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# Attainability of the Hashin–Shtrikman bounds for two-phase well-ordered composites with a nonlinear phase

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## ARTICLE INFO

## Article history:

Received 25 June 2013

Accepted after revision 8 October 2013

Available online 24 October 2013

## Keywords:

Composites

Homogenization

Bounds

Nonlinearity

## ABSTRACT

The Hashin–Shtrikman (HS) bounds for two-phase well-ordered composites are known to be attained by certain sequentially laminated constructions when the constituent phases exhibit a *linear* behavior. This implies that the bounds are optimal for that class of materials. In this Note we show that the bounds are still attained by sequentially laminated constructions when one of the phases is *nonlinear*, and that, consequently, they are optimal for a larger class of materials.

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## 1. The effective behavior of two-phase nonlinear composites

The focus of this Note is on the viscoplastic response of two-phase composites exhibiting well-separated microstructural length scales. The instantaneous response of the constituent phases is characterized by dissipation potentials  $w^{(r)}$  ( $r = 1, 2$ ) such that the Cauchy stress  $\boldsymbol{\sigma}$  and the Eulerian strain rate  $\boldsymbol{\epsilon}$  tensors are related by:

$$\boldsymbol{\sigma} = \frac{\partial w}{\partial \boldsymbol{\epsilon}}(\mathbf{x}, \boldsymbol{\epsilon}), \quad w(\mathbf{x}, \boldsymbol{\epsilon}) = \sum_{r=1}^2 \theta^{(r)}(\mathbf{x}) w^{(r)}(\boldsymbol{\epsilon}) \quad (1)$$

where the characteristic functions  $\theta^{(r)}$  serve to describe the microstructure in the current configuration, being 1 if the position vector  $\mathbf{x}$  is in phase  $r$ , and 0 otherwise. It is further assumed, as usual, that the potentials are differentiable, convex, with subquadratic growth at infinity, and such that  $w^{(r)}(\boldsymbol{\epsilon}) \geq 0$  and  $w^{(r)}(\mathbf{0}) = 0$ .

Homogenization theory states that the *effective behavior* of the composite is given by the relation between the average Cauchy stress and the average strain rate over a ‘representative volume element’  $\Omega$ , and that it can be characterized by an *effective dissipation potential*  $\tilde{w}$  such that—see, for instance, Refs. [1,2]:

$$\bar{\boldsymbol{\sigma}} = \frac{\partial \tilde{w}}{\partial \bar{\boldsymbol{\epsilon}}}(\bar{\boldsymbol{\epsilon}}), \quad \tilde{w}(\bar{\boldsymbol{\epsilon}}) = \min_{\boldsymbol{\epsilon} \in \mathcal{K}(\bar{\boldsymbol{\epsilon}})} \langle w(\mathbf{x}, \boldsymbol{\epsilon}) \rangle \quad (2)$$

where  $\bar{\boldsymbol{\sigma}} \equiv \langle \boldsymbol{\sigma} \rangle$  and  $\bar{\boldsymbol{\epsilon}} \equiv \langle \boldsymbol{\epsilon} \rangle$  with  $\langle \cdot \rangle$  denoting volume averaging over  $\Omega$ , and  $\mathcal{K}(\bar{\boldsymbol{\epsilon}})$  is the set of admissible fields  $\boldsymbol{\epsilon}$ , such that there exists a continuous velocity field  $\mathbf{u}$  satisfying  $\boldsymbol{\epsilon} = \nabla \otimes_s \mathbf{u}$  in  $\Omega$  and  $\mathbf{u} = \bar{\boldsymbol{\epsilon}} \mathbf{x}$  on  $\partial\Omega$ . The optimal velocity field in (2) solves the viscoplasticity equations in  $\Omega$  with a constitutive law (1).

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## 2. Upper bound of the Hashin–Shtrikman type

Given a set of potentials  $w^{(r)}$  and of characteristic functions  $\theta^{(r)}$ , an upper bound for  $\tilde{w}$  can be derived by introducing a set of functions defined by:

$$v^{(r)}(\mathbf{L}_0^{(r)}) \doteq \sup_{\boldsymbol{\varepsilon}} \left[ w^{(r)}(\boldsymbol{\varepsilon}) - \frac{1}{2} \boldsymbol{\varepsilon} \cdot \mathbf{L}_0^{(r)} \boldsymbol{\varepsilon} \right], \quad r = 1, 2 \tag{3}$$

where  $\mathbf{L}_0^{(r)}$  are fourth-order viscosity tensors. Then,

$$w^{(r)}(\boldsymbol{\varepsilon}) \leq \left[ \frac{1}{2} \boldsymbol{\varepsilon} \cdot \mathbf{L}_0^{(r)} \boldsymbol{\varepsilon} + v^{(r)}(\mathbf{L}_0^{(r)}) \right] \tag{4}$$

for any  $\boldsymbol{\varepsilon}$  and  $\mathbf{L}_0^{(r)}$ , and therefore,

$$\tilde{w}(\bar{\boldsymbol{\varepsilon}}) \leq \inf_{\mathbf{L}_0^{(r)} \geq 0} \left[ \frac{1}{2} \bar{\boldsymbol{\varepsilon}} \cdot \tilde{\mathbf{L}}_0 \bar{\boldsymbol{\varepsilon}} + \sum_{r=1}^2 c^{(r)} v^{(r)}(\mathbf{L}_0^{(r)}) \right] \tag{5}$$

where the  $c^{(r)} = \langle \theta^{(r)}(\mathbf{x}) \rangle$  denote the volume fractions of each phase  $r$  in  $\Omega$ —such that  $0 \leq c^{(r)} \leq 1$ ,  $c^{(1)} + c^{(2)} = 1$ —, and  $\tilde{\mathbf{L}}_0$  is the effective viscosity tensor of a linear composite with the same microstructure  $\theta^{(r)}$  as the nonlinear composite, but with a local linear behavior characterized by viscosity tensors  $\mathbf{L}_0^{(r)}$ . Various derivations of this bound have been given by Ponte Castañeda [3,4], Willis [5], Talbot & Willis [6], and Idiart & Ponte Castañeda [7], among others.

The first term in (5) can be bounded from above by the linear Hashin–Shtrikman bounds of Willis [8]. These bounds are a generalization of the original bounds of Hashin & Shtrikman [9], and can be written in terms of an effective viscosity tensor:

$$\tilde{\mathbf{L}}_{\text{HS}} = \left( \sum_{r=1}^2 c^{(r)} [\mathbf{I} + (\mathbf{L}_0^{(r)} - \mathbf{L}_0) \mathbf{P}_0] \right)^{-1} \left( \sum_{r=1}^2 c^{(r)} [\mathbf{I} + (\mathbf{L}_0^{(r)} - \mathbf{L}_0) \mathbf{P}_0]^{-1} \mathbf{L}_0^{(r)} \right) \tag{6}$$

where

$$\mathbf{P}_0 = \int_{|\mathbf{n}|=1} \mathbf{H}_0(\mathbf{n}) \nu(\mathbf{n}) \, d\mathbf{n}, \quad H_{0ijkl} = K_{ik}^{-1}(\mathbf{n}) n_j n_l |(ij)(kl), \quad K_{ik} = L_{0ijkl} n_j n_l \tag{7}$$

is a microstructural tensor that depends on the  $H$ -measure  $\nu(\mathbf{n})$  of the composite microstructure.  $H$ -measures are geometrical objects—introduced as such by Tartar [10] and Gérard [11]—that depend on the two-point microstructural correlations  $\langle \theta^{(r)}(\mathbf{x}) \theta^{(s)}(\mathbf{x} + \mathbf{z}) \rangle$ ; they quantify in the phase space the lack of compactness of weakly converging sequences of characteristic functions  $[\theta^{(r)}(\mathbf{x}) - c^{(r)}]$ —see Ref. [12]—and provide a partial characterization of the microstructural oscillations along different directions in the physical space. Explicit expressions of  $\nu(\mathbf{n})$  for different types of microstructures can be found, for instance, in [8,12,13]. In any event, the effective viscosity tensor (6) is such that:

$$\tilde{\mathbf{L}}_0 \leq \tilde{\mathbf{L}}_{\text{HS}} \quad \text{for any } \mathbf{L}_0 \text{ satisfying } \mathbf{L}_0^{(r)} \leq \mathbf{L}_0 \text{ for all } r \tag{8}$$

inequalities that can be interpreted in the sense of quadratic forms. Introducing (6) in (5) leads to a nonlinear upper bound of the Hashin–Shtrikman type of the form [6,7]:

$$\tilde{w}(\bar{\boldsymbol{\varepsilon}}) \leq \tilde{w}_{\text{HS}}(\bar{\boldsymbol{\varepsilon}}) \doteq \inf_{\mathbf{L}_0^{(r)} \geq 0} \left[ \frac{1}{2} \bar{\boldsymbol{\varepsilon}} \cdot \tilde{\mathbf{L}}_{\text{HS}} \bar{\boldsymbol{\varepsilon}} + \sum_{r=1}^2 c^{(r)} v^{(r)}(\mathbf{L}_0^{(r)}) \right] \tag{9}$$

for any  $\mathbf{L}_0$  satisfying (8). The function  $\tilde{w}_{\text{HS}}(\bar{\boldsymbol{\varepsilon}})$  bounds from above the effective potential of any composite within the class  $\mathcal{C}(w^{(r)}, c^{(r)}, \nu)$  of two-phase composites with prescribed local potentials  $w^{(r)}$ , volume fractions  $c^{(r)}$ , and  $H$ -measure  $\nu(\mathbf{n})$ .

## 3. A class of two-phase nonlinear composites with an attainable upper bound

When the local responses are *linear* and well-ordered, the bound  $\tilde{w}_{\text{HS}}$  is known to be attained for all possible values of  $c^{(r)}$  and  $\nu(\mathbf{n})$  by certain sequentially laminated constructions—see, for instance, Refs. [1,14] and references therein. The attainability of the bound when one of the phases is *nonlinear* has been demonstrated by Ponte Castañeda [4,15] for isotropic potentials  $w^{(r)}$  that depend on the first and second invariants of  $\boldsymbol{\varepsilon}$ . In this section, we show that the bound  $\tilde{w}_{\text{HS}}$  is attainable for the more general class  $\mathcal{S}(w^{(r)}, c^{(r)}, \nu) \subset \mathcal{C}(w^{(r)}, c^{(r)}, \nu)$  of two-phase composites with potentials  $w^{(r)}$  such that:

–  $w^{(1)}$  is quadratic:

$$w^{(1)}(\boldsymbol{\varepsilon}) = \frac{1}{2} \boldsymbol{\varepsilon} \cdot \mathbf{L}^{(1)} \boldsymbol{\varepsilon}, \quad \mathbf{L}^{(1)} > 0 \tag{10}$$

–  $w^{(2)}$  is square-concave<sup>1</sup> and such that

$$w^{(2)}(\boldsymbol{\varepsilon}) \leq w^{(1)}(\boldsymbol{\varepsilon}) \quad \text{for all } \boldsymbol{\varepsilon} \tag{11}$$

Thus,  $\mathcal{S}(w^{(r)}, c^{(r)}, \nu)$  is a class of two-phase composites with well-ordered phases, where the ‘more viscous’ phase exhibits a linear response. Note that both materials may exhibit anisotropic behavior.

We begin the proof by noting that for this class of composites the optimal viscosities  $\mathbf{L}_0^{(r)}$  in the bound  $\tilde{w}_{\text{HS}}$  are such that  $\mathbf{L}_0^{(1)} = \mathbf{L}^{(1)}$  and  $\mathbf{L}_0^{(2)} \leq \mathbf{L}^{(1)}$ . This follows from Lemma 2.1 in Ref. [7] and the fact that the difference  $(w^{(2)} - w^{(1)})(\boldsymbol{\varepsilon})$  is a square-concave function. Then, making use of the choice  $\mathbf{L}_0 = \mathbf{L}^{(1)}$  in  $\tilde{\mathbf{L}}_{\text{HS}}$ , the bound (9) simplifies to:

$$\tilde{w}_{\text{HS}}(\bar{\boldsymbol{\varepsilon}}) = \inf_{\mathbf{0} \leq \mathbf{L}_0^{(2)} \leq \mathbf{L}^{(1)}} \left[ \frac{1}{2} \bar{\boldsymbol{\varepsilon}} \cdot \tilde{\mathbf{L}}_{\text{HS}} \bar{\boldsymbol{\varepsilon}} + c^{(2)} \nu^{(2)}(\mathbf{L}_0^{(2)}) \right] \tag{12}$$

with

$$\tilde{\mathbf{L}}_{\text{HS}} = \mathbf{L}^{(1)} + c^{(2)} [(\mathbf{L}_0^{(2)} - \mathbf{L}^{(1)})^{-1} + c^{(1)} \mathbf{P}^{(1)}]^{-1} \tag{13}$$

where  $\mathbf{P}^{(1)}$  denotes the tensor  $\mathbf{P}_0$  defined by (7) with  $\mathbf{L}_0 = \mathbf{L}^{(1)}$ .

The attainability of the bound (12) is proved next by identifying members of the class  $\mathcal{S}(w^{(r)}, c^{(r)}, \nu)$  whose effective potential agrees exactly with (12). Making use of sequential laminations together with an iterative differential scheme, Idiart [16] has recently constructed two-phase microgeometries with prescribed volume fractions and  $H$ -measures whose effective potential can be computed exactly.<sup>2</sup> These so-called differential laminates may be interpreted as particulate composites consisting of a dispersion of inclusions of one phase distributed within a continuous matrix of the second phase, and are similar in spirit to the microgeometries considered by Ponte Castañeda [4,15] in the context of isotropic composites. When the potential of the matrix and inclusion phases comply with (10) and (11), respectively, the effective potential of these differential laminates solves the Hamilton–Jacobi equation—see Ref. [16]:

$$\frac{\partial \tilde{w}}{\partial t}(\bar{\boldsymbol{\varepsilon}}, t) + H\left(\bar{\boldsymbol{\varepsilon}}, \tilde{w}(\bar{\boldsymbol{\varepsilon}}, t), \frac{\partial \tilde{w}}{\partial \bar{\boldsymbol{\varepsilon}}}(\bar{\boldsymbol{\varepsilon}}, t)\right) = 0, \quad \tilde{w}(\bar{\boldsymbol{\varepsilon}}, 0) = w^{(2)}(\bar{\boldsymbol{\varepsilon}}) \tag{14}$$

with the Hamiltonian:

$$H(\bar{\boldsymbol{\varepsilon}}, \tilde{w}, \bar{\boldsymbol{\sigma}}) = \tilde{w} + \int_{|\mathbf{n}|=1} \max_{\mathbf{a}} \left[ \mathbf{a} \cdot \bar{\boldsymbol{\sigma}} \mathbf{n} - \frac{1}{2} (\bar{\boldsymbol{\varepsilon}} + \mathbf{a} \otimes_s \mathbf{n}) \cdot \mathbf{L}^{(1)} (\bar{\boldsymbol{\varepsilon}} + \mathbf{a} \otimes_s \mathbf{n}) \right] \nu(\mathbf{n}) \, \text{d}\mathbf{n} \tag{15}$$

where  $\nu(\mathbf{n})$  is the  $H$ -measure of the sequentially laminated microgeometry, and the integration with respect to the time-like variable  $t$  must be carried out over the interval  $[0, -\ln c^{(2)}]$ , where  $c^{(2)} = 1 - c^{(1)}$  is the volume fraction of the inclusion phase. Implicit in this result is the assumption that the effective potential is differentiable with respect to  $\bar{\boldsymbol{\varepsilon}}$  and  $t$ . This is expected to be the case since the local potentials have been assumed to be differentiable.

Upon carrying out the maximization in the Hamiltonian (15), the Hamilton–Jacobi equation (14) takes the form:

$$\frac{\partial \Delta \tilde{w}}{\partial t} + \Delta \tilde{w} + \frac{\partial \Delta \tilde{w}}{\partial \bar{\boldsymbol{\varepsilon}}} \cdot \mathbf{P}^{(1)} \frac{\partial \Delta \tilde{w}}{\partial \bar{\boldsymbol{\varepsilon}}} = 0, \quad \Delta \tilde{w}(\bar{\boldsymbol{\varepsilon}}, 0) = \Delta w^{(2)}(\bar{\boldsymbol{\varepsilon}}) \tag{16}$$

where  $\Delta(\cdot) \equiv (\cdot) - (1/2) \bar{\boldsymbol{\varepsilon}} \cdot \mathbf{L}^{(1)} \bar{\boldsymbol{\varepsilon}}$ , and  $\mathbf{P}^{(1)}$  is, once again, the tensor  $\mathbf{P}_0$  defined by (7) with  $\mathbf{L}_0 = \mathbf{L}^{(1)}$ . The solution to this equation can be written as:

$$\tilde{w}(\bar{\boldsymbol{\varepsilon}}, t) = \inf_{\mathbf{0} \leq \mathbf{L}_0^{(2)} \leq \mathbf{L}^{(1)}} \left[ \frac{1}{2} \bar{\boldsymbol{\varepsilon}} \cdot \tilde{\mathbf{L}}_0(\mathbf{L}_0^{(2)}, t) \bar{\boldsymbol{\varepsilon}} + e^{-t} \nu^{(2)}(\mathbf{L}_0^{(2)}) \right] \tag{17}$$

where the function  $\nu^{(2)}(\mathbf{L}_0^{(2)})$  is given by (3), and

$$\tilde{\mathbf{L}}_0(\mathbf{L}_0^{(2)}, t) = \mathbf{L}^{(1)} + e^{-t} [(\mathbf{L}_0^{(2)} - \mathbf{L}^{(1)})^{-1} + (1 - e^{-t}) \mathbf{P}^{(1)}]^{-1} \tag{18}$$

<sup>1</sup> A potential  $w^{(r)}$  is square-concave if there exists a concave function  $f^{(r)}$  in the space of symmetric fourth-order tensors such that  $f^{(r)}(\boldsymbol{\varepsilon} \otimes \boldsymbol{\varepsilon}) = w^{(r)}(\boldsymbol{\varepsilon})$  for all  $\boldsymbol{\varepsilon}$ —see Refs. [2,7]. This class of potentials includes, for instance, subquadratic power-law potentials commonly used to describe viscoplastic solids.

<sup>2</sup> Earlier results for smaller classes of material systems were available from the works of deBotton & Hariton [17] and deBotton [18].

It should be emphasized at this point that  $v^{(2)}$  is a non-smooth function of  $\mathbf{L}_0^{(2)}$ , and so  $\tilde{w}$  as given by expression (17) may not always be differentiable in  $\bar{\boldsymbol{\varepsilon}}$  and  $t$ ; thus, we restrict attention to differential laminates for which  $\tilde{w}$  is differentiable. That (17) satisfies the differential equation (16)<sub>1</sub> can be shown by direct substitution, making use of the identities:

$$\frac{\partial \tilde{w}}{\partial \bar{\boldsymbol{\varepsilon}}}(\bar{\boldsymbol{\varepsilon}}, t) = \tilde{\mathbf{L}}_0(\hat{\mathbf{L}}_0^{(2)}, t)\bar{\boldsymbol{\varepsilon}} \quad \text{and} \quad \frac{\partial \tilde{w}}{\partial t}(\bar{\boldsymbol{\varepsilon}}, t) = \frac{1}{2}\bar{\boldsymbol{\varepsilon}} \cdot \frac{\partial \tilde{\mathbf{L}}_0}{\partial t}(\hat{\mathbf{L}}_0^{(2)}, t)\bar{\boldsymbol{\varepsilon}} - e^{-t}v^{(2)}(\hat{\mathbf{L}}_0^{(2)}) \quad (19)$$

where  $\hat{\mathbf{L}}_0^{(2)}$  denotes the optimal  $\mathbf{L}_0^{(2)}$  maximizing (17). A proof of these identities follows from the facts that their right-hand sides belong to the subdifferentials of the objective function in (17) evaluated at the optimal  $\hat{\mathbf{L}}_0^{(2)}$  and that the function  $\tilde{w}$  is differentiable and convex in  $\bar{\boldsymbol{\varepsilon}}$  and  $e^{-t} \in [0, 1]$ . That (17) further satisfies the ‘initial’ condition (16)<sub>2</sub> at  $t = 0$  is ensured by the assumed square-concavity of the potential  $w^{(2)}$ .

Now, at  $t = -\ln c^{(2)}$ , the effective potential (17) agrees exactly with the bound (12); thus, *the nonlinear bound  $\tilde{w}_{HS}$  of the Hashin–Shtrikman type for the class  $\mathcal{S}(w^{(r)}, c^{(r)}, \nu)$  is attainable and optimal*. The composites attaining the bound have particulate microgeometries, with the linear material being the matrix phase and the less viscous, nonlinear material being the inclusion phase. It can be shown—see Ref. [16]—that in these composites the stress and strain rate fields within the inclusion phase are uniform, which is a well-known feature of extremal composites attaining the Hashin–Shtrikman bounds for linear materials—see Ref. [1]. A consequence of this feature is that when the inclusion concentration is infinitesimally small and the  $H$ -measure  $\nu(\mathbf{n})$  exhibits ellipsoidal symmetry, these composites reproduce exactly the effective potential of a linear matrix with a dilute dispersion of nonlinear ellipsoidal inclusions—cf. Ref. [19]. Of course, they also agree exactly to second order with the small-contrast expansion of Suquet & Ponte Castañeda [20]—see Ref. [16].

### Acknowledgements

This material is based upon work supported by the Agencia Nacional de Promoción Científica y Tecnológica through grant PICT-2011-0167.

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