(r)}(\epsilon_0^{(r)})\right\}, \tag{3.13}$$

and thus

$$\tilde U(\bar{\boldsymbol D}) \leqslant \sum_{r=1}^{n} c^{(r)}(\phi^{(r)})^*(\bar D), \tag{3.14}$$

where I have used the result that

$$(\phi^{(r)})^*(\bar D) = \inf_{\epsilon_0^{(r)}>0}\left\{\frac{1}{2\epsilon_0^{(r)}}\bar D^2 + v^{(r)}(\epsilon_0^{(r)})\right\}, \tag{3.15}$$

which follows from (2.18), together with the fact that the Legendre transform of the energy function $\phi^{(r)}$ (or complementary-energy function) is given by result (2.30), specialized to phase r. We remark that, not surprisingly, the alternative form for the bound (3.14) is precisely the result that would be obtained by applying the classical minimum complementary-energy principle directly to the nonlinear composite. Then, by dualizing this last relation, and recalling relation (2.17), I obtain

$$\tilde{W}(\bar{\boldsymbol{E}}) \geqslant \left(\sum_{r=1}^{n} c^{(r)} (\phi^{(r)})^*\right)^* (\bar{E}). \tag{3.16}$$

For a given composite, we know from the lack of a duality gap (see discussion at the end of §2*c*) between the two versions of the new variational principle that expressions (3.11) and (3.16) for the lower bound for $\tilde{W}$ must be equivalent. However, the latter form involves $n+1$ Legendre transforms, which in general may be more difficult to evaluate than the earlier equivalent prescription. For this reason, I prefer the earlier prescription, and from hereon I denote the bound implied by (3.11) (or by (3.16)) as $\tilde{W}_{\mathrm{W}-}$, and refer to it as the Weiner lower bound.

(*b*) *Hashin–Shtrikman lower bound*

In this subsection, I apply the structure developed in the previous subsection to obtain lower bounds of the Hashin–Shtrikman type for the class of nonlinear isotropic composites with prescribed volume fractions.

We begin by noting that the effective dielectric tensor of a linear isotropic heterogeneous material is isotropic (i.e. $\tilde{\boldsymbol{\epsilon}}_0 = \tilde{\epsilon}_0 \boldsymbol{I}$, where $\boldsymbol{I}$ is the identity tensor in R^d, with $d = 2$ or 3 representing the dimension of the space). Then, the Hashin–Shtrikman (H–S) lower bound $\tilde{\epsilon}_-$ for the effective dielectric constant $\tilde{\epsilon}_0$, satisfying $\tilde{\epsilon}_0 \geqslant \tilde{\epsilon}_-$, is given by the expression (see, for instance, Kohn & Milton 1988)

$$\tilde{\epsilon}_- = \left(\sum_{r=1}^{n} \frac{c^{(r)}}{\epsilon_0^{(r)} + (d-1)\,\epsilon_-}\right)^{-1} - (d-1)\,\epsilon_-, \tag{3.17}$$

where $\epsilon_- = \inf_s\{\epsilon_0^{(s)}\}$. This result may be rewritten by means of the identity (3.7), shifted by $(d-1)\,\epsilon_-$, in the form

$$\tilde{\epsilon}_- = \inf_{\substack{\omega^{(r)} \\ \bar{\omega}=0}} \left\{ \sum_{r=1}^{n} c^{(r)} \left[\epsilon^{(r)}(1-\omega^{(r)})^2 + (d-1)\,\epsilon_-(\omega^{(r)})^2\right] \right\}. \tag{3.18}$$

Next, we observe that the effective energy functions $\tilde{W}$ of the class of nonlinear composites with overall isotropy may be estimated once again from relation (3.2) of Corollary 3.1, where $\tilde{W}_0$ now represents the effective energy functions of the class of linear comparison composites with phases of dielectric constants $\epsilon_0^{(r)}$ in prescribed volume fractions $c^{(r)}$. Further, the distribution of the phases in the linear comparison composite is such that the nonlinear composite must be isotropic; therefore, the distribution of the phases in the corresponding linear comparison composite must also be isotropic. I emphasize that, while not all microstructures that are isotropic for linear composites are also isotropic in the nonlinear context (for example, whereas a linear periodic composite with cubic symmetry is isotropic, the corresponding nonlinear periodic composite is not necessarily isotropic), nonlinear isotropic microstructures must also be isotropic in the linear context (since linear is a special case of nonlinear). Thus it follows that a lower bound for the effective energy functions of the class of linear, isotropic comparison composites is also a lower bound

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for the subclass of linear comparison composites with 'nonlinearly isotropic' microstructure.

Therefore, replacing $\tilde{W}_0$ in (3.2) by the lower bound specified by (3.18) induces a lower bound for the class of nonlinear isotropic composites. The result is

$$\tilde{W}(\bar{\boldsymbol{E}}) \geqslant \sup_{\epsilon_0^{(r)}>0} \left\{ \inf_{\substack{\omega^{(r)} \\ \bar{\omega}=0}} \left\{ \sum_{r=1}^{n} c^{(r)} \left[\tfrac{1}{2}(\epsilon^{(r)}(1-\omega^{(r)})^2 + (d-1)\,\epsilon_{-}(\omega^{(r)})^2)\,\bar{E}^2 - v^{(r)}(\epsilon_0^{(r)})\right] \right\} \right\}$$

$$= \inf_{\substack{\omega^{(r)} \\ \bar{\omega}=0}} \left\{ \sup_{\epsilon_0^{(r)}>0} \left\{ \sum_{r=1}^{n} c^{(r)} \left[\tfrac{1}{2}(\epsilon^{(r)}(1-\omega^{(r)})^2 + (d-1)\,\epsilon_{-}(\omega^{(r)})^2)\,\bar{E}^2 - v^{(r)}(\epsilon_0^{(r)})\right] \right\} \right\}, \qquad (3.19)$$

where, once again, I have made use of the saddle point theorem to interchange the supremum and infimum operations. Then, making use of (3.10) and remarking that the minimum $\epsilon_0^{(r)}$ (i.e. ϵ_{-}) depends on the intensity of the applied electric field $\bar{E}$, and may occur in any given phase, I conclude that

$$\tilde{W}(\bar{\boldsymbol{E}}) \geqslant \min_{s} \left\{ \inf_{\substack{\omega^{(r)} \\ \bar{\omega}=0}} \left\{ \sum_{\substack{r=1 \\ r\neq s}}^{n} c^{(r)} \phi^{(r)}(|1-\omega^{(r)}|\,\bar{E}) \right.\right.$$

$$\left.\left. + c^{(s)}\phi^{(s)} \left[\sqrt{\left[(1-\omega^{(s)})^2 + (d-1)\frac{1}{c^{(s)}} \sum_{t=1}^{n} c^{(t)} (\omega^{(t)})^2\right]}\,\bar{E} \right] \right\} \right\}. \qquad (3.20)$$

I refer to the bound implied by this relation as the H–S lower bound for nonlinear isotropic composites with isotropic phases in prescribed volume fractions, and denote it by $\tilde{W}_{\mathrm{HS}-}$. We remark that a dual version of this result, with very similar form, may also be obtained from the complementary-energy version of the variational principle (Corollary 3.2). Note further that for the special case of two-phase composites, the nonlinear H–S lower bound reduces to

$$\tilde{W}_{\mathrm{HS}-}(\bar{\boldsymbol{E}}) = \min \left\{ \begin{array}{l} \inf\limits_{\omega} \{ c^{(1)}\phi^{(1)}(|1-c^{(2)}\omega|\,\bar{E}) + c^{(2)}\phi^{(2)}[\sqrt{[(1+c^{(1)}\omega)^2 + (d-1)\,c^{(1)}\omega^2]}\,\bar{E}] \} \\ \inf\limits_{\omega} \{ c^{(1)}\phi^{(1)}[\sqrt{[(1-c^{(2)}\omega)^2 + (d-1)\,c^{(2)}\omega^2]}\,\bar{E}] + c^{(2)}\phi^{(2)}(|1+c^{(1)}\omega|\,\bar{E}) \} \end{array} \right\}, \qquad (3.21)$$

where the absolute minimum of the two infimum problems normally depends on $\bar{\boldsymbol{E}}$.

Note that lower bounds of the Hashin–Shtrikman type for nonlinear dielectrics have been obtained earlier by Willis (1986), using the general method of Talbot & Willis (1985). This method, which is a generalization of the Hashin–Shtrikman (1962) variational principles for nonlinear problems, makes use of a linear homogeneous comparison material (unlike the heterogeneous comparison material of the new method) to obtain results that, at least in some cases, agree with the predictions for the bounds obtained via the new method proposed in this work. According to the work of Willis (1991), in a different physical context (nonlinear infinitesimal elasticity), the key to recovering the bounds predicted by the new method from the Talbot–Willis method centres around an optimal choice of the homogeneous comparison material (in the Talbot–Willis method). Thus Willis finds that the improvement in the bounds noted in Ponte Castañeda (1991*a*) (using essentially the new method) with reference to the bounds determined by Ponte Castañeda & Willis (1988) (using the Talbot–Willis method), for some special types of nonlinear composite materials, was a consequence of a non-optimal choice for the comparison

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material in the earlier work. It is anticipated that analogous results will hold for nonlinear dielectrics. However, I emphasize that the form of the bounds given in this work is different (and simpler) from the form of the bounds given by Willis (1986) (see also Talbot & Willis 1987). In particular, I find that, for the special case of two-phase nonlinear composites, the bounds given in this work involve only one optimization, whereas the bounds given by Willis (1986) involve, in general, two optimizations (including the optimization of the homogeneous comparison material). Further comparison between the two methods is given by Willis (1991) and by Ponte Castañeda (1992), but one distinct advantage of the new method utilizing the linear heterogeneous comparison material is that it can be used in conjunction with linear bounds and estimates, other than Hashin–Shtrikman bounds, to yield corresponding nonlinear bounds and estimates (assuming, of course, that such linear bounds and estimates are available). As an illustration of this feature of the new method, in the next subsection, I derive lower bounds of the Beran type for two-phase, nonlinear isotropic composites. Later, in §5*c* we also give an application to self-consistent estimates.

(*c*) *Beran lower bound*

In the context of linear, isotropic dielectric composites, bounds that are more restrictive than the Hashin–Shtrikman bounds, and contain additional microstructural information in the form of three-point correlation functions have been proposed by Beran (1965). For the case of two-phase systems, Milton (1981) has proposed a simple form of the Beran bounds that depends on the volume fraction of the phases, and on only one additional geometric parameter (that can be obtained from the three-point correlation functions). Milton's form for the lower bound on the effective dielectric constant $\tilde{\epsilon}_0$ is given by

$$\tilde{\epsilon}_- = \left(\sum_{r=1}^{2} \frac{c^{(r)}}{\epsilon_0^{(r)} + (d-1)\,\hat{\epsilon}_-}\right)^{-1} - (d-1)\,\hat{\epsilon}_-, \qquad (3.22)$$

which is identical in form to (3.17), except that $\hat{\epsilon}_-$ must be replaced by

$$\hat{\epsilon}_- = \left(\sum_{r=1}^{2} \frac{\zeta^{(r)}}{\epsilon_0^{(r)}}\right)^{-1}.$$

Thus this expression depends on the dielectric constants $\epsilon_0^{(1)}$ and $\epsilon_0^{(2)}$, the volume fractions $c^{(1)}$ and $c^{(2)}$, and the third-order geometric parameters $\zeta^{(1)}$ and $\zeta^{(2)} = 1-\zeta^{(1)}$, both lying in the interval $[0,1]$.

Substitution of this result into the lower bound approximation (3.2) of Corollary 3.1, and following a procedure very similar to that used for the Hashin–Shtrikman bound (making use of identity (3.7) twice: once for expression (3.22) and again for the expression of $\hat{\epsilon}_-$), we arrive at the following lower bound for the nonlinear energy function, namely,

$$\begin{aligned}\tilde{W}_{B-}(\bar{E}) = \inf_{\omega,\gamma}\{&c^{(1)}\phi^{(1)}(\sqrt{[(1-c^{(2)}\omega)^2+(d-1)\,c^{(2)}\zeta^{(1)}\omega^2(1-\zeta^{(2)}\gamma)^2]}\,\bar{E})+\dots\\ &+c^{(2)}\phi^{(2)}(\sqrt{[(1+c^{(1)}\omega)^2+(d-1)\,c^{(1)}\zeta^{(2)}\omega^2(1+\zeta^{(1)}\gamma)^2]}\,\bar{E})\}. \qquad (3.23)\end{aligned}$$

We remark that the corresponding nonlinear H–S lower bound (3.21) follows immediately from this result by choosing either $\zeta^{(1)} = 0$ or $\zeta^{(2)} = 1$, whichever yields the lowest value (note that the infimum problem over γ becomes trivial in either case). This is completely analogous to the corresponding result for linear two-phase composites. Finally, note that a dual version of this result, with slightly simpler form (involving only one optimization), may also be obtained by application of Corollary 3.2.

(*d*) *Upper bounds and estimates*

The determination of upper bounds for the effective energy functions $\tilde{W}$ of classes of nonlinear composites is intrinsically harder than the determination of the corresponding lower bounds. This is because approximations of the type (3.2) clearly do not work in this case; instead, we must resort to the exact versions of the variational principles. As we see below, it is possible to make use of the exact version of the variational principles to obtain the Weiner upper bound, but I was not able to make use of the exact version of the variational principles to obtain upper bounds of the Hashin–Shtrikman and Beran types. It is interesting to note that the method of Talbot & Willis (1985) exhibits the same limitation in determining nonlinear H–S upper bounds, if attention is restricted to linear comparison materials. Because of this difficulty, I provide instead 'upper estimates', or rather lower estimates for the upper bound, of the Hashin–Shtrikman and Beran types. The rationale behind this is that, in practice, the bounds are often used as estimates for the behaviour of the 'weakest' and 'strongest' composite possible with the given microstructural information. In this less rigorous interpretation, it is probably as useful to have a lower estimate for the 'weakest' material, as to have a lower estimate for the 'strongest' material, provided that the estimates are not too weak. In §§4 and 5, we pursue these ideas further.

The derivation of the Weiner upper bound is made possible by the corresponding upper bound for linear composites with arbitrarily variable dielectric coefficient $\epsilon_0(\boldsymbol{x})$. This bound may be expressed in the form

$$\tilde{W}_0(\bar{\boldsymbol{E}}) \leqslant \frac{1}{2}\left(\int_\Omega \epsilon_0(\boldsymbol{x})\,\mathrm{d}x\right)\bar{E}^2, \tag{3.24}$$

where $\tilde{W}_0$ is the effective energy function of the linear composite. Then, application of (3.24) to (2.24) of Theorem 2.1 leads to

$$\begin{aligned}\tilde{W}(\bar{\boldsymbol{E}}) &\leqslant \sup_{\epsilon_0(\boldsymbol{x})\geqslant 0}\left\{\frac{1}{2}\left(\int_\Omega \epsilon_0(\boldsymbol{x})\,\mathrm{d}x\right)\bar{E}^2 - \int_\Omega v(\boldsymbol{x},\epsilon_0(\boldsymbol{x}))\,\mathrm{d}x\right\}\\ &= \int_\Omega \sup_{\epsilon_0\geqslant 0}\left\{\frac{1}{2}\epsilon_0\bar{E}^2 - v(\boldsymbol{x},\epsilon_0)\right\}\mathrm{d}x\\ &= \int_\Omega \phi(\boldsymbol{x},\bar{E})\,\mathrm{d}x,\end{aligned} \tag{3.25}$$

which, via (3.1), leads in turn to the nonlinear Weiner upper bound ($\tilde{W}_{W+}$)

$$\tilde{W}(\bar{\boldsymbol{E}}) \leqslant \sum_{r=1}^{n} c^{(r)}\phi^{(r)}(\bar{E}). \tag{3.26}$$

This result may, of course, be obtained directly from the classical minimum energy principle; inclusion of the present alternative derivation in this paper is made only to emphasize that the exact version of the variational principles of §2*c* also lead (at least in principle, and sometimes in practice) to upper bounds. Finally, we remark that a dual alternative expression analogous to expression (3.11) for the Weiner lower bound is also possible, if use is made of Theorem 2.2 instead of Theorem 2.1.

A similar approach may be attempted for the nonlinear H–S upper bound. However, I was unable to obtain an explicit result analogous to (3.26). The reason is essentially that the evaluation of the pertinent term involving $\epsilon_+ = \max_x\{\epsilon_0(\boldsymbol{x})\}$ in the appropriate expression of the H–S upper bound for the linear isotropic comparison composite with continuously varying microstructure is extremely difficult. I do not pursue this point further, and proceed with the determination of estimates for the H–S upper bound, or 'upper estimates'. As discussed earlier, this is accomplished by application of approximation (3.2) of Corollary 3.1 to the H–S upper bounds for the linear comparison composite. The upper bound for the effective energy function of the linear comparison composite may be given in terms of the upper bound for its effective dielectric constant, namely,

$$\tilde{\epsilon}_+ = \left(\sum_{r=1}^{n} \frac{c^{(r)}}{\epsilon_0^{(r)} + (d-1)\,\epsilon_+}\right)^{-1} - (d-1)\,\epsilon_+, \qquad (3.27)$$

where $\epsilon_+ = \sup_s\{\epsilon_0^{(s)}\}$.

It is evident that the procedure of §3*b*, utilizing the lower bound (3.17) for the linear comparison composite to obtain a lower bound for the nonlinear composite, may be repeated making use of (3.27) instead of (3.17) to obtain an 'upper estimate' for the nonlinear composite. The result is the same as expressions (3.20) and (3.21) for the n-phase and two-phase nonlinear composites, respectively, with the difference that the outermost minimum operations must now be replaced by maximum operations. I denote this result by $\tilde{W}_{\mathrm{HS}+}$, and refer to it as the H–S 'upper estimate' for $\tilde{W}$. In §4, we see that $\tilde{W}_{\mathrm{HS}+}$ is not, in general, an upper bound for $\tilde{W}$. This is accomplished by computing the exact effective energy function of a specific nonlinear composite, and noting that it exceeds $\tilde{W}_{\mathrm{HS}+}$.

Finally, note that the same comments that we have just made about the H–S upper bounds also apply to the Beran upper bounds. Therefore, I provide here only an 'upper estimate' of the Beran type. Such an estimate is made possible by Milton's expression for the upper bound of the effective dielectric constant of the linear comparison composite, $\tilde{\epsilon}_+$, which is given by expression (3.22), but with $\hat{\epsilon}_-$ replaced by

$$\hat{\epsilon}_+ = \sum_{r=1}^{2} \zeta^{(r)} \epsilon_0^{(r)}.$$

Then, the corresponding form for the 'upper estimate' (not in general an upper bound, for the same reason as for the Hashin–Shtrikman upper estimate) of the class of nonlinear isotropic composites with prescribed volume fractions and third-order statistical information (in the form of $\zeta^{(1)}$) is given by

$$\tilde{W}_{B+}(\bar{E}) = \inf_{\omega}\{c^{(1)}\phi^{(1)}(\surd[(1-c^{(2)}\omega)^2 + (d-1)\,c^{(2)}\zeta^{(1)}\omega^2]\,\bar{E}) + \\ \ldots + c^{(2)}\phi^{(2)}(\surd[(1+c^{(1)}\omega)^2 + (d-1)\,c^{(1)}\zeta^{(2)}\omega^2]\,\bar{E})\}. \qquad (3.28)$$

This bound also reduces to the corresponding Hashin–Shtrikman 'upper estimate', by letting either $\zeta^{(1)} = 0$, or $\zeta^{(2)} = 1$, whichever yields the highest value.

4. Exact estimates

In the previous section, I made use of the variational principles of §2*c* (Theorems 2.1 and 2.2) to obtain bounds on the effective behaviour of nonlinear composites. I first studied the case of generally anisotropic composites and discovered that the

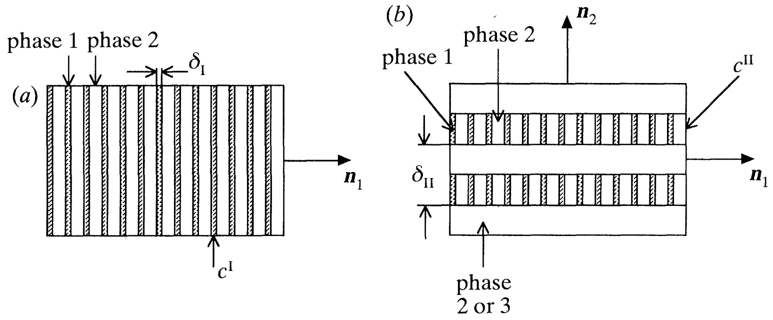

Figure 1. (*a*) A rank-I laminate and (*b*) a rank-II laminate.

well-known Weiner bounds for nonlinear composites could be recovered from these variational principles. I then considered the special, but important, case of isotropic composites, and was able to determine new lower bounds, but only 'upper estimates', in general, for the effective energy functions of these nonlinear composites. This was accomplished by making use of the Hashin–Shtrikman and Beran bounds for linear, isotropic comparison materials, with the same distribution of phases as the nonlinear composites, in the variational principles. The question then arises as to the quality of the lower bounds, and also, as to whether the 'upper estimates' could in fact be rigorous bounds. In this section, I attempt to answer these and other related questions by analysing the effective properties of sequentially laminated materials. This class of materials, although of little practical value, has proved to be of great theoretical value in the study of linear composites. For instance, in the particular case of two-phase linear dielectric composites, it has been shown by Tartar (1985) and Lurie & Cherkaev (1984, 1986) that sequentially laminated materials can be constructed attaining the corresponding linear Hashin–Shtrikman bounds. Here, I attempt a similar study for nonlinear sequentially laminated materials. I find that the variational principles of §2*c* are specially suited to the study of this class of nonlinear materials.

A sequentially laminated material (or laminate, for short) 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, in such a way as to produce hierarchical microstructures of increasing complexity. The 'rank' of the laminate refers to the number of layering operations required to reach the final iterated microstructure. Thus, as shown in figure 1*a*, a rank-I (I use roman numerals to denote rank) laminate is obtained by mixing layers of two homogeneous phases (1 and 2) to obtain a simple laminate with layering direction $\boldsymbol{n}_1$. A rank-II laminate (see figure 1*b*) is obtained by layering the rank-I laminate with a third phase (3), or alternatively with one of the original phases (say 2), in a different layering direction $\boldsymbol{n}_2$. In general, $\boldsymbol{n}_1$ and $\boldsymbol{n}_2$ can take on any orientation, but in figure 1*b* they are depicted as being orthogonal. Higher rank laminates are obtained by iterating this procedure (see Milton 1986). One important observation in connection with this procedure is that the length scale of the embedded laminates is assumed to be small compared with the length scale of the embedding laminates (for example, in figure 1*b*, $\delta_I \ll \delta_{II} \ll 1$). This assumption derives from the fact that the effective properties of simple laminates can be computed exactly, and hence, by treating the iterated laminate as a simple laminate with the embedded laminate replaced by a homogeneous material with the effective properties of the embedded laminate, the effective properties of the iterated laminate can also be computed exactly. An equivalent way of stating this result is that, under the assumption of widely separated length scales for the different layers composing the iterated laminate, the fields will be essentially constant within each elemental layer, provided that the boundary conditions applied to the laminate are uniform. This feature, greatly simplifies the computation of effective properties, thereby making sequentially laminated materials very useful constructions.

In this and in the next two paragraphs, I review some of the important results concerning linear laminates. I first note that the effective energy function of a simple laminate (which, as mentioned previously, can be computed exactly) lies within and attains (for specific orientations of the applied fields) the Weiner bounds. Thus, at least in the linear case, the Weiner bounds on the effective energy-function of arbitrarily anisotropic linear composites are known to be sharp (i.e. they can be attained by specific microstructures). In §4a, I demonstrate that the same result holds for the nonlinear Weiner bounds.

In the context of two-phase linear dielectrics, it is known that only iterated laminates of rank greater than, or equal to, the dimension of the underlying physical space ($d = 2$, or 3) can have isotropic properties. These isotropic laminates are obtained by choosing the relative volume fractions and the layering directions of each of the embedded laminates in such a way that the effective dielectric tensor of the iterated laminate is isotropic, while the absolute volume fractions of the constituent phases remain fixed. For details, the reader is referred to Tartar (1985), where it is additionally demonstrated that the result of this calculation is independent of the rank of the iterated laminate (as long as it is greater than d), and equal to the corresponding Hashin–Shtrikman bounds. The lower bound is obtained by letting the phase with the highest dielectric constant occupy one of the two phases in the (innermost) rank-I laminate, and by choosing the other phase (with the lower dielectric constant) to play the role of the homogeneous phase at each of the other layering operations. This yields a composite material with the phase with the highest dielectric constant serving as the inclusion phase, and the other phase acting as the matrix phase (for a picture of this construction in two dimensions, let phase 3 be the same as phase 2 in figure 1b). Intuitively, such a construction should produce a material with a low effective dielectric constant. In fact, it yields a material with the lowest possible dielectric constant since, as mentioned above, it attains the H–S lower bound. Conversely, the upper bound for the effective dielectric constant is obtained by exchanging the roles of the two phases in the above construction.

One source of potential concern in connection with sequentially laminated materials is the fact that the resulting inclusions are 'flat', whereas in practice the inclusions are often equi-axed. However, this concern can be easily dispelled by noting that iterated laminates can be used to model arbitrarily close the properties of any two-phase microstructure (Milton 1986). For instance, the so-called composite-spheres (concentric-spheres) model of Hashin–Shtrikman (1962) (with inclusions of spherical shape) possesses exactly the same effective properties as an isotropic iterated laminate with the same volume fractions (i.e. they both saturate the Hashin–Shtrikman bounds for isotropic microstructures). Similarly, Tartar (1985) proposed a confocal ellipsoidal microstructure to demonstrate the optimality of the anisotropic Hashin–Shtrikman bounds developed by Murat & Tartar (1985) and Lurie & Cherkaev (1984, 1986). Thus, a one-to-one correspondence may be established between the confocal ellipsoidal and sequentially laminated microstructures in this case also. It may initially seem that the concentric spheres (or confocal ellipsoids) microgeometry is to be preferred over the iterated laminate construction, but in fact the sequentially laminated microgeometry has several advantages over the composite-spheres microgeometry. First, the sequentially laminated microstructure involves a finite number of length scales in contrast with the composite-spheres microstructure which involves an infinite number of length scales (because the composite spheres must be chosen to cover all sizes to fill space). Second, the laminated microstructure, unlike the composite-spheres microstructure, generalizes to other more complex systems such as elasticity, and to nonlinear systems, as we see in §4*b*.

As discussed previously, the main advantage of sequentially laminated composites is that, when subject to uniform boundary conditions, the fields are piecewise constant within the composite (whether the phases in the composite are linear or nonlinear), except in small boundary layer regions at the interfaces separating laminates of different rank. However, the 'widely separated length scales' hypothesis makes the effect of these boundary layer regions negligible on the effective behaviour of the laminate. For example, the assumption that $\delta_{\mathrm{I}} \ll \delta_{\mathrm{II}} \ll 1$, in the two-dimensional microstructure of figure 1*b*, leads to different constant fields in each of the regions labelled 1, 2 and 3 (even if there are only two phases). For a rigorous treatment of this issue in the linear case, the reader is referred to Tartar (1985). Because of this feature of sequentially laminated materials, it is useful to state the following corollaries of Theorems 2.1 and 2.2.

Corollary 4.1. *Suppose that we have an n-phase nonlinear composite with local energy function w characterized by* (3.1), *additionally satisfying the hypotheses of Theorem* 2.1, *such that the distribution of fields within the composite is piecewise constant* (*with perhaps different constants in different portions of the same phase*). *We assume that the distribution of fields is given by the characteristic functions* $\hat{\chi}^{(s)}$ ($s = 1, \ldots, m$), *such that, for example,*

$$\boldsymbol{E}(\boldsymbol{x}) = \sum_{s=1}^{m} \hat{\chi}^{(s)}(\boldsymbol{x})\, \boldsymbol{E}^{(s)},$$

with each $\boldsymbol{E}^{(s)}$ *constant. In general, the characteristic functions* $\hat{\chi}^{(s)}$ *and the characteristic functions defining the microstructure of the composite* $\chi^{(r)}$ ($r = 1, \ldots, n$) *are different* ($m \geqslant n$). *Then, the effective energy function of the nonlinear composite may be expressed in the form*

$$\tilde{W}(\bar{\boldsymbol{E}}) = \sup_{\epsilon_0^{(s)} > 0} \left\{ \tilde{W}_0(\bar{\boldsymbol{E}}) - \sum_{s=1}^{m} \hat{c}^{(s)} v^{(s)}(\epsilon_0^{(s)}) \right\}, \tag{4.1}$$

where $\tilde{W}_0$ *is the effective energy function of a linear comparison composite with m phases of dielectric constant* $\epsilon_0^{(s)}$ *in volume fractions* $\hat{c}^{(s)}$, *such that the dielectric coefficient of the comparison composite is given by*

$$\epsilon_0(\boldsymbol{x}) = \sum_{s=1}^{m} \hat{\chi}^{(s)}(\boldsymbol{x})\, \epsilon_0^{(s)},$$

and where the functions $v^{(s)}$ *are obtained by specializing relation* (2.23) *to the sth region.*

For example, in the rank-II laminate of figure 1*b*, the linear comparison composite is made up of three phases, denoted 1, 2 and 3, even if the corresponding nonlinear composite is made up of only two phases (nonlinear phases 2 and 3 are equal).

Proof. This result is obtained directly from Theorem 2.1 by noticing that, because the fields are piecewise constant with distribution given by characteristic functions $\chi^{(s)}$, the piecewise constant distribution of comparison dielectric coefficients (with the same characteristic functions) is the exact solution for $\epsilon_0(\boldsymbol{x})$ in the variational principle (2.24). To see this, note that, within a given phase (characterized by some $\chi^{(r)}$), if $\boldsymbol{E}$ is constant (say $\boldsymbol{E}^{(s)}$) over some subregion of the given phase (characterized by $\hat{\chi}^{(s)}$) then the optimal $\epsilon_0(\boldsymbol{x})$ in relation (2.24) for the local energy function w is constant over that subregion (say $\epsilon_0^{(s)}$). Therefore the integrals in (2.24) may be evaluated exactly, and expression (4.1) results.

Corollary 4.2. *Suppose that we have an n-phase nonlinear composite with local complementary-energy function w^*, characterized by the dual version of* (3.1), *and additionally satisfying the hypotheses of Theorem* 2.2, *such that the distribution of fields within the composite is piecewise constant* (*with perhaps a different constant in different portions of the same phase*). *We assume that the distribution of fields is given by the characteristic functions $\hat{\chi}^{(s)}$* ($s = 1, \ldots, m$) ($m \geqslant n$), *such that, for example,*

$$\boldsymbol{D}(\boldsymbol{x}) = \sum_{s=1}^{m} \hat{\chi}^{(s)}(\boldsymbol{x})\,\boldsymbol{D}^{(s)},$$

with $\boldsymbol{D}^{(s)}$ constant. Then, the effective complementary-energy function of the nonlinear composite may be expressed in the form

$$\tilde{U}(\bar{\boldsymbol{D}}) = \inf_{\epsilon_0^{(s)}>0}\left\{\tilde{U}_0(\bar{\boldsymbol{D}}) + \sum_{s=1}^{m} \hat{c}^{(s)} v^{(s)}(\epsilon_0^{(s)})\right\}, \qquad (4.2)$$

where $\tilde{U}_0$ is the effective complementary-energy function of a linear comparison composite with m phases of dielectric constant $\epsilon_0^{(s)}$ in volume fractions $\hat{c}^{(s)}$, such that the dielectric coefficient of the comparison composite is given by

$$\epsilon_0(\boldsymbol{x}) = \sum_{s=1}^{m} \hat{\chi}^{(s)}(\boldsymbol{x})\,\epsilon_0^{(s)},$$

and where the functions $v^{(s)}$ are obtained by specializing relation (2.23) *to the sth region.*

Proof. The demonstration of this result is analogous to that of the previous result.

(*a*) *Simple laminates*

In this subsection, I determine the effective energy functions of simple (one layering direction) nonlinear laminates by applying the well-known corresponding results for linear laminates to the above corollaries of the variational principles of §2*c*. This calculation has been discussed previously by Ponte Castañeda (1992) for the case of two-phase (rank-I) laminates; here, I investigate n-phase laminates made up of nonlinear isotropic phases with potentials $\phi^{(r)}$ in volume fractions $c^{(r)}$, distributed randomly in layers perpendicular to a fixed orientation $\boldsymbol{n}$. The corresponding linear comparison laminate with dielectric constants $\epsilon_0^{(r)}$ in volume fractions $c^{(r)}$ has anisotropic effective dielectric tensor

$$\tilde{\boldsymbol{\varepsilon}}_0 = \left(\sum_{r=1}^{n} c^{(r)}\epsilon_0^{(r)}\right)(\boldsymbol{I}-\boldsymbol{n}\otimes\boldsymbol{n}) + \left(\sum_{r=1}^{n} \frac{c^{(r)}}{\epsilon_0^{(r)}}\right)^{-1}\boldsymbol{n}\otimes\boldsymbol{n}, \qquad (4.3)$$

where $\boldsymbol{n}$ is the lamination direction, and $\otimes$ denotes the tensor product of two vectors.

Thus, according to (4.1), the effective energy function of the nonlinear laminate may be computed from the relation

$$\tilde{W}(\bar{\boldsymbol{E}}) = \sup_{\epsilon_0^{(r)} \geqslant 0} \left\{ \tfrac{1}{2}(\tilde{\boldsymbol{\varepsilon}}_0 \bar{\boldsymbol{E}}) \cdot \bar{\boldsymbol{E}} - \sum_{r=1}^{n} c^{(r)} v^{(r)}(\epsilon_0^{(r)}) \right\}, \tag{4.4}$$

where $\tilde{\boldsymbol{\varepsilon}}_0$ is given by (4.1), and where I have used the fact that the fields for a laminated composite material (even if it is nonlinear), subject to uniform boundary conditions, are constant within each phase (i.e. $\hat{\chi}^{(r)} = \chi^{(r)}$). Then, making use of the identity (3.7), and interchanging the sup and the inf by invoking the appropriate version of the saddle point theorem, I arrive at

$$\tilde{W}(\bar{\boldsymbol{E}}) = \inf_{\substack{\omega^{(r)} \\ \bar{\omega}=0}} \left\{ \sum_{r=1}^{n} c^{(r)} \sup_{\epsilon_0^{(r)} \geqslant 0} \{ \tfrac{1}{2}\epsilon_0^{(r)} [\bar{E}^2 - (\bar{\boldsymbol{E}} \cdot \boldsymbol{n})^2 + (1-\omega^{(r)})^2 (\bar{\boldsymbol{E}} \cdot \boldsymbol{n})^2] - v^{(r)}(\epsilon_0^{(r)}) \} \right\}, \tag{4.5}$$

which by means of (3.10) finally reduces to

$$\tilde{W}(\bar{\boldsymbol{E}}) = \inf_{\substack{\omega^{(r)} \\ \bar{\omega}=0}} \left\{ \sum_{r=1}^{n} c^{(r)} \phi^{(r)} [\surd[\bar{E}^2 - (\bar{\boldsymbol{E}} \cdot \boldsymbol{n})^2 + (1-\omega^{(r)})^2 (\bar{\boldsymbol{E}} \cdot \boldsymbol{n})^2]] \right\}. \tag{4.6}$$

It is not difficult to see that the maximum value of $\tilde{W}$ in the above expression is obtained when $\bar{\boldsymbol{E}}$ is perpendicular to $\boldsymbol{n}$ yielding the Weiner upper bound $\tilde{W}_{\mathrm{W}+}$, as given by (3.26). Conversely, the minimum value of $\tilde{W}$ is obtained whenever $\bar{\boldsymbol{E}}$ is parallel to $\boldsymbol{n}$ yielding the Weiner lower bound $\tilde{W}_{\mathrm{W}-}$, as given by (3.11).

This demonstrates that the Weiner bounds on the effective energy function are sharp within the class of nonlinear anisotropic composite materials with prescribed volume fractions, and that these bounds are attained by simple rank-I laminates. Thus, for anisotropic composites, the linear and nonlinear theories are similar. As we see in the next subsection, the same is not true for nonlinear isotropic composites.

(b) *Iterated laminates*

To motivate the study of nonlinear iterated laminates, I first consider the corresponding linear problem in some detail. For the most part, we will restrict our attention to the case of two-phase composites. It is well known that rank-d laminates with orthogonal layering directions attain the isotropic Hashin–Shtrikman bounds for the effective dielectric constant of two-phase materials (in d dimensions). For example, a two-phase, orthogonal rank-II laminate in two dimensions is depicted in figure 1*b*. We note that phase 1 occupies relative volume fraction c^I in the (embedded) rank-I laminate and overall volume fraction $c^{(1)} = c^I c^{II}$ in the composite, where c^{II} is the volume fraction occupied by the rank-I laminate within the rank-II laminate. The rest of the composite is made up of phase 2 (we are letting phase 3 be equal to phase 2 in figure 1*b*). Its effective dielectric tensor is obtained by first computing the effective dielectric tensor of the embedded rank-I laminate with layering orientation $\boldsymbol{n}_1$, and then using this result in the computation of the effective dielectric tensor of the rank-II laminate with layering direction $\boldsymbol{n}_2$ (where $\boldsymbol{n}_2$ is orthogonal to $\boldsymbol{n}_1$). Thus the dielectric constants in the $\boldsymbol{n}_1$ and $\boldsymbol{n}_2$ (principal) directions of the rank-II laminate are, respectively,

$$\tilde{\epsilon}_1^{II} = c^{II} \tilde{\epsilon}_1^{I} + (1-c^{II})\, \epsilon^{(2)} \quad \text{and} \quad \tilde{\epsilon}_2^{II} = [c^{II} (\tilde{\epsilon}_2^{I})^{-1} + (1-c^{II})\, (\epsilon^{(2)})^{-1}]^{-1}, \tag{4.7}$$

where $\tilde{\epsilon}_1^{I}$ and $\tilde{\epsilon}_2^{I}$ are the effective dielectric constants of the rank-I laminate in the $\boldsymbol{n}_1$

and $\boldsymbol{n}_2$ directions. Then, the condition for isotropy is obtained by choosing c^{I} (c^{II} is in turn given by $c^{(1)} = c^{I}c^{II}$) such that $\tilde{\epsilon}_1^{II} = \tilde{\epsilon}_2^{II} = \tilde{\epsilon}$. The result of this calculation for the effective dielectric constant of the rank-II isotropic laminate is

$$\tilde{\epsilon} = \left(\sum_{r=1}^{2} \frac{c^{(r)}}{\epsilon_0^{(r)} + \epsilon^{(2)}} \right)^{-1} - \epsilon^{(2)}. \tag{4.8}$$

This result is in agreement with the H–S lower bound given by (3.17) for $d = 2$, if we assume that $\epsilon^{(1)} > \epsilon^{(2)}$. Otherwise, the above expression would yield the corresponding H–S upper bound. Thus, the isotropic laminate with the largest dielectric constant as the inclusion phase (and the smallest as the matrix phase) leads to the lowest effective dielectric constant for the composite, and *vice versa*.

Clearly, this procedure generalizes to three dimensions by considering a rank-III laminate, obtained by layering the rank-II laminate in volume fraction c^{III} again with phase 2 in direction $\boldsymbol{n}_3$ (orthogonal to $\boldsymbol{n}_1$ and $\boldsymbol{n}_2$). Then, the inclusion phase (1) occupies volume fractions c^{I} and $c^{I}c^{II}$ in the embedded rank-I and rank-II laminates, respectively, and volume fraction $c^{(1)} = c^{I}c^{II}c^{III}$ in the rank-III laminate. The condition of isotropy in this case leads to two equations to be solved for c^{I} and c^{II}. Again, the result of this calculation is in agreement with the H–S bounds (3.17) and (3.27). I emphasize that, within the class of linear, isotropic iterated laminates, there exist a large (in fact, infinite) number of laminates of different ranks attaining the H–S bounds. The rank-d laminates with orthogonal layering directions, discussed above, are the simplest such composites.

The above discussion suggests that the same identification may be possible between the nonlinear H–S bounds given in §3 and nonlinear rank-d laminates. However, this identification fails even in the simplest non-trivial case ($d = 2$). To see this, note that the effective energy function of the two-phase, nonlinear rank-II laminate with the same microstructure as the corresponding linear, isotropic rank-II laminate may be expressed in the form

$$\tilde{W}_{\mathrm{L}}(\bar{\boldsymbol{E}}) = \sup_{\epsilon_0^{(1)},\, \epsilon_0^{(2)},\, \epsilon_0^{(3)} \geqslant 0} \{\tilde{W}_0^{(3)}(\bar{\boldsymbol{E}}) - c^{(1)}v^{(1)}(\epsilon_0^{(1)}) - (c^{II} - c^{(1)})\, v^{(2)}(\epsilon_0^{(2)}) - (1 - c^{II})\, v^{(2)}(\epsilon_0^{(3)})\}, \tag{4.9}$$

where $\tilde{W}_0^{(3)}(\bar{\boldsymbol{E}})$ is the effective energy function of a three-phase, linear rank-II laminate (see figure 1*b*), with comparison dielectric constants $\epsilon_0^{(1)}, \epsilon_0^{(2)}, \epsilon_0^{(3)}$, and where c^{II} has been specified previously. Note that $\tilde{W}_0^{(3)}(\bar{\boldsymbol{E}})$ is not, in general, isotropic (unless $\epsilon_0^{(2)} = \epsilon_0^{(3)}$), and therefore $\tilde{W}_{\mathrm{L}}$ is also not isotropic. The reason in this case for the three distinct dielectric constants for the linear comparison material, even though we are dealing with a two-phase nonlinear material, is again that for the nonlinear rank-II laminate the fields will be different constants in phase 2, depending on whether we are dealing with the portion of phase 2 that is embedded within the rank-I laminate (in volume fraction $c^{II} - c^{(1)}$), or we are referring to the portion of phase 2 in the rank-II laminate that is not part of the rank-I laminate (the volume fraction of this portion of phase 2 is $1 - c^{II}$). Thus to obtain an exact result for the effective energy function of the nonlinear laminate, a three-dimensional optimization is required (see Corollary 4.1).

On the other hand, if instead of distinguishing between the two portions of the nonlinear phase 2 subject to different constant fields, and carrying out the optimization over the three independent comparison dielectric constants, we carry out the optimization over two independent variables $\epsilon_0^{(1)}, \epsilon_0^{(2)}$ (as suggested by (3.2) of

Corollary 3.1), a lower bound approximation for the effective energy function of the two-phase nonlinear laminate is obtained, namely,

$$\tilde{W}_{\mathrm{L}}(\bar{\boldsymbol{E}}) \geqslant \sup_{\epsilon_0^{(1)},\, \epsilon_0^{(2)} \geqslant 0} \{\tilde{W}_0^{(2)}(\bar{\boldsymbol{E}}) - c^{(1)} v^{(1)}(\epsilon_0^{(1)}) - (1-c^{(1)})\, v^{(2)}(\epsilon_0^{(2)})\}, \tag{4.10}$$

where $\tilde{W}_0^{(2)}(\bar{\boldsymbol{E}})$ corresponds to the effective energy function of the two-phase, isotropic linear rank-II laminate described at the beginning of this subsection (where the two linear phases are made to correspond exactly to the two nonlinear phases). Note that this result also follows directly from the exact result (4.9) by letting $\epsilon_0^{(3)} = \epsilon_0^{(2)}$; the inequality arising because the sup over the smaller set in (4.10) leads to a result that is in general smaller than the exact result of (4.9) over the larger set.

However, more importantly, note that, as demonstrated at the beginning of this subsection (see (4.8)), $\tilde{W}_0^{(2)}(\bar{\boldsymbol{E}})$ corresponds exactly to the Hashin–Shtrikman bounds for two-phase, isotropic linear composites, and hence the right-hand side of inequality (4.10) corresponds to the Hashin–Shtrikman bounds (and estimates) for two-phase, isotropic nonlinear composites. Thus, in general, we do not have equality between the nonlinear H–S bounds, and the nonlinear rank-II laminates. These observations apply also to the three-dimensional composites: in this case, as we will see, the exact computation of the nonlinear rank-III laminate involves a four-dimensional optimization, whereas the corresponding evaluation of the nonlinear H–S bounds of §3 involve only a two-dimensional optimization.

Thus we have reached the important conclusion that, in general, the H–S bounds (and estimates) of the previous section for two-phase nonlinear composites are not attained by rank-d iterated laminates with the same microstructure as the corresponding optimal linear laminates, and hence the nonlinear H–S bounds are probably not sharp. In fact, as we have seen, the nonlinear rank-d iterated laminates with the same microstructure as the corresponding optimal linear laminates are not even isotropic in general. This motivates the question of whether rank-d iterated laminates with different microstructures may be designed for overall isotropy in the nonlinear context. In general, we expect that the answer to this question is negative since it is known that d planes of symmetry are not sufficient to ensure overall isotropy in d dimensions. However, in the balance of this section, I show that nonlinear 'isotropic' rank-d (iterated) laminates may be constructed in some restricted sense. I also compute their properties, and in §5 compare them with the nonlinear H–S bounds and estimates of §3, for the special case of two-phase composites, where one of the phases has either zero, or infinite dielectric constant. The results for these extreme situations (we expect that the differences will be proportional to the contrast between the phases) within the class of two-phase nonlinear isotropic composites are expected to serve as a test of the conjecture that the results for the 'isotropic' rank-d laminates are not far from the actual (unknown) extremal effective properties (or optimal bounds) of two-phase nonlinear isotropic composites.

To determine the effective energy functions of the 'isotropic' nonlinear laminates, we first develop a simpler form for the effective energy functions of general nonlinear iterated laminates (i.e. simpler than (4.9)). Thus, to compute the effective energy function of nonlinear rank-d laminates ($d = 2, 3$), with layering directions $\boldsymbol{n}_1, \ldots, \boldsymbol{n}_d$, we begin by specifying the result of §4a to the rank-I laminate depicted in figure 1a. Denoting by c^I the volume fraction of phase 1 with potential $\phi^{(1)}$ in the rank-I laminate, and noting that $1-c^I$ is the corresponding volume fraction of phase 2 with potential $\phi^{(2)}$, we find that the effective energy function of the nonlinear, rank-I laminate is given by

$$\tilde{W}_I(\bar{\boldsymbol{E}}) = \inf_{\substack{\omega_I^{(1)},\, \omega_I^{(2)} \\ \bar{\omega}_I = 0}} \{c^I \phi^{(1)}(s^{(1)}) + (1-c^I)\,\phi^{(2)}(s^{(2)})\}, \tag{4.11}$$

where $\bar{\omega}_I = c^I \omega_{\mathrm{I}}^{(1)} + (1-c^I)\,\omega_I^{(2)}$, and where

$$s^{(1)} = \surd[\bar{E}^2 - (\bar{\boldsymbol{E}}\cdot\boldsymbol{n}_1)^2 + (1-\omega_I^{(1)})^2(\bar{\boldsymbol{E}}\cdot\boldsymbol{n}_1)^2],$$
$$s^{(2)} = \surd[\bar{E}^2 - (\bar{\boldsymbol{E}}\cdot\boldsymbol{n}_1)^2 + (1-\omega_I^{(2)})^2(\bar{\boldsymbol{E}}\cdot\boldsymbol{n}_1)^2].$$

Next, we consider the rank-II laminate obtained by mixing layers of the rank-I laminate with layers of a third phase characterized by energy function $\phi^{(3)}$, in relative (to the rank-II laminate) volume fractions c^{II} and $1-c^{II}$, respectively. The new lamination direction $\boldsymbol{n}_2$ is orthogonal to $\boldsymbol{n}_1$, as shown in figure 1*b*. Then, applying Corollary 4.1 to the resulting microstructure (a three-phase material with constant fields over each isotropic phase), we arrive, via (4.9) (with thus far arbitrary c^{II}), to the following energy function for the nonlinear rank-II laminate in dimension $d \geqslant 2$, namely,

$$\tilde{W}_{II}(\bar{\boldsymbol{E}}) = \inf_{\substack{\omega_I^{(1)},\, \omega_I^{(2)},\, \omega_{II}^{(1)},\, \omega_{II}^{(2)} \\ \bar{\omega}_I = \bar{\omega}_{II} = 0}} \{c^{II} c^I \phi^{(1)}(s^{(1)}) + c^{II}(1-c^I)\,\phi^{(2)}(s^{(2)}) + (1-c^{II})\,\phi^{(3)}(s^{(3)})\}, \tag{4.12}$$

where $\bar{\omega}_{II} = c^{II}\omega_{II}^{(1)} + (1-c^{II})\,\omega_{II}^{(2)}$, and where

$$s^{(1)} = \surd[\bar{E}^2 - (\bar{\boldsymbol{E}}\cdot\boldsymbol{n}_1)^2 - (\bar{\boldsymbol{E}}\cdot\boldsymbol{n}_2)^2 + (1-\omega_I^{(1)})^2(\boldsymbol{E}\cdot\boldsymbol{n}_1)^2 + (1-\omega_{II}^{(1)})^2(\bar{\boldsymbol{E}}\cdot\boldsymbol{n}_2)^2],$$
$$s^{(2)} = \surd[\bar{E}^2 - (\bar{\boldsymbol{E}}\cdot\boldsymbol{n}_1)^2 - (\bar{\boldsymbol{E}}\cdot\boldsymbol{n}_2)^2 + (1-\omega_I^{(2)})^2(\bar{\boldsymbol{E}}\cdot\boldsymbol{n}_1)^2 + (1-\omega_{II}^{(1)})^2(\bar{\boldsymbol{E}}\cdot\boldsymbol{n}_2)^2],$$
$$s^{(3)} = \surd[\bar{E}^2 - (\bar{\boldsymbol{E}}\cdot\boldsymbol{n}_2)^2 + (1-\omega_{II}^{(2)})^2(\bar{\boldsymbol{E}}\cdot\boldsymbol{n}_2)^2],$$

with $c^{II}c^I = c^{(1)}$. I emphasize that we cannot make use of Corollary 4.1 to treat the rank-II laminate as a simple laminate made up of the rank-I laminate (with effective energy function given by (4.11)) and phase 3, the reason being that the rank-I laminate is anisotropic and Corollary 4.1 does not hold. Instead, we must treat the rank-II laminate as a three-phase composite; in this case, we are assured of the isotropy of all three phases and may take advantage of Corollary 4.1.

A corresponding result may be obtained for a two-phase, nonlinear rank-II laminate by letting $\phi^{(3)} = \phi^{(2)}$ in (4.12). As discussed previously, even in this case, the result for $\tilde{W}_{II}$ is generally anisotropic, but in two dimensions ($d = 2$) it may be used to obtain an isotropic result, for each value of $\bar{E}$, by an appropriate choice of c^{II} (not the fixed choice that makes the corresponding linear rank-II laminate isotropic). This choice is accomplished by requiring that c^{II} ($0 \leqslant c^{II} \leqslant 1$) and $\bar{E}_1 = \bar{\boldsymbol{E}}\cdot\boldsymbol{n}_1$ satisfy the relations determined by

$$\left.\frac{\partial \tilde{W}_{II}}{\partial \bar{E}_1}\right|_{\bar{E}_1^2 + \bar{E}_2^2 = \bar{E}^2} = 0 \quad \text{and} \quad \frac{\partial \tilde{W}_{II}}{\partial c^{II}} = 0, \tag{4.13}$$

where the derivative in the first relation is subject to the constraint $\bar{E}_1^2 + \bar{E}_2^2 = \bar{E}^2$, and that in the second assumes that $\bar{E}$ is fixed. These conditions follow by performing a Taylor series expansion of (4.12) in c^{II} and $\bar{E}_1$, and requiring that the expansion yield the same result for any choice of c^{II} and $\bar{E}_1$. Physically, this corresponds to selecting a microstructure (by choosing c^{II}) – with fixed overall volume fractions of the phases – for each value of $\bar{E}$ ensuring that $\tilde{W}_{II}$ is independent of the direction of

$\bar{\boldsymbol{E}}$. Thus, the resulting energy function, $\tilde{W}^i_{II}$, is isotropic. However, $\tilde{W}^i_{II}$ does not correspond to a fixed microstructure, but rather to a family of (anisotropic) microstructures: one for each value of applied electric field. Therefore, $\tilde{W}^i_{II}$ should be thought of as the effective energy function of an approximation to the class of nonlinear isotropic microstructures. From now on, for simplicity, I refer to the result $\tilde{W}^i_{II}$, defined by (4.12) together with (4.13), as the effective energy function of the 'isotropic' nonlinear laminate. Similar comments will apply for rank-III laminates in three dimensions, which are discussed next.

The effective energy function of a nonlinear rank-III laminate is obtained by computing the effective behaviour of a simple laminate made up of layers of the rank-II laminate (just discussed) and of layers of a fourth phase with energy function $\phi^{(4)}$ in volume fractions c^{III} and $1-c^{III}$, respectively. The new layering direction $\boldsymbol{n}_3$ is chosen to be orthogonal to both $\boldsymbol{n}_2$ and $\boldsymbol{n}_1$. Then, the effective energy function of the nonlinear rank-III laminate may again be obtained by means of Corollary 4.1 (in this case, the fields are constant over each of the four phases), with the result that

$$\tilde{W}_{III}(\bar{\boldsymbol{E}}) = \inf_{\substack{\omega_I^{(1)},\omega_I^{(2)},\omega_{II}^{(1)},\omega_{II}^{(2)},\omega_{III}^{(1)},\omega_{III}^{(2)} \\ \bar{\omega}_I=\bar{\omega}_{II}=\bar{\omega}_{III}=0}} \{c^{III}c^{II}c^{I}\phi^{(1)}(s^{(1)})+c^{III}c^{II}(1-c^{I})\,\phi^{(2)}(s^{(2)})+$$

$$\ldots+c^{III}(1-c^{II})\,\phi^{(3)}(s^{(3)})+(1-c^{III})\,\phi^{(4)}(s^{(4)})\}, \qquad (4.14)$$

where $\bar{\omega}_{III} = c^{III}\omega^{(1)}_{III}+(1-c^{III})\,\omega^{(2)}_{III}$,

$$s^{(1)} = \surd[\bar{E}^2-\bar{E}_1^2-\bar{E}_2^2-\bar{E}_3^2+(1-\omega_I^{(1)})^2\bar{E}_1^2+(1-\omega_{II}^{(1)})^2\bar{E}_2^2+(1-\omega_{III}^{(1)})^2\bar{E}_3^2],$$

$$s^{(2)} = \surd[\bar{E}^2-\bar{E}_1^2-\bar{E}_2^2-\bar{E}_3^2+(1-\omega_I^{(2)})^2\bar{E}_1^2+(1-\omega_{II}^{(1)})^2\bar{E}_2^2+(1-\omega_{III}^{(1)})^2\bar{E}_3^2],$$

$$s^{(3)} = \surd[\bar{E}^2-\bar{E}_2^2-\bar{E}_3^2+(1-\omega_{II}^{(2)})^2\bar{E}_2^2+(1-\omega_{III}^{(1)})^2\bar{E}_3^2],$$

$$s^{(4)} = \surd[\bar{E}^2-\bar{E}_3^2+(1-\omega_{III}^{(2)})^2\bar{E}_3^2],$$

and where I have made use of the notations $\bar{E}_i = \boldsymbol{E}\cdot\boldsymbol{n}_i\,(i=1,2,3)$.

Once again, the effective energy function of a two-phase, nonlinear rank-III laminate may be obtained by letting $\phi^{(4)}=\phi^{(3)}=\phi^{(2)}$ in (4.14). Then, in three dimensions ($d=3$), expression (4.14) for the rank-III laminate may be used to obtain an isotropic energy function by letting $c^{III}c^{II}c^{I}=c^{(1)}$; and choosing c^{II} and c^{III} ($0\leqslant c^{II}, c^{III}\leqslant 1$), and $\bar{E}_1, \bar{E}_2, \bar{E}_3$ with $\bar{E}_1^2+\bar{E}_2^2+\bar{E}_3^2=\bar{E}^2$ such that the relations implicit in

$$\left.\frac{\partial\tilde{W}_{III}}{\partial\bar{E}_1}\right|_{\bar{E}_1^2+\bar{E}_2^2+\bar{E}_3^2=\bar{E}^2} = \left.\frac{\partial\tilde{W}_{III}}{\partial\bar{E}_2}\right|_{\bar{E}_1^2+\bar{E}_2^2+\bar{E}_3^2=\bar{E}^2} = 0, \quad \frac{\partial\tilde{W}_{III}}{\partial c^{II}} = \frac{\partial\tilde{W}_{III}}{\partial c^{III}} = 0 \qquad (4.15)$$

are satisfied. The resulting isotropic energy function $\tilde{W}^i_{III}$, corresponding to a nonlinear 'isotropic' rank-III composite, may be used to model the behaviour of nonlinear isotropic composites in three dimensions. In the next section, I compare this and the previous results for the rank-II laminates with the bounds and estimates of §3*b* for isotropic nonlinear composites, with some special choices for their constituent phases.

5. Application to two-phase composites

To illustrate the power of the methods described in the previous two sections for determining bounds and estimates for the effective properties of nonlinear composites, I investigate in this section two simple, but important, examples. They both involve a nonlinear matrix with isotropic potential $\phi^{(2)}=\phi$ (subject to the restrictions of Theorem 2.1), and an inclusion phase with either infinite dielectric constant, or alternatively, with zero dielectric constant. In the first case, we have that $\phi^{(1)} = 0$ if $E = 0$, or $\phi^{(1)} = \infty$ otherwise; and in the second, we have that $\phi^{(1)} = 0$ regardless of the value of E. Thus I compute bounds for these nonlinear composites applying the results of §3, and compare them with exact estimates for 'isotropic' laminates based on the results of §4.

(*a*) *The nonlinear material with perfectly conducting inclusions*

The first observation is that the upper bounds for this class of composite materials, corresponding to geometries with the unbounded dielectric material in the matrix phase, are unbounded. Thus we concentrate our efforts in determining lower bounds and minimal estimates for the effective properties of this class of nonlinear materials.

According to relation (3.12), the Weiner lower bound for this nonlinear material is given by

$$\tilde{W}_{\mathrm{W}-}(\bar{\boldsymbol{E}}) = (1-c)\,\phi(\bar{E}/(1-c)), \tag{5.1}$$

where I have used the fact that $\omega = (1-c)^{-1}$ solves the infimum problem in (3.12). Here c is the volume fraction of the perfectly conducting inclusions. The corresponding H–S lower bound is obtained by specializing relation (3.21) and noting that once again $\omega = (1-c)^{-1}$ solves the optimization problem. The result may be written in the form

$$\tilde{W}_{\mathrm{HS}-}(\bar{\boldsymbol{E}}) = (1-c)\,\phi\left(\frac{\surd[1+(d-1)\,c]}{1-c}\bar{E}\right). \tag{5.2}$$

Note that for $d > 1$, $\tilde{W}_{\mathrm{HS}-} > \tilde{W}_{\mathrm{W}-}$ and the H–S bound for isotropic materials is sharper than the Weiner bound for generally anisotropic materials (which also holds, in particular, for isotropic materials). Note that the above bounds hold for arbitrary nonlinear matrix satisfying the growth conditions mentioned at the end of §2*c* (superquadratic growth for ϕ). We note further that the above results for the H–S bounds can also be obtained by the method of Talbot & Willis (1985), although the specific results described above have apparently not yet appeared in the published literature. Analogous results in nonlinear infinitesimal elasticity have been given by Ponte Castañeda & Willis (1988) and improved by Ponte Castañeda (1991*a*, *b*). I do not consider the Beran bounds in this section, because they require more geometric information than the volume fractions of the phases.

Next, I obtain results for the nonlinear 'isotropic' laminates by considering the two- and three-dimensional laminates separately. The effective energy function of the two-dimensional 'isotropic' laminate is obtained directly from relations (4.12) and (4.13) with $\phi^{(2)} = \phi^{(3)} = \phi$, and by noticing that $\omega_I^{(1)} = 1$ and $\omega_{II}^{(1)} = 1$ solve the optimization problem in this case. This leads to the result that

$$\tilde{W}^i_{II}(\bar{\boldsymbol{E}}) = \underset{0\leqslant c^{II}\leqslant 1}{\underset{c^{II}}{\operatorname{stat}}}\ \underset{\bar{E}_1^2+\bar{E}_2^2=\bar{E}^2}{\underset{\bar{E}_1}{\operatorname{stat}}} \left\{(c^{II}-c)\,\phi\left[\left(\frac{c^{II}}{c^{II}-c}\right)\bar{E}_1\right]+(1-c^{II})\,\phi\left[\sqrt{\left[\bar{E}_1^2+\left(\frac{\bar{E}_2}{1-c^{II}}\right)^2\right]}\right]\right\}, \tag{5.3}$$

where I have rewritten the isotropy conditions (4.13) in the form of stationary optimization processes. In general, these optimizations will need to be carried out numerically, but we will see in §5*c* that further simplification can be achieved for special choices of ϕ. Note also that a quadratic choice for ϕ leads to the exact H–S lower bound for a two-phase, linear isotropic composite.

The corresponding three-dimensional result for the effective energy function of the nonlinear, isotropic rank-III laminate is obtained in a similar fashion from (4.14) and (4.15), and is given by

$$\tilde{W}^{i}_{III}(\bar{\boldsymbol{E}}) = \underset{\substack{c^{II},\, c^{III} \\ 0 \leqslant c^{II},\, c^{III} \leqslant 1}}{\operatorname{stat}} \; \underset{\substack{\bar{E}_1,\, \bar{E}_2 \\ \bar{E}_1^2+\bar{E}_2^2+\bar{E}_3^2=\bar{E}^2}}{\operatorname{stat}} \left\{ (c^{III}c^{II}-c)\, \phi\left[\left(\frac{c^{III}c^{II}}{c^{III}c^{II}-c}\right)\bar{E}_1\right] + \right.$$

$$\left. \ldots + c^{III}(1-c^{II})\, \phi\left[\sqrt{\left[\bar{E}_1^2 + \left(\frac{\bar{E}_2}{1-c^{II}}\right)^2\right]}\right] + (1-c^{III})\, \phi\left[\sqrt{\left[\bar{E}_1^2+\bar{E}_2^2+\left(\frac{\bar{E}_3}{1-c^{III}}\right)^2\right]}\right]\right\}. \tag{5.4}$$

This result can also be shown to reduce to the corresponding linear H–S lower bound for a quadratic choice for ϕ.

(*b*) *The nonlinear material with perfectly insulating inclusions*

In this case, note that the lower bounds will of necessity be trivial because the choice of the perfect insulator as the matrix phase leads to perfectly insulating effective behaviour for the composite. Thus I attempt to obtain upper bounds, and maximal estimates for the effective energy function of this class of composite material. The Weiner upper bound, corresponding to arbitrarily anisotropic composites, is obtained from (3.26), which yields the result

$$\tilde{W}_{\mathrm{W}+}(\bar{\boldsymbol{E}}) = (1-c)\, \phi(\bar{E}), \tag{5.5}$$

where c now stands for the volume of the perfectly insulating inclusions. The Hashin–Shtrikman upper 'estimate' (not a rigorous bound) is obtained from (3.21) with the min replaced by a max. The result is

$$\tilde{W}_{\mathrm{HS}+}(\bar{\boldsymbol{E}}) = (1-c)\, \phi\left(\sqrt{\left(\frac{d-1}{d-1+c}\right)}\bar{E}\right). \tag{5.6}$$

Although, as already mentioned, we do not expect this result to yield an upper bound in general, it does reduce to the well-known H–S upper bound in the case of a linear matrix.

The exact estimate for the two-dimensional, nonlinear 'isotropic' laminate is obtained from expressions (4.12) and (4.13) with $\phi^{(2)} = \phi^{(3)} = \phi$. In this case, the result is not as explicit as the corresponding result in §5*a*, and takes the form

$$\tilde{W}^{i}_{II}(\bar{\boldsymbol{E}}) = \underset{\substack{c^{II} \\ 0 \leqslant c^{II} \leqslant 1}}{\operatorname{stat}} \; \underset{\substack{\bar{E}_1 \\ \bar{E}_1^2+\bar{E}_2^2=\bar{E}^2}}{\operatorname{stat}} \; \inf_{\omega^{II}} \{(c^{II}-c)\, \phi(s^{(2)}) + (1-c^{II})\, \phi(s^{(3)})\}, \tag{5.7}$$

where

$$s^{(2)} = (1+c^{II}\omega_{II})\, \bar{E}_2,$$

$$s^{(3)} = \sqrt{\{\bar{E}_1^2 + [1-(1-c^{II})\, \omega_{II}]^2 \bar{E}_2^2\}}.$$

Further simplification is available for special choices of ϕ, as we see in §5*c*. Also, this result reduces to the H–S upper bound in the limit of linear behaviour for the matrix.

The result for the effective energy function of the three-dimensional, nonlinear 'isotropic' laminate is obtained analogously, with the result that

$$\tilde{W}^{i}_{III}(\bar{\boldsymbol{E}}) = \underset{\substack{c^{II},\, c^{III} \\ 0 \leqslant c^{II},\, c^{III} \leqslant 1}}{\operatorname{stat}} \; \underset{\substack{\bar{E}_1,\, \bar{E}_2 \\ \bar{E}_1^2+\bar{E}_2^2+\bar{E}_3^2=\bar{E}^2}}{\operatorname{stat}} \; \inf_{\omega^{II},\, \omega^{III}} \{(c^{III}c^{II}-c)\, \phi(s^{(2)}) +$$

$$\ldots + c^{III}(1-c^{II})\, \phi(s^{(3)}) + (1-c^{III})\, \phi(s^{(4)})\}, \tag{5.8}$$

where
$$s^{(2)} = \surd\{(1+c^{II}\omega_{II})^2\bar{E}_2^2+(1+c^{III}\omega_{III})^2\bar{E}_3^2\},$$
$$s^{(3)} = \surd\{\bar{E}_1^2+[1-(1-c^{II})\,\omega_{II}]^2\bar{E}_2^2+(1+c^{III}\omega_{III})^2\bar{E}_3^2\},$$
$$s^{(4)} = \surd\{\bar{E}_1^2+\bar{E}_2^2+[1-(1-c^{III})\,\omega_{III}]^2\bar{E}_3^2\}.$$

Again no further simplification of this result seems possible for general ϕ, but for the special case considered in the following subsection for pure-power behaviour for the matrix, a simpler form of the result is available.

(*c*) *Special results for pure-power materials*

In this subsection, I specialize the results for the two special types of nonlinear composites of §§5*a* and *b* by considering pure-power behaviour for the matrix material. This type of behaviour for the matrix, characterized by the energy-density function
$$\phi(E) = (n+1)^{-1}\epsilon E^{n+1} \tag{5.9}$$
(where $n \geqslant 1$ is the power and ϵ is the nonlinear dielectric constant), has the advantage that it yields the same type of behaviour for the isotropic composite materials with perfectly conducting, or perfectly insulating, inclusions. Thus, for these two types of isotropic composites, we have that
$$\tilde{W}(\bar{\boldsymbol{E}}) = (n-1)^{-1}\tilde{\epsilon}\bar{E}^{n+1}, \tag{5.10}$$
where $\tilde{\epsilon}$ denotes the effective nonlinear dielectric constant of the composite. For the anisotropic composites, the form of the effective energy will be different in general, but at least the Weiner bounds will also be of the form (5.10). We can then characterize the behaviour of the Weiner, Hashin–Shtrikman and 'isotropic' laminates for the class of composites studied in §§5*a* and *b* in terms of this effective nonlinear dielectric constant, appropriately normalized by the dielectric constant of the matrix material (i.e. $\tilde{\epsilon}/\epsilon$).

The results for the bounds and estimates of two-dimensional composites are not essentially different from the results for three-dimensional composites. For this reason, I only present results here for the three-dimensional composites. I begin by depicting the results for the Weiner lower bound, the H–S lower bound and the 'isotropic' laminate estimate, as given by relations (5.1), (5.2) and (5.4), respectively, for the nonlinear material with perfectly conducting inclusions. These results, expressed in terms of the effective nonlinear dielectric constant, are given respectively by
$$\left.\begin{aligned}\tilde{\epsilon}_{\mathrm{W}-}/\epsilon &= (1-c)^{-n}.\\ \tilde{\epsilon}_{\mathrm{HS}-}/\epsilon &= (1+2c)^{(n+1)/2}/(1-c)^n,\\ \frac{\tilde{\epsilon}_{III}}{\epsilon} &= \sup_{\substack{x,\,y\\ 0\leqslant x,\,y\leqslant 1\\ xy\geqslant c}}\left\{(1-y)^{2n/(n-1)}+\left(\frac{2-y}{y}\right)^{(n+1)/(n-1)}\left[(xy-c)^{2n/(n-1)}\right.\right.\\ &\qquad\left.\left.\times\left(\frac{2-x}{x}\right)^{(n+1)/(n-1)}+[(1-x)\,y]^{2n/(n-1)}\right]\right\}^{(1-n)/2}.\end{aligned}\right\} \tag{5.11}$$

Note that the expressions for the bounds are remarkably simple. On the other hand, the expression for the exact estimate for the 'isotropic' laminate involves a two-dimensional optimization, which must be solved numerically. It is interesting to note that $(\tilde{\epsilon}_{III}/\epsilon)^{1/n} \to (1-8c+12c^{\frac{4}{3}}-6c^{\frac{5}{3}}+c^2)^{-\frac{1}{2}}$ as $n\to\infty$, which is different from, but close to, $(\tilde{\epsilon}_{\mathrm{HS}-}/\epsilon)^{1/n}$ in the limit as $n\to\infty$.

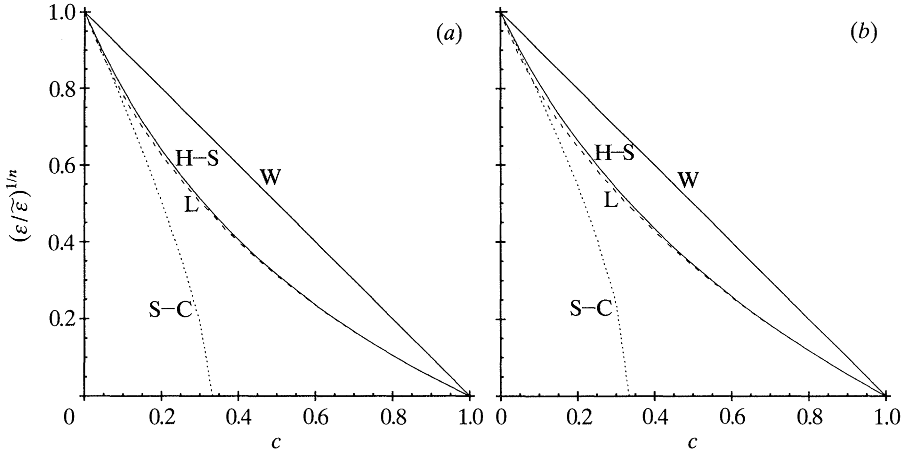

Figure 2. Plot of the effective nonlinear dielectric constant as a function of the volume fraction of perfectly conducting inclusions for (*a*) a weakly nonlinear matrix ($n = 3$) and (*b*) a strongly nonlinear matrix ($n = 10$).

The results of these computations for the effective dielectric constant of the nonlinear 'isotropic' laminate as functions of the volume fraction c of the perfectly conducting inclusions for two values of n (3 and 10, corresponding to a weakly nonlinear and a strongly nonlinear material, respectively) are given in figure 2*a* and *b* (they are denoted by L). In these plots, we have also included for comparison the results for the bounds (W and H–S), as well as self-consistent (S–C) estimates obtained in the same way as the nonlinear H–S bounds, but making use of the linear self-consistent estimate instead of the linear H–S bounds in relation (3.3). This self-consistent result takes the form

$$\tilde{\epsilon}_{\mathrm{SC}}/\epsilon = (1-c)^{-(n-1)/2}(1-3c)^{-(n+1)/2}. \qquad (5.12)$$

All of the above results are depicted in terms of the reciprocal of the effective dielectric constant to an appropriate power for plotting convenience. Thus the lower bounds appear as upper bounds in these plots. By comparing the Weiner and H–S bounds, we observe that the H–S bounds are significantly tighter than the Weiner bounds. This is not surprising since the Weiner bounds must apply for generally anisotropic materials, which is a larger class of materials than the class of isotropic materials for which the H–S bounds must hold. Recall, however, that the Weiner bounds were shown to be sharp within the class of arbitrarily anisotropic composites. Comparing the H–S bounds with the exact estimates for the 'isotropic' laminates (L), and observing that they are very close, even for the larger value of n, is suggestive that the H–S bounds, although probably not optimal, are close to the optimal bounds. Such optimal bounds would be expected to lie between the exact estimates for the 'isotropic' laminates, which are approximations to actual microstructures within the class of two-phase isotropic materials, and the H–S bounds, which are rigorous bounds in this case. Finally, we note that the self-consistent estimates lie below the bounds, as they should, and that they are close to the bounds for small volume fractions of the perfectly conducting inclusions, but diverge away from the bounds for moderate volume fractions reaching an early percolation limit at $c = \frac{1}{3}$.

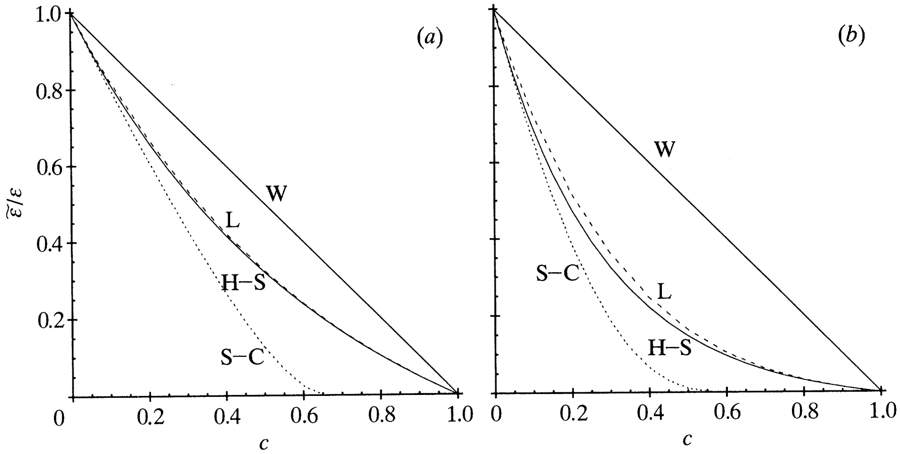

Figure 3. Plot of the effective nonlinear dielectric constant as a function of the volume fraction of perfectly insulating inclusions for (*a*) a weakly nonlinear matrix ($n = 3$) and (*b*) a strongly nonlinear matrix ($n = 10$).

Next, we consider the corresponding results for the nonlinear material with perfectly insulating inclusions in three space dimensions. These results for the Weiner upper bound, the H–S 'upper estimate', the self-consistent estimate and the exact estimate for the 'isotropic' laminate are given, respectively, by the relations

$$\left.\begin{aligned}
\tilde{\epsilon}_{\mathrm{W}+}/\epsilon &= (1-c),\\
\tilde{\epsilon}_{\mathrm{HS}+}/\epsilon &= (1-c)/(1+\tfrac{1}{2}c)^{(n+1)/2},\\
\tilde{\epsilon}_{\mathrm{SC}}/\epsilon &= (1-\tfrac{3}{2}c)^{(n+1)/2}/(1-c)^{(n-1)/2},\\
\frac{\tilde{\epsilon}_{III}}{\epsilon} &= \sup_{\substack{x,\,y\\ 0\leqslant x,\,y\leqslant 1\\ xy\geqslant c}} \left\{\frac{[(1-x)\,y+(1-y)\,p]^{(n+1)/2}}{[(xy-c)\,q^{(n+1)/(n-1)}+(1-x)\,y+(1-y)\,p^{(n+1)/(n-1)}]^{(n-1)/2}}\right\}
\end{aligned}\right\}\tag{5.13}$$

where p solves the quadratic equation

$$\tfrac{1}{2}[(1-y)/(1-x)]\,(2-x)\,p^2-[\tfrac{1}{2}xy+2(1-y)]\,p+(1-x)\,(1-y)=0,$$

and

$$q=\left(\frac{xy}{xy-c}\right)\left(\frac{1-x}{2-x}\right).$$

These results are plotted as functions of the volume fraction of inclusions c in figure 3*a* and *b* for two values of n (3 and 10). We observe that, as expected, the H–S 'upper estimates' for the isotropic composite lie well below the Weiner bounds for the arbitrarily anisotropic composites. On the other hand, we find that the exact estimates for the nonlinear 'isotropic' laminates (L) lie above the H–S 'upper estimates'. This verifies our expectation, proposed in §4, that the H–S 'upper estimates' are probably not in general upper bounds. This is because the 'isotropic' laminates correspond to (approximations of) specific microstructures within the class of isotropic composite materials, and if the H–S estimates were rigorous bounds for this class of composite materials, they would have to lie above all possible isotropic microstructures, and, in particular, they would be expected to lie above the 'isotropic' laminates. Nevertheless, we find that the effective dielectric constants of the 'isotropic' laminates are not far from the H–S 'upper estimates'. Thus, we anticipate that the optimal upper bounds for this class of materials would probably not be too distant from the 'isotropic' laminate results, and hence not too far from the H–S 'upper estimates'. Then, the H–S 'upper estimates', although not bounds, would provide simple estimates for the extremal properties of this class of nonlinear composites. We add that the self-consistent estimates agree with the H–S estimates for low values of c, but they reach an early percolation limit at $c = \frac{2}{3}$.

Finally, we note that through the well-known connections between the H–S bounds and the Maxwell–Garnett approximation, and between the self-consistent and symmetric effective-medium estimates in the context of linear conductivity, we could equally well refer to our nonlinear H–S bounds as nonlinear Maxwell–Garnett estimates, and to the self-consistent estimates as nonlinear effective-medium estimates.

6. Concluding remarks

To summarize, in this paper, I have made use of the variational principles proposed by Ponte Castañeda (1992) (Theorems 2.1 and 2.2) to develop bounds and estimates for the effective properties of nonlinear dielectrics. These variational principles can generally be used in either one of two ways: they can be used approximately to yield bounds for whole classes of microstructures (and for specific microstructures), or they can be used exactly to determine the effective properties of certain special microstructures. Prior work by the author, in different physical contexts (Ponte Castañeda 1991*a*, *b*), has made use of the approximate versions of the new variational principles to determine bounds for the effective properties of nonlinear composites. The present work is the first to consider the exact use of the variational principles in the computation of the effective properties of specific microstructures.

In §3, which deals with the approximate use of the variational principles to obtain bounds (Corollaries 3.1 and 3.2), I was able to recover the classical bounds of Weiner for generally anisotropic nonlinear composites. More importantly, I also obtained bounds of the Hashin–Shtrikman type for nonlinear isotropic composites by direct implementation of the corresponding linear Hashin–Shtrikman bounds into the new variational principles. Bounds of the Hashin–Shtrikman type for nonlinear dielectrics have also been proposed by Willis (1986) and by Talbot & Willis (1985), starting from an appropriate generalization of the Hashin–Shtrikman variational principles for nonlinear problems. However, the new method has the distinct advantage that it is not limited to bounds of the Hashin–Shtrikman type, and can be used to yield other bounds (and estimates), such as higher-order bounds (and self-consistent estimates), by direct application of the corresponding higher-order linear bounds (and estimates). Thus I was able to obtain – for the first time – bounds of the Beran type for two-phase, nonlinear isotropic composites. On the other hand, note that the method of Talbot & Willis has the advantage that it applies to anisotropic behaviour for the phases, whereas the new method, as developed thus far, applies only to isotropic phases. However, generalizations of the new method to anisotropic behaviour are certainly possible, and these will be explored elsewhere. One weakness of both methods is that the bounds generated are only one-sided. Additionally, we note that the final form of the expressions for the nonlinear Weiner and Hashin–Shtrikman bounds and estimates obtained in this work is new and simpler than previously available forms.

In §§4 and 5, we studied the application of the exact versions of the new variational principles (Corollaries 4.1 and 4.2) to compute exact estimates for the effective properties of special classes of nonlinear composite materials. These special materials, called sequentially laminated materials, are obtained as their name implies by a sequence of lamination procedures, and have the advantage that their effective behaviour can be controlled to a large extent by appropriate choices of the lamination directions, and relative volume fractions of the phases within each elemental layer. Thus, in particular, we show that a simple laminate leads to the most anisotropic composite possible within the class of arbitrarily anisotropic nonlinear composites with prescribed volume fractions, in the sense that the effective energy of the simple laminate attains the Weiner bounds (for special choices of the applied electric field). Analogous, but not quite as strong, results are obtained for the class of isotropic composites, by considering 'isotropic' laminates (in the sense described in §4*b*). In this case, we found in §5*c* that the 'isotropic' laminates do not quite attain, but lie below, the nonlinear Hashin–Shtrikman lower bounds for isotropic composites. On the other hand, we also verified that the Hashin–Shtrikman 'upper estimates' cannot be rigorous upper bounds in general (since they do not encompass the nonlinear 'isotropic' laminates), but never the less they are probably not far from the optimal upper bounds. It is expected that sequentially laminated microstructures, which have been found in recent years to play an important role in the linear theories, will also continue to play an important role in the development of the nonlinear theories. They have many advantages over other standard types of models because their effective properties can be computed exactly, and also because they can be given generalizations in other fields, where other models may be too difficult to apply.

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Appendix

In this appendix, I give a simple proof of the identity (3.7) used throughout the body of the paper. I begin by introducing a Lagrange multiplier to account for the zero-average constraint on the optimization variables, $\omega^{(r)}$. Thus,

$$\inf_{\substack{\omega^{(r)}\\ \bar{\omega}=0}} \left\{ \sum_{r=1}^{n} c^{(r)}\epsilon^{(r)}(1-\omega^{(r)})^2 \right\} = \inf_{\omega^{(r)}} \left\{ \sum_{r=1}^{n} c^{(r)}\epsilon^{(r)}(1-\omega^{(r)})^2 + \sup_{\lambda} \left\{ \lambda \sum_{r=1}^{n} c^{(r)}\omega^{(r)} \right\} \right\}, \qquad \text{(A 1)}$$

and, by duality, we have

$$\inf_{\substack{\omega^{(r)}\\ \bar{\omega}=0}} \left\{ \sum_{r=1}^{n} c^{(r)}\epsilon^{(r)}(1-\omega^{(r)})^2 \right\} = \sup_{\lambda} \left\{ \inf_{\omega^{(r)}} \left\{ \sum_{r=1}^{n} c^{(r)}\epsilon^{(r)}(1-\omega^{(r)})^2 + \lambda \sum_{r=1}^{n} c^{(r)}\omega^{(r)} \right\} \right\}. \qquad \text{(A 2)}$$

The infimum over the $\omega^{(r)}$ is satisfied by

$$\omega^{(r)} = 1 - \lambda/2\epsilon^{(r)}, \qquad \text{(A 3)}$$

which finally leads to

$$\inf_{\substack{\omega^{(r)}\\ \bar{\omega}=0}} \left\{ \sum_{r=1}^{n} c^{(r)}\epsilon^{(r)}(1-\omega^{(r)})^2 \right\} = \sup_{\lambda} \left\{ \sum_{r=1}^{n} c^{(r)} \left\{ \lambda - \frac{\lambda^2}{4\epsilon^{(r)}} \right\} \right\} = \sup_{\lambda} \left\{ \lambda - \frac{\lambda^2}{4} \sum_{r=1}^{n} \frac{c^{(r)}}{\epsilon^{(r)}} \right\} = \left(\sum_{r=1}^{n} \frac{c^{(r)}}{\epsilon^{(r)}} \right)^{-1}. \qquad \text{(A 4)}$$

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