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Fully optimized second-order variational estimates for the macroscopic response and field statistics in viscoplastic crystalline composites

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A variational method is developed to estimate the macroscopic constitutive response of composite materials consisting of aggregates of viscoplastic single-crystal grains and other inhomogeneities. The method derives from a stationary variational principle for the macroscopic stress potential of the viscoplastic composite in terms of the corresponding potential of a linear comparison composite (LCC), whose viscosities and eigenstrain rates are the trial fields in the variational principle. The resulting estimates for the macroscopic response are guaranteed to be exact to second order in the heterogeneity contrast, and to satisfy known bounds. In addition, unlike earlier 'second-order' methods, the new method allows optimization with respect to both the viscosities and eigenstrain rates, leading to estimates that are fully stationary and exhibit no duality gaps. Consequently, the macroscopic response and field statistics of the nonlinear composite can be estimated directly from the suitably optimized LCC, without the need for difficult-to-compute correction terms. The method is applied to a simple example of a porous single crystal, and the results are found to be more accurate than earlier estimates.

1. Introduction

Macroscopic samples of metals and minerals usually appear in the form of aggregates of large numbers of one or more types of single-crystal grains and other inhomogeneities, such as voids and cracks, which are

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distributed with random positions and orientations in the sample. It is then of scientific and technological value to be able to characterize the *effective* or *average* response of such macroscopic material samples from the properties of their constituents and known statistical information about their distribution, or microstructure. In addition, it is also of interest to be able to extract information about the statistics of the stress and strain fields in the constituents of these composite materials. These problems—which are difficult in general—become especially challenging when the physical mechanisms of deformation are nonlinear, as in metal-forming operations or polar ice flows. The objective of this work is to develop homogenization techniques to characterize the macroscopic response and field statistics in viscoplastic polycrystals and composites.

The simplest and perhaps most commonly used homogenization procedure in polycrystalline plasticity is the uniform strain-rate approximation of Taylor [1]. In the specific context of viscoplastic polycrystals, Hutchinson [2] has shown that the Taylor approximation provides a rigorous upper bound incorporating first-order statistical information in the form of the orientation distribution function. In addition, this author made use of the 'incremental' self-consistent approximation of Hill [3] to generate improved estimates by incorporating additional information about the average shape of the grains.

Improved bounds of the Hashin–Shtrikman [4] type for viscoplastic polycrystals were first developed by Dendievel *et al.* [5], making use of a generalization of the Hashin–Shtrikman variational principles for nonlinear media due to Willis [6,7], as well as by deBotton & Ponte Castañeda [8] by means of the variational method of Ponte Castañeda [9]. This second method makes use of a linear comparison composite (LCC), whose properties are determined by means of a suitable variational principle, leading to a 'secant' linearization of the constitutive response of the grains, evaluated at the *second moments* of the stresses in the grains [10,11]. In addition, the linear comparison variational method has the advantage that it allows the use of other types of bounds and estimates for the LCC. In particular, Nebozhyn & Gilormini [12] proposed 'variational linear comparison' self-consistent estimates for viscoplastic polycrystals and demonstrated that they improved on the earlier 'incremental' self-consistent estimates [3] by showing that these last estimates violate rigorous bounds for sufficiently small rate-sensitivity exponents, especially when the single-crystal grains are highly anisotropic.

More accurate estimates for the macroscopic response of viscoplastic polycrystals were given by Liu & Ponte Castañeda [13] (see also [14]) by means of the 'second-order' homogenization method of Ponte Castañeda [15] and Liu & Ponte Castañeda [16]. The 'second-order' method makes use of more general LCCs incorporating suitably selected eigenstrain rates, leading to an improved 'generalized secant' approximation of the nonlinear constitutive relations and ensuring that the resulting estimates are exact to second order in the heterogeneity contrast, and thus in agreement with the perturbation expansions of Suquet & Ponte Castañeda [17]. In spite of the improved accuracy of the 'second-order' homogenization estimates [16] relative to the earlier 'variational' estimates [8], the 'second-order' estimates have certain undesirable features, arising from the lack of optimality of the eigenstrains, which hamper their efficient application in practice. These include the facts that the macroscopic constitutive relation and fields statistics cannot be obtained directly from the LCC, and the existence of a 'duality gap' (i.e. the estimates resulting from the primary and complementary variational statements are different)—which strongly suggests the possibility of further improvements in the accuracy of its predictions. In this work, we propose a new variational method, where both the viscosities and eigenstrain rates of the constituent phases of the LCC are generated by consistent optimization procedures. This leads to 'full stationarity' for the resulting estimates, which are still exact to second order in the contrast, but have all the advantages of the earlier 'variational' estimates in that the macroscopic constitutive relation and fields statistics of the nonlinear composite can be conveniently expressed in terms of the corresponding quantities for the suitably optimized LCC. Finally, we show explicitly by means of a simple application for a porous single crystal that the new 'fully optimized' second-order estimates do not exhibit a duality gap and provide more accurate estimates than the earlier second-order estimates [16].

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In this paper, scalars are denoted by italic Roman or Greek letters (e.g. a, α), vectors by boldface Roman letters ($\mathbf{b}$), second-order tensors by boldface italic Roman or boldface Greek letters ($\boldsymbol{C}$, $\boldsymbol{\alpha}$) and fourth-order tensors by double-struck letters ($\mathbb{P}$).

2. Theoretical background on nonlinear homogenization

In this work, we are interested in heterogeneous materials occupying regions of space Ω that comprised several (anisotropic) crystalline and possibly other phases, which in turn occupy subregions $\Omega^{(r)}$ ($r=1,\ldots,N$) of Ω that are distributed randomly in location and orientation. The phases could correspond to different orientations of the same crystalline material, or they could correspond to altogether different materials. An example of the first type of heterogeneous material would be a standard polycrystal consisting of aggregates of grains of the same single-crystal material with varying orientations. An example of the second type would be a porous single crystal consisting of voids, or vacuous inclusions, that are distributed uniformly in a single-crystal matrix with given uniform orientation. More general examples would include two-phase polycrystals, porous polycrystals and inclusion-hardened polycrystals. Here, we will make use of indicator functions $\chi^{(r)}$, defined to be equal to 1 if the position vector $\mathbf{x}$ is in $\Omega^{(r)}$ and zero otherwise, to describe the location of the various phases. Note that $\sum_{r=1}^{N}\chi^{(r)}(\mathbf{x})=1$. Furthermore, we make use of the symbols $\langle . \rangle$ and $\langle . \rangle^{(r)}$ to denote volume averages over the composite (Ω) and over phase r ($\Omega^{(r)}$), respectively.

For simplicity, the constitutive behaviour of the single-crystal phases will be taken to be viscoplastic. Then, for a given stress $\boldsymbol{\sigma}$, the *local* constitutive response is defined by

$$\boldsymbol{\epsilon}=\frac{\partial u}{\partial \boldsymbol{\sigma}},\quad u(\mathbf{x},\boldsymbol{\sigma})=\sum_{r=1}^{N}\chi^{(r)}(\mathbf{x})u^{(r)}(\boldsymbol{\sigma}),\quad u^{(r)}(\boldsymbol{\sigma})=\sum_{k=1}^{K^{(r)}}\phi_{(k)}^{(r)}(\tau_{(k)}^{(r)}), \tag{2.1}$$

where $\boldsymbol{\epsilon}$ is the Eulerian strain rate, and u and $u^{(r)}$ are the stress potentials for the composite and for the rth crystalline phase, respectively. The convex functions $\phi_{(k)}^{(r)}$ ($k=1,\ldots,K^{(r)}$) characterize the response of the $K^{(r)}$ slip systems in phase r, consisting of a certain crystalline material with orientation $\mathbf{Q}^{(r)}$, and depend on the resolved shear (or Schmid) stresses

$$\tau_{(k)}^{(r)}=\boldsymbol{\sigma}\cdot\boldsymbol{\mu}_{(k)}^{(r)},\quad \text{where } \boldsymbol{\mu}_{(k)}^{(r)}=\frac{1}{2}(\boldsymbol{n}_{(k)}^{(r)}\otimes\boldsymbol{m}_{(k)}^{(r)}+\boldsymbol{m}_{(k)}^{(r)}\otimes\boldsymbol{n}_{(k)}^{(r)}). \tag{2.2}$$

Here $\boldsymbol{\mu}_{(k)}^{(r)}$ is the second-order tensor obtained from the symmetrized dyadic product of the unit vectors $\boldsymbol{n}_{(k)}^{(r)}$, normal to the slip plane, and $\boldsymbol{m}_{(k)}^{(r)}$, along the slip direction, of the kth system in a crystal with orientation $\mathbf{Q}^{(r)}$. Note that the Schmid tensors $\boldsymbol{\mu}_{(k)}^{(r)}$ for a one-material polycrystal are related to the corresponding tensors $\boldsymbol{\mu}_{(k)}$ for a 'reference' crystal via $\boldsymbol{\mu}_{(k)}^{(r)}=\mathbf{Q}^{(r)^T}\boldsymbol{\mu}_{(k)}\mathbf{Q}^{(r)}$. A commonly used model for the slip potentials of a given single crystal is given by the power-law form:

$$\phi_{(k)}(\tau)=\frac{\gamma_0(\tau_0)_{(k)}}{n_{(k)}+1}\left|\frac{\tau}{(\tau_0)_{(k)}}\right|^{n_{(k)}+1}, \tag{2.3}$$

where $m_{(k)}=1/n_{(k)}$ ($0\le m_{(k)}\le 1$) and $(\tau_0)_{(k)}>0$ are, respectively, the strain-rate sensitivity and reference flow stress of the kth slip system, and γ_0 is a reference strain rate. Note that the limits as $n_{(k)}$ tends to 1 and ∞ are of special interest, since they correspond to linearly viscous and rigid–ideally plastic behaviour. Of course, the properties $n_{(k)}$ and $(\tau_0)_{(k)}$ could also depend on the type of material for composites with multiple material phases.

Assuming the appropriate *separation of length scales*, the effective behaviour of the composite can be described by the *effective stress potential* (e.g. [2,11])

$$\tilde{U}(\bar{\boldsymbol{\sigma}})=\min_{\boldsymbol{\sigma}\in\mathcal{S}}\langle u(\mathbf{x},\boldsymbol{\sigma})\rangle=\min_{\boldsymbol{\sigma}\in\mathcal{S}}\sum_{r=1}^{N}c^{(r)}\langle u^{(r)}(\boldsymbol{\sigma})\rangle^{(r)}, \tag{2.4}$$

where the scalars $c^{(r)} = \langle \chi^{(r)} \rangle$ denote the volume fractions of the given phases and

$$\mathcal{S} = \{\boldsymbol{\sigma},\ \operatorname{div} \boldsymbol{\sigma} = \mathbf{0} \text{ in } \Omega, \boldsymbol{\sigma}\mathbf{n} = \bar{\boldsymbol{\sigma}}\mathbf{n} \text{ on } \partial\Omega\} \tag{2.5}$$

denotes the set of statically admissible stresses. Note that $\langle \boldsymbol{\sigma} \rangle = \bar{\boldsymbol{\sigma}}$ is the *macroscopic stress* in the composite, and it can be related to the *macroscopic strain rate* $\bar{\boldsymbol{\epsilon}} = \langle \boldsymbol{\epsilon} \rangle$ via the macroscopic constitutive relation

$$\bar{\boldsymbol{\epsilon}} = \frac{\partial \tilde{U}}{\partial \bar{\boldsymbol{\sigma}}}. \tag{2.6}$$

Assuming convexity of the stress potentials $u^{(r)}$ of the phases, it is also possible to define a dual formulation in terms of the strain-rate potentials $w^{(r)}$ for the phases, which may be obtained by means of the Legendre transformation (see below). The dual formulation is described by the macroscopic strain-rate potential $\tilde{W}$, which is, in turn, the Legendre transform of $\tilde{U}$ and is such that $\bar{\boldsymbol{\sigma}} = \partial \tilde{W}/\partial \bar{\boldsymbol{\epsilon}}$. In general, the effective potentials $\tilde{U}$ and $\tilde{W}$ are difficult to compute, because they involve sets of nonlinear partial differential equations with randomly oscillating coefficients. In this work, new approximations will be developed for these potentials by making use of variational principles for suitably defined LCCs. The resulting approximations will be required to satisfy the bounds of deBotton & Ponte Castañeda [8] and to be exact to second order in the contrast [17].

In addition to the macroscopic constitutive relation for a nonlinear composite, it is also of interest to characterize the statistics of the stress and strain-rate fields in the composite, as described, in particular, by the first and second moments of the fields in the phases of the composite. Thus, the first moments, or averages, of the stress and strain-rate fields over phase r are defined via $\bar{\boldsymbol{\sigma}}^{(r)} = \langle \boldsymbol{\sigma} \rangle^{(r)}$ and $\bar{\boldsymbol{\epsilon}}^{(r)} = \langle \boldsymbol{\epsilon} \rangle^{(r)}$, and are such that $\bar{\boldsymbol{\sigma}} = \sum_{r=1}^{N} c^{(r)} \bar{\boldsymbol{\sigma}}^{(r)}$ and $\bar{\boldsymbol{\epsilon}} = \sum_{r=1}^{N} c^{(r)} \bar{\boldsymbol{\epsilon}}^{(r)}$. Similarly, the second moments of the stress and strain rate over phase r are given by $\langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)}$ and $\langle \boldsymbol{\epsilon} \otimes \boldsymbol{\epsilon} \rangle^{(r)}$. Here, we will also make use of the statistical quantities

$$\bar{\tau}^{(r)}_{(k)} = \bar{\boldsymbol{\sigma}}^{(r)} \cdot \boldsymbol{\mu}^{(r)}_{(k)}, \quad \bar{\bar{\tau}}^{(r)}_{(k)} = \boldsymbol{\mu}^{(r)}_{(k)} \cdot \langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)} \boldsymbol{\mu}^{(r)}_{(k)} \quad \text{and} \quad \mathrm{SD}^{(r)}(\tau^{(r)}_{(k)}) = \sqrt{\bar{\bar{\tau}}^{(r)}_{(k)} - (\bar{\tau}^{(r)}_{(k)})^2}, \tag{2.7}$$

corresponding to the phase average, second moment and standard deviation of the resolved shear stresses over slip system k in phase r.

Analytical expressions for the first and second moments of the stress and strain-rate fields in the phases of nonlinear composites have been given by Idiart & Ponte Castañeda [18]. The idea is to perturb the potential of the given phase by means of suitable terms that are linear, or quadratic in the appropriate field, and to make use of the above variational formulation to generate corresponding estimates for the homogenized potentials of the perturbed problem, which can then be differentiated with respect to the coefficients of the perturbing terms. Thus, the first moment, or average, of the stress field in phase r of the nonlinear composite may be obtained via the identity (see proposition 3.1 of [18])

$$\bar{\boldsymbol{\sigma}}^{(r)} = \frac{1}{c^{(r)}} \, \partial_{\boldsymbol{\eta}^{(r)}} \tilde{U}_{\boldsymbol{\eta}} \Big|_{\boldsymbol{\eta}^{(r)} = \mathbf{0}}, \tag{2.8}$$

where $\boldsymbol{\eta}^{(r)}$ is a constant, symmetric, second-order tensor, and $\tilde{U}_{\boldsymbol{\eta}}$ denotes the effective potential of a composite with (perturbed) local potential

$$u_{\boldsymbol{\eta}}(\mathbf{x}, \boldsymbol{\sigma}) = \sum_{s=1}^{N} \chi^{(s)}(\mathbf{x}) u^{(s)}(\boldsymbol{\sigma}) + \chi^{(r)}(\mathbf{x}) \boldsymbol{\eta}^{(r)} \cdot \boldsymbol{\sigma}. \tag{2.9}$$

Similarly, the second moment of the stress field in phase r of the nonlinear composite may be obtained via the identity (see corollary 3.3 of [18])

$$\langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)} = \frac{2}{c^{(r)}} \, \partial_{\mathbb{M}^{(r)}} \tilde{U}_{\mathbb{M}} \Big|_{\mathbb{M}^{(r)} = \mathbb{O}}, \tag{2.10}$$

where $\mathbb{M}^{(r)}$ is a constant, positive semi-definite, fourth-order tensor with major and minor symmetries, and $\tilde{U}_{\mathbb{M}}$ denotes the effective potential of a composite with (perturbed) local potential

$$u_{\mathbb{M}}(\boldsymbol{\sigma}) = \sum_{s=1}^{N} \chi^{(s)}(\mathbf{x}) u^{(s)}(\boldsymbol{\sigma}) + \chi^{(r)}(\mathbf{x}) \frac{1}{2} \boldsymbol{\sigma} \cdot \mathbb{M}^{(r)} \boldsymbol{\sigma}. \tag{2.11}$$

In addition, the first moment, or average, of the strain-rate field in phase r of the nonlinear composite may be obtained via (see proposition 3.4 of [18])

$$\bar{\boldsymbol{\epsilon}}^{(r)} = -\frac{1}{c^{(r)}} \, \partial_{\boldsymbol{\tau}^{(r)}} \tilde{U}_{\boldsymbol{\tau}} \Big|_{\boldsymbol{\tau}^{(r)}=\mathbf{0}}, \tag{2.12}$$

where $\boldsymbol{\tau}^{(r)}$ is a constant, symmetric, second-order tensor, and $\tilde{U}_{\boldsymbol{\tau}}$ denotes the effective potential of a composite with (perturbed) local potential given by

$$u_{\boldsymbol{\tau}}(\mathbf{x}, \boldsymbol{\sigma}) = \sum_{\substack{s=1 \\ s \neq r}}^{N} \chi^{(s)}(\mathbf{x}) u^{(s)}(\boldsymbol{\sigma}) + \chi^{(r)}(\mathbf{x}) u^{(r)}(\boldsymbol{\sigma} - \boldsymbol{\tau}^{(r)}), \tag{2.13}$$

and similarly for the second moment of the strain-rate field.

Idiart & Ponte Castañeda [18] have shown that, when the 'variational' (secant) linear comparison method [9] is used to estimate the effective potential of the nonlinear heterogeneous material in terms of the potential of a certain LCC, the first and second moments of the fields over the phases can be estimated directly from the corresponding moments in the LCC. On the other hand, Idiart & Ponte Castañeda [18] have also shown that, when the 'generalized secant' second-order method [15] is used to estimate the effective potential of the nonlinear heterogeneous material, the resulting estimates for the first and second moments of the fields over the phases of the nonlinear heterogeneous material do not, in general, coincide with the corresponding first and second moments in the LCC; certain additional correction terms are needed on account of the lack of full stationarity in the 'generalized secant' linear comparison method [15]. One of the primary objectives of this work is to develop fully optimized estimates of the generalized secant type for crystalline aggregates, such that the first and second moments of the fields over the phases of the nonlinear heterogeneous material can also be estimated directly from the corresponding moments in the LCC—without the need for correction terms. This will resolve an outstanding issue with the generalized secant estimates of Liu & Ponte Castañeda [16], which were obtained by an appropriate adaptation of the work of Ponte Castañeda [15] for polycrystalline aggregates (see also [19]).

3. Fully optimized linear comparison estimates

In this section, a 'second-order' variational technique is derived to estimate the effective behaviour and field fluctuations in composites with viscoplastic crystalline phases. For this purpose, we first derive a generalization of the 'linear comparison' transformation [8,9] for the stress potentials of the nonlinear phases. It is recalled that the linear comparison transformation expresses the potential of the nonlinear material in terms of a quadratic comparison potential (corresponding to linear constitutive behaviour), where the viscosity of this comparison medium is determined by a suitable optimization procedure. Building on earlier work [15,16], the generalization consists in the use of a comparison potential including both a linear term as well as the quadratic term, where the coefficients of such a comparison potential correspond to certain eigenstrain rates and viscosities to be determined by a suitable optimization process. We then make use of this transformation to express the macroscopic potential for the nonlinear composite in terms of the macroscopic potential for an LCC with the same microstructure as the original nonlinear composite, but with linear properties characterized by the eigenstrains and viscosities of the slip systems in the various phases. This allows the use of homogenization estimates for the LCC to generate corresponding estimates for the original nonlinear composite by means of a suitable optimization procedure over the properties of the LCC. Different from the earlier 'second-order' estimates [15,16], the new 'second-order' estimates can be fully optimized over the properties of the relevant LCCs. This endows the new estimates with some advantageous and useful properties, including the facts that the macroscopic constitutive relation and the field statistics of the nonlinear composite can be obtained directly from those of the LCC.

(a) Generalized Legendre transformation

We first consider an even, scalar-valued, (strictly) convex function ϕ of a scalar variable τ, such that $\phi(0)=0$ (e.g. the power-law function defined in (2.3)). Then, under appropriate smoothness hypotheses, its Legendre transform is defined by the single-valued function

$$\phi^*(\gamma)=\underset{\tau}{\mathrm{stat}}\{\tau\gamma-\phi(\tau)\}, \tag{3.1}$$

and is such that $\phi^{**}=\phi$, or

$$\phi(\tau)=\underset{\gamma}{\mathrm{stat}}\{\tau\gamma-\phi^*(\gamma)\}. \tag{3.2}$$

Furthermore, the respective optimality (i.e. stationarity) conditions $\gamma=\phi'(\tau)$ and $\tau=\phi^{*\prime}(\gamma)$ are the *unique* inverses of each other. In fact, as is well known, the Legendre transformation can be defined even when the function is not smooth, but still convex. In this case, the stationary operation in expression (3.1) is replaced by a supremum operation, and then ϕ^* is usually referred to as the Legendre–Fenchel transform of ϕ.

In this work, we are interested in expressing the macroscopic potential of a nonlinear composite in terms of a linear composite with a quadratic potential. With this goal in mind, we consider the function

$$\psi(\tau)=\phi(\tau)-\frac{\lambda}{2}\tau^2, \tag{3.3}$$

where the original function ϕ has been 'shifted' by a quadratic function $\frac{\lambda}{2}\tau^2$, and where λ is a parameter to be determined further below.

As illustrated in figure 1*a* for the special case where $\phi(\tau)=\tau^4/4$, the shifted function is not expected to be convex in general. However, following Sewell [20], it is still possible to define the Legendre transform of this function via the expression

$$\psi^*(\gamma)=\underset{\tau}{\mathrm{stat}}\{\tau\,\gamma-\psi(\tau)\}=\underset{\tau}{\mathrm{stat}}\left\{\tau\,\gamma+\frac{\lambda}{2}\tau^2-\phi(\tau)\right\}, \tag{3.4}$$

although it should be remarked that the stationarity condition

$$\gamma=\psi'(\tau)=\phi'(\tau)-\lambda\tau \tag{3.5}$$

may have multiple solutions for τ as a function of γ, as depicted in figure 1*b* for the example of figure 1*a*. In such cases, as depicted in figure 1*c*, the Legendre transform ψ^* will be a multiple-valued function for a range of values of the variable γ. Because of the multi-valued character of the function ψ^*, it is necessary to exercise care in the computation of the double-Legendre transform ψ^{**}. Nevertheless, it can be shown [20] that, with the proper branch selection, the duality result ($\psi^{**}=\psi$) still survives in this case.

Thus, using Legendre duality, we can write that

$$\phi(\tau)-\frac{\lambda}{2}\tau^2=\psi(\tau)=\underset{\gamma}{\mathrm{stat}}\{\tau\gamma-\psi^*(\gamma)\}, \tag{3.6}$$

where the 'correct' branch of the multiple-valued function ψ^* must be used to ensure equality.

Now, for given τ, we assume that λ is chosen such that the three solutions of the equation (3.5) are labelled and ordered as

$$\check{\tau}\le\tau\le\hat{\tau}. \tag{3.7}$$

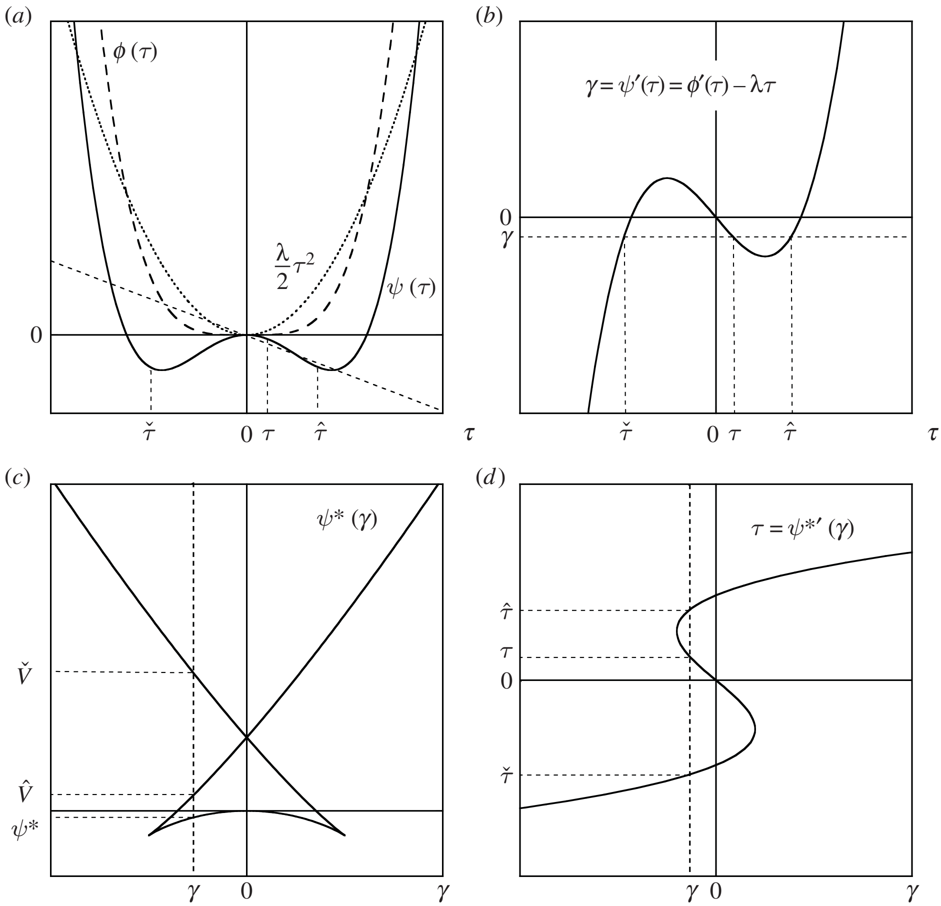

Figure 1. Legendre transform of the non-convex function ψ and duality issues. (*a*) The 'shifted' function ψ in expression (3.3) for the special case of $\phi(\tau) = \tau^4/4$ and $\lambda = 3$ for the quadratic shift. (*b*) The stationarity condition for the Legendre transform $\gamma = \phi'(\tau) - \lambda\tau$. (*c*) The Legendre transform $\psi^*(\gamma)$ is 'swallow-tailed' and multi-valued. Error functions $\check{V}(\lambda, \gamma)$ and $\hat{V}(\lambda, \gamma)$. (*d*) The corresponding stationarity conditions for the Legendre transform of ψ^*, namely $\tau = \psi^{*\prime}(\gamma)$.

Then, for reasons that will become evident further below, it is useful to introduce the functions

$$\check{V}(\lambda, \gamma) = \operatorname*{stat}_{\check{\tau}} \left\{ \gamma\check{\tau} + \frac{\lambda}{2}\check{\tau}^2 - \phi(\check{\tau}) \right\} \quad \text{and} \quad \hat{V}(\lambda, \gamma) = \operatorname*{stat}_{\hat{\tau}} \left\{ \gamma\hat{\tau} + \frac{\lambda}{2}\hat{\tau}^2 - \phi(\hat{\tau}) \right\}, \tag{3.8}$$

corresponding to the two 'other' branches of the multiple-valued function ψ^* (i.e. not the 'correct' branch of ψ^* recovering the value of the function of the left-hand side of expression (3.6) at τ). Note that the labels $\check{\cdot}$ and $\hat{\cdot}$ on the functions $\check{V}$ and $\hat{V}$ are used, respectively, to emphasize the fact that these functions are evaluated at the (generally) different stationary points $\check{\tau}$ and $\hat{\tau}$, such that $\check{\tau} \leq \tau \leq \hat{\tau}$ (cf. figure 1*b*), satisfying the conditions

$$\phi'(\check{\tau}) - \lambda\check{\tau} = \gamma = \phi'(\hat{\tau}) - \lambda\hat{\tau}. \tag{3.9}$$

In the context of these expressions, it should be emphasized that τ is the 'correct' value of the inverse of γ in (3.5), in the sense that it is consistent with expression (3.6). Therefore, as illustrated in figure 1*c*, the functions $\check{V}$ and $\hat{V}$ are different functions of the variable γ (and also different from $\psi^*(\gamma)$).

Next, writing expression (3.6) in the form

$$\phi(\tau) - \frac{\lambda}{2}\tau^2 = \underset{\gamma}{\text{stat}}\{\tau\,\gamma - \alpha\psi^*(\gamma) - (1-\alpha)\psi^*(\gamma)\}, \tag{3.10}$$

where α is some appropriately selected 'weight factor' such that $0 < \alpha < 1$, it is possible to generate the 'relaxed' estimate

$$\phi(\tau) - \frac{\lambda}{2}\tau^2 \approx \underset{\gamma}{\text{stat}}\{\tau\,\gamma - \alpha\check{V}(\lambda,\gamma) - (1-\alpha)\hat{V}(\lambda,\gamma)\}, \tag{3.11}$$

consisting in the replacement of the 'correct' branch of ψ^* (evaluated at τ) by the two 'other' branches of the function ψ^*, namely $\check{V}$ and $\hat{V}$ (which are evaluated at the other two stationary points, $\check{\tau}$ and $\hat{\tau}$, such that $\check{\tau} \le \tau \le \hat{\tau}$). Of course, this comes at the cost of losing the exact equality in expression (3.10), and this is why we have used the approximate equality in expression (3.11). Nevertheless, the equality can still be recovered by a further optimization procedure over the variable λ, as we will see next. Thus, moving the term $(\lambda/2)\tau^2$ to the right-hand side of the expression (3.11), so that

$$\phi(\tau) \approx \underset{\gamma}{\text{stat}}\left\{\tau\,\gamma + \frac{\lambda}{2}\tau^2 - \alpha\check{V}(\lambda,\gamma) - (1-\alpha)\hat{V}(\lambda,\gamma)\right\}, \tag{3.12}$$

it is seen that we can now also optimize the right-hand side with respect to λ to obtain the result

$$\phi(\tau) = \underset{\lambda}{\text{stat}}\left\{\underset{\gamma}{\text{stat}}\left[\tau\,\gamma + \frac{\lambda}{2}\tau^2 - \alpha\check{V}(\lambda,\gamma) - (1-\alpha)\hat{V}(\lambda,\gamma)\right]\right\}, \tag{3.13}$$

where we have again written an equality because the stationarity conditions in this expression ensure equality, regardless of the value of α. To see this, it is noted that the stationarity conditions with respect to γ and λ reduce to

$$\tau = \alpha\check{\tau} + (1-\alpha)\hat{\tau} \quad \text{and} \quad \tau^2 = \alpha\check{\tau}^2 + (1-\alpha)\hat{\tau}^2, \tag{3.14}$$

respectively, which can be combined to obtain the result

$$\alpha(1-\alpha)(\hat{\tau} - \check{\tau})^2 = 0. \tag{3.15}$$

It follows from this last result that the stationarity conditions in the estimate (3.13) for the function ϕ require that $\hat{\tau} = \check{\tau} = \tau$, whenever $0 < \alpha < 1$, and imply equality in expression (3.13).

We will make use of the representation (3.13) for the slip potentials $\phi^{(r)}_{(k)}$, defining the potentials of the single-crystal phases $u^{(r)}$ in expressions (2.1), in the corresponding expression (2.4) for the effective stress potentials $\tilde{U}$ for the viscoplastic composite. As will be seen, the use of expression (3.13) will allow the generation of fully optimized second-order (FO-SO) estimates for $\tilde{U}$ in terms of the effective stress potential $\tilde{U}_L$ of an LCC. In preparation for this, we first define the LCC more precisely.

(b) The linear comparison composite

By analogy with expressions (2.1) for the stress potentials of the phases of the nonlinear composite, we introduce stress potentials for the phases of the LCC by means of the expressions

$$u^{(r)}_L(\boldsymbol{\sigma}) = \sum_{k=1}^{K^{(r)}} \phi_L{}^{(r)}_{(k)}(\tau^{(r)}_{(k)}), \tag{3.16}$$

where it is recalled that the $\tau^{(r)}_{(k)}$ are the resolved shear stresses defined by expression (2.2), and the functions $\phi_L{}^{(r)}_{(k)}$ are quadratic slip potentials defined by

$$\phi_L{}^{(r)}_{(k)}(\tau) = \frac{1}{4\mu^{(r)}_{(k)}}\,\tau^2 + \gamma^{(r)}_{(k)}\tau, \tag{3.17}$$

in terms of a 'slip viscosity' $\mu_{(k)}^{(r)}$ and a 'slip eigenstrain rate' $\gamma_{(k)}^{(r)}$ ($r=1,\ldots,N$; $s=1,\ldots,K^{(r)}$). It is easy to see that the phase potential $u_L^{(r)}$ can then be put in the form

$$u_L^{(r)}(\boldsymbol{\sigma}) = \tfrac{1}{2}\boldsymbol{\sigma}\cdot\mathbb{M}^{(r)}\boldsymbol{\sigma} + \boldsymbol{\gamma}^{(r)}\cdot\boldsymbol{\sigma}, \tag{3.18}$$

by letting

$$\mathbb{M}^{(r)} = \sum_{k=1}^{K^{(r)}} \frac{1}{2\mu_{(k)}^{(r)}}\boldsymbol{\mu}_{(k)}^{(r)}\otimes\boldsymbol{\mu}_{(k)}^{(r)} \quad \text{and} \quad \boldsymbol{\gamma}^{(r)} = \sum_{k=1}^{K^{(r)}} \gamma_{(k)}^{(r)}\boldsymbol{\mu}_{(k)}^{(r)}, \tag{3.19}$$

where it is recalled that the $\boldsymbol{\mu}_{(k)}^{(r)}$ ($r=1,\ldots,N$; $s=1,\ldots,K^{(r)}$) correspond to the slip tensors that have been defined in the context of expression (2.2).

Differentiation of expression (3.18) with respect to $\boldsymbol{\sigma}$ shows that the constitutive relation of this material is indeed linear, i.e. $\boldsymbol{\epsilon} = \mathbb{M}^{(r)}\boldsymbol{\sigma} + \boldsymbol{\gamma}^{(r)}$. Note that this constitutive relation is mathematically analogous to that for a thermoelastic composite [21,22]. Then, defining the stress potential for the LCC via

$$u_L(\mathbf{x},\boldsymbol{\sigma}) = \sum_{r=1}^{N}\chi^{(r)}(\mathbf{x})u_L^{(r)}(\boldsymbol{\sigma}), \tag{3.20}$$

we can write the effective, or homogenized, stress potential for the composite as

$$\tilde{U}_L(\bar{\boldsymbol{\sigma}}) = \min_{\boldsymbol{\sigma}\in\mathcal{S}}\langle u_L(\mathbf{x},\boldsymbol{\sigma})\rangle = \min_{\boldsymbol{\sigma}\in\mathcal{S}}\sum_{r=1}^{N} c^{(r)}\langle u_L^{(r)}(\boldsymbol{\sigma})\rangle^{(r)}. \tag{3.21}$$

It is well known that the effective potential $\tilde{U}_L$ may be re-expressed in the form

$$\tilde{U}_L(\bar{\boldsymbol{\sigma}}) = \frac{1}{2}\bar{\boldsymbol{\sigma}}\cdot\tilde{\mathbb{M}}\bar{\boldsymbol{\sigma}} + \tilde{\boldsymbol{\gamma}}\cdot\bar{\boldsymbol{\sigma}} + \frac{1}{2}\tilde{g}, \tag{3.22}$$

where $\tilde{\mathbb{M}}$, $\tilde{\boldsymbol{\gamma}}$ and $\tilde{g}$ are the effective viscous compliance, effective eigenstrain rate and effective potential at zero stress, respectively [21,22]. Of course, the values of these effective variables will depend not only on the properties of the phases, but also on the specific microstructure. In this work, it will be assumed that estimates, such as the Willis estimate [23,24], or the self-consistent estimates [23,25], are available for this problem, depending on the specific microstructure (or class of microstructures) of interest. The idea then is to make use of such linear estimates to generate corresponding estimates for composites with nonlinear properties (as defined in the previous section), exhibiting identical microstructures, or classes of microstructures.

It is also well known (e.g. [11]) that the first and second moments of the stress in the LCC can be obtained from $\tilde{U}_L$ via the identities

$$\langle\boldsymbol{\sigma}\rangle^{(r)} = \frac{1}{c^{(r)}}\frac{\partial\tilde{U}_L}{\partial\boldsymbol{\gamma}^{(r)}} \quad \text{and} \quad \langle\boldsymbol{\sigma}\otimes\boldsymbol{\sigma}\rangle^{(r)} = \frac{2}{c^{(r)}}\frac{\partial\tilde{U}_L}{\partial\mathbb{M}^{(r)}}, \tag{3.23}$$

which are consistent with the more general forms (2.8) and (2.10) for nonlinear composites. In this work, we make use of the variables $\bar{\tau}_{(k)}^{(r)}$, $\bar{\bar{\tau}}_{(k)}^{(r)}$ and $\mathrm{SD}^{(r)}(\tau_{(k)}^{(r)})$, as defined by relations (2.7), to denote the average, second moment and standard deviation, respectively, of the resolved shear stresses over slip system k over phase r in the LCC.

(c) Stationary variational estimates

Returning now to the nonlinear composite with stress potentials $u^{(r)}$ given by expressions (2.1) in terms of the slip potentials $\phi_{(k)}^{(r)}$, we make use of expressions (3.13) for the general function ϕ

to write

$$
\begin{aligned}
u^{(r)}(\boldsymbol{\sigma}) &= \sum_{k=1}^{K^{(r)}} \phi_{(k)}^{(r)}(\tau_{(k)}^{(r)}) \\
&= \sum_{k=1}^{K^{(r)}} \operatorname*{stat}_{\mu_{(k)}^{(r)},\gamma_{(k)}^{(r)}} \left\{ \tau_{(k)}^{(r)}\gamma_{(k)}^{(r)} + \frac{1}{4\mu_{(k)}^{(r)}}(\tau_{(k)}^{(r)})^2 - \alpha^{(r)}\check{V}(\mu_{(k)}^{(r)},\gamma_{(k)}^{(r)}) - (1-\alpha^{(r)})\hat{V}(\mu_{(k)}^{(r)},\gamma_{(k)}^{(r)}) \right\} \\
&= \operatorname*{stat}_{\substack{\mu_{(n)}^{(r)},\gamma_{(n)}^{(r)},\\ n=1,\ldots,K^{(r)}}} \sum_{k=1}^{K^{(r)}} \left\{ \tau_{(k)}^{(r)}\gamma_{(k)}^{(r)} + \frac{1}{4\mu_{(k)}^{(r)}}(\tau_{(k)}^{(r)})^2 - \alpha^{(r)}\check{V}(\mu_{(k)}^{(r)},\gamma_{(k)}^{(r)}) - (1-\alpha^{(r)})\hat{V}(\mu_{(k)}^{(r)},\gamma_{(k)}^{(r)}) \right\},
\end{aligned}
$$

where it is recalled that the $\alpha^{(r)}$ are constant 'weight factors' between 0 and 1, and where we have identified the variables λ in expressions (3.13) with the 'slip viscosities' $\mu_{(k)}^{(r)}$, i.e. $\lambda \to 1/(2\mu_{(k)}^{(r)})$, so that, in particular, the functions $\check{V}_{(k)}^{(r)}$ and $\hat{V}_{(k)}^{(r)}$ in equation (3.8) are now written as

$$
\check{V}_{(k)}^{(r)}(\mu_{(k)}^{(r)},\gamma_{(k)}^{(r)}) = \operatorname*{stat}_{\check{\tau}_{(k)}^{(r)}} \left\{ \gamma_{(k)}^{(r)}\check{\tau}_{(k)}^{(r)} + \frac{1}{4\mu_{(k)}^{(r)}}(\check{\tau}_{(k)}^{(r)})^2 - \phi_{(k)}^{(r)}(\check{\tau}_{(k)}^{(r)}) \right\}, \tag{3.24}
$$

and similarly for $\hat{V}_{(k)}^{(r)}$.

By recalling that $\tau_{(k)}^{(r)} = \boldsymbol{\sigma}\cdot\boldsymbol{\mu}_{(k)}^{(r)}$, as well as expressions (3.18) and (3.19) defining the stress potential $u_L^{(r)}$ of phase r of the LCC, it is noted that the above expression for $u^{(r)}$ can be rewritten more compactly as

$$
u^{(r)}(\boldsymbol{\sigma}) = \operatorname*{stat}_{\substack{\mu_{(n)}^{(r)},\gamma_{(n)}^{(r)},\\ n=1,\ldots,K^{(r)}}} \left\{ u_L^{(r)}(\boldsymbol{\sigma}) - \sum_{k=1}^{K^{(r)}} V_{(k)}^{(r)}(\mu_{(k)}^{(r)},\gamma_{(k)}^{(r)}) \right\}, \tag{3.25}
$$

where we have also introduced the short-hand notation

$$
V_{(k)}^{(r)}(\mu_{(k)}^{(r)},\gamma_{(k)}^{(r)}) = \alpha^{(r)}\check{V}_{(k)}^{(r)}(\mu_{(k)}^{(r)},\gamma_{(k)}^{(r)}) + (1-\alpha^{(r)})\hat{V}_{(k)}^{(r)}(\mu_{(k)}^{(r)},\gamma_{(k)}^{(r)}) \tag{3.26}
$$

for the weighted average of the functions $\check{V}_{(k)}^{(r)}$ and $\hat{V}_{(k)}^{(r)}$.

It follows, by substituting expressions (3.25) for the potentials $u^{(r)}$ in expression (2.4) for the effective potential $\tilde{U}$ of the nonlinear composite, and interchanging the order of the minimum and stationary operations, that

$$
\tilde{U}(\bar{\boldsymbol{\sigma}}) = \operatorname*{stat}_{\mu_{(n)}^{(s)}(\mathbf{x}),\gamma_{(n)}^{(s)}(\mathbf{x})} \left\{ \tilde{U}_L(\bar{\boldsymbol{\sigma}}) - \sum_{r=1}^{N} c^{(r)} \sum_{k=1}^{K^{(r)}} \langle V_{(k)}^{(r)}(\mu_{(k)}^{(r)},\gamma_{(k)}^{(r)})\rangle^{(r)} \right\}, \tag{3.27}
$$

where it is noted that the slip viscosities $\mu_{(n)}^{(s)}$ and eigenstrain rates $\gamma_{(n)}^{(s)}$ $(s=1,\ldots,N, n=1,\ldots,K^{(r)})$ appearing both in the effective potential $\tilde{U}_L$ of the LCC and in the 'error' function $V_{(k)}^{(r)}$ are now functions of position $\mathbf{x}$ within the individual phases of the composite. In this context, however, it should be emphasized that the trial fields $\mu_{(n)}^{(s)}(\mathbf{x})$ and $\gamma_{(n)}^{(s)}(\mathbf{x})$ in the variational principle (3.27) for $\tilde{U}$ are not subjected to any differential constraints. This is unlike the trial field $\boldsymbol{\sigma}$ in the variational statement (2.4) for $\tilde{U}$, which is required to satisfy the equilibrium equation in (2.5). However, the equilibrium equation is still enforced in the variational estimate (3.27), by means of the effective potential $\tilde{U}_L$ for the LCC, as given by (3.21). This variational statement could therefore be viewed as a generalization of the maximum principles of deBotton & Ponte Castañeda [8], where there is an additional optimization over the eigenstrain rates $\gamma_{(n)}^{(s)}(\mathbf{x})$, besides the optimization over the slip viscosity fields $\mu_{(n)}^{(s)}(\mathbf{x})$ (labelled $\alpha_{(n)}^{(s)}$ in the earlier work). While the new variational statement (3.27) is not an extremum principle, it has the capability of generating more accurate estimates as will be seen next.

Thus, it is noted that, since the trial fields $\mu^{(s)}_{(n)}(\mathbf{x})$ and $\gamma^{(s)}_{(n)}(\mathbf{x})$ in the variational principle (3.27) for $\tilde{U}$ are not subjected to any differential constraints, they can be chosen to be uniform in each phase, that is, $\mu^{(s)}_{(n)}(\mathbf{x}) = \mu^{(s)}_{(n)}$ and $\gamma^{(s)}_{(n)}(\mathbf{x}) = \gamma^{(s)}_{(n)}$. It follows that the macroscopic potential can be approximated as

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) \approx \underset{\mu^{(s)}_{(n)}, \gamma^{(s)}_{(n)}}{\text{stat}} \left\{ \tilde{U}_L(\bar{\boldsymbol{\sigma}}) - \sum_{r=1}^{N} c^{(r)} \sum_{k=1}^{K^{(r)}} V^{(r)}_{(k)}(\mu^{(r)}_{(k)}, \gamma^{(r)}_{(k)}) \right\}, \tag{3.28}$$

where $\tilde{U}_L$ now corresponds to the effective potential for the LCC defined by expressions (3.18) to (3.21) with constant per phase properties, and it is recalled that the functions $V^{(r)}_{(k)}$ correspond to the weighted average of the error functions, as defined by expressions (3.24) and (3.26). Building on the earlier remarks in connection with the variational statement (3.27), it is noted that the new estimate (3.28) can be shown to reduce to the stationary variational estimate (3.15) in [8], when the weight factors $\alpha^{(r)}$ and the variables $\gamma^{(s)}_{(n)}$ are all set equal to 0. In other words, the new stationary estimate (3.28) provides a generalization of the earlier estimates of deBotton & Ponte Castañeda [8], consisting in the use of a more general LCC. In addition, the estimate (3.28) also provides a generalization of the second-order estimates of Liu & Ponte Castañeda [16], essentially consisting in the use of two error functions $V^{(r)}$ (instead of one). As will be seen next, the use of such additional error functions will enable the generation of estimates for $\tilde{U}$ that are stationary with respect to *both* the variables $\mu^{(s)}_{(n)}$ and $\gamma^{(s)}_{(n)}$. This is unlike the corresponding estimates of Liu & Ponte Castañeda [16], which could only be made stationary with respect to *either* the $\mu^{(s)}_{(n)}$ *or* the $\gamma^{(s)}_{(n)}$ (but not both). Loosely speaking, the use of only one error function $V^{(r)}$ in the earlier estimate of Liu & Ponte Castañeda [16] had the implication that the optimization conditions for the variables $\mu^{(s)}_{(n)}$ and $\gamma^{(s)}_{(n)}$ led to conflicting requirements for the variables $\hat{\tau}^{(r)}$, by identifying them with both the first and second moments of the stress in the phases of the LCC—conditions which cannot be simultaneously satisfied when there are fluctuations of the stress field in the phases of the composite.

Next, we spell out the stationarity conditions associated with the estimate (3.28). We begin with the 'inner' stat problems for the variables $\check{\tau}^{(r)}$ and $\hat{\tau}^{(r)}$, implicit in the definitions (e.g. (3.24)) for the functions $\check{V}^{(r)}$ and $\hat{V}^{(r)}$, respectively. These conditions can be written in the form

$$(\phi^{(r)}_{(k)})'(\check{\tau}^{(r)}_{(k)}) - \frac{1}{2\mu^{(r)}_{(k)}}\check{\tau}^{(r)}_{(k)} = \gamma^{(r)}_{(k)} = (\phi^{(r)}_{(k)})'(\hat{\tau}^{(r)}_{(k)}) - \frac{1}{2\mu^{(r)}_{(k)}}\hat{\tau}^{(r)}_{(k)}, \tag{3.29}$$

where ϕ' has been used to denote the derivative of ϕ. Note that these two conditions imply that

$$(\phi^{(r)}_{(k)})'(\hat{\tau}^{(r)}_{(k)}) - (\phi^{(r)}_{(k)})'(\check{\tau}^{(r)}_{(k)}) = \frac{1}{2\mu^{(r)}_{(k)}}(\hat{\tau}^{(r)}_{(k)} - \check{\tau}^{(r)}_{(k)}), \tag{3.30}$$

which can be viewed as a 'generalized secant' linearization [16].

Next, we consider the 'outer' optimization problems with respect to the variables $\gamma^{(s)}_{(n)}$ and $\mu^{(s)}_{(n)}$ in the variational estimate (3.28). When taking derivatives of the term $\tilde{U}_L$ with respect to these variables, we make use of expressions (3.19) and of the chain rule to write the derivatives with respect to $\gamma^{(s)}_{(n)}$ and $\mu^{(s)}_{(n)}$, respectively, in terms of the first and second moments of the stress in the LCC via expressions (3.23). In addition, due to the stationarity operations with respect to the variables $\check{\tau}^{(r)}_{(k)}$ and $\hat{\tau}^{(r)}_{(k)}$ in the definitions (e.g. (3.24)) of the functions $\check{V}^{(r)}_{(k)}$ and $\hat{V}^{(r)}_{(k)}$, respectively, these variables can be treated as being 'fixed' in the context of the optimization with respect to the variables $\gamma^{(s)}_{(n)}$ and $\mu^{(s)}_{(n)}$. Thus, by making use of the chain rule, the stationarity condition with respect to the slip eigenstrain-rate variables $\gamma^{(s)}_{(n)}$ can be written as

$$\alpha^{(r)}\check{\tau}^{(r)}_{(k)} + (1-\alpha^{(r)})\hat{\tau}^{(r)}_{(k)} = \bar{\boldsymbol{\sigma}}^{(r)} \cdot \boldsymbol{\mu}^{(r)}_{(k)} = \bar{\tau}^{(r)}_{(k)}, \tag{3.31}$$

where it is emphasized that $\bar{\boldsymbol{\sigma}}^{(r)} = \langle \boldsymbol{\sigma} \rangle^{(r)}$ is the average of the stress field in phase r in the problem (3.21) for the LCC, which may be computed in terms of $\tilde{U}_L$ via the identity $(3.23)_1$. Also, as a consequence, the quantities $\bar{\tau}^{(r)}_{(k)}$ are functions of the properties of the LCC, as determined by the variables $\gamma^{(s)}_{(n)}$ and $\mu^{(s)}_{(n)}$ $(s = 1, \dots, N; n = 1, \dots, K^{(r)})$. Similarly, it is found that the stationarity conditions with respect to the slip compliance $\mu^{(s)}_{(n)}$ can be written as

$$\alpha^{(r)}(\check{\tau}^{(r)}_{(k)})^2 + (1 - \alpha^{(r)})(\hat{\tau}^{(r)}_{(k)})^2 = \boldsymbol{\mu}^{(r)}_{(k)} \cdot \langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)} \boldsymbol{\mu}^{(r)}_{(k)} = \bar{\bar{\tau}}^{(r)}_{(k)}, \tag{3.32}$$

where, again, it is emphasized that the variable $\langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)}$ corresponds to the second moment of the stress field in phase r of the LCC, which may be computed via identity $(3.23)_2$, and is therefore also a function of all the variables $\gamma^{(s)}_{(n)}$ and $\mu^{(s)}_{(n)}$. Thus, it can then be seen that the introduction of the two error functions evaluated at two different stationary points $\check{\tau}^{(r)}_{(k)}$ and $\hat{\tau}^{(r)}_{(k)}$ provides the capability to simultaneously satisfy conditions on the first and second moments of the stress fields—something that could not be achieved with the use of a single stationary point, as first attempted by Liu & Ponte Castañeda [16]. This provides the justification for the use of the more complex two-variable optimization expression (3.13) for the slip potentials $\phi^{(r)}_{(k)}$ instead of the simpler one-variable optimization form (3.6).

It is also worth mentioning that straightforward algebra shows that the conditions (3.31) and (3.32) can be combined to obtain the following result for the standard deviations of the resolved shear stress fluctuations in the phases, as defined by expressions (2.7), namely

$$\sqrt{\alpha^{(r)}(1 - \alpha^{(r)})}|\hat{\tau}^{(r)}_{(k)} - \check{\tau}^{(r)}_{(k)}| = \mathrm{SD}^{(r)}(\tau^{(r)}_{(k)}). \tag{3.33}$$

The stationarity conditions (3.29), (3.31) and (3.32) provide, at least in principle, a sufficient number of equations to solve for the $4\sum_{r=1}^{N} K^{(r)}$ unknown variables $\check{\tau}^{(s)}_{(n)}$, $\hat{\tau}^{(s)}_{(n)}$, $\gamma^{(s)}_{(n)}$ and $\mu^{(s)}_{(n)}$. The solutions of these equations can then be used to generate *fully stationary* variational estimates for $\tilde{U}$ via expression (3.28). (In this context, it should be noted that the resulting estimates for $\tilde{U}$ are sensitive to the values of the weight factors $\alpha^{(r)}$; however, the selection of these weight factors does not affect their stationarity properties and will be dictated by certain symmetry requirements to be discussed below in the context of specific cases.) As will be seen next, the new variational estimates (3.28) exhibit several interesting and useful properties as a consequence of their stationarity properties.

Simplified expression for the macroscopic stress potential. Use of the stationarity conditions (3.31) and (3.32) in expression (3.28) for the effective potential $\tilde{U}$ of the nonlinear composite can be easily seen to lead to the simplified result

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) = \sum_{r=1}^{N} c^{(r)} \sum_{k=1}^{K^{(r)}} [\alpha^{(r)} \phi^{(r)}_{(k)}(\check{\tau}^{(r)}_{(k)}) + (1 - \alpha^{(r)}) \phi^{(r)}_{(k)}(\hat{\tau}^{(r)}_{(k)})], \tag{3.34}$$

where we have replaced the approximate equality by an equality, for simplicity. Thus, it can be seen that the final result for the effective potential $\tilde{U}$ of the nonlinear composite involves a weighted average of the slip potentials evaluated at the stress variables $\check{\tau}^{(r)}_{(k)}$ and $\hat{\tau}^{(r)}_{(k)}$, which, as we have seen, depend on the first and second moments of the resolved shear stresses in the LCC defined above. Note that this alternative form for the estimate (3.28) is different from analogous simplified forms for $\tilde{U}$ due to deBotton & Ponte Castañeda [8] (see also [11,26]) and Liu & Ponte Castañeda [16], using the variational bounding procedure and earlier second-order methods, respectively.

Macroscopic constitutive relation via the linear comparison composite. Due to the stationarity of the variables $\check{\tau}^{(r)}_{(k)}$ and $\hat{\tau}^{(r)}_{(k)}$ in the definitions (e.g. (3.24)) of the functions $\check{V}^{(r)}_{(k)}$ and $\hat{V}^{(r)}_{(k)}$, respectively, as well as the stationarity of the variables $\gamma^{(s)}_{(n)}$ and $\mu^{(s)}_{(n)}$, all these variables can be treated as being fixed in the variational estimate (3.28) for the effective potential $\tilde{U}$ of the nonlinear composite.

Therefore, by means of expression (3.22) for $\tilde{U}_L$, and of the chain rule in expression (2.6), it can be shown that the macroscopic constitutive relation for the nonlinear composite reduces to

$$\bar{\boldsymbol{\epsilon}} = \tilde{\mathbb{M}}\bar{\boldsymbol{\sigma}} + \tilde{\boldsymbol{\gamma}}, \tag{3.35}$$

where $\tilde{\mathbb{M}}$ and $\tilde{\boldsymbol{\gamma}}$ are, respectively, the effective compliance and eigenstrain rate of the LCC. Of course, specific estimates for $\tilde{\mathbb{M}}$ and $\tilde{\boldsymbol{\gamma}}$, in terms of the properties of the optimized LCC, will depend on the microstructure of the composite [22]. The interest in the property (3.35) of the fully stationary estimates lies in the fact that this expression is much easier to evaluate than the derivative of expression (3.28) for the effective potential $\tilde{U}$, since the effective potential $\tilde{U}_L$ of the LCC, as defined by expression (3.22), already requires the computation of $\tilde{\mathbb{M}}$ and $\tilde{\boldsymbol{\gamma}}$. This simplification is especially significant for problems with large numbers of phases, such as polycrystals [16]. In addition, this property also shows that the fully stationary estimates are exact to second order in the contrast, provided that the corresponding estimate for the LCC is also exact to second order in the contrast.

Field statistics. As already mentioned, a general procedure is available [26] for computing the moments of the stress and strain-rate field in the phases of the nonlinear composite by means of suitably perturbed nonlinear problems. In particular, the first moment, or average, of the stress field in phase r of the nonlinear composite with stress potentials $u^{(s)}$ may be obtained via expression (2.8), where $\boldsymbol{\eta}^{(r)}$ is a constant, symmetric, second-order tensor, and $\tilde{U}_{\eta}$ denotes the effective potential of a composite with (perturbed) local potential given by (2.9). When applied to the LCC, as defined by expressions (3.21) and (3.18), this result implies that

$$\bar{\boldsymbol{\sigma}}_L^{(r)} = \frac{1}{c^{(r)}}\,\partial_{\boldsymbol{\eta}^{(r)}}\tilde{U}_{L_\eta}\Big|_{\boldsymbol{\eta}^{(r)}=\mathbf{0}}, \tag{3.36}$$

where we have used the subscript L in $\bar{\boldsymbol{\sigma}}_L^{(r)}$ to emphasize that it corresponds to the average stress over phase r of the LCC, and where $\tilde{U}_{L_\eta}$ denotes the effective potential of a composite with (perturbed) local potential

$$u_{L_{\boldsymbol{\eta}}}(\mathbf{x},\boldsymbol{\sigma}) = \sum_{s=1}^{N}\chi^{(s)}(\mathbf{x})u_L^{(s)}(\boldsymbol{\sigma}) + \chi^{(r)}(\mathbf{x})\boldsymbol{\eta}^{(r)}\cdot\boldsymbol{\sigma}. \tag{3.37}$$

Then, noting that

$$\tilde{U}_{\boldsymbol{\eta}}(\bar{\boldsymbol{\sigma}}) = \operatorname*{stat}_{\mu^{(s)}_{(n)},\gamma^{(s)}_{(n)}}\left\{\tilde{U}_{L_\eta}(\bar{\boldsymbol{\sigma}}) - \sum_{r=1}^{N}c^{(r)}\sum_{k=1}^{K^{(r)}}V^{(r)}_{(k)}(\mu^{(r)}_{(k)},\gamma^{(r)}_{(k)})\right\}, \tag{3.38}$$

where the functions $V^{(r)}_{(k)}$, as defined by (3.26), are independent of the perturbation parameters $\boldsymbol{\eta}^{(r)}$, and using the chain rule and the stationarity with respect to the variables $\mu^{(r)}_{(k)}$ and $\gamma^{(r)}_{(k)}$, it is easy to see that the quantities $\partial_{\boldsymbol{\eta}^{(r)}}\tilde{U}_{\boldsymbol{\eta}}$ and $\partial_{\boldsymbol{\eta}^{(r)}}\tilde{U}_{L_\eta}$ (evaluated at the optimal values of the $\mu^{(r)}_{(k)}$ and $\gamma^{(r)}_{(k)}$) are identical. By setting $\boldsymbol{\eta}^{(r)} = 0$, it is concluded that

$$\bar{\boldsymbol{\sigma}}^{(r)} = \bar{\boldsymbol{\sigma}}_L^{(r)}. \tag{3.39}$$

In other words, the phase averages of the stress field in the nonlinear composite can be estimated directly from the phase averages of the stress field in the (optimized) LCC. Note that these quantities are required anyway for the computation of the fully stationary estimate (3.28) via the stationarity conditions (3.31), and therefore no additional calculations are needed to obtain the phase averages of the stress in the nonlinear composite.

A completely analogous calculation starting with expression (2.10) for the second moment of the stress in the nonlinear composite can be used to show that the second moments of the stress fields in the nonlinear composite can be consistently estimated from the LCC, that is,

$$\langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)} = \langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle_L^{(r)}. \tag{3.40}$$

In addition, by proposition 3.4 of [26], the first moment, or average, of the strain-rate field in phase r of the nonlinear composite may be obtained via expression (2.12), where $\boldsymbol{\tau}^{(r)}$ is a constant, symmetric, second-order tensor, and $\tilde{U}_{\boldsymbol{\tau}}$ denotes the effective potential of a composite with (perturbed) local potential given by expression (2.13). When specialized to the LCC, as defined by expressions (3.21) and (3.18), this result implies that

$$\bar{\boldsymbol{\epsilon}}_L^{(r)} = -\frac{1}{c^{(r)}}\, \partial_{\boldsymbol{\tau}^{(r)}} \tilde{U}_{L_{\boldsymbol{\tau}}}\Big|_{\boldsymbol{\tau}^{(r)}=\mathbf{0}}, \tag{3.41}$$

where $\tilde{U}_{L_{\boldsymbol{\tau}}}$ denotes the effective potential of a composite with (perturbed) local potential $u_{L_{\boldsymbol{\tau}}}$ given by expression (2.13) with $u^{(s)}$ replaced by $u_L^{(s)}$. As was the case for the phase averages of the stress field, it is easy to see that the fully stationary estimate for $\tilde{U}_{\boldsymbol{\tau}}$ in terms of $\tilde{U}_{L_{\boldsymbol{\tau}}}$ involves error functions $\check{V}_{(k)}^{(r)}$ and $\hat{V}_{(k)}^{(r)}$, which are independent of the variables $\boldsymbol{\tau}^{(r)}$, and is stationary with respect to the variables $\mu_{(k)}^{(r)}$ and $\gamma_{(k)}^{(r)}$, leading to the conclusion—via the chain rule—that $\partial_{\boldsymbol{\tau}}^{(r)} \tilde{U}_{\boldsymbol{\tau}} = \partial_{\boldsymbol{\tau}}^{(r)} \tilde{U}_{L_{\boldsymbol{\tau}}}$, and therefore that

$$\bar{\boldsymbol{\epsilon}}^{(r)} = \bar{\boldsymbol{\epsilon}}_L^{(r)}. \tag{3.42}$$

Thus, the phase averages of the strain-rate field in the nonlinear composite can also be estimated directly from the corresponding phase averages of the strain-rate field in the LCC. It should be noted in this context that the results (3.42) for the phase averages of the strain rate are consistent with the corresponding result (3.35) for the macroscopic strain rate, since

$$\bar{\boldsymbol{\epsilon}} = \sum_{s=1}^{N} c^{(s)} \bar{\boldsymbol{\epsilon}}^{(s)} = \sum_{s=1}^{N} c^{(s)} \bar{\boldsymbol{\epsilon}}_L^{(s)} = \tilde{\mathbb{M}} \bar{\boldsymbol{\sigma}} + \tilde{\boldsymbol{\gamma}}. \tag{3.43}$$

It is also possible to similarly show by means of corollary 3.7 of [26] that the second moments of the strain-rate field in the nonlinear composite can be consistently estimated from the LCC, that is, $\langle \boldsymbol{\epsilon} \otimes \boldsymbol{\epsilon} \rangle^{(r)} = \langle \boldsymbol{\epsilon} \otimes \boldsymbol{\epsilon} \rangle_L^{(r)}$.

In summary, the full stationarity of the variational estimate (3.28) implies that not only can the macroscopic constitutive relation for the nonlinear composite be estimated directly from the corresponding constitutive relation of the LCC, but, in addition, so can the first and second moments of the stress and strain-rate fields in the nonlinear composite. Since the 'fully stationary' estimate (3.28) already requires the computation of the first and second moments of the stress field in the LCC, the fact that these quantities actually also provide estimates for the corresponding first and second moments of the stress field in the actual nonlinear composite is very convenient, in particular, given that the direct computation from the appropriate perturbed potentials is not straightforward. For example, it has been shown [26] that the first and second moments of the stress and strain-rate fields in the earlier versions of the 'tangent' and 'generalized secant' second-order estimates [15,27,28]—which are not stationary with respect to the variables $\mu_{(k)}^{(r)}$ or $\gamma_{(k)}^{(r)}$, respectively—involve additional terms which are difficult to compute, except for certain simplifications for *special* choices of these parameters. On the other hand, the present fully stationary version of the second-order estimates is similar to the variational bounds [8,9], for which the first and second moments of the stress and strain-rate fields in the LCC can be used to directly estimate the corresponding moments of the fields in the actual nonlinear composite. This is a feature that certainly makes worthwhile the additional complexities associated with the use of multiple error functions in the stationary estimates (3.28).

Legendre duality. As we have seen, it is possible to generate consistent estimates for the macroscopic constitutive relation of the composite, as well as for the phase averages and second moments of the stress and strain-rate fields, directly from the optimized LCC. In this sense, the variational statement (3.28) for the effective stress potential $\tilde{U}$ of the nonlinear composite in terms of the corresponding estimate (3.22) for the effective stress potential $\tilde{U}_L$ of the LCC is completely general. However, it is well known [22] that the linear problem for $\tilde{U}_L$ has a completely equivalent representation in terms of the strain-rate potential $\tilde{W}_L$, which is generated by Legendre duality: $\tilde{W}_L = \tilde{U}_L^*$. For this reason, it is of interest to construct the Legendre dual of the variational statement (3.28) for $\tilde{U}$. Thus, we write

$$\begin{aligned}
\tilde{W}(\bar{\boldsymbol{\epsilon}}) &= \underset{\bar{\boldsymbol{\sigma}}}{\mathrm{stat}}\{\bar{\boldsymbol{\sigma}} \cdot \bar{\boldsymbol{\epsilon}} - \tilde{U}(\bar{\boldsymbol{\sigma}})\} \\
&= \underset{\bar{\boldsymbol{\sigma}}}{\mathrm{stat}} \left\{ \bar{\boldsymbol{\sigma}} \cdot \bar{\boldsymbol{\epsilon}} - \underset{\mu^{(s)}_{(n)}, \gamma^{(s)}_{(n)}}{\mathrm{stat}} \left[\tilde{U}_L(\bar{\boldsymbol{\sigma}}) - \sum_{r=1}^{N} c^{(r)} \sum_{k=1}^{K^{(r)}} V^{(r)}_{(k)}(\mu^{(r)}_{(k)}, \gamma^{(r)}_{(k)}) \right] \right\} \\
&= \underset{\mu^{(s)}_{(n)}, \gamma^{(s)}_{(n)}}{\mathrm{stat}} \left\{ \underset{\bar{\boldsymbol{\sigma}}}{\mathrm{stat}}[\bar{\boldsymbol{\sigma}} \cdot \bar{\boldsymbol{\epsilon}} - \tilde{U}_L(\bar{\boldsymbol{\sigma}})] + \sum_{r=1}^{N} c^{(r)} \sum_{k=1}^{K^{(r)}} V^{(r)}_{(k)}(\mu^{(r)}_{(k)}, \gamma^{(r)}_{(k)}) \right\},
\end{aligned}$$

where the order of the stationary operations over the macroscopic stress $\bar{\boldsymbol{\sigma}}$ and the properties $\mu^{(r)}_{(k)}$ and $\gamma^{(r)}_{(k)}$ of the LCC has been interchanged. The result can then be rewritten in the form

$$\tilde{W}(\bar{\boldsymbol{\epsilon}}) = \underset{\mu^{(s)}_{(n)}, \gamma^{(s)}_{(n)}}{\mathrm{stat}} \left\{ \tilde{W}_L(\bar{\boldsymbol{\epsilon}}) + \sum_{r=1}^{N} c^{(r)} \sum_{k=1}^{K^{(r)}} V^{(r)}_{(k)}(\mu^{(r)}_{(k)}, \gamma^{(r)}_{(k)}) \right\}, \tag{3.44}$$

where $\tilde{W}_L$ is the effective strain-rate potential of the LCC, which is obtained via the Legendre transform of the corresponding effective stress potential $\tilde{U}_L$. The result can be written as [21,22]

$$\tilde{W}_L(\bar{\boldsymbol{\epsilon}}) = \tfrac{1}{2}\bar{\boldsymbol{\epsilon}} \cdot \tilde{\mathbb{L}}\bar{\boldsymbol{\epsilon}} + \tilde{\boldsymbol{\tau}} \cdot \bar{\boldsymbol{\epsilon}} + \tilde{f}, \tag{3.45}$$

where $\tilde{\mathbb{L}}$, $\tilde{\boldsymbol{\tau}}$ and $\tilde{f}$ are the effective viscosity, effective eigenstress and effective energy at zero strain, respectively. They are related to the effective viscous compliance $\tilde{\mathbb{M}}$, effective eigenstrain rate $\tilde{\boldsymbol{\gamma}}$ and effective energy at zero stress $\tilde{g}$ via the relations

$$\tilde{\mathbb{L}} = (\tilde{\mathbb{M}})^{-1}, \quad \tilde{\boldsymbol{\tau}} = -(\tilde{\mathbb{M}})^{-1}\tilde{\boldsymbol{\gamma}} \quad \text{and} \quad \tilde{f} = -\tilde{g} + \frac{1}{2}\tilde{\boldsymbol{\gamma}} \cdot (\tilde{\mathbb{M}})^{-1}\tilde{\boldsymbol{\gamma}}. \tag{3.46}$$

Finally, it is noted that it is possible to rewrite the result (3.44) in other forms making use of Legendre duality for the properties of the phases, but we will not pursue this here, for brevity.

4. Applications for porous single crystals

In this section, our objective is to illustrate the main features of the fully optimized second-order (FO-SO) homogenization technique by means of an example. For this purpose, we consider a special class of (two-phase) porous materials with 'particulate' microstructures [29], consisting of aligned cylindrical pores ($r=2$) that are distributed randomly and isotropically in a viscoplastic single-crystal matrix phase ($r=1$). We assume that the laboratory frame of reference is described by the orthonormal set of vectors $\mathbf{e}_i$ ($i=1,2,3$), such that the symmetry axes of the cylindrical pores are aligned with $\mathbf{e}_3$. We further assume that the behaviour of the crystalline matrix is characterized by an *incompressible* stress potential $u^{(1)}$ of the form (2.1), where the slip potentials $\psi^{(1)}_{(k)}$ are of the power-law type (2.3), with the same viscous exponents and flow stresses, i.e. $n_{(k)} = n$, and $(\tau_0)_{(k)} = \tau_0$ for all k. On the other hand, the Schmid tensors $\boldsymbol{\mu}_{(k)}$ are taken to be of the form $(2.2)_2$, with slip directions $\mathbf{m}_{(k)} = \mathbf{e}_3$ (for all k) and slip normals given by $\mathbf{n}_{(k)} = \cos\theta_{(k)}\mathbf{e}_1 + \sin\theta_{(k)}\mathbf{e}_2$. In this work, we will focus on two different types of anisotropies: (i) *square* ($K=2$), $\theta_{(k)} = 0, \pi/2$, and (ii) *hexagonal* ($K=3$), $\theta_{(k)} = 0, \pm\pi/3$. The porous material is subjected to anti-plane loadings, and the relevant viscoplastic boundary value problem then becomes a two-dimensional vectorial problem, where the non-zero components of the stress and strain-rate vectors, namely σ_{13}, σ_{23}, ϵ_{13} and ϵ_{23}, are functions of x_1 and x_2 only.

The fact that the viscous exponent n is the same for all the slip systems leads to the effective potential of the polycrystal $\tilde{U}$ being a homogeneous function of degree $n+1$ on the average stress $\bar{\boldsymbol{\sigma}}$. Furthermore, because of the anti-plane loading, the effective stress potential can be written as

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) = \frac{\gamma_0 \tilde{\tau}_0}{1+n}\left(\frac{\bar{\tau}_e}{\tilde{\tau}_0}\right)^{1+n}, \tag{4.1}$$

where $\bar{\tau}_e = \sqrt{(1/2)\,\bar{\boldsymbol{\sigma}}_d \cdot \bar{\boldsymbol{\sigma}}_d} = (\bar{\sigma}_{13}^2 + \bar{\sigma}_{23}^2)^{1/2}$ is the macroscopic equivalent stress, and $\tilde{\tau}_0$ is the *effective flow stress*, which depends on the porosity $c = c^{(2)}$, the nonlinearity n, and the direction of loading $\bar{\phi} = \tan^{-1}(\bar{\sigma}_{23}/\bar{\sigma}_{13})$, thus completely characterizing the effective response of the porous material. It is noted that, for the particular class of composites considered here, the potential $\tilde{U}$ exhibits the same symmetries as the matrix potential $u^{(1)}$, and therefore $\tilde{\tau}_0$ is a periodic function of $\bar{\phi}$ with period π/K. Thus, it suffices to restrict attention to loading directions in the range $|\bar{\phi}| \le \pi/(2K)$. Note that the values $\bar{\phi} = 0$ and $\bar{\phi} = \pm\pi/(2K)$ correspond, respectively, to loadings directed along a slip system and a 'corner' of the matrix phase.

For additional simplicity, we consider here only loadings that are aligned with one of the slip systems ($\bar{\phi} = 0$). In this case, the symmetry of the problem is such that it suffices to consider the matrix material of the LCC to be characterized by an eigenstrain rate $\boldsymbol{\gamma}_{(1)}$ and a viscous compliance tensor of the form

$$\mathbb{M}^{(1)} = \frac{1}{2\lambda}\mathbb{E} + \frac{1}{2\mu}\mathbb{F}, \tag{4.2}$$

where $\mathbb{E}$ and $\mathbb{F}$ are eigentensors given by $\mathbb{E} = \boldsymbol{\mu}_{(1)} \otimes \boldsymbol{\mu}_{(1)}$ and $\mathbb{F} = \mathbb{K} - \mathbb{E}$, with $\boldsymbol{\mu}_{(1)} = (1/\sqrt{2})(\mathbf{e}_1 \otimes \mathbf{e}_3 + \mathbf{e}_3 \otimes \mathbf{e}_1)$, and $\mathbb{K}$ denoting the out-of-plane projection of the fourth-order identity tensor. In addition, due to the assumed symmetry of the loading, it follows from (3.19) that

$$\frac{1}{2\lambda} = \sum_{k=1}^{K} \frac{\cos^2\theta_{(k)}}{2\mu_{(k)}^{(1)}}, \quad \frac{1}{2\mu} = \sum_{k=1}^{K} \frac{\sin^2\theta_{(k)}}{2\mu_{(k)}^{(1)}} \quad \text{and} \quad \boldsymbol{\gamma}^{(1)} = \sum_{k=1}^{K} \gamma_{(k)}^{(1)} \boldsymbol{\mu}_{(k)}. \tag{4.3}$$

Note that the degree of anisotropy of the viscous compliance tensor $\mathbb{M}^{(1)}$ is characterized by the ratio $k = \lambda/\mu$, which takes the value 1 for isotropic tensors and 0 or infinity for strongly anisotropic tensors. The behaviour of the LCC can then be characterized by estimates of the Willis type [23,24], which for the above-described porous microstructures takes the form [30]

$$\tilde{U}_L(\bar{\boldsymbol{\sigma}}) = \frac{1}{2}\bar{\boldsymbol{\sigma}} \cdot \tilde{\mathbb{M}}\bar{\boldsymbol{\sigma}} + \boldsymbol{\gamma}_{(1)} \cdot \bar{\boldsymbol{\sigma}}, \tag{4.4}$$

where $\tilde{\mathbb{M}}$ has the same form as $\mathbb{M}^{(1)}$, as given by (4.2), but with effective viscosities

$$\tilde{\lambda} = \frac{1-c}{1+c\sqrt{k}}\lambda \quad \text{and} \quad \tilde{\mu} = \frac{1-c}{1+c/\sqrt{k}}\mu. \tag{4.5}$$

It is easy to verify that the average stress over the matrix is given by $\bar{\boldsymbol{\sigma}}^{(1)} = (1-c)^{-1}\bar{\boldsymbol{\sigma}}$, and to compute the second moment of the stress in the matrix $\langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(1)}$. Using this information, the stationarity conditions (3.31) and (3.32) can be easily used to obtain the expressions

$$\frac{\check{\tau}_{(k)}^{(1)}}{\bar{\tau}_e^{(1)}} = \cos\theta_{(k)} - \sqrt{\frac{c}{2}\left(\frac{1-\alpha}{\alpha}\right)\left(\sqrt{k}\cos^2\theta_{(k)} + \frac{1}{\sqrt{k}}\sin^2\theta_{(k)}\right)} \tag{4.6}$$

and

$$\frac{\hat{\tau}_{(k)}^{(1)}}{\bar{\tau}_e^{(1)}} = \cos\theta_{(k)} + \sqrt{\frac{c}{2}\left(\frac{\alpha}{1-\alpha}\right)\left(\sqrt{k}\cos^2\theta_{(k)} + \frac{1}{\sqrt{k}}\sin^2\theta_{(k)}\right)}, \tag{4.7}$$

where $\bar{\tau}_e^{(1)} = \bar{\tau}_e/(1-c)$. Also, it follows, respectively, from the stationarity conditions (3.29) and from the generalized secant conditions (3.30) that

$$\gamma_{(k)}^{(1)} = \frac{1}{\tau_0^n}\left|\hat{\tau}_{(k)}^{(1)}\right|^{n-1}\hat{\tau}_{(k)}^{(1)} - \frac{1}{2\mu_{(k)}^{(1)}}\hat{\tau}_{(k)}^{(1)} \quad \text{and} \quad \frac{1}{2\mu_{(k)}^{(1)}} = \frac{1}{\tau_0^n}\frac{\left|\hat{\tau}_{(k)}^{(1)}\right|^{n-1}\hat{\tau}_{(k)}^{(1)} - \left|\check{\tau}_{(k)}^{(1)}\right|^{n-1}\check{\tau}_{(k)}^{(1)}}{\hat{\tau}_{(k)}^{(1)} - \check{\tau}_{(k)}^{(1)}}. \tag{4.8}$$

An equation for k may then be obtained by substituting expressions (4.6) and (4.7) into $(4.8)_2$ for $\mu_{(k)}^{(1)}$, and then plugging the result into the expression

$$k = \frac{\lambda}{\mu} = \sum_{k=1}^{K}\frac{\sin^2\theta_{(k)}}{2\mu_{(k)}^{(1)}}\left(\sum_{k=1}^{K}\frac{\cos^2\theta_{(k)}}{2\mu_{(k)}^{(1)}}\right)^{-1}, \tag{4.9}$$

which, in turn, is obtained by means of expressions (4.3). Note that k is independent of $\bar{\tau}_e$ and depends only on n, as well as on the geometry of the slip systems (through the angles $\theta_{(k)}$). Having obtained the value of k, the values of the $\check{\tau}_{(k)}^{(1)}$ and $\hat{\tau}_{(k)}^{(1)}$ can be easily computed from (4.6) and (4.7), and then the values of $\gamma_{(k)}^{(1)}$ and $\mu_{(k)}^{(1)}$ can also be computed via (4.8). The constitutive behaviour of the LCC then follows from expressions (4.4) and (4.5), and, as already noted, the macroscopic response and field statistics of the nonlinear porous composite can be computed from the corresponding response and statistics of the LCC via expressions (3.35) and (3.39), (3.40), (3.42), etc.

However, a simple expression for the effective flow stress of the nonlinear porous material may be obtained from expression (3.34) and is given by

$$\frac{\tilde{\tau}_0}{(1-c)\tau_0} = \left\{\sum_{k=1}^{K}\left[\alpha\left|\frac{\check{\tau}_{(k)}^{(1)}}{\bar{\tau}_e^{(1)}}\right|^{n+1} + (1-\alpha)\left|\frac{\hat{\tau}_{(k)}^{(1)}}{\bar{\tau}_e^{(1)}}\right|^{n+1}\right]\right\}^{-1/n}. \tag{4.10}$$

Finally, it is noted that, although in principle it is possible to make other choices for the value of α, in the results to be presented next, α will be set equal to $1/2$, which is the most symmetric choice.

Figure 2 presents plots of the above-described FO-SO estimates for the effective flow stress $\tilde{\tau}_0$ for power-law, porous crystalline materials, as a function of the strain-rate sensitivity $m = 1/n$, for a fixed porosity ($c = 0.25$). The results are normalized by the flow stress τ_0 of the matrix slip systems and are given for two different matrix materials: (*a*) 'square' symmetry ($K = 2$) and (*b*) 'hexagonal' symmetry ($K = 3$). The results are compared with several bounds and estimates for the effective flow stress of the porous crystals, as described next. The Taylor bound is obtained by making use of a uniform strain rate ($\boldsymbol{\epsilon} = \bar{\boldsymbol{\epsilon}}$) in the expression for the effective potential $\tilde{W}$, and incorporates only information on the porosity c. The variational (VAR) upper bounds are obtained by means of the variational linear comparison method of Idiart & Ponte Castañeda [29], making use of the Hashin–Shtrikman–Willis bounds [4,23] for the appropriate LCC, while the relaxed variational (RVAR) upper bounds are obtained by means of the relaxed version of the variational linear comparison method proposed by deBotton & Ponte Castañeda [8], making use of the same bounds for the LCC. Note again that these two bounding methods give the same predictions for $K = 2$, but somewhat different answers for $K = 3$. The partially optimized second-order (PO-SO) estimates were generated by means of the method proposed by Liu & Ponte Castañeda [16] for the stress potential $\tilde{U}$, as well as a dual version for the strain-rate potential $\tilde{W}$ that was recently proposed by Idiart & Vincent [19], using estimates of the Willis type for the relevant LCC. It should be emphasized in this context that these two estimates (PO-SO-U and PO-SO-W) are different for all values of m except $m = 0$ and 1, due to the lack of stationarity with respect to the eigenstrain rates in these estimates. Finally, results are also shown for the estimates of Idiart [31] for sequentially laminated microstructures (LAMs). These microstructures are also 'particulate' in character, with a well-defined matrix

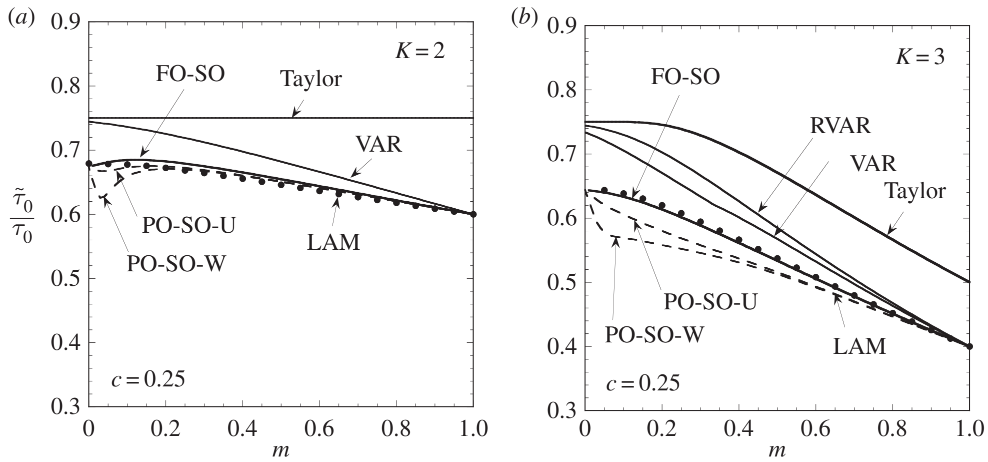

Figure 2. Predictions of the FO-SO method for the effective flow stress $\tilde{\tau}_0$, normalized by the flow stress of the matrix τ_0, for power-law, porous crystalline materials, as a function of the strain-rate sensitivity $m = 1/n$, for a given porosity ($c = 0.25$) and two different crystal symmetries: (*a*) 'square' ($K = 2$) and (*b*) 'hexagonal' ($K = 3$). The macroscopic stress is directed along a slip system ($\bar{\phi} = 0$). Comparisons are given with various bounds and estimates (refer to text for description).

and inclusion phases (i.e. the crystalline and vacuous phases, respectively). They have the property that, in the special case of linearly viscous behaviour ($m = 1$), they agree exactly with the Hashin–Shtrikman–Willis bounds [23] for statistically isotropic microstructures (in the plane), as well as with the Willis-type estimates [24] for statistically isotropic distributions of circular voids.

Having described the various bounds and estimates, we can now make the following observations concerning the new fully optimized (FO-SO) estimates developed in this work. First, the FO-SO estimates satisfy all available upper bounds, including the RVAR bounds [8], as well as the tighter (for $K = 3$ only) VAR bounds [29], which provide generalizations of the Hashin–Shtrikman–Willis bounds for the nonlinear porous composites. Note that these bounds agree with the Hashin–Shtrikman–Willis bounds for $m = 1$. Second, the FO-SO estimates are in very good agreement with—but are not exactly identical to—the LAM estimates [31] for the full range of values of m. Once again, these results are known to agree exactly in the linear case ($m = 1$), but are not expected to be in perfect agreement for other values of m, since they correspond to somewhat different microstructures. The microstructures for the *exact* LAM results shown in figure 2 have isotropic two-point statistics in the transverse plane; however, the voids are not circular, and their centres are distributed anisotropically (albeit in a way that leads to isotropic two-point statistics). On the other hand, the FO-SO estimates make use of *approximate* estimates for the LCC, which do correspond to circular voids whose centres are distributed with isotropic two-point statistics in the transverse plane. Third, the new FO-SO estimates are fairly similar to the earlier PO-SO estimates for $K = 2$, except for fairly low values of the strain-rate sensitivity ($m < 0.2$), and less similar for $K = 3$, when significant differences are observed for values of $m < 0.5$. In particular, the FO-SO estimates do not exhibit the 'bellies' exhibited by the PO-SO estimates for small values of m, when the differences can reach values of about 10%. Interestingly, however, the FO-SO estimates agree exactly with both versions of the PO-SO estimates not only for $m = 1$, but also for $m = 0$. The difference in the predictions of the FO-SO and PO-SO methods for $0 < m < 1$ is expected in view of the fact that the eigenstrain rates in the PO-SO methods are not optimized, leading to a duality gap (i.e. the U and W versions are different), whereas the new FO-SO estimates are fully optimized and therefore exhibit no duality gap. However, it is interesting to further remark that the new FO-SO estimates agree with the PO-SO estimates in the limit as $m \to 0$, when the duality gap in the PO-SO estimates shrinks to zero.

5. Concluding remarks

In this paper, we have developed a variational method for estimating the macroscopic properties of nonlinear composites with viscoplastic crystalline phases in terms of the corresponding properties of suitably designed LCCs. The phases of the LCC are characterized by certain slip viscosities and eigenstrain rates, which in turn play the role of trial fields in a variational statement for the stress potential of the viscoplastic composites. When fully optimized, these slip viscosities and eigenstrain rates are found to be determined by a certain 'generalized secant' linearization of the slip potentials of the nonlinear crystals, and to depend on the first and second moments of the resolved shear stresses in the LCC, which in turn depend on the microstructure of the nonlinear composite. The resulting 'fully optimized' estimates are exact to second order in the contrast and satisfy all known bounds. In addition, they exhibit several desirable and useful properties that were missing in earlier estimates of the second-order type, such as the estimates of Liu & Ponte Castañeda [16], due to the lack of 'full stationarity' with respect to the slip viscosities and eigenstrain rates in the LCC. These properties, which were already present in the earlier—albeit less accurate—variational bounds of deBotton & Ponte Castañeda [8], include the following: (i) the macroscopic response and field statistics of the nonlinear composite can be obtained directly from the corresponding macroscopic response and field statistics in the LCC, thus greatly simplifying the computation of these quantities, and (ii) the lack of a 'duality gap', leading to more accurate predictions. By way of an example, the new method was applied to the simple case of a porous single crystal, and the results were compared with known bounds and earlier estimates, including the exact estimates of Idiart & Vincent [19] for porous crystals with 'sequentially laminated' microstructures [31]. In this context, it should be noted that, while both methods give very similar predictions for this particular example, the new method developed in this work would apply to more general multi-phase composites, including polycrystals, while the method of Idiart [31] is so far restricted to two-phase systems with 'particulate' microstructures. Finally, it should also be noted that, although the new homogenization method developed in this work was presented in the specific context of viscoplastic crystalline composites, the general ideas of the method are expected to be useful for a large number of applications, where homogenized nonlinear properties can be given appropriate variational formulations. These will be explored in future publications.

Competing interests. We declare we have no competing interests.

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