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Variational estimates for the effective response and field statistics in thermoelastic composites with intra-phase property fluctuations

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Abstract

Composites, Thermoelasticity, Variational methods. In this work, variational estimates are provided for the macroscopic response, as well as for the first and second moments of the stress and strain fields, in thermoelastic composites with non-uniform distributions of the thermal stress and elastic moduli in the constituent phases. These estimates are obtained in terms of a 'comparison composite' with uniform phase properties depending on the first and second moments of a certain combination of the given intra-phase thermal stresses and modulus field distributions. Under certain hypotheses, these estimates can be shown to lead to upper and lower bounds for the free energy of the composite, which reduce to standard results when the intra-phase fluctuations vanish. An illustrative application is given for rigidly reinforced composites with a non-uniform distribution of the thermal stress in the matrix phase.

1 Introduction

There are many problems of current interest which can be described as 'thermoelastic' composites, where the 'thermal' stress (also called 'eigenstress' or 'transformation stress' in the literature), or the elasticity tensor, or both, need not be uniform within a given phase. One standard example is provided by residual stresses in polycrystals (Noyan & Schadler (1994)), where due to the forming process, non-uniform distributions of the

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stresses may develop within the grains of the polycrystal, even when the applied macroscopic stress vanishes. A second example is provided by elasto-(visco)plastic composites (see Lahellec & Suquet (2007a)) where the (visco)plastic strain field may be heterogeneous in a given phase. As a final example, it is noted that the 'incremental' linearization scheme proposed by Hill (1965) for estimating the macroscopic behavior of plastic polycrystals involves the 'tangent' modulus of the grains, which is non-uniform in the grains, due to the inhomogeneity of the stress and strain fields that are produced in the grains of the polycrystal under loading.

Motivated by these examples, we consider a representative volume element V of a composite material made of different linear thermoelastic phases. The stress-strain relation in an individual phase reads as:

$$\boldsymbol{\sigma}(\boldsymbol{x}) = \boldsymbol{L}(\boldsymbol{x}) : \boldsymbol{\varepsilon}(\boldsymbol{x}) + \boldsymbol{\tau}(\boldsymbol{x}), \tag{1}$$

where both the elastic moduli $\boldsymbol{L}(\boldsymbol{x})$ and the polarization field $\boldsymbol{\tau}(\boldsymbol{x})$ depend on the position $\boldsymbol{x}$ and *are non uniform per phase.*

The question addressed in this study is to estimate the macroscopic behavior, as well as the first and the second moments of the strain and stress fields, $\boldsymbol{\varepsilon}$ and $\boldsymbol{\sigma}$, in a thermoelastic composite with phase constitutive response described by relation (1)–utilizing only partial statistical information about $\boldsymbol{L}$ and $\boldsymbol{\tau}$ in each phase (assumed to be statistically uniform in the composite). To the best of the authors' knowledge, the exact response to this question is not known in general. However it is known in the special case where the elastic moduli $\boldsymbol{L}$ and the polarization field $\boldsymbol{\tau}$ are uniform in each phase r, *i.e.* piecewise uniform through the entire volume element V. Quite naturally, our approach to the problem is to replace the initial thermoelastic composite with nonuniform moduli and nonuniform polarization field by a *linear (thermoelastic) comparison composite (LCC)* with piecewise uniform elastic moduli $\boldsymbol{L}_0^{(r)}$ and piecewise uniform polarization stress $\boldsymbol{\tau}_0^{(r)}$.

To this end, a very natural first choice is to replace the nonuniform fields $\boldsymbol{\tau}(\boldsymbol{x})$ and $\boldsymbol{L}(\boldsymbol{x})$ by their average, or first moment, over each individual phase:

$$\boldsymbol{L}_0^{(r)} = \langle \boldsymbol{L} \rangle^{(r)}, \quad \boldsymbol{\tau}_0^{(r)} = \langle \boldsymbol{\tau} \rangle^{(r)}. \tag{2}$$

However, even in the one-dimensional case where calculations can be carried out explicitly (see appendix), this simple choice does not yield the exact result for the second moment of the stress and strain fields.

The criterion adopted here to determine the improved estimates for the quantities $\boldsymbol{L}_0^{(r)}$ and $\boldsymbol{\tau}_0^{(r)}$ in the comparison composite is based on a variational approach for a 'comparison composite,' which follows closely the variational approach of Ponte Castañeda (1991) for nonlinear composites (see also Ponte Castañeda & Suquet (1998) and Ponte Castañeda & Willis (1999), and references therein).

2 Effective energies and elementary bounds

Select a volume element V of the nonhomogeneous material under consideration. Although neither the elastic moduli or the polarizations are piecewise uniform, we shall assume that there exists an underlying geometrical decomposition V in the form of N subdomains $V^{(r)}|_{r=1,..,N}$ which is a natural microstructure. An example of such a situation corresponds to a polycrystalline material where the grains are well identified, even though the plastic strain $\boldsymbol{\gamma}$ can be nonuniform even within each grain. The characteristic function and the volume fraction of $V^{(r)}$ are denoted by $\chi^{(r)}(\boldsymbol{x})$ and $c^{(r)}$, respectively. The angular brackets, $\langle . \rangle$ and $\langle . \rangle^{(r)}$, will be used to denote the spatial averages over V and $V^{(r)}$, respectively. Total and partial averages will be denoted by an upper bar:

$$\overline{\boldsymbol{\varepsilon}} = \langle \boldsymbol{\varepsilon} \rangle, \quad \overline{\boldsymbol{\varepsilon}}^{(r)} = \langle \boldsymbol{\varepsilon} \rangle^{(r)}$$

2.1 Effective potentials

The constitutive relation (1) derives from a (convex) thermoelastic energy w which reads at point $\boldsymbol{x}$ as:

$$w(\boldsymbol{x}, \boldsymbol{\varepsilon}) = \frac{1}{2}\boldsymbol{\varepsilon} : \boldsymbol{L}(\boldsymbol{x}) : \boldsymbol{\varepsilon} + \boldsymbol{\tau}(\boldsymbol{x}) : \boldsymbol{\varepsilon}. \tag{3}$$

The local stress-strain relation (1) can be inverted and derives from a dual potential denoted by u, conjugate to w in the sense of convex dual functions:

$$u(\boldsymbol{x}, \boldsymbol{\sigma}) = \sup_{\boldsymbol{\varepsilon}} \left\{ \boldsymbol{\sigma} : \boldsymbol{\varepsilon} - w(\boldsymbol{x}, \boldsymbol{\varepsilon}) \right\}. \tag{4}$$

A straightforward calculation shows that:

$$u(\boldsymbol{x}, \boldsymbol{\sigma}) = \frac{1}{2}\boldsymbol{\sigma} : \boldsymbol{M}(\boldsymbol{x}) : \boldsymbol{\sigma} + \gamma(\boldsymbol{x}) : \boldsymbol{\sigma} + \frac{1}{2}\gamma(\boldsymbol{x}) : \boldsymbol{L}(\boldsymbol{x}) : \gamma(\boldsymbol{x}), \tag{5}$$

where

$$\boldsymbol{M}(\boldsymbol{x}) = \boldsymbol{L}^{-1}(\boldsymbol{x}), \quad \gamma(\boldsymbol{x}) = -\boldsymbol{M}(\boldsymbol{x}) : \boldsymbol{\tau}(\boldsymbol{x}). \tag{6}$$

Although the elastic moduli and the polarizations are not piecewise uniform, it will be convenient to use a different notation for the restriction of w and u to $V^{(r)}$:

$$w^{(r)}(\boldsymbol{x}, \boldsymbol{\varepsilon}) = w(\boldsymbol{x}, \boldsymbol{\varepsilon})\chi^{(r)}(\boldsymbol{x}), \quad u^{(r)}(\boldsymbol{x}, \boldsymbol{\sigma}) = u(\boldsymbol{x}, \boldsymbol{\sigma})\chi^{(r)}(\boldsymbol{x}).$$

The effective potential, or energy function, corresponding to w is given by

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}}) = \inf_{\boldsymbol{\varepsilon} \ \in \mathcal{K}(\overline{\boldsymbol{\varepsilon}})} \langle w(\boldsymbol{x}, \boldsymbol{\varepsilon}) \rangle, \tag{7}$$

where $\mathcal{K}(\overline{\boldsymbol{\varepsilon}})$ is the set of fields $\boldsymbol{\varepsilon}(\boldsymbol{x})$ such that there exists a displacement field $\boldsymbol{v}(\boldsymbol{x})$ satisfying

$$\boldsymbol{\varepsilon}(\boldsymbol{x}) = \frac{1}{2}(\boldsymbol{\nabla v} + {}^{\mathsf{T}}\boldsymbol{\nabla v}) \text{ and } \boldsymbol{v}(\boldsymbol{x}) = \overline{\boldsymbol{\varepsilon}}.\boldsymbol{x} \ \text{ on } \partial V. \tag{8}$$

The complementary effective potential $\widetilde{u}$ is characterized as:

$$\widetilde{u}(\overline{\boldsymbol{\sigma}}) = \inf_{\boldsymbol{\sigma} \in \mathcal{S}(\overline{\boldsymbol{\sigma}})} \langle u(\boldsymbol{x}, \boldsymbol{\sigma}) \rangle, \tag{9}$$

where $\mathcal{S}(\overline{\boldsymbol{\sigma}})$ is the set of fields $\boldsymbol{\sigma}(\boldsymbol{x})$ such that

$$\operatorname{div}(\boldsymbol{\sigma}) = \boldsymbol{0}, \quad \langle \boldsymbol{\sigma} \rangle = \overline{\boldsymbol{\sigma}}.$$

Note that $\widetilde{u}$ and $\widetilde{w}$ are dual convex potentials (Suquet (1987) in the linear case and Willis (1989) in general) satisfying in particular:

$$\widetilde{u}(\overline{\boldsymbol{\sigma}}) = \sup_{\overline{\boldsymbol{\varepsilon}}} \left\{ \overline{\boldsymbol{\sigma}} : \overline{\boldsymbol{\varepsilon}} - \widetilde{w}(\overline{\boldsymbol{\varepsilon}}) \right\}. \tag{10}$$

Note also that the macroscopic stress $\overline{\boldsymbol{\sigma}}$ and macroscopic strain $\overline{\boldsymbol{\varepsilon}}$ are related by the constitutive relations

$$\overline{\boldsymbol{\sigma}} = \frac{\partial \widetilde{w}}{\partial \overline{\boldsymbol{\varepsilon}}} (\overline{\boldsymbol{\varepsilon}}), \quad \text{and} \quad \overline{\boldsymbol{\varepsilon}} = \frac{\partial \widetilde{u}}{\partial \overline{\boldsymbol{\sigma}}} (\overline{\boldsymbol{\sigma}}). \tag{11}$$

2.2 Elementary bounds

The Voigt and Reuss bounds correspond to the particular choice $\boldsymbol{\varepsilon} = \overline{\boldsymbol{\varepsilon}}$ in (7) and $\boldsymbol{\sigma} = \overline{\boldsymbol{\sigma}}$ in (9) respectively:

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}}) \leq \widetilde{w}^{\mathrm{Voigt}}(\overline{\boldsymbol{\varepsilon}}) = \frac{1}{2} \overline{\boldsymbol{\varepsilon}} : \langle \boldsymbol{L} \rangle : \overline{\boldsymbol{\varepsilon}} + \langle \boldsymbol{\tau} \rangle : \overline{\boldsymbol{\varepsilon}},$$

$$\widetilde{u}(\overline{\boldsymbol{\sigma}}) \leq \widetilde{u}^{\mathrm{Reuss}}(\overline{\boldsymbol{\sigma}}) = \frac{1}{2} \overline{\boldsymbol{\sigma}} : \langle \boldsymbol{M} \rangle : \overline{\boldsymbol{\sigma}} - \overline{\boldsymbol{\sigma}} : \langle \boldsymbol{M} : \boldsymbol{\tau} \rangle + \frac{1}{2} \langle \boldsymbol{\tau} : \boldsymbol{M} : \boldsymbol{\tau} \rangle.$$

A straightforward calculation gives the dual of $\widetilde{u}^{\mathrm{Reuss}}$ as:

$$\begin{aligned} \widetilde{w}^{\mathrm{Reuss}}(\overline{\boldsymbol{\varepsilon}}) &= \tfrac{1}{2} \overline{\boldsymbol{\varepsilon}} : \langle \boldsymbol{M} \rangle^{-1} : \overline{\boldsymbol{\varepsilon}} + \overline{\boldsymbol{\varepsilon}} : \langle \boldsymbol{M} \rangle^{-1} : \langle \boldsymbol{M} : \boldsymbol{\tau} \rangle \\ &+ \tfrac{1}{2} \langle \boldsymbol{M} : \boldsymbol{\tau} \rangle : \langle \boldsymbol{M} \rangle^{-1} : \langle \boldsymbol{M} : \boldsymbol{\tau} \rangle - \tfrac{1}{2} \langle \boldsymbol{\tau} : \boldsymbol{M} : \boldsymbol{\tau} \rangle. \end{aligned}$$

These bounds can be given a general form:

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}}) = \frac{1}{2} \overline{\boldsymbol{\varepsilon}} : \widetilde{\boldsymbol{L}} : \overline{\boldsymbol{\varepsilon}} + \widetilde{\boldsymbol{\tau}} : \overline{\boldsymbol{\varepsilon}} + \widetilde{f}(\boldsymbol{\tau}), \tag{12}$$

where for the Voigt bound:

$$\widetilde{\boldsymbol{L}} = \widetilde{\boldsymbol{L}}^{\mathrm{Voigt}} = \langle \boldsymbol{L} \rangle, \quad \widetilde{\boldsymbol{\tau}} = \widetilde{\boldsymbol{\tau}}^{\mathrm{Voigt}} = \langle \boldsymbol{\tau} \rangle, \quad \widetilde{f}(\boldsymbol{\tau}) = \widetilde{f}^{\mathrm{Voigt}}(\boldsymbol{\tau}) = 0, \tag{13}$$

and for the Reuss bound:

$$\left. \begin{aligned} \widetilde{\boldsymbol{L}}^{\mathrm{Reuss}} &= \langle \boldsymbol{M} \rangle^{-1}, \quad \widetilde{\boldsymbol{\tau}}^{\mathrm{Reuss}} = \langle \boldsymbol{M} \rangle^{-1} : \langle \boldsymbol{M} : \boldsymbol{\tau} \rangle, \\ \widetilde{f}^{\mathrm{Reuss}}(\boldsymbol{\tau}) &= \tfrac{1}{2} \langle \boldsymbol{\tau} : \boldsymbol{M} \rangle : \langle \boldsymbol{M} \rangle^{-1} : \langle \boldsymbol{M} : \boldsymbol{\tau} \rangle - \tfrac{1}{2} \langle \boldsymbol{\tau} : \boldsymbol{M} : \boldsymbol{\tau} \rangle. \end{aligned} \right\} \tag{14}$$

2.3 General structure of the exact energy

We now come back to the general problem

$$\boldsymbol{\sigma}(\boldsymbol{x}) = \boldsymbol{L}(\boldsymbol{x}) : \boldsymbol{\varepsilon}(\boldsymbol{x}) + \boldsymbol{\tau}(\boldsymbol{x}), \quad \mathrm{div}\,(\boldsymbol{\sigma}(\boldsymbol{x})) = 0, \quad \boldsymbol{\varepsilon} \in \mathcal{K}(\overline{\boldsymbol{\varepsilon}}). \tag{15}$$

The tensors $\overline{\boldsymbol{\varepsilon}}$ and $\boldsymbol{\tau}(\boldsymbol{x})$ being given, the solution $\boldsymbol{\varepsilon}$ of this problem can be obtained by the superposition principle. Consider first the case where $\boldsymbol{\tau}$ is identically $\mathbf{0}$. Problem (15) is then a standard elasticity problem and its solution can be expressed by means of the elastic strain-localization tensor $\boldsymbol{A}(\boldsymbol{x})$ as

$$\boldsymbol{\varepsilon}(\boldsymbol{x}) = \boldsymbol{A}(\boldsymbol{x}) : \overline{\boldsymbol{\varepsilon}}. \tag{16}$$

Consider next the case where $\overline{\boldsymbol{\varepsilon}} = \mathbf{0}$ and $\boldsymbol{\tau}(\boldsymbol{x})$ is arbitrary. The solution of (15) can be expressed by means of the nonlocal elastic Green operator $\boldsymbol{\Gamma}(\boldsymbol{x}, \boldsymbol{x'})$ of the nonhomogeneous elastic medium as:

$$\boldsymbol{\varepsilon}(\boldsymbol{x}) = -\boldsymbol{\Gamma} * \boldsymbol{\tau}(\boldsymbol{x}), \quad \text{where} \quad \boldsymbol{\Gamma} * \boldsymbol{\tau}(\boldsymbol{x}) \overset{def}{=} \frac{1}{|V|} \int_V \boldsymbol{\Gamma}(\boldsymbol{x}, \boldsymbol{x'}) : \boldsymbol{\tau}(\boldsymbol{x'}) \, d\boldsymbol{x'}. \tag{17}$$

It follows from the superposition principle that the solution of (15) reads as:

$$\boldsymbol{\varepsilon}(\boldsymbol{x}) = \boldsymbol{A}(\boldsymbol{x}) : \overline{\boldsymbol{\varepsilon}} - \boldsymbol{\Gamma} * \boldsymbol{\tau}(\boldsymbol{x}). \tag{18}$$

Using this form of the strain field in the effective energy

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}}) = \frac{1}{2} \langle \boldsymbol{\varepsilon} : \boldsymbol{L} : \boldsymbol{\varepsilon} \rangle + \langle \boldsymbol{\varepsilon} : \boldsymbol{\tau} \rangle,$$

yields an expression which can be simplified by noting that:

$$\langle \boldsymbol{\tau} : \boldsymbol{\Gamma} * \boldsymbol{\tau} \rangle = \langle (\boldsymbol{\Gamma} * \boldsymbol{\tau}) : \boldsymbol{L} : (\boldsymbol{\Gamma} * \boldsymbol{\tau}) \rangle, \tag{19}$$

and

$$\langle (\boldsymbol{\Gamma} * \boldsymbol{\tau}) : \boldsymbol{L} : \boldsymbol{A} : \overline{\boldsymbol{\varepsilon}} \rangle = 0. \tag{20}$$

The relation (19) results from the fact that $\boldsymbol{\Gamma} * \boldsymbol{\tau}$ is a compatible strain field with average $\mathbf{0}$ and that $-\boldsymbol{L} : \boldsymbol{\Gamma} * \boldsymbol{\tau} + \boldsymbol{\tau}$ is a divergence-free stress field. Therefore, it follows from Hill's lemma that:

$$\langle (\boldsymbol{\Gamma} * \boldsymbol{\tau}) : (-\boldsymbol{L} : \boldsymbol{\Gamma} * \boldsymbol{\tau} + \boldsymbol{\tau}) \rangle = 0,$$

hence (19). Similarly, the relation (20) results from the fact that $\boldsymbol{L} : \boldsymbol{A} : \overline{\boldsymbol{\varepsilon}}$ is divergence-free stress field and that $\boldsymbol{\Gamma} * \boldsymbol{\tau}$ is a compatible strain field with average $\mathbf{0}$. In addition, since $\boldsymbol{A}(\boldsymbol{x}) : \overline{\boldsymbol{\varepsilon}}$ is a compatible strain field, and $\left\langle \boldsymbol{A}^T \right\rangle = \boldsymbol{I}$, it follows that

$$\left\langle \overline{\boldsymbol{\varepsilon}} : \boldsymbol{A}^T : \boldsymbol{L} : \boldsymbol{A} : \overline{\boldsymbol{\varepsilon}} \right\rangle = \overline{\boldsymbol{\varepsilon}} : \left\langle \boldsymbol{A}^T \right\rangle : \langle \boldsymbol{L} : \boldsymbol{A} \rangle : \overline{\boldsymbol{\varepsilon}} = \overline{\boldsymbol{\varepsilon}} : \langle \boldsymbol{L} : \boldsymbol{A} \rangle : \overline{\boldsymbol{\varepsilon}}. \tag{21}$$

Then, using the relations (19), (20), and (21), the effective energy $\widetilde{w}$ can be put in the form (12) with:

$$\widetilde{\boldsymbol{L}} = \langle \boldsymbol{L} : \boldsymbol{A} \rangle, \quad \widetilde{\boldsymbol{\tau}} = \left\langle \boldsymbol{A}^T : \boldsymbol{\tau} \right\rangle, \quad \widetilde{f}(\boldsymbol{\tau}) = -\frac{1}{2} \langle \boldsymbol{\tau} : \boldsymbol{\Gamma} * \boldsymbol{\tau} \rangle. \tag{22}$$

It is useful to note that, since $\boldsymbol{\Gamma} * \boldsymbol{\tau}$ is a strain field with average 0, one has, for every divergence-free stress field $\boldsymbol{\sigma}^*$:

$$\langle \boldsymbol{\tau} : \boldsymbol{\Gamma} * \boldsymbol{\tau} \rangle = \langle (\boldsymbol{\tau} - \boldsymbol{\sigma}^*) : \boldsymbol{\Gamma} * (\boldsymbol{\tau} - \boldsymbol{\sigma}^*) \rangle. \tag{23}$$

This last relation shows that $\widetilde{f}$ is in fact due to the part of $\boldsymbol{\tau}$ which is not in equilibrium.

Remarks:

(1) It follows from (22) and (19) that

$$\widetilde{f}(\boldsymbol{\tau}) = -\frac{1}{2} \langle (\boldsymbol{\Gamma} * \boldsymbol{\tau}) : \boldsymbol{L} : (\boldsymbol{\Gamma} * \boldsymbol{\tau}) \rangle \leq 0. \tag{24}$$

In other words, $\widetilde{f}$ is always negative. In addition $\widetilde{f} = 0$ if and only if $\boldsymbol{\Gamma} * \boldsymbol{\tau} = \boldsymbol{0}$, which is the case only when $\boldsymbol{\tau}$ is a divergence-free field.

(2) Assume that $\boldsymbol{\tau}$ is *not* a divergence-free field. Taking $\lambda \boldsymbol{\tau}$ in the effective energy (7) and letting λ go to $+\infty$ ($\overline{\boldsymbol{\varepsilon}}$ being fixed), it is seen that the exact effective energy goes to $-\infty$ as $\lambda^2 \widetilde{f}(\boldsymbol{\tau})$.

(3) When $\overline{\boldsymbol{\varepsilon}} = \boldsymbol{0}$, the Voigt and Reuss bounds read:

$$\widetilde{f}^{\text{Reuss}}(\boldsymbol{\tau}) \leq \widetilde{f}(\boldsymbol{\tau}) \leq \widetilde{f}^{\text{Voigt}}(\boldsymbol{\tau}). \tag{25}$$

The term $\widetilde{f}^{\text{Voigt}}(\boldsymbol{\tau})$ is identically 0 and the second inequality in (25) is nothing else than the fact that $\widetilde{f}(\boldsymbol{\tau})$ is always negative (already derived by different means in (24)). On the other hand , the Reuss bound does reproduce the divergence of the $\widetilde{f}(\lambda \boldsymbol{\tau})$ as λ goes to $+\infty$. In this connection, note that the energy term $\widetilde{f}^{\text{Reuss}}(\boldsymbol{\tau})$ can be alternatively written

$$\widetilde{f}^{\text{Reuss}}(\boldsymbol{\tau}) = -\frac{1}{2} \langle \boldsymbol{\tau}^* : \boldsymbol{M} : \boldsymbol{\tau}^* \rangle, \quad \boldsymbol{\tau}^* = \boldsymbol{\tau} - \langle \boldsymbol{M} \rangle^{-1} : \langle \boldsymbol{M} : \boldsymbol{\tau} \rangle = \boldsymbol{\tau} - \widetilde{\boldsymbol{\tau}}^{\text{Reuss}}.$$

Thus, the Reuss bound on $\widetilde{w}$ can diverge as $-\lambda^2$ when λ goes to $+\infty$, except when $\boldsymbol{\tau}^* = 0$ which corresponds to a completely uniform polarization $\boldsymbol{\tau}$.

(4) It is recalled (Laws (1973); Willis (1981)) here for future reference that when $\boldsymbol{L}(\boldsymbol{x})$ and $\boldsymbol{\tau}(\boldsymbol{x})$ are constant per phase such that

$$\boldsymbol{L}(\boldsymbol{x}) = \sum_{r=1}^{N} \chi^{(r)}(\boldsymbol{x}) \boldsymbol{L}^{(r)}, \quad \text{and} \quad \boldsymbol{\tau}(\boldsymbol{x}) = \sum_{r=1}^{N} \chi^{(r)}(\boldsymbol{x}) \boldsymbol{\tau}^{(r)}, \tag{26}$$

where $\boldsymbol{L}^{(r)}$ and $\boldsymbol{\tau}^{(r)}$ are constant, the effective properties (22) associated with the effective energy (12) reduce to

$$\widetilde{\boldsymbol{L}} = \sum_{r=1}^{N} c^{(r)} \boldsymbol{L}^{(r)} : \overline{\boldsymbol{A}}^{(r)}, \quad \widetilde{\boldsymbol{\tau}} = \sum_{r=1}^{N} c^{(r)} \, \overline{\boldsymbol{A}}^{(r)T} : \boldsymbol{\tau}^{(r)}, \quad \widetilde{f}(\boldsymbol{\tau}) = \frac{1}{2} \sum_{r=1}^{N} c^{(r)} \, \overline{\mathbf{a}}^{(r)} : \boldsymbol{\tau}^{(r)}, \tag{27}$$

where $\overline{\boldsymbol{A}}^{(r)} = \langle \boldsymbol{A} \rangle^{(r)}$ and $\overline{\mathbf{a}}^{(r)} = -\langle \boldsymbol{\Gamma} * \boldsymbol{\tau} \rangle^{(r)}$ are strain concentration tensors such that the average of the strain over phase r is given by $\langle \boldsymbol{\varepsilon} \rangle^{(r)} = \overline{\boldsymbol{A}}^{(r)} \overline{\boldsymbol{\varepsilon}} + \overline{\mathbf{a}}^{(r)}$. In particular, for a two-phase composite, it is known (Levin (1967)) that $\widetilde{\boldsymbol{\tau}}$ and $\widetilde{f}$ may be obtained directly in terms of $\widetilde{\boldsymbol{L}}$ via

$$\widetilde{\boldsymbol{\tau}} = \langle \boldsymbol{\tau} \rangle + (\widetilde{\boldsymbol{L}} - \langle \boldsymbol{L} \rangle) : \left(\boldsymbol{L}^{(1)} - \boldsymbol{L}^{(2)} \right)^{-1} : \left(\boldsymbol{\tau}^{(1)} - \boldsymbol{\tau}^{(2)} \right), \tag{28}$$

$$\widetilde{f}(\boldsymbol{\tau}) = \frac{1}{2} \left(\boldsymbol{\tau}^{(1)} - \boldsymbol{\tau}^{(2)} \right) : \left(\boldsymbol{L}^{(1)} - \boldsymbol{L}^{(2)} \right)^{-1} : (\widetilde{\boldsymbol{L}} - \langle \boldsymbol{L} \rangle) : \left(\boldsymbol{L}^{(1)} - \boldsymbol{L}^{(2)} \right)^{-1} : \left(\boldsymbol{\tau}^{(1)} - \boldsymbol{\tau}^{(2)} \right). \tag{29}$$

3 Bounds on the effective potentials through linear comparison composites

3.1 Lower bound on the primal energy

Introduce a *linear comparison composite (LCC)* with piecewise constant elastic moduli and polarization:

$$w_0(\boldsymbol{x}, \boldsymbol{\varepsilon}) = \frac{1}{2} \boldsymbol{\varepsilon} : \boldsymbol{L}_0(\boldsymbol{x}) : \boldsymbol{\varepsilon} + \boldsymbol{\tau}_0(\boldsymbol{x}) : \boldsymbol{\varepsilon}, \tag{30}$$

where

$$\boldsymbol{L}_0(\boldsymbol{x}) = \sum_{r=1}^{N} \chi^{(r)}(\boldsymbol{x}) \boldsymbol{L}_0^{(r)}, \quad \boldsymbol{\tau}_0(\boldsymbol{x}) = \sum_{r=1}^{N} \chi^{(r)}(\boldsymbol{x}) \boldsymbol{\tau}_0^{(r)}, \tag{31}$$

and let

$$w_0^{(r)}(\boldsymbol{\varepsilon}) = \frac{1}{2} \boldsymbol{\varepsilon} : \boldsymbol{L}_0^{(r)} : \boldsymbol{\varepsilon} + \boldsymbol{\tau}_0^{(r)} : \boldsymbol{\varepsilon}. \tag{32}$$

Assume that

$$\boldsymbol{\Delta} \boldsymbol{L}^{(r)}(\boldsymbol{x}) = \boldsymbol{L}(\boldsymbol{x}) - \boldsymbol{L}_0^{(r)} \geq 0, \tag{33}$$

where the inequality in (33) is to be understood in the sense of positive semi-definite quadratic forms and set:

$$\boldsymbol{\Delta} \boldsymbol{\tau}^{(r)}(\boldsymbol{x}) = \boldsymbol{\tau}(\boldsymbol{x}) - \boldsymbol{\tau}_0^{(r)}. \tag{34}$$

Consider the infimum problem:

$$\inf_{\boldsymbol{e} \in \mathbb{R}_s^{3\times 3}} \left\{ w^{(r)}(\boldsymbol{x}, \boldsymbol{e}) - w_0^{(r)}(\boldsymbol{e}) \right\}.$$

Note that due to assumption (33), the function $w^{(r)} - w_0^{(r)}$ is convex .Therefore, the above infimum problem has a solution and one can define the 'error' function $\mathcal{V}^{(r)}$ between $w^{(r)}$ and $w_0^{(r)}$ as:

$$\begin{aligned} \mathcal{V}^{(r)}(\boldsymbol{x}, \boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}) &= \inf_{\boldsymbol{e} \in \mathbb{R}_s^{3\times 3}} \left\{ w^{(r)}(\boldsymbol{x}, \boldsymbol{e}) - w_0^{(r)}(\boldsymbol{e}) \right\} \\ &= -\frac{1}{2} \boldsymbol{\Delta} \boldsymbol{\tau}^{(r)}(\boldsymbol{x}) : (\boldsymbol{\Delta} \boldsymbol{L}^{(r)}(\boldsymbol{x}))^{-1} : \boldsymbol{\Delta} \boldsymbol{\tau}^{(r)}(\boldsymbol{x}). \end{aligned} \tag{35}$$

Then, for every $\boldsymbol{\tau}_0^{(r)}$ and $\boldsymbol{L}_0^{(r)}$ satisfying (33), one has:

$$w^{(r)}(\boldsymbol{x}, \boldsymbol{\varepsilon}) \geq w_0^{(r)}(\boldsymbol{\varepsilon}) + \mathcal{V}^{(r)}(\boldsymbol{x}, \boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}). \tag{36}$$

By definition, $\widetilde{w}(\overline{\boldsymbol{\varepsilon}})$ is such that

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}}) = \inf_{\boldsymbol{\varepsilon} \ \in \mathcal{K}(\overline{\boldsymbol{\varepsilon}})} \langle w(\boldsymbol{x}, \boldsymbol{\varepsilon}) \rangle = \inf_{\boldsymbol{\varepsilon} \ \in \mathcal{K}(\overline{\boldsymbol{\varepsilon}})} \sum_{r=1}^{N} c^{(r)} \Big\langle w^{(r)}(\boldsymbol{x}, \boldsymbol{\varepsilon}) \Big\rangle^{(r)}.$$

According to (36), the following inequality holds for every $\boldsymbol{\varepsilon} \in \mathcal{K}(\overline{\boldsymbol{\varepsilon}})$ and for every $\boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}$ satisfying (33):

$$\langle w(\boldsymbol{x}, \boldsymbol{\varepsilon}) \rangle \geq \sum_{r=1}^{N} c^{(r)} \Big\langle w_0^{(r)}(\boldsymbol{\varepsilon}) + \mathcal{V}^{(r)}(\boldsymbol{x}, \boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}) \Big\rangle^{(r)}. \tag{37}$$

Taking the infimum of the two sides of (37) with respect to $\boldsymbol{\varepsilon}$, one has

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}}) \geq \widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) + \langle \mathcal{V}(\boldsymbol{x}, \boldsymbol{\tau}_0, \boldsymbol{L}_0) \rangle, \tag{38}$$

for every $\boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}$ satisfying (33), where

$$\widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) = \inf_{\boldsymbol{\varepsilon} \ \in \mathcal{K}(\overline{\boldsymbol{\varepsilon}})} \langle w_0(\boldsymbol{x}, \boldsymbol{\varepsilon}) \rangle = \sum_{r=1}^{N} c^{(r)} \Big\langle w_0^{(r)}(\boldsymbol{\varepsilon}) \Big\rangle^{(r)},$$

and

$$\langle \mathcal{V}(\boldsymbol{x}, \boldsymbol{L}_0, \boldsymbol{\tau}_0) \rangle = \sum_{r=1}^{N} c^{(r)} \Big\langle \mathcal{V}^{(r)}(\boldsymbol{x}, \boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}) \Big\rangle^{(r)}. \tag{39}$$

Finally, making use of (35) in (39), and taking the supremum of (38) over all possible $\boldsymbol{\tau}_0^{(r)}$ and $\boldsymbol{L}_0^{(r)}$, one obtains the result:

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}}) \geq \sup_{\boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}} \left\{ \widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) - \frac{1}{2} \sum_{r=1}^{N} c^{(r)} \Big\langle \boldsymbol{\Delta\tau}^{(r)}(\boldsymbol{x}) : (\boldsymbol{\Delta L}^{(r)}(\boldsymbol{x}))^{-1} : \boldsymbol{\Delta\tau}^{(r)}(\boldsymbol{x}) \Big\rangle^{(r)} \right\}. \tag{40}$$

Stationarity conditions may be written for the supremum problem (40). Thus, stationarity with respect to $\boldsymbol{\tau}_0^{(r)}$ gives:

$$\langle \boldsymbol{\varepsilon}_0 \rangle^{(r)} = -\Big\langle \Big(\boldsymbol{\Delta L}^{(r)} \Big)^{-1} : \boldsymbol{\Delta\tau} \Big\rangle^{(r)}, \tag{41}$$

where $\boldsymbol{\varepsilon}_0$ denotes the strain field in the LCC, while stationarity with respect to $\boldsymbol{L}_0^{(r)}$leads to

$$\langle \boldsymbol{\varepsilon}_0 \otimes \boldsymbol{\varepsilon}_0 \rangle^{(r)} = \Big\langle \Big(\boldsymbol{\Delta L}^{(r)} \Big)^{-1} : \boldsymbol{\Delta\tau} \otimes \Big(\boldsymbol{\Delta L}^{(r)} \Big)^{-1} : \boldsymbol{\Delta\tau} \Big\rangle^{(r)}. \tag{42}$$

The two equations (41) and (42) are to be solved for $\boldsymbol{\tau}_0^{(r)}|_{r=1,..,N}$ and $\boldsymbol{L}_0^{(r)}|_{r=1,..,N}$, keeping in mind that $\boldsymbol{\varepsilon}_0$ depends on these unknowns. This system of nonlinear coupled equations may have one solution, or several solutions, or no solution.

Let us assume that there exists a solution for the $\boldsymbol{L}_0^{(r)}$'s and $\boldsymbol{\tau}_0^{(r)}$'s which satisfies the assumed positivity condition (33). In order to have the best possible lower bound one has to make sure that the stationarity point in (40) is a maximum. For this purpose, it is sufficient to prove that the function $\widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) + \langle \mathcal{V}(\boldsymbol{x}, \boldsymbol{\tau}_0, \boldsymbol{L}_0)\rangle$ is a concave function of the unknowns $(\boldsymbol{\tau}_0^{(s)}, \boldsymbol{L}_0^{(s)})|_{s=1,..,N}$.

To show this, first note that, for every $\boldsymbol{\varepsilon}$, $w_0^{(r)}(\boldsymbol{\varepsilon})$ is an affine, and therefore concave, function of $(\boldsymbol{L}_0^{(r)}, \boldsymbol{\tau}_0^{(r)})$. Similarly it follows from (35) that, under the inequality (33), $\mathcal{V}^{(r)}$ is a concave function of $(\boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)})$. Consequently the function $w_0^{(r)}(\boldsymbol{\varepsilon}) + \mathcal{V}^{(r)}(\boldsymbol{x}, \boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)})$ is, for every $\boldsymbol{\varepsilon}$, a concave function of $(\boldsymbol{L}_0^{(r)}, \boldsymbol{\tau}_0^{(r)})$. Therefore,

$$\sum_{r=1}^{N} c^{(r)} \left\langle w_0^{(r)}(\boldsymbol{\varepsilon}) + \mathcal{V}^{(r)}(\boldsymbol{x}, \boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}) \right\rangle^{(r)}$$

is a concave function of the unknowns $(\boldsymbol{\tau}_0^{(s)}, \boldsymbol{L}_0^{(s)})|_{s=1,..,N}$. Then taking the infimum over $\boldsymbol{\varepsilon}$ of these concave functions is again a concave function which proves that $\widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) + \langle \mathcal{V}(\boldsymbol{x}, \boldsymbol{\tau}_0, \boldsymbol{L}_0)\rangle$ is a concave function of the unknowns $(\boldsymbol{\tau}_0^{(s)}, \boldsymbol{L}_0^{(s)})|_{s=1,..,N}$. Therefore, every stationary point for the right-hand-side of (40) is a supremum provided that the solutions to (41) and (42) satisfy the inequality (33).

Comments:

(1) An upper bound for the energy can be obtained in a similar way when

$$\boldsymbol{\Delta L}^{(r)}(\boldsymbol{x}) = \boldsymbol{L}(\boldsymbol{x}) - \boldsymbol{L}_0^{(r)} \leq 0. \tag{43}$$

The modifications to be made to the lower bound procedure go as follows. First the infimum in (35) is changed into a supremum. Therefore the inequalities in (36), (37) and (38) are reversed. The bound (40) becomes an *upper bound*:

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}}) \leq \inf_{\boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}} \left\{ \widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) + \langle \mathcal{V}(\boldsymbol{x}, \boldsymbol{\tau}_0, \boldsymbol{L}_0)\rangle \right\}, \tag{44}$$

and the stationarity conditions (41) and (42) remain unchanged. In order to prove that the stationary points of the right-hand-side of (44) are in fact infima, it is sufficient to show that $\widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) + \langle \mathcal{V}(\boldsymbol{x}, \boldsymbol{\tau}_0, \boldsymbol{L}_0)\rangle$ is a convex function of $(\boldsymbol{\tau}_0^{(s)}, \boldsymbol{L}_0^{(s)})|_{s=1,..,N}$. The arguments follow closely those used to prove that it is a concave function of these arguments when the inequality (33) holds.

(2) Define the fluctuations $\boldsymbol{C}^{(r)}(\boldsymbol{e})$ of a field $\boldsymbol{e}$ in phase r as:

$$\boldsymbol{C}^{(r)}(\boldsymbol{e}) = \left\langle \left(\boldsymbol{e} - \langle \boldsymbol{e}\rangle^{(r)}\right) \otimes \left(\boldsymbol{e} - \langle \boldsymbol{e}\rangle^{(r)}\right)\right\rangle^{(r)}. \tag{45}$$

Then, making use of (41), the stationarity condition (42) can be rewritten as:

$$\boldsymbol{C}^{(r)}\left(\boldsymbol{\varepsilon}_0\right) = \boldsymbol{C}^{(r)}\left(\boldsymbol{\Delta L}^{-1} : \boldsymbol{\Delta \tau}\right), \tag{46}$$

In other words, the two stationarity conditions amount to equating the per phase averages and fluctuations of the fields $\boldsymbol{\varepsilon}_0$ and $\boldsymbol{\Delta L}^{-1} : \boldsymbol{\Delta \tau}$.

(3) When the elastic moduli are uniform per phase and denoted by $\boldsymbol{L}^{(r)}$, the stationarity conditions take a simpler form:

$$\boldsymbol{\Delta L}^{(r)} : \langle \boldsymbol{\varepsilon}_0 \rangle^{(r)} = -\langle \boldsymbol{\Delta \tau} \rangle^{(r)}, \ \left\langle \left(\boldsymbol{\Delta L}^{(r)} : \boldsymbol{\varepsilon}_0 \right) \otimes \left(\boldsymbol{\Delta L}^{(r)} : \boldsymbol{\varepsilon}_0 \right) \right\rangle^{(r)} = \langle \boldsymbol{\Delta \tau} \otimes \boldsymbol{\Delta \tau} \rangle^{(r)}. \tag{47}$$

where $\boldsymbol{\Delta L}^{(r)} = \boldsymbol{L}^{(r)} - \boldsymbol{L}_0^{(r)}$.

(4) The right-hand side of (38) can be considered as an approximation of the actual potential $\widetilde{w}$. Taking advantage of the stationarity of the variational lower bound (40) (or the corresponding upper bound (44)) with respect to the variables $\boldsymbol{L}_0^{(r)}$ and $\boldsymbol{\tau}_0^{(r)}$, it is easy to show that the macroscopic stress-strain relation of the original composite, as determined by (11), leads to the approximation

$$\overline{\boldsymbol{\sigma}} = \widetilde{\boldsymbol{L}}_0 \overline{\boldsymbol{\varepsilon}} + \widetilde{\boldsymbol{\tau}}_0, \tag{48}$$

where the tensors $\widetilde{\boldsymbol{L}}_0$ and $\widetilde{\boldsymbol{\tau}}_0$ have to be evaluated at the optimal values of $\boldsymbol{L}_0^{(r)}$ and $\boldsymbol{\tau}_0^{(r)}$ resulting from expressions (41) and (42).

3.2 Dual approach

A completely parallel approach can be developed with the dual potential u defined by (4) and the expression of which is given by (5). The dual potential for the linear comparison composite has a similar expression:

$$u_0^{(r)}(\boldsymbol{\sigma}) = \sup_{\boldsymbol{\varepsilon}} \left\{ \boldsymbol{\sigma} : \boldsymbol{\varepsilon} - w_0^{(r)}(\boldsymbol{\varepsilon}) \right\} = \frac{1}{2} \boldsymbol{\sigma} : \boldsymbol{M}_0^{(r)} : \boldsymbol{\sigma} + \boldsymbol{\gamma}_0^{(r)} : \boldsymbol{\sigma} + \frac{1}{2} \boldsymbol{\gamma}_0^{(r)} : \boldsymbol{L}_0^{(r)} : \boldsymbol{\gamma}_0^{(r)}, \tag{49}$$

where

$$\boldsymbol{M}_0^{(r)} = \left(\boldsymbol{L}_0^{(r)} \right)^{-1}, \quad \boldsymbol{\gamma}_0^{(r)} = -\boldsymbol{M}_0^{(r)} : \boldsymbol{\tau}_0^{(r)}. \tag{50}$$

Now, assuming that (33) holds, the primal effective potential $\widetilde{w}$ admits the lower bound (40), which by means of (10), gives an upper bound on the dual effective potential $\widetilde{u}$. More specifically, for every $\boldsymbol{\tau}^{(r)}$ and $\boldsymbol{L}^{(r)}$:

$$\widetilde{u}(\overline{\boldsymbol{\sigma}}) \leq \sup_{\overline{\boldsymbol{\varepsilon}}} \left\{ \overline{\boldsymbol{\sigma}} : \overline{\boldsymbol{\varepsilon}} - \widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) - \langle \mathcal{V}(\boldsymbol{x}, \boldsymbol{\tau}_0, \boldsymbol{L}_0) \rangle \right\} \leq \widetilde{u}_0(\overline{\boldsymbol{\sigma}}) - \langle \mathcal{V}(\boldsymbol{x}, \boldsymbol{\tau}_0, \boldsymbol{L}_0) \rangle,$$

and therefore

$$\widetilde{u}(\overline{\boldsymbol{\sigma}}) \leq \inf_{\boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}} \left\{ \widetilde{u}_0(\overline{\boldsymbol{\sigma}}) - \langle \mathcal{V}(\boldsymbol{x}, \boldsymbol{\tau}_0, \boldsymbol{L}_0) \rangle \right\}. \tag{51}$$

The error function $\mathcal{V}^{(r)}$ between $w^{(r)}$ and $w_0^{(r)}$, given by (35), is also (up to a sign) the error function between $u^{(r)}$ and $u_0^{(r)}$. Indeed, we have that

$$
\begin{aligned}
\mathcal{V}^{(r)}(\boldsymbol{x},\boldsymbol{\tau}_0^{(r)},\boldsymbol{L}_0^{(r)}) &= \inf_{\boldsymbol{\varepsilon}}\left\{w^{(r)}(\boldsymbol{x},\boldsymbol{\varepsilon}) - w_0^{(r)}(\boldsymbol{\varepsilon})\right\} \\
&= \inf_{\boldsymbol{\varepsilon}}\left\{w^{(r)}(\boldsymbol{x},\boldsymbol{\varepsilon}) - \sup_{\boldsymbol{\sigma}}\left\{\boldsymbol{\sigma}:\boldsymbol{\varepsilon} - u_0^{(r)}(\boldsymbol{x},\boldsymbol{\sigma})\right\}\right\} \\
&= \inf_{\boldsymbol{\sigma}}\left\{\inf_{\boldsymbol{\varepsilon}}\left\{w^{(r)}(\boldsymbol{x},\boldsymbol{\varepsilon}) - \boldsymbol{\sigma}:\boldsymbol{\varepsilon}\right\} + u_0^{(r)}(\boldsymbol{x},\boldsymbol{\sigma})\right\} \\
&= \inf_{\boldsymbol{\sigma}}\left\{u_0^{(r)}(\boldsymbol{x},\boldsymbol{\sigma}) - u^{(r)}(\boldsymbol{x},\boldsymbol{\sigma})\right\} \\
&= -\sup_{\boldsymbol{\sigma}}\left\{u^{(r)}(\boldsymbol{x},\boldsymbol{\sigma}) - u_0^{(r)}(\boldsymbol{x},\boldsymbol{\sigma})\right\}.
\end{aligned} \tag{52}
$$

The relations (50) can be inverted to express $\boldsymbol{\tau}_0^{(r)}$ and $\boldsymbol{L}_0^{(r)}$ in terms of $\boldsymbol{\gamma}_0^{(r)}$ and $\boldsymbol{M}_0^{(r)}$. Consequently, $\mathcal{V}^{(r)}$ can be considered as a function of the strain polarization $\boldsymbol{\gamma}_0^{(r)}$ and of the elastic compliance $\boldsymbol{M}_0^{(r)}$. In fact, it is easy to show that

$$
\mathcal{V}^{(r)}(\boldsymbol{x},\boldsymbol{\gamma}_0^{(r)},\boldsymbol{M}_0^{(r)}) = \frac{1}{2}\boldsymbol{\Delta\gamma}^{(r)}(\boldsymbol{x}):(\boldsymbol{\Delta M}^{(r)}(\boldsymbol{x}))^{-1}:\boldsymbol{\Delta\gamma}^{(r)}(\boldsymbol{x}) + \frac{1}{2}\boldsymbol{\gamma}_0^{(r)}:\boldsymbol{L}_0^{(r)}:\boldsymbol{\gamma}_0^{(r)}, \tag{53}
$$

where $\boldsymbol{\Delta M}^{(r)}(\boldsymbol{x}) = \boldsymbol{M}(\boldsymbol{x}) - \boldsymbol{M}_0^{(r)} \leq 0$ and $\boldsymbol{\Delta\gamma}^{(r)}(\boldsymbol{x}) = \boldsymbol{\gamma}(\boldsymbol{x}) - \boldsymbol{\gamma}_0^{(r)}$.

Therefore, the bound (51) can be alternatively written as:

$$
\widetilde{u}(\overline{\boldsymbol{\sigma}}) \leq \inf_{\boldsymbol{\gamma}_0^{(r)},\boldsymbol{M}_0^{(r)}}\left\{\widetilde{u}_0(\overline{\boldsymbol{\sigma}}) - \frac{1}{2}\sum_{r=1}^{N} c^{(r)}\left\langle\boldsymbol{\Delta\gamma}^{(r)}(\boldsymbol{x}):(\boldsymbol{\Delta M}^{(r)}(\boldsymbol{x}))^{-1}:\boldsymbol{\Delta\gamma}^{(r)}(\boldsymbol{x})\right\rangle^{(r)}\right\}, \tag{54}
$$

where the last term in the definition (49) of $u_0^{(r)}$ should be dropped in the computation of $\widetilde{u}_0$ in the right hand side of this expression, since it cancels out exactly with the second term in expression (53) for the functions $V^{(r)}$.

It should be noted that the lower bound (54) for $\widetilde{u}$ could have been obtained more directly by following a procedure completely analogous to the one outlined earlier for the upper bound (44) for the primal effective energy $\widetilde{w}$ under the hypothesis (43).

Also, in complete analogy to what was stated in remark 4 of the previous subsection, it can be shown that the macroscopic constitutive relation for the original composite may be expressed in the form

$$
\overline{\boldsymbol{\varepsilon}} = \widetilde{\boldsymbol{M}}_0\overline{\boldsymbol{\sigma}} + \widetilde{\boldsymbol{\gamma}}_0, \tag{55}
$$

where the tensors $\widetilde{\boldsymbol{M}}_0$ and $\widetilde{\boldsymbol{\gamma}}_0$ have to be evaluated at the optimal values of $\boldsymbol{M}_0^{(r)}$ and $\boldsymbol{\gamma}_0^{(r)}$ resulting from stationarity conditions associated with the optimization problem defined by the right hand side of expression (54).

3.3 No strain fluctuation in one of the phases of the LCC

In certain special cases, it can happen that the strain field $\boldsymbol{\varepsilon}_0$ in one of the phases of the LCC is uniform, which implies that its fluctuations $\boldsymbol{C}^{(r)}(\boldsymbol{\varepsilon}_0)$ vanish. This is the case in all phases of a laminate. This is also the case in one of the phases of composites for which one of the Hashin-Shtrikman bounds is an accurate approximation. When this happens, the stationarity equation (46) requires that the LCC be chosen in such a way that the fluctuations of $\boldsymbol{\Delta L}^{-1} : \boldsymbol{\tau}$ vanish in the phase where $\boldsymbol{\varepsilon}_0$ has no fluctuation. In other words, one would have to choose $\boldsymbol{L}_0^{(r)}$ in such a way that $\boldsymbol{\Delta L}^{-1} : \boldsymbol{\tau}$ is uniform in this phase which is impossible in general. Consider the case where the elastic moduli are piecewise uniform. Then the condition (46) can be rewritten as

$$\boldsymbol{C}^{(r)}\left(\boldsymbol{\Delta L} : \boldsymbol{\varepsilon}_0\right) = \boldsymbol{C}^{(r)}\left(\boldsymbol{\Delta \tau}\right). \tag{56}$$

When the fluctuations of $\boldsymbol{\varepsilon}_0$ vanish in one phase, the left-hand-side of (56) vanishes, whereas

$$\boldsymbol{C}^{(r)}\left(\boldsymbol{\Delta \tau}\right) = \boldsymbol{C}^{(r)}\left(\boldsymbol{\tau}\right) + \left(\langle \boldsymbol{\tau}\rangle^{(r)} - \boldsymbol{\tau}_0^{(r)}\right) \otimes \left(\langle \boldsymbol{\tau}\rangle^{(r)} - \boldsymbol{\tau}_0^{(r)}\right),$$

which cannot vanish except when $\boldsymbol{\tau}$ is uniform in this phase. The stationarity equation (46) has no solution in this particular phase. This means that the extremum in (40) or (44) is attained on the boundary of the admissible domain for $\boldsymbol{L}_0^{(r)}$, which in turn suggests identifying the elastic moduli in this phase with the original piecewise constant elastic moduli. As will be shown next, this choice combined with other choices in the other phases, leads to an *upper bound* for the effective energy of the composite.

For definiteness, assume that the strain field in the LCC has fluctuations in all phases except in phase 1. Set:

$$\boldsymbol{L}_0^{(1)} = \boldsymbol{L}^{(1)}, \quad \boldsymbol{\tau}_0^{(1)} = \langle \boldsymbol{\tau}\rangle^{(1)}. \tag{57}$$

The other $\boldsymbol{L}_0^{(r)}$ ($r \neq 1$) are searched such that $\boldsymbol{\Delta L}^{(r)} \leq 0$. With the choice (57), one has in phase 1,

$$w_0^{(1)}(\boldsymbol{\varepsilon}) = w^{(1)}(\boldsymbol{x}, \boldsymbol{\varepsilon}) + (\boldsymbol{\tau}_0^{(1)} - \boldsymbol{\tau}(\boldsymbol{x})) : \boldsymbol{\varepsilon},$$

whereas in the other phases ($r \neq 1$)

$$w_0^{(r)}(\boldsymbol{x}, \boldsymbol{\varepsilon}) \geq w^{(r)}(\boldsymbol{x}, \boldsymbol{\varepsilon}) - \mathcal{V}^{(r)}(\boldsymbol{x}, \boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}).$$

Therefore, denoting by $\boldsymbol{\varepsilon}_0$ the strain field in the LCC:

$$\begin{aligned}
\widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) &= \langle w_0(\boldsymbol{\varepsilon}_0)\rangle = \sum_{r=1}^{N} c^{(r)} \left\langle w_0^{(r)}(\boldsymbol{\varepsilon}_0)\right\rangle^{(r)} \\
&\geq c^{(1)}\left(\left\langle w^{(1)}(\boldsymbol{x}, \boldsymbol{\varepsilon}_0)\right\rangle^{(1)} - \left\langle (\boldsymbol{\tau}_0^{(1)} - \boldsymbol{\tau}) : \boldsymbol{\varepsilon}_0\right\rangle^{(1)}\right) \\
&\quad + \sum_{r=2}^{N} c^{(r)}\left(\left\langle w^{(r)}(\boldsymbol{\varepsilon}_0)\right\rangle^{(r)} - \left\langle \mathcal{V}^{(r)}(\boldsymbol{x}, \boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)})\right\rangle^{(r)}\right)
\end{aligned}$$

But since $\boldsymbol{\varepsilon}_0$ has no fluctuation in phase 1 and since $\boldsymbol{\tau}_0^{(1)}$ is the average of $\boldsymbol{\tau}$ over phase 1, the above result reduces to

$$\widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) \geq \langle w(\boldsymbol{\varepsilon}_0)\rangle - \sum_{r=2}^{N} c^{(r)} \left\langle \mathcal{V}^{(r)}(\boldsymbol{x}, \boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)})\right\rangle^{(r)}.$$

Finally, we obtain

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}}) \leq \langle w(\boldsymbol{\varepsilon}_0)\rangle \leq \inf_{\boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}} \left\{ \widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) + \sum_{r=2}^{N} c^{(r)} \left\langle \mathcal{V}^{(r)}(\boldsymbol{x}, \boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)})\right\rangle^{(r)} \right\},$$

which is an upper bound for the effective potential $\widetilde{w}$ under the hypothesis that $\boldsymbol{\Delta L}^{(r)} \leq 0$ $(r \neq 1)$.

4 Statistics of the fields

The aim of this section is to show that the first and second moments of the strain and stress fields in the actual non-piecewise uniform composite can be approximated at a good degree of accuracy by the first and second moments of the fields in the LCC.

Following Idiart & Ponte Castañeda (2007), the first and second moments of the strain and stress field in the actual non piecewise-uniform composite can be obtained by perturbing the initial potentials by terms which are piecewise-uniform in each phase. More specifically, consider the following perturbed potentials:

$$w_{\boldsymbol{t}}(\boldsymbol{x}, \boldsymbol{\varepsilon}) = \sum_{r=1}^{N} \chi^{(r)}(\boldsymbol{x}) w_{\boldsymbol{t}}^{(r)}(\boldsymbol{x}, \boldsymbol{\varepsilon}), \quad w_{\boldsymbol{t}}^{(r)}(\boldsymbol{x}, \boldsymbol{\varepsilon}) = w^{(r)}(\boldsymbol{x}, \boldsymbol{\varepsilon}) + \boldsymbol{t}^{(r)} : \boldsymbol{\varepsilon}, \tag{58}$$

and

$$w_{\boldsymbol{\lambda}}(\boldsymbol{x}, \boldsymbol{\varepsilon}) = \sum_{r=1}^{N} \chi^{(r)}(\boldsymbol{x}) w_{\boldsymbol{\lambda}}^{(r)}(\boldsymbol{x}, \boldsymbol{\varepsilon}), \quad w_{\boldsymbol{\lambda}}^{(r)}(\boldsymbol{x}, \boldsymbol{\varepsilon}) = w^{(r)}(\boldsymbol{x}, \boldsymbol{\varepsilon}) + \frac{1}{2}\boldsymbol{\varepsilon} : \boldsymbol{\lambda}^{(r)} : \boldsymbol{\varepsilon}, \tag{59}$$

where the $\boldsymbol{t}^{(r)}$'s and the $\boldsymbol{\lambda}^{(r)}$'s are constant second-order and fourth-order tensors respectively. Then, following Idiart & Ponte Castañeda (2007):

$$\langle \boldsymbol{\varepsilon} \rangle^{(r)} = \frac{1}{c^{(r)}} \frac{\partial \widetilde{w}_{\boldsymbol{t}}(\overline{\boldsymbol{\varepsilon}})}{\partial \boldsymbol{t}^{(r)}}|_{\boldsymbol{t}=\boldsymbol{0}}, \quad \langle \boldsymbol{\varepsilon} \otimes \boldsymbol{\varepsilon} \rangle^{(r)} = \frac{2}{c^{(r)}} \frac{\partial}{\partial \boldsymbol{\lambda}^{(r)}} \widetilde{w}_{\boldsymbol{\lambda}}(\overline{\boldsymbol{\varepsilon}})|_{\boldsymbol{\lambda}=\boldsymbol{0}}. \tag{60}$$

Similar relations apply to the first and second moments of the strain in the linear comparison composite.

Using the variational estimate (40) for the effective potential $\widetilde{w}$ amounts to making the following approximation:

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}}) \simeq \sup_{\boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}} \left\{ \widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) + \langle \mathcal{V}(\boldsymbol{x}, \boldsymbol{\tau}_0, \boldsymbol{L}_0) \rangle \right\}.$$

Note that the perturbation by a piecewise uniform polarization $\boldsymbol{t}^{(r)}$ or piecewise uniform moduli $\boldsymbol{\lambda}^{(r)}$, does not affect the error function $\mathcal{V}^{(r)}$. In other words, from the inequality (36)

$$w^{(r)}(\boldsymbol{x}, \boldsymbol{\varepsilon}) \geq w_0^{(r)}(\boldsymbol{\varepsilon}) + \mathcal{V}^{(r)}(\boldsymbol{x}, \boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}),$$

one obtains

$$w_{\boldsymbol{t}}^{(r)}(\boldsymbol{x}, \boldsymbol{\varepsilon}) \geq w_{0,\boldsymbol{t}}^{(r)}(\boldsymbol{\varepsilon}) + \mathcal{V}^{(r)}(\boldsymbol{x}, \boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}),$$

with the same function $\mathcal{V}^{(r)}$ as above. Consequently, the lower bound (40) still applies to the perturbed potential and can be taken as an approximation for the effective potential:

$$\widetilde{w}_{\boldsymbol{t}}(\overline{\boldsymbol{\varepsilon}}) \simeq \sup_{\boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}} \left\{ \widetilde{w}_{0,\boldsymbol{t}}(\overline{\boldsymbol{\varepsilon}}) + \langle \mathcal{V}(\boldsymbol{x}, \boldsymbol{\tau}_0, \boldsymbol{L}_0) \rangle \right\}. \tag{61}$$

Taking the derivative with respect to $\boldsymbol{t}^{(r)}$ of both sides of this equality, and noting that $\langle \mathcal{V} \rangle$ does not depend on $\boldsymbol{t}$, we obtain that

$$\langle \boldsymbol{\varepsilon} \rangle^{(r)} \simeq \langle \boldsymbol{\varepsilon}_0 \rangle^{(r)}. \tag{62}$$

The same procedure applied to $\widetilde{w}_{\boldsymbol{\lambda}}$ and $\widetilde{w}_{0,\boldsymbol{\lambda}}$ shows that

$$\langle \boldsymbol{\varepsilon} \otimes \boldsymbol{\varepsilon} \rangle^{(r)} \simeq \langle \boldsymbol{\varepsilon}_0 \otimes \boldsymbol{\varepsilon}_0 \rangle^{(r)}. \tag{63}$$

As for the moments of the stress field, they are obtained by suitable perturbations of the dual potential u. These perturbations read as:

$$u_{\boldsymbol{\eta}}(\boldsymbol{x}, \boldsymbol{\sigma}) = \sum_{r=1}^{N} \chi^{(r)}(\boldsymbol{x}) u_{\boldsymbol{\eta}}^{(r)}(\boldsymbol{x}, \boldsymbol{\sigma}), \quad u_{\boldsymbol{\eta}}^{(r)}(\boldsymbol{x}, \boldsymbol{\sigma}) = u^{(r)}(\boldsymbol{x}, \boldsymbol{\sigma}) + \boldsymbol{\eta}^{(r)} : \boldsymbol{\sigma}, \tag{64}$$

and

$$u_{\boldsymbol{\mu}}(\boldsymbol{x}, \boldsymbol{\sigma}) = \sum_{r=1}^{N} \chi^{(r)}(\boldsymbol{x}) u_{\boldsymbol{\mu}}^{(r)}(\boldsymbol{x}, \boldsymbol{\sigma}), \quad u_{\boldsymbol{\mu}}^{(r)}(\boldsymbol{x}, \boldsymbol{\sigma}) = u^{(r)}(\boldsymbol{x}, \boldsymbol{\sigma}) + \frac{1}{2} \boldsymbol{\sigma} : \boldsymbol{\mu}^{(r)} : \boldsymbol{\sigma}. \tag{65}$$

Then,

$$\langle \boldsymbol{\sigma} \rangle^{(r)} = \frac{1}{c^{(r)}} \frac{\partial \widetilde{u}_{\boldsymbol{\eta}}}{\partial \boldsymbol{\eta}^{(r)}}(\overline{\boldsymbol{\sigma}})|_{\boldsymbol{\eta}=\boldsymbol{0}}, \quad \langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)} = \frac{2}{c^{(r)}} \frac{\partial \widetilde{u}_{\boldsymbol{\mu}}}{\partial \boldsymbol{\mu}^{(r)}}(\overline{\boldsymbol{\sigma}})|_{\boldsymbol{\mu}=\boldsymbol{0}}. \tag{66}$$

It is often convenient to compute the above moments of the stress for a given macroscopic strain $\overline{\boldsymbol{\varepsilon}}$, by means of the primal potential w. Following Idiart & Ponte Castañeda (2007), one can introduce the Legendre transforms $w_{\boldsymbol{\eta}}$ and $w_{\boldsymbol{\mu}}$ of $u_{\boldsymbol{\eta}}$ and $u_{\boldsymbol{\mu}}$ and note (as in proposition 3.4 and corollary 3.7 of Idiart & Ponte Castañeda (2007)) that:

$$\langle \boldsymbol{\sigma} \rangle^{(r)} = -\frac{1}{c^{(r)}} \frac{\partial \widetilde{w}_{\boldsymbol{\eta}}}{\partial \boldsymbol{\eta}^{(r)}}(\overline{\boldsymbol{\varepsilon}})|_{\boldsymbol{\eta}=\boldsymbol{0}}, \quad \langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)} = -\frac{2}{c^{(r)}} \frac{\partial \widetilde{w}_{\boldsymbol{\mu}}}{\partial \boldsymbol{\mu}^{(r)}}(\overline{\boldsymbol{\sigma}})|_{\boldsymbol{\mu}=\boldsymbol{0}}. \tag{67}$$

Similar relations hold for the first and second moment of the stress field in the comparison composite. It should be noted that the error function $\mathcal{V}^{(r)}$ between $w_{\boldsymbol{\eta}}^{(r)}$ and $w_{0,\boldsymbol{\eta}}^{(r)}$ is, up to a sign, the same as the error function between $u_{\boldsymbol{\eta}}^{(r)}$ and $u_{0,\boldsymbol{\eta}}^{(r)}$ (see (52)) and, since these potentials are obtained from $u^{(r)}$ and $u_0^{(r)}$ by the same perturbation term, this error function is the original function $\mathcal{V}^{(r)}$ between $w^{(r)}$ and $w_0^{(r)}$. This leads to variational estimates for the perturbed potentials:

$$\widetilde{w}_{\boldsymbol{\eta}}(\overline{\boldsymbol{\varepsilon}}) \simeq \sup_{\boldsymbol{\tau}_0^{(r)}, \boldsymbol{L}_0^{(r)}} \left\{ \widetilde{w}_{0,\boldsymbol{\eta}}(\overline{\boldsymbol{\varepsilon}}) + \langle \mathcal{V}(\boldsymbol{x}, \boldsymbol{\tau}_0, \boldsymbol{L}_0) \rangle \right\}, \tag{68}$$

with a similar relation for $\widetilde{w}_{\boldsymbol{\mu}}$. Taking the derivative of the above relation with respect to $\boldsymbol{\eta}^{(r)}$ (which does not appear in $\langle \mathcal{V} \rangle$) and of the similar relation for $\widetilde{w}_{\boldsymbol{\mu}}$ with respect to $\boldsymbol{\mu}$, gives:

$$\overline{\boldsymbol{\sigma}}^{(r)} \simeq \overline{\boldsymbol{\sigma}}_0^{(r)}, \quad \langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)} \simeq \langle \boldsymbol{\sigma}_0 \otimes \boldsymbol{\sigma}_0 \rangle^{(r)}. \tag{69}$$

In conclusion, the variational estimates for the first and second moments of the local strain and stress fields coincide with those in the LCC.

5 Application to rigidly reinforced composites

With the objective of illustrating the general results of the previous sections, we provide in this section an application to rigidly reinforced composites with a prescribed distribution of non-uniform polarization field in the matrix phase. For simplicity, the microstructure will be taken to consist of aligned, cylindrical fibers of the rigid phase distributed randomly in a uniform elastic matrix material. Assuming that the modulus tensor of the matrix phase is invariant under a reflection about the fiber direction, the three-dimensional elasticity problem is known (Hill (1964)) to reduce to uncoupled anti-plane strain and generalized plane strain problems. Motivated by this result, we restrict our attention here to the two-dimensional problem defined by plane strain loading of the fiber-reinforced composite in the transverse plane. The matrix elasticity tensor will be further taken to be incompressible and to have the special orthogonal anisotropy (see Ponte Castañeda (2002b)) defined by

$$\boldsymbol{L}^{(1)} = 2\lambda \boldsymbol{E} + 2\mu \boldsymbol{F}, \tag{70}$$

where $\boldsymbol{E}$ and $\boldsymbol{F}$ are orthogonal projection operators, such that $\boldsymbol{E} : \boldsymbol{F} = \boldsymbol{F} : \boldsymbol{E} = \mathbf{0}$, $\boldsymbol{E} : \boldsymbol{E} = \boldsymbol{E}$, $\boldsymbol{F} : \boldsymbol{F} = \boldsymbol{F}$, $\boldsymbol{E} + \boldsymbol{F} = \mathbf{K}$, and $\mathbf{K}$ is the identity in the space of incompressible, fourth-order tensors in two dimensions. Note that a general representation for $\boldsymbol{E}$ may be given in terms of deviatoric, second-order tensors $\hat{\boldsymbol{\epsilon}}_{\|}$ with $\hat{\boldsymbol{\epsilon}}_{\|} : \hat{\boldsymbol{\epsilon}}_{\|} = 1$, such that $\boldsymbol{E} = \hat{\boldsymbol{\epsilon}}_{\|} \otimes \hat{\boldsymbol{\epsilon}}_{\|}$. In addition, it is assumed that the matrix is subjected to a distribution of polarization $\boldsymbol{\tau}^{(1)}(\mathbf{x})$ with prescribed phase average $\overline{\boldsymbol{\tau}}^{(1)}$, and fluctuations $\boldsymbol{C}^{(1)}(\boldsymbol{\tau})$.

The computation of the various variational estimates requires a 'comparison' composite with the same microstructure as the above-described composite, where the matrix phase now has a modulus tensor

$$\boldsymbol{L}_0^{(1)} = 2\lambda_0 \boldsymbol{E} + 2\mu_0 \boldsymbol{F}, \tag{71}$$

and a constant polarization $\boldsymbol{\tau}_0^{(1)}$, while the fiber phase is still rigid. As discussed in remark 4 of section 2, the effective energy of a two-phase composite with constant-per-phase moduli and polarization is completely determined (via expressions (28) and (29)) by the effective modulus tensor $\widetilde{\boldsymbol{L}}_0$ of the corresponding two-phase elastic composite. However, for the case of rigid fibers, the results simplify dramatically, and we obtain

$$\widetilde{\boldsymbol{\tau}}_0 = \boldsymbol{\tau}_0^{(1)}, \quad \text{and} \quad \widetilde{f}_0 = 0. \tag{72}$$

Further assuming that the fibers have circular cross-section and are distributed isotropically in the transverse plane, the following estimate of Hashin-Shtrikman type is available (see Ponte Castañeda & Willis (1995)) for the elasticity tensor of the comparison composite

$$\widetilde{\boldsymbol{L}}_0 = \boldsymbol{L}_0^{(1)} + \frac{c}{1-c}\left(\mathbf{P}_0^{(1)}\right)^{-1}, \tag{73}$$

where c is the volume fraction of the fibers, and the Eshelby-type, microstructural tensor $\mathbf{P}_0^{(1)}$ is determined by

$$\mathbf{P}_0^{(1)} = \frac{1}{4\pi}\int_{|\boldsymbol{\xi}|\,=\,1} \mathbf{H}_0^{(1)}(\boldsymbol{\xi})\; dS(\boldsymbol{\xi}), \tag{74}$$

where $H^{(1)}_{0\,ijkh}(\boldsymbol{\xi}) = N^{(1)}_{0\,ik}\,\xi_j\xi_h|_{(ij)(kh)}$, with $\mathbf{N}^{(1)}_0 = {\mathbf{K}^{(1)}_0}^{-1}$, and $K^{(1)}_{0\,ik} = L^{(1)}_{0\,ijkh}\xi_j\xi_h$. The expression (73) is in general a lower bound for the effective stiffness of all isotropic rigidly-reinforced composites. In addition it is known to be attained for specific microstructures and it will assumed in the sequel that the composite under consideration has such a microstructure. Then, making use of expression (71), it follows (Ponte Castañeda (2002b)) that

$$\mathbf{P}^{(1)}_0 = \frac{1}{2(\lambda_0+\sqrt{\mu_0\lambda_0})}\boldsymbol{E} + \frac{1}{2(\mu_0+\sqrt{\mu_0\lambda_0})}\boldsymbol{F}, \tag{75}$$

and also that

$$\widetilde{\boldsymbol{L}}_0 = \frac{2\lambda_0}{1-c}\left(1+c\sqrt{\frac{\mu_0}{\lambda_0}}\right)\boldsymbol{E} + \frac{2\mu_0}{1-c}\left(1+c\sqrt{\frac{\lambda_0}{\mu_0}}\right)\boldsymbol{F}. \tag{76}$$

In conclusion, assuming that the applied strain $\overline{\boldsymbol{\varepsilon}}$ is co-axial with the material anisotropy tensor $\hat{\boldsymbol{\epsilon}}_{\|}$, and defining the equivalent strain via $\overline{\varepsilon}^2_e = (2/3)\overline{\boldsymbol{\varepsilon}} : \overline{\boldsymbol{\varepsilon}}$, the energy function of the comparison composite is given by

$$\widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) = \frac{3}{2}\left(\frac{\lambda_0}{1-c}\right)\left(1+c\sqrt{\frac{\mu_0}{\lambda_0}}\right)\overline{\varepsilon}^2_e + \boldsymbol{\tau}^{(1)}_0 : \overline{\boldsymbol{\varepsilon}}, \tag{77}$$

which is seen to be a function of λ_0, μ_0 and $\boldsymbol{\tau}^{(1)}_0$. In addition, the phase average and fluctuations of the strain may be easily computed (see Idiart et al. (2006)), and are given by

$$\langle\boldsymbol{\varepsilon}_0\rangle^{(1)} = \frac{1}{1-c}\overline{\boldsymbol{\varepsilon}}_0, \quad \text{and} \quad \boldsymbol{C}^{(1)}(\boldsymbol{\varepsilon}) = \frac{3}{4}\frac{c}{(1-c)^2}\left(\sqrt{\frac{\mu_0}{\lambda_0}}\boldsymbol{E} + \sqrt{\frac{\lambda_0}{\mu_0}}\boldsymbol{F}\right)\overline{\varepsilon}^2_e. \tag{78}$$

The result (77) may be interpreted as a lower bound for fiber-reinforced composites with the special classes of microstructures described earlier. Alternatively, it can be viewed as an exact estimate for those members of the above class that attain this bound, since it is known that the bound is attainable (Milton (2002)). In the discussion below, we take this second point of view and make use of the estimate (77), together with the theory developed in section 3, to obtain 'upper' and 'lower' bounds for the corresponding effective energy of the fiber-reinforced composite with a non-uniform distribution of polarization in the matrix phase.

Thus, assuming that $\lambda_0 \leq \lambda$ and $\mu_0 \leq \mu$ (so that $\boldsymbol{L}^{(1)}_0 \leq \boldsymbol{L}^{(1)}$), it follows from expression (40) that

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}}) \geq \sup_{\boldsymbol{\tau}^{(1)}_0, \boldsymbol{L}^{(1)}_0} \left\{ \widetilde{w}_0(\overline{\boldsymbol{\varepsilon}}) - \frac{(1-c)}{2}\left\langle\left(\boldsymbol{\tau}^{(1)}-\boldsymbol{\tau}^{(1)}_0\right) : \left(\boldsymbol{L}^{(1)}-\boldsymbol{L}^{(1)}_0\right)^{-1} : \left(\boldsymbol{\tau}^{(1)}-\boldsymbol{\tau}^{(1)}_0\right)\right\rangle^{(1)}\right\}. \tag{79}$$

Then, using expressions (70) and (71) for $\boldsymbol{L}^{(1)}$ and $\boldsymbol{L}^{(1)}_0$, respectively, and the pertinent stationarity conditions (41) and (42) (or (46)), we arrive at the following equations for $\boldsymbol{\tau}^{(1)}_0$, λ_0 and μ_0, namely,

$$\boldsymbol{\tau}^{(1)}_0 = \overline{\boldsymbol{\tau}}^{(1)} + \frac{2(\lambda-\lambda_0)}{1-c}\overline{\boldsymbol{\varepsilon}}, \tag{80}$$

$$\frac{3c}{(1-c)^2}\sqrt{\frac{\mu_0}{\lambda_0}}\,(\lambda-\lambda_0)^2\,\overline{\varepsilon}_e^2=\left\langle\left(\boldsymbol{\tau}^{(1)}-\overline{\boldsymbol{\tau}}^{(1)}\right):\boldsymbol{E}:\left(\boldsymbol{\tau}^{(1)}-\overline{\boldsymbol{\tau}}^{(1)}\right)\right\rangle^{(1)}, \tag{81}$$

$$\frac{3c}{(1-c)^2}\sqrt{\frac{\lambda_0}{\mu_0}}\,(\mu-\mu_0)^2\,\overline{\varepsilon}_e^2=\left\langle\left(\boldsymbol{\tau}^{(1)}-\overline{\boldsymbol{\tau}}^{(1)}\right):\boldsymbol{F}:\left(\boldsymbol{\tau}^{(1)}-\overline{\boldsymbol{\tau}}^{(1)}\right)\right\rangle^{(1)}. \tag{82}$$

When solutions are found to the system of the two equations (81) and (82) for $\lambda_0\leq\lambda$ and $\mu_0\leq\mu$ (note that $\boldsymbol{\tau}_0^{(1)}$ is determined in terms of λ_0 from (80)), and the results are used in the right-hand-side of (79), a 'lower' bound is obtained for the effective energy $\widetilde{w}$ of the fiber-reinforced composite with a non-uniform distribution of polarization in the matrix phase. On the other hand, according to what was stated in remark 1 of section 3, when solutions of the system (81) and (82) with $\lambda_0\geq\lambda$ and $\mu_0\geq\mu$ are obtained, the expression in the right-hand-side of (79) provides an 'upper' bound for the effective energy $\widetilde{w}$ of the fiber-reinforced composite with a non-uniform distribution of polarization in the matrix phase. In addition, the corresponding estimates for the macroscopic stress may be obtained from expression (48) such that

$$\overline{\boldsymbol{\sigma}}=\frac{2}{1-c}\left(\lambda+c\sqrt{\mu_0\lambda_0}\right)\overline{\boldsymbol{\varepsilon}}+\overline{\boldsymbol{\tau}}^{(1)}, \tag{83}$$

where we have already made use of expression (80) for $\boldsymbol{\tau}_0^{(1)}$ to simplify the result. Thus, this expression provides a 'lower' estimate (not necessarily a bound) for the stress when solutions such that $\lambda_0\leq\lambda$ and $\mu_0\leq\mu$ are obtained from expressions (81) and (82), as well as an 'upper' estimate (again not necessarily a bound) for the stress when solutions such that $\lambda_0\geq\lambda$ and $\mu_0\geq\mu$ are obtained from expressions (81) and (82).

Given that our interest here is to explore the effect of fluctuations in the distribution of the polarization field, we will assume in the illustrative example below that the matrix phase is isotropic, so that $\lambda=\mu$, and that the polarization field has zero average, so that $\overline{\boldsymbol{\tau}}^{(1)}=\mathbf{0}$, and is co-axial with the direction defined by the applied field $\overline{\boldsymbol{\varepsilon}}$, so that $\boldsymbol{\tau}^{(1)}(\mathbf{x})=\tau^{(1)}(\mathbf{x})\hat{\boldsymbol{\epsilon}}_\parallel$. Then, the 'orthogonal' component ($\boldsymbol{F}$ projection) of the polarization fluctuations vanishes, and defining the 'parallel' component of the polarization via $\tau_\parallel=\boldsymbol{\tau}:\hat{\boldsymbol{\epsilon}}_\parallel$, the parallel fluctuations are described by

$$\overline{\overline{\tau}}_\parallel=\sqrt{\left\langle\tau_\parallel^2\right\rangle^{(1)}}. \tag{84}$$

Under these further assumptions, it follows from (82) that $\mu_0=\mu$, and from (81) that the value of $\lambda_0\leq\mu$ for the 'lower' bound is determined from the solution of the equation

$$\frac{\sqrt{3c}}{1-c}\left(1-\frac{\lambda_0}{\mu}\right)\left(\frac{\mu}{\lambda_0}\right)^{1/4}=\frac{\overline{\overline{\tau}}_\parallel}{\mu\overline{\varepsilon}_e}. \tag{85}$$

The corresponding value of $\lambda_0\geq\mu$ for the 'upper' bound is determined by replacing the factor $1-\lambda_0/\mu$ by $\lambda_0/\mu-1$ in the same equation. Then, the expression (79) for the lower bound on the effective energy of the composite with the non-uniform distribution of polarization specializes to

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}})\geq\frac{1}{1-c}\left[1+\frac{c}{2}\left(\frac{\lambda_0}{\mu}-1\right)\left(\frac{\mu}{\lambda_0}\right)^{1/2}+c\left(\frac{\lambda_0}{\mu}\right)^{1/2}\right]\left(\frac{3}{2}\mu\overline{\varepsilon}_e^2\right), \tag{86}$$

which is a 'lower' bound for the effective energy when the branch of (85) with $\lambda_0 \leq \mu$ is used. On the other hand, an 'upper' bound is provided by the same expression when the $\lambda_0 \geq \mu$ branch is used instead. In addition, the expression for the macroscopic stress (83) reduces to

$$\overline{\sigma}_e = \frac{3\mu}{1-c}\left[1 + c\left(\frac{\lambda_0}{\mu}\right)^{1/2}\right]\overline{\varepsilon}_e, \tag{87}$$

where again the $\lambda_0 \leq \mu$ and $\lambda_0 \geq \mu$ branches of (85) must be used for the stress associated with the 'lower' and 'upper' bounds on the energy (not necessarily bound on the stress). Finally, it is pointed out that estimates for the average and fluctuations of the strain in the matrix phase may be obtained via expressions (78) for the comparison composite with $\mu_0 = \mu$ and the appropriate choice of λ_0.

For comparison purposes, we also include here the corresponding specialization for the Reuss lower bound for this special case of a rigidly reinforced two-dimensional composite, as given by

$$\widetilde{w}^{Reuss}(\overline{\boldsymbol{\varepsilon}}) = \frac{3}{2}\frac{\mu}{1-c}\,\overline{\varepsilon}_e^2 - \frac{1-c}{4\mu}\,\overline{\overline{\tau}}_{\|}^2. \tag{88}$$

Note that this bound grows quadratically with the applied strain $\overline{\varepsilon}_e$—and decreases quadratically with the polarization fluctuations $\overline{\overline{\tau}}_{\|}$. This behavior should be contrasted with the corresponding behavior of the 'lower' and 'upper' bounds, which as discussed in more detail in the next paragraph, is *non-quadratic* in both the applied strain and polarization fluctuations. In addition, we provide the corresponding specialization of the effective energy making use of the 'first moment' approximation (2)

$$\widetilde{w}(\overline{\boldsymbol{\varepsilon}}) = \frac{3\mu}{2}\frac{1+c}{1-c}\,\overline{\varepsilon}_e^2, \tag{89}$$

which is seen to be independent of the polarization fluctuations in view of the fact that $\overline{\boldsymbol{\tau}}^{(1)} = \mathbf{0}$ in this particular example. Note that this estimate lies between the bounds, and agrees exactly with them when the polarization fluctuations vanish.

The results for the above-described composite are shown in Fig. 1 for a fiber volume fraction of 25%. Part (a) shows the value of λ_0, normalized by μ, for the 'lower' and 'upper' bounds, as functions of the dimensionless parameter $\overline{\overline{\tau}}_{\|}/\left(\mu\overline{\varepsilon}_e\right)$. It can be seen that $\lambda_0 \to \mu$ not only when the polarization fluctuations $\overline{\overline{\tau}}_{\|}$ vanish, but also when the applied strain $\overline{\varepsilon}_e$ increases. On the other hand, λ_0 tends to its extreme values of zero and infinity, when either the fluctuations increase or the applied strain decreases. Parts (b) and (c) show the new 'lower' and 'upper' bounds, as determined by (86), as well as the Reuss lower bound (88) and 'first moment' estimate (89) for the effective energy, normalized with respect to $3\overline{\overline{\tau}}_{\|}^2/(2\mu)$ and $(3/2)\mu\overline{\varepsilon}_e^2$, as functions of $\mu\overline{\varepsilon}_e/\overline{\overline{\tau}}_{\|}$ and its inverse $\overline{\overline{\tau}}_{\|}/\left(\mu\overline{\varepsilon}_e\right)$, respectively. The first of these plots illustrates the behavior of the effective energy as a function of the applied strain for fixed polarization fluctuations, while the second shows the corresponding behavior of the effective energy as a function of the polarization fluctuations, for fixed applied strain. Thus, it can be seen that while the new 'lower' bound improves on the Reuss lower bound only for sufficiently large strains, the opposite behavior is observed as a function of the polarization fluctuations. In fact, it can be shown that the new 'lower' bound tends to the Reuss lower bound as $\mu\overline{\varepsilon}_e/\overline{\overline{\tau}}_{\|} \to 0$, so that both lower bounds on the effective energy are seen to tend to $-\infty$ quadratically with the polarization fluctuations, in agreement with the comments provided in remark 2 of section 2. On the other hand, for the microstructures for which the HS estimate (73) is attained, the new 'upper' bound is significantly tighter than the classical Voigt bound (which corresponds to a much larger class of microstructures and happens to be infinite in this case). Thus, for fixed polarizations, the range of possible behaviors—as determined by the bounds on the effective energy of the composite—remains bounded as the strain increases, although the differences continue to increase in magnitude as the polarization fluctuations increase. We also observe that the 'first moment' estimate satisfies the bounds, but as already mentioned, it is completely insensitive to the polarization fluctuations. Finally, part (d) show plots of the macroscopic stress, normalized by the applied strain, as a function of $\overline{\overline{\tau}}_{\|}/\left(\mu\bar{\varepsilon}_e\right)$. Although the results for the 'upper' and 'lower' estimates are not necessarily bounds, it can be seen that the stress (unlike the effective energy) associated with the 'lower' bound does not decrease without a bound with increasing polarization fluctuations, tending to the Reuss lower estimate as the polarization fluctuations increase. This makes it clear that the quadratic drop in the effective energy with the polarization fluctuations arises from the polarization terms in the energy and not from the strain terms.

6 Concluding remarks

In this work, variational approximations have been developed to estimate the macroscopic response, as well as the phase averages and covariance of the fluctuations of the stress and strain fields in elastic composite materials with non-uniform distributions of polarization and/or moduli. The approximation consists in the replacement of the actual composite by a 'comparison' composite with uniform, per-phase polarization and moduli that are determined by a suitably designed variational statement. When the moduli of the actual composite are uniform per phase, the resulting estimates for the macroscopic behavior are found to depend on the first and second moments of the polarization distributions in each phase. On the other hand, it has been shown that the first and second moments of the stress and strain fields in the actual composite may be estimated consistently from the corresponding first and second moments of the stress and strain fields in the comparison composite. Furthermore, when the modulus tensor of the comparison composite is chosen to be greater (lower) than the modulus of the actual composite (in the sense of quadratic forms), the resulting estimate for the effective free energy of the composite is shown to be an upper (lower) bound. Although this was not explored in this work, it is also possible to make other choices for the phase moduli of the comparison composite (see Ponte Castañeda (2002a) and Lahellec & Suquet (2007b)), also leading to stationary variational estimates, but which may not necessarily be bounds. Such possibilities are application-dependent and will continue to be explored in future work.

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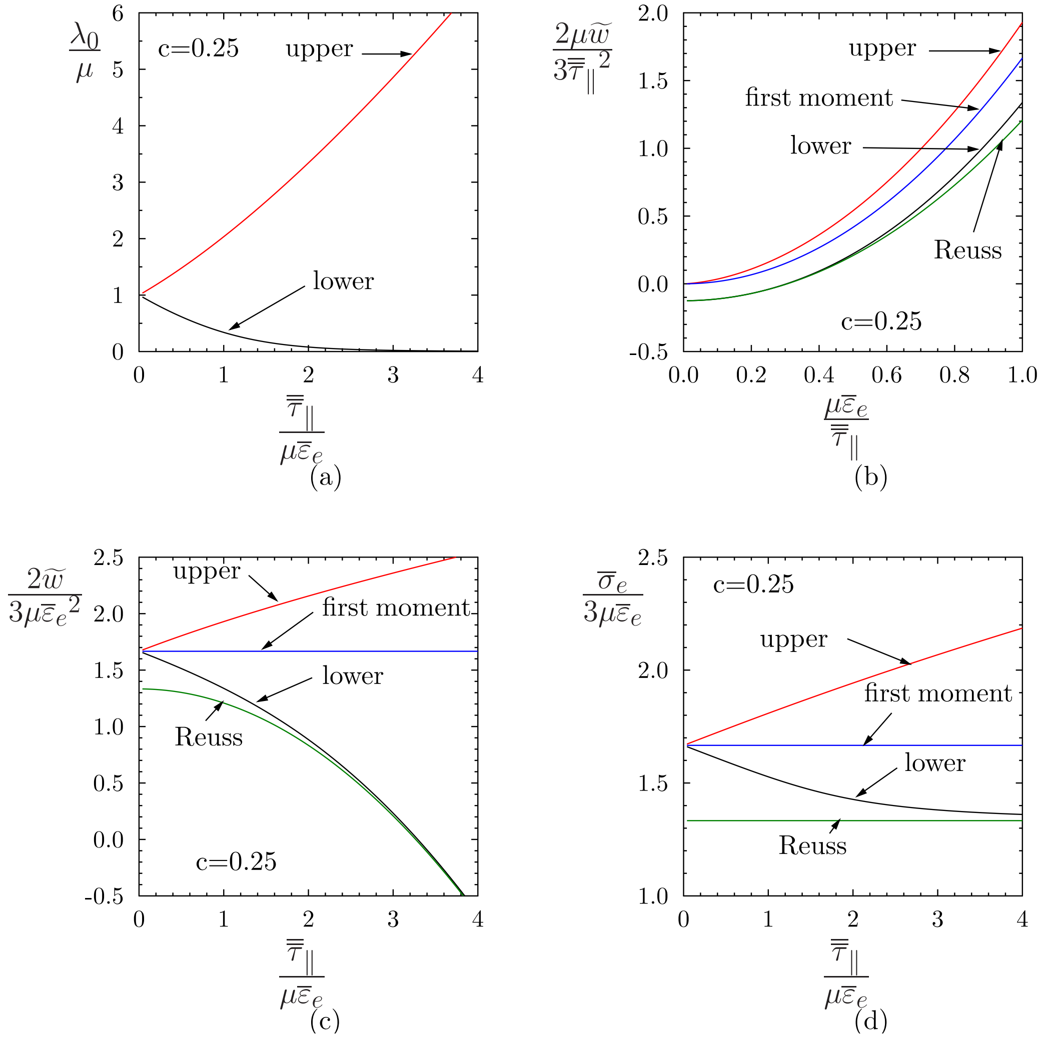

Fig. 1. (a) Solutions for λ_0 of 'comparison' composite for 'lower' and 'upper' bounds, as function of applied strain $\bar{\varepsilon}_e$ and parallel polarization fluctuation $\overline{\overline{\tau}}_{\|}$. (b) Comparison of the new 'lower' and 'upper' bound for the effective energy with the Reuss lower bound and 'first moment' estimate, plotted as functions of the applied strain (for fixed polarization fluctuation). (c) Comparison of the new 'lower' and 'upper' bound for the effective energy with the Reuss lower bound and 'first moment' estimate, plotted as functions of the polarization fluctuation (for fixed applied strain). (d) Comparison of the new 'lower' and 'upper' estimates with the Reuss and 'first moment' estimates for the normalized stress, as functions of the polarization fluctuation (for fixed applied strain).

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Appendix: Unidimensional case

In dimension 1, the constitutive relations read as

$$\sigma(x) = \ell(x) : \varepsilon(x) + \tau(x)$$

where both ℓ and τ may have fluctuations within a single phase. The equilibrium equations, the compatibility conditions and the boundary conditions take the form

$$\frac{d\sigma}{dx} = 0, \quad \varepsilon(x) = \frac{du}{dx}(x), \quad u(0) = 0, \quad u(L) = \overline{\varepsilon}L.$$

It follows from the equilibrium equation that the Reuss model is exact $\sigma(x) = \overline{\sigma}$. Therefore:

$$\widetilde{w}(\overline{\varepsilon}) = \frac{1}{2}\frac{1}{\langle m\rangle}\overline{\varepsilon}^2 + \frac{\langle m\tau\rangle}{\langle m\rangle}\overline{\varepsilon} + \frac{1}{2}\frac{\langle m\tau\rangle^2}{\langle m\rangle} - \frac{1}{2}\left\langle m\tau^2\right\rangle$$

and

$$\overline{\sigma} = \frac{1}{\langle m\rangle}\overline{\varepsilon} + \frac{\langle m\tau\rangle}{\langle m\rangle}.$$

In the LCC with piecewise uniform moduli and polarization fields, there is no strain fluctuation in the phases. Therefore the variational procedure is not applicable in this case and a direct approach is preferable. A piecewise uniform LCC will give the exact energy and the exact overall stress iff

$$\langle m_0\rangle = \langle m\rangle, \quad \langle m_0\tau_0\rangle = \langle m\tau\rangle, \quad \left\langle m_0\tau_0^2\right\rangle = \left\langle m\tau^2\right\rangle. \tag{90}$$

These 3 equations have many different piecewise solutions $m_0^{(r)}$ and $\tau_0^{(r)}$. Let us give one of them for which the elastic moduli in the LCC coincide with those in the actual composite (where the elastic moduli are assumed to be piecewise uniform). The LCC is searched such that $m_0^{(r)} = m^{(r)}$. (90) gives:

$$\left.\begin{aligned} c^{(1)}m^{(1)}\tau_0^{(1)} + c^{(2)}m^{(2)}\tau_0^{(2)} &= \langle m\tau\rangle \\ c^{(1)}m^{(1)}(\tau_0^{(1)})^2 + c^{(2)}m^{(2)}(\tau_0^{(2)})^2 &= \langle m\tau^2\rangle \end{aligned}\right\}$$

This system has two solutions:

$$\tau_0^{(1)} = \frac{\langle m\tau\rangle}{\langle m\rangle} \pm \sqrt{\frac{c^{(2)}m^{(2)}}{c^{(1)}m^{(1)}}\left[\frac{\langle m\tau^2\rangle}{\langle m\rangle} - \frac{\langle m\tau\rangle^2}{\langle m\rangle^2}\right]}, \quad \tau_0^{(2)} = \frac{\langle m\tau\rangle}{\langle m\rangle} - \pm\sqrt{\frac{c^{(1)}m^{(1)}}{c^{(2)}m^{(2)}}\left[\frac{\langle m\tau^2\rangle}{\langle m\rangle} - \frac{\langle m\tau\rangle^2}{\langle m\rangle^2}\right]},$$

where the $\pm$ signs are the same in both expressions. These solutions differ from $\langle\tau\rangle^{(1)}$ and $\langle\tau\rangle^{(2)}$ respectively.