Contributions in mechanics of materials

Exact results for weakly nonlinear composites and implications for homogenization methods

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Abstract. Weakly nonlinear composite conductors are characterized by position-dependent dissipation potentials expressible as an additive composition of a quadratic potential and a nonquadratic potential weighted by a small parameter. This additive form carries over to the effective dissipation potential of the composite when expanded to first order in the small parameter. However, the first-order correction of this asymptotic expansion depends only on the zeroth-order values of the local fields, namely, the local fields within the perfectly linear composite conductor. This asymptotic expansion is exploited to derive the exact effective conductivity of a composite cylinder assemblage exhibiting weak nonlinearity of the power-law type (i.e., power law with exponent $m = 1 + \delta$, such that $|\delta| \ll 1$), and found to be identical (to first order in δ) to the corresponding asymptotic result for sequentially laminated composites of infinite rank. These exact results are used to assess the capabilities of more general nonlinear homogenization methods making use of the properties of optimally selected linear comparison composites.

Keywords. Homogenization, Nonlinear, Heterogeneous materials, Exact solution, Composite materials, Effective conductivity.

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1. Introduction

The development of multiscale approximations for the mechanical and physical properties of disordered solids greatly benefits from the identification of specific material systems whose properties at different length scales can be linked exactly, as they provide guidance and useful benchmarks for evaluating the relative merits of competing schemes. Several classes of such solvable systems have been identified when the constitutive phases exhibit *linear* responses. These include, for instance, sequential laminates (e.g., [1–3]), dilute dispersions (e.g., [4]), assemblages of neutral coated inclusions (e.g., [5–8]), symmetric materials (e.g., [9, 10]), and iterated composites (e.g., [11–13]). Comparatively fewer results are available when the constitutive phases exhibit *nonlinear* responses. Such nonlinear results are typically generated by partially extending the above linear results, and often require more involved mathematical descriptions. Examples include sequential laminates (e.g., [14, 15]), symmetric materials (e.g., [16]), and iterated composites (e.g., [17, 18]). Now, these descriptions can simplify considerably when the constitutive responses are *weakly nonlinear* (e.g., [19–22]). But, despite this observation, results for weakly nonlinear composites are seldomly exploited in the development of homogenization methods.

The purpose of this paper is to report exact results for the electrical conductivity of two-phase composites exhibiting weak nonlinearity. Such composites are characterized by position-dependent dissipation potentials expressible as an additive composition of a quadratic potential and a nonquadratic potential weighted by a small parameter. This additive form carries over to the effective dissipation potential of the composite when expanded to first order in the small parameter. However, the first-order correction of this asymptotic expansion depends only on the zeroth-order values of the local fields, namely, the local fields within the perfectly linear composite conductor. This asymptotic expansion is exploited to derive the exact effective conductivity of a composite cylinder assemblage (CCA) exhibiting weak nonlinearity of the power-law type. More specifically, we consider conductors with phase constitutive relation of the type $J = \epsilon E^m$, where J is the current, E is the electric field, ϵ is the nonlinear conductivity and $m = 1 + \delta$, with $|\delta| \ll 1$. The exact asymptotic result (to first order in δ) for these weakly nonlinear composites, which has not previously been reported in the literature, is then used to assess the capabilities of various nonlinear homogenization methods.

These nonlinear homogenization methods blossomed from ideas initiated by Willis [23], who proposed a generalization of the variational bounds of [24] for linear composites, which could also handle nonlinear composites. The first application of these bounds of the Hashin–Shtrikman (HS) type to nonlinear composites was pursued by Talbot and Willis [25]. A more general variational approach consisting in the use of an optimally chosen "linear comparison composite" (LCC) was advanced by Ponte Castañeda [26, 27] (see also [28] for the special case of power-law materials). This approach is not only capable of delivering bounds of the HS type, but can also be used to generate three-point bounds and other estimates, such as self-consistent-type estimates, by means of corresponding bounds and estimates for linear composites. Because of their bounding properties, these "variational linear comparison" estimates have been shown to deliver significantly improved estimates. For example, Gilormini [29] showed that the "classical" extension [30, 31] of the self-consistent method [32] for nonlinear composites violates the rigorous bounds of [26]. In addition, Suquet [33] provided a reinterpretation of the variational linear comparison method of [26] in terms of the second moments of the fields in the phases of the LCC, thus providing an alternative explanation for the improvements observed relative to the classical homogenization schemes using only the averages or first moments of the fields in the phases. However, it was found that these bounds and estimates were not able to reproduce the exact estimates of [34] for small heterogeneity contrast. For these reasons, Ponte Castañeda [35] proposed an alternative "tangent second-order" (TSO) variational approach making use of more general LCCs,

which, while not yielding bounds, was capable of reproducing exactly to second order in the contrast the asymptotic expansions of [34]. In spite of giving improved results for small contrast, the TSO estimates were not as robust as the variational estimates for strongly nonlinear composites exhibiting strong heterogeneity contrast. More recently, Ponte Castañeda [36] provided a fully optimized second-order (FOSO) homogenization method combining the advantages of the variational and TSO methods, which makes use of both the first and second moments of the fields and has the additional advantage of allowing the estimation of these moments in the nonlinear composite directly from the corresponding moments in the LCC [37]. In implementing the FOSO method, one must choose the specific value, a weight-factor α which is not dictated by the underlying variational procedure. Out of convenience, Ponte Castañeda [36] chose a value of $\alpha = 0.5$, and the choice was adopted by Furer and Ponte Castañeda [38], who suggested that the "optimal" value of α would depend, among other things, on the microstructure, nonlinearity, and properties of the phases. The exact results reported in this work are used to assess this choice. Finally, it is also shown that the exact estimate of [15] for the corresponding class of sequentially laminated nonlinear composite conductors also agrees exactly to first order in the weakly nonlinear limit with the CCA result.

2. Effective behavior of weakly nonlinear composites

We consider material systems made up of two distinct constitutive phases. The electrical conductivity of each phase is characterized by a dissipation potential $w^{(r)}$ ($r = 1, 2$), such that the current density $\mathbf{J}$ and the electric field intensity $\mathbf{E}$ are related by

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

where the characteristic functions $\chi^{(r)}$ serve to describe the microstructure, being 1 if the position vector $\mathbf{x}$ is in phase r, and 0 otherwise, so that $\chi^{(1)}(\mathbf{x}) + \chi^{(2)}(\mathbf{x}) = 1$. The potentials $w^{(r)}$ are assumed to be strictly convex and bounded from below. The focus is on material systems where the size of the characteristic particle size is much smaller than the size of the specimen. In that case, the functions $\chi^{(r)}$ exhibit rapid oscillations and homogenization theory states that the *overall response* of the composite is given by the relation between the average current density and the average electric field over a "representative volume element" Ω. Then, letting $\langle \cdot \rangle$ denote the volume average over Ω, and letting $\bar{\mathbf{J}} = \langle \mathbf{J} \rangle$ and $\bar{\mathbf{E}} = \langle \mathbf{E} \rangle$, the overall response can be characterized by the *effective potential* $\tilde{w}$, such that (e.g., [39])

$$\bar{\mathbf{J}} = \frac{\partial \tilde{w}}{\partial \bar{\mathbf{E}}}(\bar{\mathbf{E}}), \quad \tilde{w}(\bar{\mathbf{E}}) = \min_{\mathbf{E} \in \mathcal{K}(\bar{\mathbf{E}})} \langle w(\mathbf{x}, \mathbf{E}) \rangle, \tag{2}$$

where $\mathcal{K}(\bar{\mathbf{E}})$ is the set of admissible fields $\mathbf{E}(\mathbf{x})$, such that there is a continuous scalar field φ in a suitable functional space satisfying $\mathbf{E} = -\nabla \varphi$ in Ω and $\varphi = -\bar{\mathbf{E}} \cdot \mathbf{x}$ on $\partial \Omega$. In view of these differential constraint, the minimizer $\mathbf{E}$ must be such that the associated current density field $\mathbf{J}$ be divergence free, i.e., $\nabla \cdot \mathbf{J} = 0$ within Ω. Dual variational formulations are available involving the Legendre transform of $\tilde{w}(\bar{\mathbf{E}})$, as given by $\tilde{u}(\bar{\mathbf{J}}) = \tilde{w}^*(\bar{\mathbf{E}})$.

This constitutive framework can be used to model weakly nonlinear as well as strongly nonlinear behaviors. Weakly nonlinear materials are hereby characterized by dissipation potentials of the form

$$w^{(r)}(\mathbf{E}) = w_0^{(r)}(\mathbf{E}) + \delta\, w^{(r)}(\mathbf{E}), \tag{3}$$

where $w_0^{(r)}$ is quadratic but $w^{(r)}$ is not, $w_0^{(r)}$ and $w^{(r)}$ are both bounded from below, and δ is a small parameter. The effective response of this weakly nonlinear composite can then be determined, at least formally, by expanding the effective potential (2) to first order in δ. Denoting

by $\mathbf{E}_\delta$ and $\mathbf{E}_0$ the electric field intensities attaining the minimum in $(2)_2$ for nonzero δ and for $\delta = 0$, respectively,

$$\widetilde{w}(\overline{\mathbf{E}}) = \langle w(\mathbf{x}, \mathbf{E}_\delta) \rangle = \langle w_0(\mathbf{x}, \mathbf{E}_0) \rangle + \delta \left\langle w(\mathbf{x}, \mathbf{E}_0) + \frac{\partial w_0}{\partial \mathbf{E}}(\mathbf{x}, \mathbf{E}_0) \cdot \frac{\partial \mathbf{E}_\delta}{\partial \delta}\bigg|_{\delta=0} \right\rangle + O(\delta^2). \tag{4}$$

Then, noting that $\partial w_0 / \partial \mathbf{E}(\mathbf{x}, \mathbf{E}_0)$ is the current density field $\mathbf{J}_0$ within the composite for $\delta = 0$, and is therefore divergence free, and that $\partial \mathbf{E}_\delta / \partial \delta|_{\delta=0} = -\nabla \partial \varphi_\delta / \partial \delta|_{\delta=0}$, Hill's lemma implies that [40]

$$\left\langle \frac{\partial w_0}{\partial \mathbf{E}}(\mathbf{x}, \mathbf{E}_0) \cdot \frac{\partial \mathbf{E}_\delta}{\partial \delta}\bigg|_{\delta=0} \right\rangle = \left\langle \frac{\partial w_0}{\partial \mathbf{E}}(\mathbf{x}, \mathbf{E}_0) \right\rangle \cdot \left\langle \frac{\partial \mathbf{E}_\delta}{\partial \delta}\bigg|_{\delta=0} \right\rangle = \left\langle \frac{\partial w_0}{\partial \mathbf{E}}(\mathbf{x}, \mathbf{E}_0) \right\rangle \cdot \frac{\partial \overline{\mathbf{E}}}{\partial \delta}\bigg|_{\delta=0} = 0, \tag{5}$$

since $\langle \mathbf{E}_\delta \rangle = \overline{\mathbf{E}}$ and the expansion is carried out for fixed $\overline{\mathbf{E}}$. The effective potential of the weakly nonlinear composite is thus given by

$$\widetilde{w}(\overline{\mathbf{E}}) = \langle w_0(\mathbf{x}, \mathbf{E}_0) \rangle + \delta \langle w(\mathbf{x}, \mathbf{E}_0) \rangle + O(\delta^2), \tag{6}$$

and therefore its evaluation to first order in δ requires knowledge of the *linear* fields only. This observation has been exploited in earlier works [19, 20, 22] to study composites exhibiting weakly nonlinear responses of the polynomial type. In the next section, results are derived for a different class of composites exhibiting weakly nonlinear responses of the power-law type.

3. Composite cylinder assemblages with power-law dissipation

We consider a special class of cylindrical dispersions consisting of nonoverlapping homothetic composite cylinders of infinite sizes filling up the entire material volume, each composed of an exterior cylinder coating made up of the matrix phase and an interior concentric cylinder made up of the inclusion phase. These microstructures were introduced by HS [5] and are referred to as CCAs. When the phases exhibit an isotropic linear response, the assemblage exhibits an in-plane isotropic linear effective response that can be determined exactly by solving the field equations within a single composite cylinder subject to uniform boundary conditions (e.g., [5, 41]). When the phases exhibit a nonlinear response, on the other hand, the computation of the exact response requires the solution of the field equations throughout the dispersion. In view of the expansion (6), however, the linear fields within a single composite cylinder still furnish the exact effective response when the phases exhibit a weakly nonlinear response, at least to first order. Henceforth, we compute the expansion (6) for assemblages characterized by isotropic dissipation potentials of the power-law form

$$w^{(r)}(\mathbf{E}) = \frac{\epsilon^{(r)}}{m+1} E^{m+1}, \tag{7}$$

where $E = |\mathbf{E}|$ represents the magnitude of $\mathbf{E}$, $\epsilon^{(r)}$ denotes the conductivity of phase r, and the exponent $m > 0$ characterizes the degree of nonlinearity. For simplicity, the exponent m is taken to be the same for both phases. A weakly nonlinear response is reproduced by taking $m = 1 + \delta$ and expanding the power-law potentials to first order in δ:

$$w^{(r)}(\mathbf{E}) = \frac{\epsilon^{(r)}}{2} E^2 + \delta \frac{\epsilon^{(r)}}{4} E^2 (\ln E^2 - 1) + O(\delta^2). \tag{8}$$

This expression is indeed of the form (3) with

$$w_0^{(r)}(\mathbf{E}) = \frac{\epsilon^{(r)}}{2} E^2 \quad \text{and} \quad w^{(r)}(\mathbf{E}) = \frac{\epsilon^{(r)}}{4} E^2 (\ln E^2 - 1). \tag{9}$$

Now, it is well known that when both phases exhibit the same nonlinear exponent m, the effective potential is also of the power-law type with the same nonlinear exponent m (see, for instance, [40]). In view of the overall (in-plane) isotropy of the assemblage, we can thus write

$$\widetilde{w}(\overline{\mathbf{E}}) = \frac{\widetilde{\epsilon}}{m+1} \overline{E}^{m+1}, \tag{10}$$

where $\overline{E} = |\overline{\mathbf{E}}|$. The effective conductivity $\widetilde{\epsilon}$ depends on the local conductivities $\epsilon^{(r)}$, the microstructural morphology, and, more importantly, it also depends on the exponent m. Thus,

$$\widetilde{w}(\overline{\mathbf{E}}) = \frac{\widetilde{\epsilon}_0}{2}\overline{E}^2 + \delta\frac{\overline{E}^2}{4}[2\widetilde{\epsilon}_1 + \widetilde{\epsilon}_0(\ln\overline{E}^2 - 1)] + O(\delta^2), \tag{11}$$

where the constants $\widetilde{\epsilon}_0$ and $\widetilde{\epsilon}_1$ are such that $\widetilde{\epsilon} = \widetilde{\epsilon}_0 + \delta\,\widetilde{\epsilon}_1 + O(\delta^2)$, and characterize completely the effective response. Expressions (6), (9) and (11) imply that

$$\widetilde{\epsilon}_0\overline{E}^2 = \langle\epsilon(\mathbf{x})E_0^2\rangle \quad\text{and}\quad 2\overline{E}^2[\widetilde{\epsilon}_1 + \widetilde{\epsilon}_0\ln\overline{E}] = \langle\epsilon(\mathbf{x})E_0^2\ln E_0^2\rangle, \tag{12}$$

which furnish the effective constants $\widetilde{\epsilon}_0$ and $\widetilde{\epsilon}_1$ in terms of the linear electric field distribution. Within any composite cylinder of the assemblage occupying a domain Ω_c, the linear electric potential is solution to the field equations (see, for instance, [41])

$$-\nabla\cdot[\epsilon(\mathbf{x})\nabla\varphi_0] = 0 \quad\text{in }\Omega_c, \quad \varphi_0 = -\overline{\mathbf{E}}\cdot\mathbf{x} \quad\text{on }\partial\Omega_c, \quad \varphi_0 \in H^1(\Omega_c), \tag{13}$$

where $\epsilon(\mathbf{x}) = \chi^{(1)}(\mathbf{x})\epsilon^{(1)} + \chi^{(2)}(\mathbf{x})\epsilon^{(2)}$. In terms of a polar coordinate system with origin at the center of the cylinder and polar axis colinear with the applied electric field $\overline{\mathbf{E}}$, the solution to these equations can be written as (see, for instance, [41])

$$\varphi_0(\mathbf{x}) = \begin{cases} \overline{E}\,a_1 r\cos\theta & 0 \leq r \leq r_i \text{ (inclusion phase)} \\ \overline{E}\left(a_2 r + \dfrac{b_2}{r}\right)\cos\theta & r_i \leq r \leq r_o \text{ (matrix phase)}, \end{cases} \tag{14}$$

where r and θ are, respectively, the radial and angular coordinates, r_i and r_o are, respectively, the inner and outer radii of the matrix coating, and a_1, a_2, b_2 are constants given by

$$a_1 = -\frac{2\epsilon^{(2)}}{(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})}, \tag{15}$$

$$a_2 = -\frac{(\epsilon^{(1)} + \epsilon^{(2)})}{(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})}, \tag{16}$$

$$b_2 = \frac{(\epsilon^{(1)} - \epsilon^{(2)})}{(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})}r_i^2. \tag{17}$$

In these expressions, $c = (r_i/r_o)^2$ denotes the volume fraction of inclusions. The linear electric field intensity is thus given by

$$E_0^2(\mathbf{x}) = |\nabla\varphi_0(\mathbf{x})|^2 = \overline{E}^2 \times \begin{cases} a_1^2 & 0 \leq r \leq r_i \\ \left(a_2 - \dfrac{b_2}{r^2}\right)^2\cos^2\theta + \left(a_2 + \dfrac{b_2}{r^2}\right)^2\sin^2\theta & r_i \leq r \leq r_o. \end{cases} \tag{18}$$

Introducing the field distribution (18) in $(12)_1$, we have that

$$\widetilde{\epsilon}_0 = \frac{1}{\pi r_o^2}\int_0^{r_i}\int_0^{2\pi}\epsilon^{(1)}a_1^2 r\,\mathrm{d}r\,\mathrm{d}\theta + \frac{1}{\pi r_o^2}\int_{r_i}^{r_o}\int_0^{2\pi}\epsilon^{(2)}\left[\left(a_2 - \frac{b_2}{r^2}\right)^2\cos^2\theta + \left(a_2 + \frac{b_2}{r^2}\right)^2\sin^2\theta\right]r\,\mathrm{d}r\,\mathrm{d}\theta, \tag{19}$$

and upon integration we obtain

$$\widetilde{\epsilon}_0 = \epsilon^{(2)}\frac{(\epsilon^{(1)} + \epsilon^{(2)}) + c(\epsilon^{(1)} - \epsilon^{(2)})}{(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})}, \tag{20}$$

which is the effective conductivity of the linear CCA [5, 41].

In turn, introducing the field distribution (18) in (12)$_2$, we have that

$$2\widetilde{\epsilon}_1 + \widetilde{\epsilon}_0 \ln \overline{E}^2 = \frac{1}{\pi r_o^2} \int_0^{r_i} \int_0^{2\pi} \epsilon^{(1)} a_1^2 (\ln a_1^2 + \ln \overline{E}^2) \, r \, dr \, d\theta$$
$$+ \frac{1}{\pi r_o^2} \int_{r_i}^{r_o} \int_0^{2\pi} \epsilon^{(2)} \left[\left(a_2 - \frac{b_2}{r^2} \right)^2 \cos^2 \theta + \left(a_2 + \frac{b_2}{r^2} \right)^2 \sin^2 \theta \right]$$
$$\times \left(\ln \left[\left(a_2 - \frac{b_2}{r^2} \right)^2 \cos^2 \theta + \left(a_2 + \frac{b_2}{r^2} \right)^2 \sin^2 \theta \right] + \ln \overline{E}^2 \right) r \, dr \, d\theta, \qquad (21)$$

which in view of (19) simplifies to

$$\widetilde{\epsilon}_1 = \frac{1}{\pi r_o^2} \int_0^{r_i} \int_0^{2\pi} \frac{\epsilon^{(1)}}{2} a_1^2 \ln a_1^2 \, r \, dr \, d\theta + \frac{1}{\pi r_o^2} \int_{r_i}^{r_o} \int_0^{2\pi} \frac{\epsilon^{(2)}}{2} \left[\left(a_2 - \frac{b_2}{r^2} \right)^2 \cos^2 \theta + \left(a_2 + \frac{b_2}{r^2} \right)^2 \sin^2 \theta \right]$$
$$\times \ln \left[\left(a_2 - \frac{b_2}{r^2} \right)^2 \cos^2 \theta + \left(a_2 + \frac{b_2}{r^2} \right)^2 \sin^2 \theta \right] r \, dr \, d\theta, \qquad (22)$$

and upon integration yields (see Appendix A for details)

$$\widetilde{\epsilon}_1 = c \epsilon^{(1)} a_1^2 \ln |a_1| + (1-c) \epsilon^{(2)} \left[a_2^2 \ln |a_2| + c(1 + \ln |a_2|) \left(\frac{b_2}{r_i^2} \right)^2 \right]. \qquad (23)$$

Replacing the various constants by their expressions (15)–(17), we finally obtain

$$\widetilde{\epsilon}_1 = \widetilde{\epsilon}_0 \ln \left[\frac{(\epsilon^{(1)} + \epsilon^{(2)})}{(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})} \right]$$
$$+ \frac{c \epsilon^{(2)}}{[(\epsilon^{(1)} + \epsilon^{(2)}) - c(\epsilon^{(1)} - \epsilon^{(2)})]^2} \left(4 \epsilon^{(1)} \epsilon^{(2)} \ln \left[\frac{2 \epsilon^{(2)}}{\epsilon^{(1)} + \epsilon^{(2)}} \right] + (1-c)(\epsilon^{(1)} - \epsilon^{(2)})^2 \right). \qquad (24)$$

For later reference, it is noted that in the limiting case of perfectly insulating inclusions ($\epsilon^{(1)} = 0$), the exact results (3) and (24) simplify to

$$\frac{\widetilde{\epsilon}_0}{\epsilon^{(2)}} = \frac{1-c}{1+c} \quad \text{and} \quad \frac{\widetilde{\epsilon}_1}{\epsilon^{(2)}} = \frac{1-c}{1+c} \left[\frac{c}{1+c} - \ln(1+c) \right]. \qquad (25)$$

4. Infinite-rank laminates with power-law dissipation

It is well known that material systems with disparate classes of underlying microgeometries are most likely to exhibit different effective responses when the local behavior is nonlinear, even if exhibiting coincident effective responses when the local behavior is linear. Motivated by this general rule, we consider here material systems with two-dimensional microgeometries belonging to a fairly rich class confected in [15]. These microgeometries are of "particulate" type and are constructed by resorting to a differential scheme in combination with two-phase sequential laminations of infinite rank, and are such that when the constituent phases and the inclusion distribution are isotropic, and the local constitutive response is linear, their effective conductivity agrees exactly with that of the CCAs considered in the previous section. More generally, the effective dissipation potential of these infinite-rank laminates is given by

$$\widetilde{w}(\overline{\mathbf{E}}) = \widehat{w}(\overline{\mathbf{E}}, -\ln c), \qquad (26)$$

where $c = c^{(2)}$ is the volume fraction of the inclusion phase, and $\widehat{w}(\overline{\mathbf{E}}, t)$ is solution to the Hamilton–Jacobi equation

$$\frac{\partial \widehat{w}}{\partial t}(\overline{\mathbf{E}}, t) + \widehat{w} + H \left(\overline{\mathbf{E}}, \frac{\partial \widehat{w}}{\partial \overline{\mathbf{E}}} \right) = 0 \quad \text{with } \widehat{w}(\overline{\mathbf{E}}, 0) = w^{(1)}(\overline{\mathbf{E}}) \qquad (27)$$

and Hamiltonian

$$H(\overline{\mathbf{E}}, \overline{\mathbf{J}}) = \max_{a(\mathbf{n})} \langle a \mathbf{n} \cdot \overline{\mathbf{J}} - w^{(2)}(\overline{\mathbf{E}} + a\mathbf{n}) \rangle_v. \qquad (28)$$

In this last expression, $\langle \cdot \rangle_v = \int_{|\mathbf{n}|=1} (\cdot) v(\mathbf{n}) \, \mathrm{d}s(\mathbf{n})$ is an orientational average weighted by the reduced H-measure $v(\mathbf{n})$ of the microgeometry. For statistically isotropic composites with power-law dissipation potentials, this formalism generates an exact result for the effective conductivity given by (see [42])

$$\widetilde{\epsilon} = \widehat{\epsilon}(-\ln c), \tag{29}$$

where $\widehat{\epsilon}(t)$ is solution to the ordinary differential equation

$$\frac{\mathrm{d}\widehat{\epsilon}}{\mathrm{d}t} + h(\widehat{\epsilon}) = 0 \quad \text{with } \widehat{\epsilon}(0) = \epsilon^{(1)} \tag{30}$$

and

$$h(z) = z + \frac{1}{2\pi} \int_0^{2\pi} ((m+1)z \, a_*(\theta, z) \cos\theta - \epsilon^{(2)} [1 + 2a_*(\theta, z) \cos\theta + a_*(\theta, z)^2]^{(m+1)/2}) \, \mathrm{d}\theta. \tag{31}$$

In this last expression, the function $a_*(\theta, z)$ is solution to the optimality condition

$$z \cos\theta - \epsilon^{(2)} (a_*(\theta, z) + \cos\theta)(1 + 2a_*(\theta, z) \cos\theta + a_*(\theta, z)^2)^{(m-1)/2} = 0. \tag{32}$$

This differential equation can be written in integral form as

$$\int_{\epsilon^{(1)}}^{\widetilde{\epsilon}} \frac{\mathrm{d}z}{h(z)} = \ln c. \tag{33}$$

We now set $m = 1 + \delta$ and $\widetilde{\epsilon} = \widetilde{\epsilon}_0 + \delta \, \widetilde{\epsilon}_1 + O(\delta^2)$, and proceed with the expansion of (33) to first order in δ:

$$\int_{\epsilon^{(1)}}^{\widetilde{\epsilon}_0 + \delta \, \widetilde{\epsilon}_1 + O(\delta^2)} \frac{\mathrm{d}z}{h_0(z) + \delta \, h_1(z) + O(\delta^2)} = \int_{\epsilon^{(1)}}^{\widetilde{\epsilon}_0} \frac{\mathrm{d}z}{h_0(z)} + \delta \left[\frac{\widetilde{\epsilon}_1}{h_0(\widetilde{\epsilon}_0)} - \int_{\epsilon^{(1)}}^{\widetilde{\epsilon}_0} \frac{h_1(z)}{h_0(z)^2} \, \mathrm{d}z \right] + O(\delta^2) = \ln c, \tag{34}$$

where use has been made of Leibniz's rule; thus,

$$\int_{\epsilon^{(1)}}^{\widetilde{\epsilon}_0} \frac{\mathrm{d}z}{h_0(z)} = \ln c \quad \text{and} \quad \widetilde{\epsilon}_1 = h_0(\widetilde{\epsilon}_0) \int_{\epsilon^{(1)}}^{\widetilde{\epsilon}_0} \frac{h_1(z)}{h_0(z)^2} \, \mathrm{d}z. \tag{35}$$

The first identity furnishes the linear conductivity $\widetilde{\epsilon}_0$, while the second identity furnishes the first-order correction $\widetilde{\epsilon}_1$. These identities require the expansion of the integrand $h(z)$, which in turn requires the expansion of the function $a_*(\theta)$ as given by (32). The latter is given by

$$a_*(\theta, z) = \frac{z - \epsilon^{(2)}}{\epsilon^{(2)}} \cos\theta - \delta \frac{z}{\epsilon^{(2)}} \ln \left[\sin^2\theta + \frac{z}{\epsilon^{(2)}} \cos^2\theta \right] \cos\theta + O(\delta^2), \tag{36}$$

while the former is given by

$$h_0(z) = \frac{z^2 - \epsilon^{(2)2}}{2\epsilon^{(2)}} \quad \text{and} \quad h_1(z) = \frac{z^2 - \epsilon^{(2)2}}{4\epsilon^{(2)}} - \frac{z^2 + \epsilon^{(2)2}}{2\epsilon^{(2)}} \ln \left(\frac{z + \epsilon^{(2)}}{2\epsilon^{(2)}} \right). \tag{37}$$

In this last expression, use has been made of the integrals (49). Introducing $(37)_1$ into $(35)_1$ and evaluating the integral we obtain

$$\frac{\widetilde{\epsilon}_0 - \epsilon^{(2)}}{\widetilde{\epsilon}_0 + \epsilon^{(2)}} \frac{\epsilon^{(1)} + \epsilon^{(2)}}{\epsilon^{(1)} - \epsilon^{(2)}} = c. \tag{38}$$

The solution to this equation for the linear effective conductivity $\widetilde{\epsilon}_0$ agrees exactly with (3). In turn, introducing (37) and (38) into $(35)_2$ and evaluating the integral we obtain an expression that agrees exactly with expression (24) for $\widetilde{\epsilon}_1$. In conclusion, these infinite-rank laminates exhibit the same effective response as the CCAs of the previous section not only when the phases exhibit linear behavior but also when the phases exhibit weakly nonlinear behavior of the power-law type (to first order in the nonlinearity perturbation parameter δ). The fact that the same weakly nonlinear conductivity is displayed by the infinite-rank laminates as for the CCAs suggests that the effective parameter $\widetilde{\epsilon}_1$ may be only modestly sensitive to microgeometrical details (as is the case for the effective parameter $\widetilde{\epsilon}_0$ for linear composites).

5. Estimates for weakly nonlinear composites based on linear comparison composites

This section provides the relevant formulas for the various nonlinear homogenization estimates considered in this work, after being specialized to weakly nonlinear, power-law, composite conductors in two dimensions. In all cases, the effective properties of the LCC are determined via the estimates of [5] for two-phase composite conductors (specialized to 2D). Therefore, all the nonlinear estimates recover the exact conductivity of the CCA of the previous section when the phases exhibit a linear response.

First, it is recalled that variational (VAR) bounds of the HS type for two-phase nonlinear conductors have been given by Ponte Castañeda [27]. Expressions for the VAR bounds for weakly nonlinear composites were obtained by Ponte Castañeda [43] for general nonlinear correction $w^{(r)}$, which can be specialized for power-law conductors by means of expression $(9)_2$ for $w^{(r)}$. The form of the resulting correction $\widetilde{\epsilon}_1$ for general two-phase composites is somewhat too cumbersome to include here and it will simply be noted that the result does not recover the exact result for the CCA given by expression (24), although of course it still provides a bound. For the special case when the inclusions are assumed to be perfectly insulating ($\epsilon^{(1)} \to 0$), the result for the effective nonlinear conductivity (for power-law behavior) simplifies considerably to the expression

$$\frac{\widetilde{\epsilon}^{\text{VAR}}}{\epsilon^{(2)}} = \frac{1-c}{(1+c)^{(m+1)/2}},$$ (39)

while the corresponding result for the weakly nonlinear correction reduces to

$$\frac{\widetilde{\epsilon}_1^{\text{VAR}}}{\epsilon^{(2)}} = -\frac{1}{2}\frac{1-c}{1+c}\ln(1+c),$$ (40)

which is indeed seen to be different from the corresponding exact expression $(25)_2$ for the CCA.

Next, it is recalled that the TSO estimates for nonlinear composite conductors were given by Ponte Castañeda [44]. Two different results were obtained depending on whether one starts with the primal formulation ($\bar{w}$), as given by (2), or by the dual formulation obtained by means of the Legendre transformation ($\bar{u} = \bar{w}^*$). The result for the primal formulation ($\bar{w}$) is given by

$$\widetilde{\epsilon}^{\text{TSO}} = \sum_{r=1}^{2} c^{(r)}\epsilon^{(r)}\left[E^{(r)(1+m)} + \frac{1+m}{2}E^{(r)m}(1-E^{(r)})\right],$$ (41)

where $cE^{(1)} + (1-c)E^{(2)} = 1$ and $E^{(2)}$ is solution to

$$1 - \frac{\epsilon^{(1)}}{\epsilon^{(2)}}\frac{1-(1-c)E^{(2)}}{(cE^{(2)})^m} = \frac{1-E^{(2)}}{cE^{(2)}}\sqrt{m}.$$ (42)

The solution to this equation admits the following asymptotic expansion in the small parameter $\delta = m - 1$:

$$E^{(2)} = \frac{\epsilon^{(1)}+\epsilon^{(2)}}{(\epsilon^{(1)}+\epsilon^{(2)})-c(\epsilon^{(1)}-\epsilon^{(2)})} + \delta\frac{c}{2}\frac{\epsilon^{(2)}-\epsilon^{(1)}-4\epsilon^{(1)}\ln\left[\frac{\epsilon^{(1)}+\epsilon^{(2)}}{2}\right]}{[(\epsilon^{(1)}+\epsilon^{(2)})-c(\epsilon^{(1)}-\epsilon^{(2)})]^2}\epsilon^{(2)} + O(\delta^2).$$ (43)

Upon introducing this expression into (41) and expanding in δ, it can be verified that the TSO estimate reproduces the exact correction $\widetilde{\epsilon}_1$ for the conductivity of the weakly nonlinear CCA, as given (24). The corresponding result for the dual formulation ($\bar{u}$) is similar in form to the above result for the primal ($\bar{w}$) formulation and will not be repeated here, for brevity. However, it should be noted that while the primal and dual TSO estimates are not identical (i.e., there is a duality gap), it can be shown that the dual formulation also leads to the exact result for the weakly nonlinear correction $\widetilde{\epsilon}_1$ of the CCA.

Finally, we consider the FOSO estimates of [36]. The FOSO method—like the VAR method— does not have a duality gap, but the final form of the estimate depends on the formulation (primal or dual). In fact, the initial form of the potentials for the LCC in the primal ($\bar{w}$) and dual ($\bar{u}$)

formulations were chosen by Ponte Castañeda [36] to be slightly inconsistent, for simplicity of the calculations, and as a consequence of this "nonsymmetric" choice of the LCC, the FOSO delivers two different sets of estimates depending on whether the derivation is based on electric field potentials (w), giving rise to the FOSO(W) formulation, or on the current density potentials (u), giving rise to the so-called FOSO(U) formulation. In spite of the inconsistency in the predictions of the two formulations, which is due to the lack of symmetry in the form of the energy potential of the LCC, it should be emphasized that both formulations are fully consistent and do not exhibit a duality gap. By using a more symmetric choice for the potential of the LCC, Furer and Ponte Castañeda [38] proposed a "symmetric" version of the FOSO method, which predicts the same effective response whether one starts from the primal ($\tilde{w}$) or dual ($\tilde{u}$) formulations. As will be seen below, all three formulations deliver the same estimates in the weakly nonlinear limit.

Since the expressions for the different versions of the FOSO estimates are a bit complicated, we will consider here only the case of perfectly insulating inclusions ($\epsilon^{(1)} \to 0$), which corresponds to the case of infinite heterogeneity contrast (recall that the results are exact to second order in the heterogeneity contrast) and should be representative of more general cases. The expressions for general nonlinearity are provided in Appendix B. As already mentioned, in the weakly nonlinear limit, all three estimates are identical to first order in $\delta = m - 1$ with correction given by

$$\frac{\widetilde{\epsilon}_1^{\text{FOSO}}}{\epsilon^{(2)}} = -\frac{1-c}{1+c}\ln(1+c) + \frac{(1-c)}{2(1+c)^2}[(1-\alpha)\widehat{j}^2\ln(\widehat{j}^2) + \alpha\,\breve{j}^2\ln(\breve{j}^2)], \tag{44}$$

where

$$\widehat{j}^2 = (1-\alpha)\left(1 + \sqrt{2c}\sqrt{\frac{\alpha}{1-\alpha}} + \frac{\alpha c}{1-\alpha}\right), \quad \breve{j}^2 = \alpha\left(1 - \sqrt{2c}\sqrt{\frac{1-\alpha}{\alpha}} + \frac{(1-\alpha)c}{\alpha}\right) \tag{45}$$

and where α corresponds to a certain weight factor usually set equal to $1/2$.

6. Results and discussion

The exact results derived in Section 3 for CCAs exhibiting weak power-law nonlinearity are now exploited to assess in more detail the capabilities of the various homogenization estimates of Section 5. (Since the corresponding results of Section 4 for the infinite-rank laminates are identical to the results for CCAs, it will not be necessary to refer to those results explicitly in this section, although the fact that they are identical should of course be kept in mind.) We begin by recalling that both versions (primal and dual) of the TSO estimates reproduce the exact result for any volume fraction and heterogeneity contrast. Thus, these estimates are found to be asymptotically exact not only for weakly heterogeneous systems—to second order in the contrast—but also for weakly nonlinear systems—to first order in the nonlinearity parameter $\delta = m - 1$. However recomforting, this observation is not necessarily expected to hold for more general weakly nonlinear systems.

We now focus on the VAR and FOSO estimates, which—unlike the TSO estimates—do not reproduce the exact nonlinear correction $\widetilde{\epsilon}_1$ on the effective conductivity. However, it is recalled that the VAR estimate provides a lower bound for $\widetilde{\epsilon}_1$, while the various versions of the FOSO reduce to the same identical result for $\widetilde{\epsilon}_1$ and depend on a certain weight factor α. Figure 1 provides plots of these estimates for the case of perfectly insulating inclusions, along with the exact result. For the FOSO estimates, three different values of the parameter α are considered ($\alpha = 0.25, 0.5, 0.75$). Part (a) displays plots of the nonlinear correction $\widetilde{\epsilon}_1$ as a function of the inclusion volume fraction c, normalized by the matrix conductivity $\epsilon^{(2)}$. Among all these estimates, the VAR estimate is found to be the least accurate. This is related to the fact that the VAR estimate actually provides rigorous bounds for the effective conductivity of all composites with statistically isotropic microstructures—including for the CCAs. However, as shown recently by Furer and
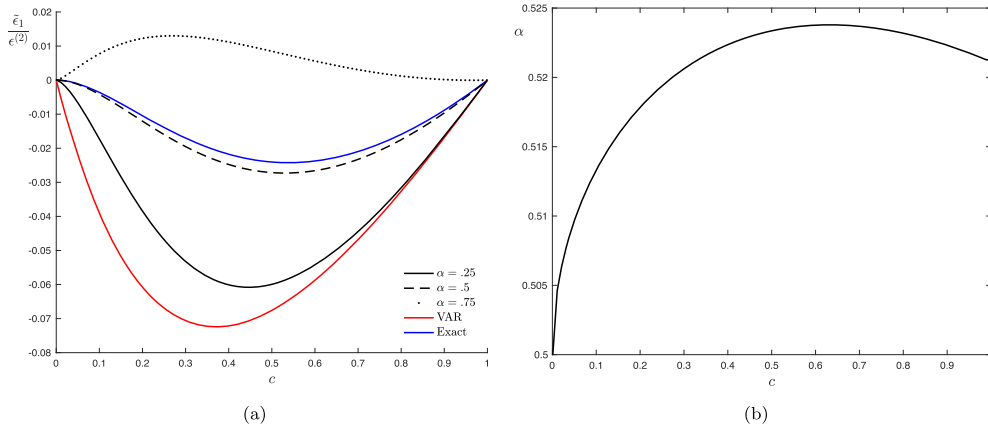
Figure 1. (a) Exact result and corresponding estimates for the weakly nonlinear correction $\widetilde{\epsilon}_1$ as a function of inclusion volume fraction c, normalized by the matrix conductivity $\epsilon^{(2)}$. (b) The value of α for which the FOSO estimate for $\widetilde{\epsilon}_1$ reproduces the exact result.

Ponte Castañeda [45], the VAR bounds also hold for microstructures that, while isotropic in the linear case, are anisotropic in the nonlinear case. Thus the VAR estimate provides an upper bound on the dissipation potential, which translates into a lower bound on the correction $\widetilde{\epsilon}_1$. The FOSO estimates are seen to comply with this bound for the three values of α considered. Interestingly, the FOSO predictions are seen to be asymmetric about $\alpha = 0.5$. Of the three values of α considered, $\alpha = 0.5$ is found to generate the most accurate predictions. Moreover, these predictions are quite close to the exact result for the entire range of inclusion volume fraction. A comparison of the analytical expressions for the exact result and the FOSO estimates with arbitrary α reveals that the FOSO estimate reproduces the exact result if α is chosen to satisfy the equation

$$
2c = (1-\alpha)\left(1 + \sqrt{2c}\sqrt{\frac{\alpha}{1-\alpha}} + \frac{\alpha c}{1-\alpha}\right)\ln\left(1 + \sqrt{2c}\sqrt{\frac{\alpha}{1-\alpha}} + \frac{\alpha c}{1-\alpha}\right)
$$
$$
+ \alpha\left(1 - \sqrt{2c}\sqrt{\frac{1-\alpha}{\alpha}} + \frac{(1-\alpha)c}{\alpha}\right)\ln\left(1 - \sqrt{2c}\sqrt{\frac{1-\alpha}{\alpha}} + \frac{(1-\alpha)c}{\alpha}\right). \tag{46}
$$

The solution to this equation is plotted in Figure 1(b) as a function of the volume fraction of inclusions c. While clearly dependent on c, this "optimal" value of α is seen to be very close to the value $\alpha = 0.5$ advocated by Furer and Ponte Castañeda [38] based on comparisons with other estimates. In fact, it can be verified that the value $\alpha = 0.5$ leads to the *exact* asymptotic result, $\widetilde{\epsilon}_1/\epsilon^{(2)} \sim -c^2/2$, in the dilute limit ($c \ll 1$). These observations for an infinitely contrasted system, and the fact that the FOSO estimates are exact for weakly contrasted systems to second order, suggest that the use of $\alpha = 0.5$ for general material systems provides a reasonable compromise between accuracy and simplicity.

Next, the influence of α on the FOSO predictions for strongly nonlinear power-law assemblages is considered. Thus, Figure 2 shows results for the effective conductivity $\widetilde{\epsilon}$, as a function of nonlinearly m, for a fixed value of $c = 0.2$. For comparison purposes, results are included for the three different versions of the FOSO methods—nonsymmetric FOSO(U) and FOSO(W) [36] and symmetric FOSO [38]—as well as the VAR bounds, the two versions of the TSO estimates (TSO(U) and TSO(W)) and the exact weakly nonlinear limit to first order in $\delta = m - 1$ (and therefore linear in m). The pairs of Figures 2(a) and (b) and Figures 2(c) and (d) show, respectively, the effective
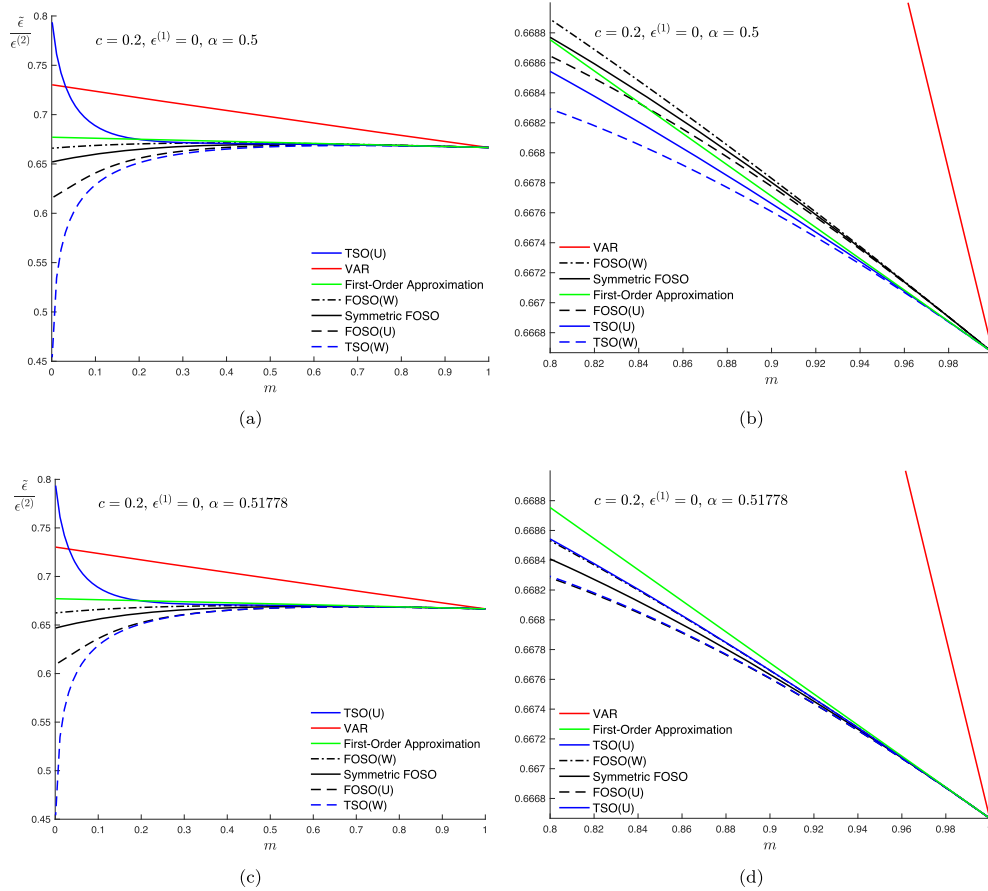
Figure 2. Estimates for the effective conductivity with α equal to (a, b) 0.5 and (c, d) the value for which the FOSO method gives the exact correction ($\alpha \approx 0.5178$). Results are presented as a function of the nonlinear m, for a fixed volume fraction $c = 0.2$ of the perfectly insulating phase.

conductivity for both the entire range of nonlinearities (left column), as well as zoomed-in versions for weak nonlinearities (right column), for $\alpha = 0.5$ (top row) and the value of α ($\alpha \approx 0.5178$) for which the FOSO estimate recovers the exact result in the weakly nonlinear limit (bottom row). It is noted that the order in which the different methods appear in each legend corresponds to the order in which the values predicted by the various methods at $m = 0$ also appear. In particular, the topmost entry in the legend is the method predicting the largest value of the effective conductivity at $m = 0$, while the bottom-most entry in the legend is the method predicting the smallest value at $m = 0$. As was remarked by Furer and Ponte Castañeda [38], the symmetric FOSO estimate always lies somewhere between the FOSO(U) and FOSO(W) estimates, with a maximum difference of about 5% between the symmetric and nonsymmetric versions for $m = 0$. Moreover, the effect of changing α on the various FOSO predictions is relatively minor; for example, as can be seen in Figures 2(a) and (c), the symmetric FOSO method predicts a value of the effective conductivity for $m = 0$ slightly above 0.65 when $\alpha = 0.5$, while it gives a value slightly below 0.65 when $\alpha \approx 0.5178$. On the other hand, the TSO estimates, which are independent of any weight factors, are very close for values of m between 0.5 and 1, but diverge dramatically in the strongly non-

linear limit as m approaches zero. In fact the TSO(U) violates the upper bound provided by the VAR estimates for values of m less than about 0.04. In addition, it is seen in Figure 2(a) that the FOSO(U) method and the TSO(W) method are rather close for values of $0.5 \leq m \leq 1$, while, in Figure 2(c), this agreement continues up to $m \approx 0.2$. In fact, the FOSO(U) estimate crosses above the TSO(W) estimate for $0.3 < m < 1$ in Figure 2(a) for $\alpha = 0.5$, but remains always below it in Figure 2(c) when $\alpha \approx 0.5178$.

The magnified results in Figures 2(b) and (d) show more clearly the effect of the choice of α on the comparisons of the FOSO estimates with the other estimates. As expected, the two TSO estimates, which give the exact weakly nonlinear behavior, lie tangent to the first-order approximation in the limit as $m \to 1$ in both figures. On the other hand, the various FOSO estimates only do so for $\alpha \approx 0.5178$, as can be seen in Figure 2(d). In fact, consistent with earlier observations for this value of α, it can also be seen in Figure 2(d) that the FOSO(U) and TSO(W) estimates appear to agree, as do the FOSO(W) and TSO(U) estimates, even for nonlinearities up to $m = 0.8$, again with the symmetric FOSO lying roughly in between the two sets of estimates. Finally, also consistent with earlier observations, all the estimates are seen to be significantly lower than the VAR bound. Again, this can be explained by the fact that the VAR bound must hold for certain nonlinearly anisotropic microstructures, including the finite-rank microstructures attaining the corresponding linear bounds [45].

7. Closing remarks

This paper provides exact asymptotic estimates for weakly nonlinear power-law composite conductors with CCA and infinite-rank laminated microstructures. It is known that both of these microstructures attain exactly the 2-D HS bounds [5], when the material behavior is linear. The objective of this work is to test more general estimates obtained by various nonlinear homogenization methods making use of corresponding estimates for LCCs against these exact results in order to assess their accuracy and capabilities. The first finding is that the TSO estimates of the HS type [44] agree to first order in the nonlinearity parameter with the exact asymptotic result. However, this surprising result has been found (details not presented in this paper) not to carry over to the corresponding 3-D conductivity results, and may therefore be little more than a coincidence. Indeed, it is known that the TSO method performs poorly for strongly nonlinear behavior, as a consequence of the fact that it neglects the effect of the field fluctuations which become very significant in the strongly nonlinear limit. In particular, the TSO estimates are known to exhibit a duality gap that increases from zero in the weakly nonlinear limit to very large values in the strong nonlinear limit.

For these reasons, improved FOSO estimates have been proposed [36], which make use of both the first and second moments of the field fluctuations in the linearization procedure, albeit at the expense of having to introduce certain weight factors α (corresponding to weights of the multiple stationary points of the error function in the LCC methods) in order to ensure that the resulting estimates do not exhibit duality gaps. Nonetheless, they have the added advantage that they allow the estimation of the macroscopic behavior, as well as of the first and second moments of the field statistics, directly from the corresponding estimates for the LCC. The second important finding of this work is that the FOSO estimates depend on the weight factors even in the weakly nonlinear limit. However, the FOSO can be made to be consistent with the weakly nonlinear limit for an appropriate choice of the weight factor α. More importantly, the resulting value for α, at least in this very special case, is found to be very close to $1/2$, which is the value initially proposed by Ponte Castañeda [36], and found to provide reasonably accurate estimates also by Furer and Ponte Castañeda [38] in other situations. In fact, the choice $\alpha = 1/2$ for the FOSO delivers the exact result for the weakly nonlinear correction to the effective conductivity

in the limit of dilute concentrations of perfectly conducting particles. Consequently, the choice of 1/2 for the weight factors α is recommended more generally, at least until this issue is better understood (see also [46]).

The third significant finding is that the VAR bounds of the HS type [27]—while still rigorous bounds for weakly nonlinear composites—are not exact to first order in the nonlinearity parameter. However, this result is not surprising in view of recent findings by Furer and Ponte Castañeda [45] showing that the VAR bound of the HS type, which makes use of the HS bounds for the corresponding LCC, must include microstructures that, while isotropic for linear material behavior, can be anisotropic when used for nonlinear composites. In fact, it was shown by Furer and Ponte Castañeda [45] that the same finite-rank sequential laminates that have been shown to attain the HS bounds in the linear case [2], also attain the VAR bounds of the HS type for certain special loadings.

Finally, it was also shown that the estimates of [15] for infinite-rank sequential laminates coincide with the corresponding exact results for the weakly nonlinear CCA microstructures—to first order in the nonlinearity parameter. In fact, we have recently verified that this is also true in 3D conductivity, where the effective properties of the composite sphere assemblage (CSA) and isotropic infinite-rank laminates agree to first order in the nonlinearity parameter. (Details are not shown here for brevity.) In any case, this final result seems to suggest that the effective behavior of weakly nonlinear composites should be relatively insensitive to the specific type of particulate microstructure involved, just as for linear composites where the CCA/CSA and sequential laminates (of various ranks) lead to identical macroscopic response.

Acknowledgments

Appendix A. Integration in the weakly nonlinear constant (22)

The integration over the angular variable θ in the second term of the right-hand side of (22) requires the evaluation of

$$\int_0^{2\pi} \left[\left(a_2 - \frac{b_2}{r^2}\right)^2 \cos^2\theta + \left(a_2 + \frac{b_2}{r^2}\right)^2 \sin^2\theta \right] \ln\left[\left(a_2 - \frac{b_2}{r^2}\right)^2 \cos^2\theta + \left(a_2 + \frac{b_2}{r^2}\right)^2 \sin^2\theta \right] \mathrm{d}\theta. \quad (47)$$

Making use of standard identities for squared trigonometric functions in the argument of the logarithm, this expression can be written as

$$\left(a_2 - \frac{b_2}{r^2}\right)^2 \int_0^{2\pi} \cos^2\theta \ln\left[a_2^2 - 2a_2 \frac{b_2}{r^2}\cos(2\theta) + \left(\frac{b_2}{r^2}\right)^2 \right] \mathrm{d}\theta$$

$$+ \left(a_2 + \frac{b_2}{r^2}\right)^2 \int_0^{2\pi} \sin^2\theta \ln\left[a_2^2 - 2a_2 \frac{b_2}{r^2}\cos(2\theta) + \left(\frac{b_2}{r^2}\right)^2 \right] \mathrm{d}\theta. \quad (48)$$

The two integrals in this expression can be evaluated by noting that for $r_i \le r \le r_o$ the ratio $|b_2/r^2/a_2| < 1$, see expressions (16) and (17), and by invoking the integrals (see integrals 4.397-14 4.397-15 in [47])

$$\int_0^{\pi/2} \cos^2\theta \ln\left[1 + 2a\cos(2\theta) + a^2 \right] \mathrm{d}\theta = -\int_0^{\pi/2} \sin^2\theta \ln\left[1 + 2a\cos(2\theta) + a^2 \right] \mathrm{d}\theta = \frac{\pi a}{4} \quad (49)$$

for $|a| < 1$. Thus, the integral (48) is

$$2\pi \left[a_2^2 + \left(\frac{b_2}{r^2}\right)^2 \right] \ln a_2^2 + 4\pi \left(\frac{b_2}{r^2}\right)^2. \quad (50)$$

In view of this last expression, the second term of the right-hand side of (22) is given by

$$
\begin{aligned}
\frac{\epsilon^{(2)}}{2} \frac{1}{\pi r_o^2} & \int_{r_i}^{r_o} 2\pi \left\{ \left[a_2^2 + \left(\frac{b_2}{r^2} \right)^2 \right] \ln a_2^2 + 2 \left(\frac{b_2}{r^2} \right)^2 \right\} r \, dr \\
&= \frac{\epsilon^{(2)}}{2} \int_c^1 \left\{ \left[a_2^2 + \left(\frac{b_2}{r_o^2} \right)^2 z^{-2} \right] \ln a_2^2 + 2 \left(\frac{b_2}{r_o^2} \right)^2 z^{-2} \right\} dz \\
&= \epsilon^{(2)} (1-c) \left[a_2^2 \ln|a_2| + c(1 + \ln|a_2|) \left(\frac{b_2}{r_i^2} \right)^2 \right].
\end{aligned}
\tag{51}
$$

Appendix B. Fully optimized second-order estimates for power-law conductors with perfectly insulating inclusions

For the case of perfectly insulating inclusions considered here, the dual ($\tilde{u}$) formulation is the easiest to work with and therefore we start with the FOSO(U) version. We then give the corresponding results for the symmetric FOSO version, which is similar in form. Finally, the results of the FOSO(W) version, which makes use of the more complex primal ($\tilde{w}$) formulation, is given.

FOSO(U) version. Converting the results of [36] for 2-D viscoplastic composites with porous inclusions to the mathematically analogous case of 2-D power-law conductors with perfectly insulating inclusions, it is found that the effective conductivity $\tilde{\epsilon}^{\mathrm{FOSO}}$, defined by (10), is given by

$$
\begin{aligned}
\frac{\tilde{\epsilon}^{\mathrm{FOSO}}}{\epsilon^{(2)}} &= (1-c) \left[\operatorname*{stat}_k \{ (1-\alpha) \hat{j}^{n+1} + \alpha \check{j}^{n+1} \} \right]^{-1/n} \\
&= (1-c) \left[\operatorname*{stat}_k f(k(n), n) \right]^{-1/n},
\end{aligned}
\tag{52}
$$

where $n = 1/m$ and

$$
\hat{j} = \sqrt{\hat{j}_\parallel^2 + \hat{j}_\perp^2}, \quad \check{j} = \sqrt{\check{j}_\parallel^2 + \check{j}_\perp^2},
$$
$$
\hat{j}_\parallel = 1 + \sqrt{\frac{\alpha}{1-\alpha}} \sqrt{\frac{c}{2}} k^{1/4}, \quad \check{j}_\parallel = 1 - \sqrt{\frac{1-\alpha}{\alpha}} \sqrt{\frac{c}{2}} k^{1/4},
$$
$$
\hat{j}_\perp = \sqrt{\frac{\alpha}{1-\alpha}} \sqrt{\frac{c}{2}} k^{-1/4}, \quad \check{j}_\perp = -\sqrt{\frac{1-\alpha}{\alpha}} \sqrt{\frac{c}{2}} k^{-1/4}.
\tag{53}
$$

In addition, $k = k(n)$ is the anisotropy ratio of the LCC, and the optimal value k^* is determined as a solution to the equation

$$
\frac{\partial f(k^*(n), n)}{\partial k} = 0.
\tag{54}
$$

Then, letting $k = k^*(n)$, as determined by the stationarity condition (54), and making use of the fact that $\partial/\partial m = -m^{-2}(\partial/\partial n)$, where $n = 1/m$, it is found that

$$
\begin{aligned}
\frac{\tilde{\epsilon}_1^{\mathrm{FOSO}}}{\epsilon^{(2)}} &= \frac{\partial}{\partial m} \frac{\tilde{\epsilon}^{\mathrm{FOSO}}}{\epsilon^{(2)}} = (1-c) \frac{\partial}{\partial m} [f(k^*(n), n)]^{-1/n} \\
&= -\frac{(1-c)}{m^2} \frac{\partial}{\partial n} [f(k^*, n)]^{-1/n} \\
&= -\frac{(1-c)}{m^2} [f(k^*, n)]^{-1/n} \left[\frac{\log(f(k^*, n))}{n^2} \right. \\
&\qquad \left. -\frac{1}{nf(k^*, n)} \left(\frac{\partial f(k^*, n)}{\partial k} \frac{\partial k^*}{\partial n} + \frac{\partial f(k^*, n)}{\partial n} \right) \right].
\end{aligned}
\tag{55}
$$

Note that the derivative with respect to n in the last line is taken with k^* held fixed, while the term $(\partial f(k^*, n))/\partial k$ vanishes on account of (54). Using the fact that $k^*(1) = 1$ and $f(1, 1) = (1 + c)$, it follows that

$$\frac{\widetilde{\epsilon}_1^{\text{FOSO}}}{\epsilon^{(2)}} = \frac{\partial}{\partial m} \left. \frac{\widetilde{\epsilon}^{\text{FOSO}}}{\epsilon^{(2)}} \right|_{m=1} = \frac{(1-c)}{(1+c)^2} [(1-\alpha)\widehat{j}^2 \log(\widehat{j}) + \alpha \breve{j}^2 \log(\breve{j})] - \frac{1-c}{1+c} \log(1+c), \quad (56)$$

where $\widehat{j}$ and $\breve{j}$ are given as in (53), with $k = 1$.

Symmetric FOSO version. The symmetric FOSO version is also given (see [38] for details) by an expression of the form (52), with $\widehat{j}_\parallel$ and $\breve{j}_\parallel$ as given in (53), but with $\widehat{j}_\perp$ and $\breve{j}_\perp$ obtained as the solution of equations

$$\widehat{j}_\perp \breve{j}_\perp ((\widehat{j}_\parallel - \breve{j}_\parallel) + k(1 - 2\alpha)(\widehat{j}_\parallel + \breve{j}_\parallel)) - 2k((1-\alpha)\breve{j}_\parallel \widehat{j}_\perp^2 - \alpha \widehat{j}_\parallel \breve{j}_\perp^2) = 0, \quad (57)$$

$$4\alpha(1-\alpha)\widehat{j}_\perp \breve{j}_\perp + (1 - 2\alpha)\left((1-\alpha)\widehat{j}_\perp^2 - \alpha \breve{j}_\perp^2\right) + \frac{c}{2\sqrt{k}} = 0, \quad (58)$$

and chosen in just a way that $\breve{j}_\perp < \widehat{j}_\perp$. Then, following the same procedure as above, and noting that in this context, when $k = 1$, the expressions for $\widehat{j}_\perp$ and $\breve{j}_\perp$ using the symmetric FOSO method is the same as those for the FOSO(U) method, we conclude that they will give the same first-order correction, given by (56).

FOSO(W) version. The implementation of the FOSO(W) version makes use of the primal ($\widetilde{w}$) formulation, which we spell out next and is a bit more complicated, as the average electric field in the phases are unknown. Nonetheless, it can be shown that an estimate for the effective conductivity can equivalently be estimated via

$$\frac{\widetilde{\epsilon}^{\text{FOSO}}}{\epsilon^{(2)}} = (1-c) \operatorname*{stat}_{k, \overline{E}^{(2)}} \{(1-\alpha)\widehat{E}^{m+1} + \alpha \breve{E}^{m+1}\}$$

$$= (1-c) \operatorname*{stat}_{k, \overline{E}^{(2)}} g(k(m), \overline{E}^{(2)}(m), m), \quad (59)$$

where $\overline{E}^{(2)}$ is the average electric field in the matrix phase. The optimal values of k^* and $\overline{E}^{(2)*}$ are then determined via the equations

$$\frac{\partial g(k^*, \overline{E}^{(2)*}, m)}{\partial k} = 0,$$

$$\frac{\partial g(k^*, \overline{E}^{(2)*}, m)}{\partial \overline{E}^{(2)}} = 0. \quad (60)$$

As above, the form of $g(k, \overline{E}^{(2)}, m)$ is different depending on whether one uses the FOSO(W) method or the symmetric FOSO method. In the former case, we have

$$\widehat{E} = \sqrt{\widehat{E}_\parallel^2 + \widehat{E}_\perp^2}, \quad \breve{E} = \sqrt{\breve{E}_\parallel^2 + \breve{E}_\perp^2},$$

$$\widehat{E}_\parallel = \overline{E}^{(2)} - (\overline{E}^{(2)} - 1)\sqrt{\frac{\alpha}{1-\alpha}} \frac{k^{-1/4}}{\sqrt{2c}}, \quad \breve{E}_\parallel = \overline{E}^{(2)} + (\overline{E}^{(2)} - 1)\sqrt{\frac{1-\alpha}{\alpha}} \frac{k^{-1/4}}{\sqrt{2c}},$$

$$\widehat{E}_\perp = -(\overline{E}^{(2)} - 1)\sqrt{\frac{\alpha}{1-\alpha}} \frac{k^{1/4}}{\sqrt{2c}}, \quad \breve{E}_\perp = (\overline{E}^{(2)} - 1)\sqrt{\frac{1-\alpha}{\alpha}} \frac{k^{1/4}}{\sqrt{2c}}, \quad (61)$$

while in the latter case, $\widehat{E}_\parallel$ and $\breve{E}_\parallel$ are the same, while $\widehat{E}_\perp$ and $\breve{E}_\perp$ are again related through

$$\widehat{E}_\perp \breve{E}_\perp (k(\widehat{E}_\parallel - \breve{E}_\parallel) + (1 - 2\alpha)(\widehat{E}_\parallel + \breve{E}_\parallel)) - 2((1-\alpha)\breve{E}_\parallel \widehat{E}_\perp^2 - \alpha \widehat{E}_\parallel \breve{E}_\perp^2) = 0, \quad (62)$$

$$4\alpha(1-\alpha)\widehat{E}_\perp \breve{E}_\perp + (1 - 2\alpha)((1-\alpha)\widehat{E}_\perp^2 - \alpha \breve{E}_\perp^2) + (E^{(2)} - 1)^2 \frac{\sqrt{k}}{2c} = 0, \quad (63)$$

and chosen so that $\breve{E}_\perp < \widehat{E}_\perp$.

Once again, when $m = 1$, so that $k = 1$, the expressions are the same whether one uses the FOSO(W) or symmetric FOSO method. Moreover, using the fact that $\overline{E}^{(2)*}(1) = (1 + c)^{-1}$, it can be shown that the FOSO(W) version leads to the same correction $\widetilde{\epsilon}_1$ as the FOSO(U) version, as given by (56).

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