



Field statistics in linearized elastic and viscous composites and polycrystals

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ABSTRACT

This paper presents a general method to estimate the first and second moments of the stress and deformation fields in linearized elastic and viscous composites and polycrystals. The methodology can be seamlessly extended by means of the 'linear comparison' variational homogenization methods (Ponte Castañeda, 1991, 2016) to extract the corresponding field statistics in composites and polycrystals with nonlinear properties, including hyperelastic composites, as well as viscoplastic composites, undergoing finite deformations. Expressions are obtained for the overall response and statistics of the deformation gradient field in hyperelastic composites with linearized (incremental) response characterized by elasticity tensors exhibiting major symmetry, but not minor symmetry. These results are then specialized for composites with fully symmetric elasticity tensors, and use is made of a well-known analogy to convert these results into corresponding estimates for the overall viscosity tensor and statistics of the velocity gradient for linearly viscous composites. Subsequently, the velocity-gradient statistics are projected onto the appropriate symmetric and anti-symmetric subspaces to extract the strain-rate and spin tensor statistics, respectively. The formulation recovers known relations for the overall response and fields statistics, but also allows for the computation of hitherto unexplored quantities, such as the second moments of the spin fluctuations in viscous composites. To illustrate the results, applications are considered for various examples, including two-phase viscous composites, as well as viscous polycrystals, with special types of isotropic and anisotropic linearized response.

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1. Introduction

The effective or overall response of nonlinear composites can be estimated by means of homogenization methods making use of linear comparison composites (LCCs), whose properties are optimized using appropriately designed variational principles (Ponte Castañeda, 1991, 2016). In addition, these methods can be used in combination with certain perturbation identities (Idiart and Ponte Castañeda, 2007a,b) to extract consistent estimates for the first and second moments of the stress and deformation fields in the nonlinear composites in terms of the corresponding estimates in the LCC. Therefore, standard estimates for linear composites and polycrystals (Willis, 1977, 1981) can be used to generate corresponding estimates for nonlinear composites and polycrystals. The simplest such nonlinear homogenization method makes use of a 'secant' linearization of the nonlinear constitutive relation for the phases, where the phase moduli for the 'elastic' LCC depend on the second moments of fields (Ponte Castañeda, 1991; Suquet, 1995; Ponte Castañeda

and Suquet, 1998). Although this method is capable of providing bounds for the overall response, it is accurate only up to first order in the heterogeneity contrast and underestimates the second moments (Moulinec and Suquet, 2003). In addition, it cannot be used for hyperelastic composites undergoing finite strains, as a consequence of the non-convexity of their stored-energy functions. To overcome these limitations, 'second-order' methods were developed by means of more general LCCs with polarizations or eigenstrains. The simplest such method is the 'tangent' second-order method, where the phase moduli of the LCC are evaluated at the first moments or averages of the fields in the phases (Ponte Castañeda, 1996; Ponte Castañeda and Willis, 1999; Ponte Castañeda and Tiberio, 2000). However, the tangent method fails to provide accurate estimates for nonlinear composites composed of phases with large contrasts (Nebozhyn and Ponte Castañeda, 1998; Leroy and Ponte Castañeda, 2001). To remedy this deficiency, improved second-order methods were developed by introducing LCCs that depend on both the averages and second moments of the fields in the phases (Ponte Castañeda, 2002; Liu and Ponte Castañeda, 2004; Lopez-Pamies and Ponte Castañeda, 2004, 2006a). In spite of providing improved results for the macroscopic response, the choice of the properties of the LCC in these improved second-order methods was not fully

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optimized and led to inconsistent results for the field statistics, especially when estimated directly from the corresponding statistics in the LCC (Idiart and Ponte Castañeda, 2007b). Finally, fully optimized second-order methods were developed (Ponte Castañeda, 2015, 2016), which allowed the computation of the macroscopic response and field statistics for nonlinear composites and polycrystals directly and consistently from the corresponding response and statistics in LCCs with the fully optimized properties.

Because of the different Lagrangian and Eulerian descriptions that are commonly used to characterize respectively elastic and viscous response, in this work, we consider, separately, linearized elastic and viscous responses. The results obtained, besides being interesting of their own accord, will provide the foundation for generating, in other works, corresponding results for non-linear elastic and viscoplastic composites by means of the fully optimized second-order methods

For hyperelastic composites with linearized or incremental response, the elasticity tensors of the phases relating the Piola–Kirchhoff (PK) stress to the deformation gradient are generally anisotropic fourth-order tensors with major, but no minor symmetries. As a consequence, it is important to generate estimates for the overall response and field statistics of the PK stress and deformation gradient for linear composites with anisotropic phases and microstructures. In particular, we develop expressions for the field statistics for linearized elastic composites with particulate microstructures, such as those with fibrous microstructures (Honeker and Thomas, 1996; Finlay et al., 1998; Quapp and Weiss, 1998; Honeker et al., 2000), as well as with polycrystalline microstructures, such as polydomain thermoplastic elastomers (Honeker and Thomas, 1996; Racherla et al., 2010).

For viscoplastic composites with linearized constitutive response undergoing finite strains, the viscosity tensor relating the Cauchy stress and Eulerian strain rate are also generally anisotropic, but exhibit both major and minor symmetries. In this work, we also generate estimates for the first and second moments of the stress and velocity gradient fields in these linear composites with anisotropic phases and microstructures. In particular, we develop expressions for the field statistics in linearly viscous composites with particulate microstructures, such as two-phase composites with isotropic (Danas et al., 2008; Furer and Ponte Castañeda, 2018) or crystalline phases (Song and Ponte Castañeda, 2017a,b), as well as linearly viscous composites with granular microstructures, such as viscoplastic polycrystals (Liu and Ponte Castañeda, 2004; Brenner et al., 2004; Lebensohn et al., 2004, 2007; Song and Ponte Castañeda, 2018a). At this point, we should mention that there are established procedures to obtain the first and second moments of the stress and strain-rate fields from the overall potentials of linear composites (Bobeth and Diener, 1986; Parton and Buryachenko, 1990; Kreher, 1990; Ponte Castañeda and Suquet, 1998). In this work, the field statistics are derived using a novel, more general procedure, which will be shown to recover results that have already been reported in the literature (Song and Ponte Castañeda, 2017a, 2018a), thus validating our procedure, but, more importantly, which can be used to generate new results for the first and second moments of the spin tensors in the phases, as well as for the cross-correlation of the strain-rate and spin fields in the phases for linearly viscous composites. These are results that are necessary for the development of more accurate models for texture evolution and recrystallization, accounting for the non-uniformity of the stress and deformation fields in composites with crystalline phases, as have been attempted recently for viscoplastic polycrystals (Petryk et al., 2008; Lebensohn et al., 2016; Zecevic et al., 2017, 2019).

The rest of the paper is organized as follows. In Section 2, we derive expressions for the overall response and field statistics

for linearized elastic composites and polycrystals with moduli exhibiting only major symmetry. Although special forms of these results have been given previously in the context of earlier applications of the second-order method to hyperelastic composites, a more complete and general set of results is given in this section, including results for N -phase composites with general particulate microstructures in Section 2.3, as well as for N -phase composites with granular microstructures (including polycrystals) in Section 2.4. In Section 3, these expressions are specialized for linearly viscous composites and polycrystals with fully symmetric phase moduli to obtain the overall response and velocity-gradient statistics for those composites. Then, these statistics are projected onto symmetric and anti-symmetric subspaces to extract the field statistics of the strain rate and spin tensor. In particular, new analytical expressions for the covariance tensor of the fluctuations of the spin, as well as the cross-covariances of the spin and the strain rate, are given in Section 3.3 for two-phase composites with particulate microstructures, as well as in Section 3.4 for N -phase composites with granular microstructures. Section 4 presents numerical applications for the field statistics in two-phase isotropic incompressible composites with particulate and granular microstructures, as well as in crystalline composites, including porous single crystals and solid polycrystals. In particular, results are given for the first time for the fluctuations of the spin fields in these two-phase composites and polycrystals. These results demonstrate that the fluctuations of the spin fields can be large – and comparable in magnitude to the corresponding fluctuations of the strain-rate field – for composites with large contrast in the phase viscosities as well as for crystalline systems with large grain anisotropy. Finally, in Section 5, we summarize the main features of our formulation and results of our work.

2. Composites with linearized hyperelastic constitutive response

The constitutive response of a hyperelastic material undergoing finite deformation, relating the first Piola–Kirchhoff (PK) stress \mathbf{S} to the deformation gradient \mathbf{F} , can be described by means of the constitutive relation

$$\mathbf{S} = \frac{\partial W}{\partial \mathbf{F}}(\mathbf{F}), \quad (1)$$

where $W(\mathbf{F})$ is the elastic stored-energy function. It is important to recall that both \mathbf{S} and \mathbf{F} are general non-symmetric second-order tensors. The incremental or linearized constitutive response (Ogden, 1997) of such a hyperelastic material is given by $\delta \mathbf{S} = \mathbb{L} \delta \mathbf{F}$, where $\delta \mathbf{F} = \mathbf{F} - \mathbf{F}'$ is the deformation gradient increment, $\delta \mathbf{S} = \mathbf{S} - \mathbf{S}'$ is the stress increment and \mathbb{L} is the incremental elasticity modulus tensor with components

$$L_{ijkl} = \frac{\partial^2 W}{\partial F_{ij} \partial F_{kl}}(\mathbf{F}'). \quad (2)$$

Note that \mathbb{L} is a fourth-order tensor that maps non-symmetric, second-order tensors into non-symmetric, second-order tensors and exhibits *major* symmetry (i.e., $L_{ijkl} = L_{klij}$), but not *minor* symmetry (i.e., $L_{ijkl} \neq L_{jikl}$). By introducing the stress polarization or eigenstress tensor $\mathbf{T} = \mathbf{S}' - \mathbb{L} \mathbf{F}'$, which is also a non-symmetric second-order tensor, the incremental relation can be alternatively written in the form

$$\mathbf{S} = \mathbb{L} \mathbf{F} + \mathbf{T}, \quad (3)$$

where \mathbb{L} is the above-defined incremental elasticity modulus tensor.

It should be emphasized here that, because of the non-uniformity of the deformation field $\mathbf{F}(\mathbf{x})$, both the elasticity tensor \mathbb{L} and the polarization \mathbf{T} are, in general, also non-uniform. This

has the implication that the linearized properties of the phases in a hyperelastic composite material are *non-uniform*—and this is a large part of the difficulty with the nonlinear homogenization problem for such hyperelastic composites. However, nonlinear homogenization methods have been developed (Ponte Castañeda and Tiberio, 2000; Lopez-Pamies and Ponte Castañeda, 2004) making use of linear comparison composites (LCCs) with *uniform* phase properties in order to estimate the macroscopic response of the (nonlinear) hyperelastic composites in terms of corresponding homogenization estimates for the macroscopic response of such *linearized* composites with *uniform* phase properties—as ‘trial’ fields in the context of suitably defined variational principles for the properties of such LCCs. For example, earlier (non-optimized) versions of the ‘second-order’ methods have been used to estimate the macroscopic response of particle- and fiber-reinforced elastomers (Ponte Castañeda and Tiberio, 2000; Lopez-Pamies and Ponte Castañeda, 2006a; Avazmohammadi and Ponte Castañeda, 2014). However, the computation of consistent estimates for the field statistics in such hyperelastic composites using the fully optimized second-order method (Ponte Castañeda, 2016) still remains largely unexplored and this fact partially provides the motivation for the results to be obtained in this section. They will be of use in future works to obtain consistent estimates for the field statistics in (nonlinear) hyperelastic composites.

Thus, in this section, we will describe how to generalize and make use of homogenization methods to generate estimates for the macroscopic behavior and field statistics of linearized composites and polycrystals with constituents exhibiting linear response of the type described by (3) with *uniform* properties $\mathbb{L}^{(r)}$ and $\mathbf{T}^{(r)}$. Note that this type of constitutive behavior is mathematically analogous to infinitesimal thermoelasticity, except that the stress and strain measures are non-symmetric, while $\mathbb{L}^{(r)}$ and $\mathbf{T}^{(r)}$ play the roles of the elasticity modulus tensor and thermal stress, respectively. As a consequence, the homogenization methods to be described below will consist of appropriate generalizations of the classical Hashin–Shtrikman–Willis and self-consistent models for linear thermoelastic composites and polycrystals (Laws, 1973; Willis, 1981) with relaxed symmetry assumptions.

2.1. Local constitutive relation and overall response

We consider composite materials occupying a volume Ω and consisting of N randomly distributed phases, each occupying subvolumes $\Omega^{(r)}$, such that $\Omega = \cup_{r=1}^N \Omega^{(r)}$. As usual, we assume ergodicity and separation of length scales, so that Ω is a representative volume element (RVE) for the composite. Its microstructure is then characterized by indicator functions $\chi^{(r)}(\mathbf{X})$, which take the value 1 if \mathbf{X} is in $\Omega^{(r)}$ and 0 otherwise. The volume average of $\chi^{(r)}(\mathbf{X})$ defines the volume fraction of phase r , i.e. $c^{(r)} = \langle \chi^{(r)}(\mathbf{X}) \rangle = |\Omega^{(r)}|/|\Omega|$. Here, $\langle \cdot \rangle$ is used to denote a volume average over the RVE, while $\langle \cdot \rangle^{(r)}$ will be similarly used to denote an average over phase r . The distribution of phases is characterized by the two-point statistics $p^{(rs)}(\mathbf{X} - \mathbf{X}') = \langle \chi^{(r)}(\mathbf{X}) \chi^{(s)}(\mathbf{X}') \rangle$, which will be assumed to be ‘ellipsoidal’ in this work (Willis, 1977).

Following the above discussion, the local constitutive relation for the composites will be taken to be of the form

$$\mathbf{S} = \frac{\partial W}{\partial \mathbf{F}}(\mathbf{X}, \mathbf{F}), \quad W(\mathbf{X}, \mathbf{F}) = \sum_{r=1}^N \chi^{(r)}(\mathbf{X}) W^{(r)}(\mathbf{F}), \quad (4)$$

where the stored-energy function of phase r is given by

$$W^{(r)}(\mathbf{F}) = \frac{1}{2} \mathbf{F} \cdot \mathbb{L}^{(r)} \mathbf{F} + \mathbf{T}^{(r)} \cdot \mathbf{F} + \frac{1}{2} f^{(r)}, \quad (5)$$

such that

$$\mathbf{S} = \mathbb{L}^{(r)} \mathbf{F} + \mathbf{T}^{(r)}, \quad (6)$$

in phase r . In these expressions, the phase elasticity tensors $\mathbb{L}^{(r)}$ will be assumed to be uniform and to exhibit major, but not minor symmetries, while the phase stress polarization $\mathbf{T}^{(r)}$ and zero deformation-gradient energy $f^{(r)}$ will also be assumed to be uniform non-symmetric second-order tensor and scalar quantities, respectively.

As shown by Hill (1972), the homogenized or effective stored-energy function for the composite is given by

$$\begin{aligned} \tilde{W}(\bar{\mathbf{F}}) &= \min_{\mathbf{F} \in \mathcal{K}(\bar{\mathbf{F}})} \langle W(\mathbf{X}, \mathbf{F}) \rangle \\ &= \min_{\mathbf{F} \in \mathcal{K}(\bar{\mathbf{F}})} \sum_{r=1}^N c^{(r)} \left[\frac{1}{2} \mathbb{L}^{(r)} \cdot \langle \mathbf{F} \otimes \mathbf{F} \rangle^{(r)} + \mathbf{T}^{(r)} \cdot \langle \mathbf{F} \rangle^{(r)} + \frac{1}{2} f^{(r)} \right], \end{aligned} \quad (7)$$

where $\mathcal{K}(\bar{\mathbf{F}}) = \{\mathbf{F} | \exists \mathbf{x} = \chi(\mathbf{X}), \text{ such that } \mathbf{F} = \nabla \chi \text{ in } \Omega \text{ and } \mathbf{x} = \bar{\mathbf{F}}\mathbf{X} \text{ on } \partial\Omega\}$, from which it follows that the relation between the macroscopic stress $\bar{\mathbf{S}} = \langle \mathbf{S} \rangle$ and the macroscopic deformation gradient $\bar{\mathbf{F}} = \langle \mathbf{F} \rangle$ is given by

$$\bar{\mathbf{S}} = \frac{\partial \tilde{W}}{\partial \bar{\mathbf{F}}}. \quad (8)$$

It should be noted that the elasticity tensors $\mathbb{L}^{(r)}$ of elastomeric phases need not be positive definite, and are usually assumed to be such that the energy functions $W^{(r)}$ are polyconvex, or at least strongly elliptic (Ball, 1977). In such cases, the macroscopic stored-energy function \tilde{W} can still be defined, although the possibility of microscopic and macroscopic instabilities cannot be ruled out (Geymonat et al., 1993; Avazmohammadi and Ponte Castañeda, 2016). In this section, the assumption will be made, for simplicity, that the elasticity tensors $\mathbb{L}^{(r)}$ are *positive definite*, so that the energy functions $W^{(r)}$ are strictly convex and the macroscopic stored-energy function \tilde{W} is always defined by a unique solution. More specific applications to elastomeric composites where this assumption must be relaxed will be provided elsewhere.

Following the analysis of Laws (1973) for linear thermoelastic composites, Ponte Castañeda and Tiberio (2000) used linearity to argue that the average of the deformation gradient over phase r in linearized elastic composites can be expressed in the form

$$\bar{\mathbf{F}}^{(r)} = \langle \mathbf{F} \rangle^{(r)} = \mathbb{A}^{(r)} \bar{\mathbf{F}} + \mathbf{a}^{(r)}, \quad (9)$$

where $\mathbb{A}^{(r)}$ and $\mathbf{a}^{(r)}$ are respectively (fully non-symmetric) fourth- and second-order phase concentration tensors, such that

$$\sum_{r=1}^N c^{(r)} \mathbb{A}^{(r)} = \mathbb{I} \quad \text{and} \quad \sum_{r=1}^N c^{(r)} \mathbf{a}^{(r)} = \mathbf{0}, \quad (10)$$

consistently with the average relation

$$\bar{\mathbf{F}} = \sum_{r=1}^N c^{(r)} \bar{\mathbf{F}}^{(r)}. \quad (11)$$

In the above expression, \mathbb{I} is the fourth-order identity tensor with components $I_{ijkl} = \delta_{ik} \delta_{jl}$.

Making use of the linearity of \mathbf{F} on $\bar{\mathbf{F}}$ (i.e., $\mathbf{F}(\mathbf{X}) = \mathbb{A}(\mathbf{X}) \bar{\mathbf{F}} + \mathbf{a}(\mathbf{X})$), it can then be shown that the macroscopic response of the composite can be described by the homogenized stored-energy function

$$\tilde{W}(\bar{\mathbf{F}}) = \frac{1}{2} \bar{\mathbf{F}} \cdot \tilde{\mathbb{L}} \bar{\mathbf{F}} + \tilde{\mathbf{T}} \cdot \bar{\mathbf{F}} + \frac{1}{2} \tilde{f}, \quad (12)$$

such that

$$\bar{\mathbf{S}} = \tilde{\mathbb{L}} \bar{\mathbf{F}} + \tilde{\mathbf{T}}, \quad (13)$$

where $\tilde{\mathbb{L}}$ is the effective elasticity tensor, $\tilde{\mathbf{T}}$ is the effective stress polarization and \tilde{f} is the effective zero deformation-gradient energy and are given by the expressions

$$\begin{aligned}\tilde{\mathbb{L}} &= \sum_{r=1}^N c^{(r)} \mathbb{L}^{(r)} \mathbb{A}^{(r)}, \quad \tilde{\mathbf{T}} = \sum_{r=1}^N c^{(r)} (\mathbb{A}^{(r)})^T \mathbf{T}^{(r)} \quad \text{and} \\ \tilde{f} &= \sum_{r=1}^N c^{(r)} (f^{(r)} + \mathbf{T}^{(r)} \cdot \mathbf{a}^{(r)}).\end{aligned}\quad (14)$$

These results follow the same derivations of their classical linear elasticity counterparts, but without the additional symmetrization steps needed to account for the symmetry of the infinitesimal strain and stress tensor (Laws, 1973). Although the major symmetry of $\tilde{\mathbb{L}}$ is not apparent from the above expression, it follows from the existence of the overall stored-energy function (7).

Under the assumption of positive definiteness of the moduli tensors $\mathbb{L}^{(r)}$, the constitutive response of the composite can be equivalently described in terms of the dual potential $U^{(r)}(\mathbf{S})$, which is defined as

$$U^{(r)}(\mathbf{S}) = \sup_{\mathbf{F}} [\mathbf{S} \cdot \mathbf{F} - W^{(r)}(\mathbf{F})] = \frac{1}{2} \mathbf{S} \cdot \mathbb{M}^{(r)} \mathbf{S} + \mathbf{G}^{(r)} \cdot \mathbf{S} + \frac{1}{2} h^{(r)}, \quad (15)$$

where the phase compliance tensor $\mathbb{M}^{(r)}$, deformation-gradient polarization $\mathbf{G}^{(r)}$ and zero stress energy $h^{(r)}$ are

$$\begin{aligned}\mathbb{M}^{(r)} &= (\mathbb{L}^{(r)})^{-1}, \quad \mathbf{G}^{(r)} = -(\mathbb{L}^{(r)})^{-1} \mathbf{T}^{(r)}, \\ h^{(r)} &= -f^{(r)} + \mathbf{T}^{(r)} \cdot (\mathbb{L}^{(r)})^{-1} \mathbf{T}^{(r)}.\end{aligned}\quad (16)$$

The macroscopic stress potential $\tilde{U}(\bar{\mathbf{S}})$ can then be defined as

$$\begin{aligned}\tilde{U}(\bar{\mathbf{S}}) &= \inf_{\mathbf{S} \in \mathcal{T}(\bar{\mathbf{S}})} \sum_{r=1}^N c^{(r)} \langle U^{(r)}(\mathbf{S}) \rangle^{(r)} \\ &= \inf_{\mathbf{S} \in \mathcal{T}(\bar{\mathbf{S}})} \sum_{r=1}^N c^{(r)} \left[\frac{1}{2} \mathbb{M}^{(r)} \cdot \langle \mathbf{S} \otimes \mathbf{S} \rangle^{(r)} + \mathbf{G}^{(r)} \cdot \langle \mathbf{S} \rangle^{(r)} + \frac{1}{2} g^{(r)} \right],\end{aligned}\quad (17)$$

where $\mathcal{T}(\bar{\mathbf{S}}) = \{\mathbf{S} | \nabla \cdot \mathbf{S} = \mathbf{0} \text{ in } \Omega \text{ and } \mathbf{S} \mathbf{n} = \bar{\mathbf{S}} \mathbf{n} \text{ on } \partial\Omega\}$, and is such that $\bar{\mathbf{F}} = \partial \tilde{U} / \partial \bar{\mathbf{S}}$.

From the linearity of the problem, it is again possible to introduce stress concentration tensors $\mathbb{B}^{(r)}$ and $\mathbf{b}^{(r)}$, such that

$$\bar{\mathbf{S}}^{(r)} = \mathbb{B}^{(r)} \bar{\mathbf{S}} + \mathbf{b}^{(r)}, \quad (18)$$

where

$$\sum_{r=1}^N c^{(r)} \mathbb{B}^{(r)} = \mathbb{I} \quad \text{and} \quad \sum_{r=1}^N c^{(r)} \mathbf{b}^{(r)} = \mathbf{0}, \quad (19)$$

consistently with the average relation $\bar{\mathbf{S}} = \sum_{r=1}^N c^{(r)} \bar{\mathbf{S}}^{(r)}$. In terms of the stress concentration tensors and phase moduli, the overall compliance $\tilde{\mathbb{M}}$, deformation-gradient polarization $\tilde{\mathbf{G}}$ and zero stress energy \tilde{h} can then be expressed as

$$\begin{aligned}\tilde{\mathbb{M}} &= \sum_{r=1}^N c^{(r)} \mathbb{M}^{(r)} \mathbb{B}^{(r)}, \quad \tilde{\mathbf{G}} = \sum_{r=1}^N c^{(r)} (\mathbb{B}^{(r)})^T \mathbf{G}^{(r)} \quad \text{and} \\ \tilde{h} &= \sum_{r=1}^N c^{(r)} (h^{(r)} + \mathbf{G}^{(r)} \cdot \mathbf{b}^{(r)}).\end{aligned}\quad (20)$$

Due to the separation of length scales hypothesis and the assumed positive definiteness of the moduli tensors, it follows that the overall stress potential $\tilde{U}(\bar{\mathbf{S}})$ is the Legendre dual of the

overall stored-energy function $\tilde{W}(\bar{\mathbf{F}})$, so that

$$\tilde{U}(\bar{\mathbf{S}}) = \sup_{\bar{\mathbf{F}}} [\bar{\mathbf{S}} \cdot \bar{\mathbf{F}} - \tilde{W}(\bar{\mathbf{F}})] = \frac{1}{2} \bar{\mathbf{S}} \cdot \tilde{\mathbb{M}} \bar{\mathbf{S}} + \tilde{\mathbf{G}} \cdot \bar{\mathbf{S}} + \frac{1}{2} \tilde{h}, \quad (21)$$

where the effective moduli in the overall stress potential are related to the corresponding effective moduli of the overall stored-energy function (7) by the expressions

$$\tilde{\mathbb{M}} = \tilde{\mathbb{L}}^{-1}, \quad \tilde{\mathbf{G}} = -\tilde{\mathbb{L}}^{-1} \tilde{\mathbf{T}}, \quad \tilde{h} = -\tilde{f} + \tilde{\mathbf{T}} \cdot \tilde{\mathbb{L}}^{-1} \tilde{\mathbf{T}}, \quad (22)$$

in full analogy to expressions (16) for the phase moduli.

2.2. Identities for the field statistics

In this section, we provide some exact relations for the field statistics in the above-defined linear elastic composites, generalizing corresponding results for infinitesimal elasticity (see, for example, Ponte Castañeda and Suquet, 1998). Thus, making use of the stationarity of the right-hand side of expression (7) for \tilde{W} , together with the generalized chain rule, it follows by differentiation with respect to $\mathbf{T}^{(r)}$ and $\mathbb{L}^{(r)}$, respectively, that the first and second moments of the deformation gradient in phases are given by

$$\bar{\mathbf{F}}^{(r)} = \frac{1}{c^{(r)}} \frac{\partial \tilde{W}}{\partial \mathbf{T}^{(r)}}(\bar{\mathbf{F}}), \quad \langle \mathbf{F} \otimes \mathbf{F} \rangle^{(r)} = \frac{2}{c^{(r)}} \frac{\partial \tilde{W}}{\partial \mathbb{L}^{(r)}}(\bar{\mathbf{F}}). \quad (23)$$

It should be noted that the first of these expressions for the phase average $\bar{\mathbf{F}}^{(r)}$ can be shown to be exactly consistent with expression (9) by making use of Eq. (12) for the overall stored-energy function, together with Eqs. (14) for the effective moduli. It also follows from Eq. (7) that

$$\tilde{W}(\bar{\mathbf{F}}) = \sum_{r=1}^N \left[\mathbb{L}^{(r)} \cdot \frac{\partial \tilde{W}}{\partial \mathbb{L}^{(r)}} + \mathbf{T}^{(r)} \cdot \frac{\partial \tilde{W}}{\partial \mathbf{T}^{(r)}} + f^{(r)} \frac{\partial \tilde{W}}{\partial f^{(r)}} \right], \quad (24)$$

which is consistent with the fact that \tilde{W} is homogeneous of degree one in the moduli $\mathbb{L}^{(r)}$, $\mathbf{T}^{(r)}$ and $f^{(r)}$.

Similarly, by taking derivatives of expression (17) for the macroscopic stress potential $\tilde{U}(\bar{\mathbf{S}})$ with respect to $\mathbf{G}^{(r)}$ and $\mathbb{M}^{(r)}$, the following identities are obtained for the first and second moments of the stress field

$$\bar{\mathbf{S}}^{(r)} = \frac{1}{c^{(r)}} \frac{\partial \tilde{U}}{\partial \mathbf{G}^{(r)}}(\bar{\mathbf{S}}), \quad \langle \mathbf{S} \otimes \mathbf{S} \rangle^{(r)} = \frac{2}{c^{(r)}} \frac{\partial \tilde{U}}{\partial \mathbb{M}^{(r)}}(\bar{\mathbf{S}}). \quad (25)$$

It is again noted that the above expression for $\bar{\mathbf{S}}^{(r)}$ is exactly consistent with expression (18), when (21) is used in conjunction with (20).

Alternatively, the stress statistics can also be derived from the deformation-gradient statistics using the local constitutive relation (6), i.e.,

$$\begin{aligned}\bar{\mathbf{S}}^{(r)} &= \mathbb{L}^{(r)} \bar{\mathbf{F}}^{(r)} + \mathbf{T}^{(r)}, \\ \langle \mathbf{S} \otimes \mathbf{S} \rangle^{(r)} &= \mathbb{L}^{(r)} \langle \mathbf{F} \otimes \mathbf{F} \rangle^{(r)} \mathbb{L}^{(r)} + (\mathbb{L}^{(r)} \bar{\mathbf{F}}^{(r)}) \otimes \mathbf{T}^{(r)} \\ &\quad + \mathbf{T}^{(r)} \otimes (\mathbb{L}^{(r)} \bar{\mathbf{F}}^{(r)}) + \mathbf{T}^{(r)} \otimes \mathbf{T}^{(r)}.\end{aligned}\quad (26)$$

The equivalence of (25) and (26) stems from the duality of the formulations based on stress potentials and stored-energy functions.

Defining the stress and deformation-gradient fluctuation covariance tensors in phase r by

$$\begin{aligned}\mathbb{C}^{(r)}(\mathbf{S}) &= \langle \mathbf{S} \otimes \mathbf{S} \rangle^{(r)} - \bar{\mathbf{S}}^{(r)} \otimes \bar{\mathbf{S}}^{(r)} \quad \text{and} \\ \mathbb{C}^{(r)}(\mathbf{F}) &= \langle \mathbf{F} \otimes \mathbf{F} \rangle^{(r)} - \bar{\mathbf{F}}^{(r)} \otimes \bar{\mathbf{F}}^{(r)},\end{aligned}\quad (27)$$

respectively, and using the major symmetry of the elasticity tensors $\mathbb{L}^{(r)}$, the above relations for the first and second moments

of the stress lead to the following identities relating the stress covariance in phase r to the deformation-gradient covariance in phase r :

$$\mathbb{C}^{(r)}(\mathbf{S}) = \mathbb{L}^{(r)} \mathbb{C}^{(r)}(\mathbf{F}) \mathbb{L}^{(r)}. \quad (28)$$

For completeness, we also define here the overall fluctuation covariance tensors $\mathbb{C}(\mathbf{S})$ and $\mathbb{C}(\mathbf{F})$ by means of

$$\begin{aligned} \mathbb{C}(\mathbf{S}) &= \sum_{r=1}^N c^{(r)} \langle \mathbf{S} \otimes \mathbf{S} \rangle^{(r)} - \bar{\mathbf{S}} \otimes \bar{\mathbf{S}} \quad \text{and} \\ \mathbb{C}(\mathbf{F}) &= \sum_{r=1}^N c^{(r)} \langle \mathbf{F} \otimes \mathbf{F} \rangle^{(r)} - \bar{\mathbf{F}} \otimes \bar{\mathbf{F}}. \end{aligned} \quad (29)$$

In the next two subsections, we use the above-described relations to provide more explicit expressions for the macroscopic response and field statistics for composites with two types of microstructures: (i) particulate microstructures consisting of inclusions embedded in a continuous matrix phase, and (ii) granular microstructures consisting of grains surrounded by other grains, such that there is no continuous matrix phase. Since the constitutive relations involve the non-symmetric PK stress and deformation gradient, the classical formulations need to be suitably generalized (e.g., Ponte Castañeda and Tiberio, 2000). In particular, we will consider here appropriate generalization of the PCW estimates of Willis (1978) and Ponte Castañeda and Willis (1995) for particulate microstructures and the self-consistent (SC) estimates of Hershey (1954), Kröner (1958) and Willis (1977) for granular microstructures.

2.3. Composites with particulate microstructure: Ponte Castañeda-Willis estimates

2.3.1. N -phase composites with particulate microstructure

Following Ponte Castañeda and Willis (1995), we consider N -phase composites with particulate microstructures consisting of $N - 1$ families of aligned ellipsoidal inclusions with elastic properties $\mathbb{L}^{(r)}$, $\mathbf{T}^{(r)}$, $f^{(r)}$ and volume fractions $c^{(r)}$ ($r = 2, \dots, N$) that are distributed randomly in a matrix phase with properties $\mathbb{L}^{(1)}$, $\mathbf{T}^{(1)}$, $f^{(1)}$ and volume fraction $c^{(1)}$. The inclusions of a given phase r are assumed to be identical in shape and orientation, as determined by the shape tensors $\mathbf{Z}_i^{(r)}$, such that $\Omega_i^{(r)} = \{\mathbf{X}, |(\mathbf{Z}_i^{(r)})^{-T} \mathbf{X}| \leq 1\}$. Moreover, the distribution of the inclusions is characterized by the two-point correlation function for the particle centers, which is assumed to possess ‘ellipsoidal symmetry,’ as defined by the shape tensor \mathbf{Z}_d .

The phase and macroscopic stored-energy functions are given by (5) and (7), respectively, with the overall properties $\bar{\mathbb{L}}$, $\bar{\mathbf{T}}$ and \bar{f} described in terms of the corresponding phase properties and concentration tensors by expressions (14). For the particulate composites described above, the concentration tensors are obtained by generalizing the formulation of Ponte Castañeda and Willis (1995) for infinitesimal linear-elastic composites to incremental linear-elastic composites with more general phase constitutive relations (6), following earlier generalizations for infinitesimal linear-elastic thermoelastics (Ponte Castañeda, 2005). The procedure is essentially the same, except that the minor symmetrization step required in infinitesimal thermoelasticity is not needed here. For that reason, the derivation will not be detailed here and we will simply state the resulting expressions for the concentration tensors for the inclusion phases ($r = 2, \dots, N$) as

$$\begin{aligned} \mathbb{A}^{(r)} &= [\mathbb{I} + \mathbb{P}_i^{(r)} \Delta \mathbb{L}^{(r)}]^{-1} \mathbb{X}, \\ \mathbf{a}^{(r)} &= [\mathbb{I} + \mathbb{P}_i^{(r)} \Delta \mathbb{L}^{(r)}]^{-1} \left[\mathbb{P}_d \sum_{s=1}^N c^{(s)} (\mathbb{A}^{(s)})^T \mathbf{T}^{(s)} - \mathbb{P}_i^{(r)} \mathbf{T}^{(r)} \right], \end{aligned} \quad (30)$$

where

$$\begin{aligned} \Delta \mathbb{L}^{(r)} &= \mathbb{L}^{(r)} - \mathbb{L}^{(1)} \quad \text{and} \\ \mathbb{X} &= \left\{ \sum_{s=1}^N c^{(s)} [\mathbb{I} + (\mathbb{P}_i^{(s)} - \mathbb{P}_d) \Delta \mathbb{L}^{(s)}] [\mathbb{I} + \mathbb{P}_i^{(s)} \Delta \mathbb{L}^{(s)}]^{-1} \right\}^{-1}. \end{aligned} \quad (31)$$

The microstructural tensors $\mathbb{P}_i^{(r)}$ and \mathbb{P}_d depend on the matrix stiffness $\mathbb{L}^{(1)}$ through the acoustic tensor $\mathbf{K}^{(1)}$ with components $K_{ik}^{(1)} = L_{ijkl}^{(1)} \zeta_j \zeta_l$, and on the inclusion shape tensor $\mathbf{Z}_i^{(r)}$ and the distribution shape tensor \mathbf{Z}_d , respectively. For example, when the particle and distribution shape tensors are identical $\mathbf{Z}_i^{(r)} = \mathbf{Z}_d = \mathbf{Z}$, $\mathbb{P}_i^{(r)} = \mathbb{P}_d = \mathbb{P}^{(1)}$, where the Cartesian components of $\mathbb{P}^{(1)}$ are given by

$$P_{ijkl}^{(1)} = \frac{\det(\mathbf{Z})}{4\pi} \int_{|\boldsymbol{\zeta}|=1} \frac{N_{ik}^{(1)} \zeta_j \zeta_l}{|\mathbf{Z}\boldsymbol{\zeta}|^3} dS_{\boldsymbol{\zeta}}, \quad (32)$$

where $\mathbf{N}^{(1)} = (\mathbf{K}^{(1)})^{-1}$. (Thus, $\mathbb{P}_i^{(r)}$ and \mathbb{P}_d are obtained by replacing \mathbf{Z} by $\mathbf{Z}_i^{(r)}$ and \mathbf{Z}_d , respectively.) Note that $P_{ijkl}^{(1)} = P_{klij}^{(1)}$ and that the results of Ponte Castañeda and Willis (1995) for infinitesimal linear-elastic composites are obtained by further symmetrizing with respect to the subscript pairs i, j .

In terms of the inclusion concentration tensors given by expressions (30) and (31), the overall moduli can then be readily obtained by means of the expressions

$$\begin{aligned} \bar{\mathbb{L}} &= \mathbb{L}^{(1)} + \sum_{r=2}^N c^{(r)} \Delta \mathbb{L}^{(r)} \mathbb{A}^{(r)}, \quad \bar{\mathbf{T}} = \mathbf{T}^{(1)} + \sum_{r=2}^N c^{(r)} (\mathbb{A}^{(r)})^T \Delta \mathbf{T}^{(r)}, \\ \bar{f} &= \bar{f} + \sum_{r=2}^N c^{(r)} \Delta \mathbf{T}^{(r)} \cdot \mathbf{a}^{(r)}, \end{aligned} \quad (33)$$

with $\Delta \mathbb{L}^{(r)}$ given by (31)₁, $\Delta \mathbf{T}^{(r)} = \mathbf{T}^{(r)} - \mathbf{T}^{(1)}$ and $\bar{f} = \sum_{r=1}^N c^{(r)} f^{(r)}$.

Next, the average deformation gradient in the inclusions $\bar{\mathbf{F}}^{(r)}$ ($r = 2, \dots, N$) can be computed by means of expressions (9), together with expressions (30) and (31) for the concentration tensors, while the average deformation gradient in the matrix phase follows from the average condition and is given by $\bar{\mathbf{F}}^{(1)} = (1/c^{(1)}) (\bar{\mathbf{F}} - \sum_{r=2}^N c^{(r)} \bar{\mathbf{F}}^{(r)})$. On the other hand, the second moment in the phases can be obtained by means of expressions (23)₂, together with (12) and (33), leading to

$$\langle \mathbf{F} \otimes \mathbf{F} \rangle^{(r)} = \frac{1}{c^{(r)}} \left[\bar{\mathbf{F}} \cdot \frac{\partial \bar{\mathbb{L}}}{\partial \mathbb{L}^{(r)}} \bar{\mathbf{F}} + 2 \frac{\partial \bar{\mathbf{T}}}{\partial \mathbb{L}^{(r)}} \cdot \bar{\mathbf{F}} + \frac{\partial \bar{f}}{\partial \mathbb{L}^{(r)}} \right], \quad (34)$$

where the differential operators $\partial \bar{\mathbb{L}} / \partial \mathbb{L}^{(r)}$, $\partial \bar{\mathbf{T}} / \partial \mathbb{L}^{(r)}$ and $\partial \bar{f} / \partial \mathbb{L}^{(r)}$ are respectively eighth-, sixth- and fourth-order tensors with Cartesian components given by

$$\frac{\partial \bar{L}_{ijkl}}{\partial L_{rsuv}^{(r)}}, \quad \frac{\partial \bar{T}_{ij}}{\partial L_{rsuv}^{(r)}} \quad \text{and} \quad \frac{\partial \bar{f}}{\partial L_{rsuv}^{(r)}}. \quad (35)$$

In this context, it should be noted that the free indices in expression (34) for $\langle F_{rs} F_{uv} \rangle^{(r)}$ are associated with the denominators $\partial L_{rsuv}^{(r)}$, so that the other indices, such as in $\partial \bar{L}_{ijkl}$ and $\partial \bar{T}_{ij}$, are summed over in the usual way:

$$\langle F_{rs} F_{uv} \rangle^{(r)} = \frac{1}{c^{(r)}} \left[\bar{F}_{ij} \frac{\partial \bar{L}_{ijkl}}{\partial L_{rsuv}^{(r)}} \bar{F}_{kl} + 2 \frac{\partial \bar{T}_{ij}}{\partial L_{rsuv}^{(r)}} \bar{F}_{ij} + \frac{\partial \bar{f}}{\partial L_{rsuv}^{(r)}} \right]. \quad (36)$$

Completely analogous expressions are available for the first and second moments of the stress in the phases.

It can be shown, by direct computation, that the second moments in the inclusions ($r = 2, \dots, N$) are given directly in terms of the phase averages by means of

$$\langle \mathbf{F} \otimes \mathbf{F} \rangle^{(r)} = \bar{\mathbf{F}}^{(r)} \otimes \bar{\mathbf{F}}^{(r)} \quad \text{and} \quad \langle \mathbf{S} \otimes \mathbf{S} \rangle^{(r)} = \bar{\mathbf{S}}^{(r)} \otimes \bar{\mathbf{S}}^{(r)}, \quad (37)$$

implying that the fluctuation covariance tensors in the inclusions are null, so that

$$\mathbb{C}^{(r)}(\mathbf{F}) = \mathbb{O} \quad \text{and} \quad \mathbb{C}^{(r)}(\mathbf{S}) = \mathbb{O}, \quad (38)$$

irrespective of the anisotropy of the phases and microstructural shape tensors. This is a consequence of the uniform polarizations in the inclusions associated with the Hashin–Shtrikman–Willis approximation that is used in the PCW estimates for particulate composites, which manifests itself through the independence of the microstructural tensors $\mathbb{P}_i^{(r)}$ and \mathbb{P}_d on the properties of the inclusions. On the other hand, the second moment of the deformation-gradient field in the matrix phase ($r = 1$) involves the derivative with respect to the matrix stiffness tensor $\mathbb{L}^{(1)}$ of the overall stiffness tensor $\tilde{\mathbb{L}}$, of the overall stress polarization $\tilde{\mathbf{T}}$ and of the overall zero deformation-gradient energy \tilde{f} —all of which exhibit additional dependence on $\mathbb{L}^{(1)}$ through the tensors $\mathbb{P}_i^{(r)}$ and \mathbb{P}_d . As a consequence, the covariance tensors of the deformation gradient and stress fields in the matrix are non-zero, but their expressions are quite complicated and will not be further detailed here. Instead, in the next subsection, we will provide more explicit expressions for the first and second moments of the fields in two-phase particulate composites.

2.3.2. Two-phase composites with particulate microstructure

In this subsection, we consider two-phase composites with particulate microstructures for which the inclusions have elastic properties $\mathbb{L}^{(2)}, \mathbf{T}^{(2)}, f^{(2)}$ and volume fraction $c^{(2)}$ and are randomly distributed within a matrix phase with properties $\mathbb{L}^{(1)}, \mathbf{T}^{(1)}, f^{(1)}$ and volume fraction $c^{(1)} = 1 - c^{(2)}$. Moreover, for simplicity, the particle and distribution shape tensors are assumed to be identical, such that $\mathbf{Z}_i^{(2)} = \mathbf{Z}_d = \mathbf{Z}$. In this case, it was shown by Ponte Castañeda and Tiberio (2000), by means of an appropriate generalization of the Levin's relations (Levin, 1992), that the concentration tensors can be written as

$$\begin{aligned} \mathbb{A}^{(2)} &= [\mathbb{I} + c^{(1)}\mathbb{P}^{(1)}\Delta\mathbb{L}]^{-1}, \\ \mathbf{a}^{(2)} &= (\Delta\mathbb{L})^{-1}(\mathbb{A}^{(2)} - \mathbb{I})^T \Delta\mathbf{T} = -c^{(1)}\mathbb{P}^{(1)}\mathbb{A}^{(2)T} \Delta\mathbf{T}, \end{aligned} \quad (39)$$

where $\Delta\mathbb{L} = \mathbb{L}^{(2)} - \mathbb{L}^{(1)}$, $\Delta\mathbf{T} = \mathbf{T}^{(2)} - \mathbf{T}^{(1)}$ and $\mathbb{P}^{(1)}$ is the microstructural tensor defined by expression (32). In addition, the effective properties (33) for the two-phase composites can be written in the simplified form

$$\begin{aligned} \tilde{\mathbb{L}} &= \mathbb{L}^{(1)} + c^{(2)}[(\mathbb{L}^{(2)} - \mathbb{L}^{(1)})^{-1} + c^{(1)}\mathbb{P}^{(1)}]^{-1}, \\ \tilde{\mathbf{T}} &= c^{(1)}\mathbf{T}^{(1)} + c^{(2)}\mathbf{T}^{(2)} + (\tilde{\mathbb{L}} - \langle\mathbb{L}\rangle)(\Delta\mathbb{L})^{-1} \Delta\mathbf{T}, \\ \tilde{f} &= c^{(1)}f^{(1)} + c^{(2)}f^{(2)} + \Delta\mathbf{T} \cdot [(\Delta\mathbb{L})^{-1}(\tilde{\mathbb{L}} - \langle\mathbb{L}\rangle)(\Delta\mathbb{L})^{-1}] \Delta\mathbf{T}. \end{aligned} \quad (40)$$

The first moment of the deformation gradient in the inclusion phase then follows trivially from $\bar{\mathbf{F}}^{(2)} = \mathbb{A}^{(2)}\bar{\mathbf{F}} + \mathbf{a}^{(2)}$, together with expressions (39), while the corresponding first moment in the matrix phase can be obtained from the average condition $\bar{\mathbf{F}}^{(1)} = (1/c^{(1)})(\bar{\mathbf{F}} - c^{(2)}\bar{\mathbf{F}}^{(2)})$. As pointed out in the more general case, the fluctuation covariance tensor in the inclusion phase is null, i.e.,

$$\mathbb{C}^{(2)}(\mathbf{F}) = \mathbb{O}. \quad (41)$$

On the other hand, the fluctuation covariance in the matrix phase is given by

$$\begin{aligned} \mathbb{C}^{(1)}(\mathbf{F}) &= -\frac{1}{c^{(2)}}(\bar{\mathbf{F}} - \bar{\mathbf{F}}^{(1)}) \otimes (\bar{\mathbf{F}} - \bar{\mathbf{F}}^{(1)}) \\ &\quad - \frac{1}{c^{(2)}} \left[(\mathbb{P}^{(1)})^{-1}(\bar{\mathbf{F}} - \bar{\mathbf{F}}^{(1)}) \right] \cdot \frac{\partial \mathbb{P}^{(1)}}{\partial \mathbb{L}^{(1)}} \left[(\mathbb{P}^{(1)})^{-1}(\bar{\mathbf{F}} - \bar{\mathbf{F}}^{(1)}) \right], \end{aligned} \quad (42)$$

where the differential operator $\partial \mathbb{P}^{(1)} / \partial \mathbb{L}^{(1)}$ has Cartesian components given by

$$\begin{aligned} \frac{\partial P_{ijkl}^{(1)}}{\partial L_{rsuv}^{(1)}} &= -\frac{\det(\mathbf{Z})}{8\pi} \int_{|\xi|=1} \frac{1}{|\mathbf{Z}\xi|^3} \left[(N_{ir}^{(1)}\xi_s)(N_{ku}^{(1)}\xi_v) \right. \\ &\quad \left. + (N_{kr}^{(1)}\xi_s)(N_{iu}^{(1)}\xi_v) \right] \xi_j \xi_l dS_\xi, \end{aligned} \quad (43)$$

(recall that $\mathbf{N}^{(1)}$ is the inverse of the acoustic tensor $\mathbf{K}^{(1)}$) and where the free indices of $\mathbb{C}_{rsuv}^{(1)}$ are matched up with those associated with $\partial L_{rsuv}^{(1)}$ in the second term in the right hand side. Thus, we have expressed the second moments of the fluctuations in the matrix phase in terms of the phase moduli, concentration tensors, overall deformation gradient as well as the derivative of the microstructural tensor $\mathbb{P}^{(1)}$ with respect to the matrix modulus $\mathbb{L}^{(1)}$. The derivation of the above expressions is straightforward and is omitted here for brevity (Lopez-Pamies and Ponte Castañeda, 2006a; Siboni and Ponte Castañeda, 2016).

The statistics of the stress follow similarly. The stress concentration tensors for the inclusions are given by

$$\begin{aligned} \mathbb{B}^{(2)} &= [\mathbb{I} + c^{(1)}\mathbb{Q}^{(1)}\Delta\mathbb{M}]^{-1}, \\ \mathbf{b}^{(2)} &= (\Delta\mathbb{M})^{-1}(\mathbb{B}^{(2)} - \mathbb{I})^T \Delta\mathbf{G} = -c^{(1)}\mathbb{Q}^{(1)}\mathbb{B}^{(2)T} \Delta\mathbf{G}, \end{aligned} \quad (44)$$

where $\Delta\mathbb{M} = \mathbb{M}^{(2)} - \mathbb{M}^{(1)}$, $\Delta\mathbf{G} = \mathbf{G}^{(2)} - \mathbf{G}^{(1)}$ and $\mathbb{Q}^{(1)}$ is the microstructural tensor, defined by $\mathbb{Q}^{(1)} = \mathbb{L}^{(1)} - \mathbb{L}^{(1)}\mathbb{P}^{(1)}\mathbb{L}^{(1)}$. The first moments of the stress are then given by

$$\bar{\mathbf{S}}^{(2)} = \mathbb{B}^{(2)}\bar{\mathbf{S}} + \mathbf{b}^{(2)} \quad \text{and} \quad \bar{\mathbf{S}}^{(1)} = \frac{1}{c^{(1)}}(\bar{\mathbf{S}} - c^{(2)}\bar{\mathbf{S}}^{(2)}). \quad (45)$$

Similar to (41) and (42), the PCW estimates for the covariance of the fluctuations of the stress field in the inclusions and matrix phase are respectively given by

$$\mathbb{C}^{(2)}(\mathbf{S}) = \mathbb{O}, \quad (46)$$

and

$$\begin{aligned} \mathbb{C}^{(1)}(\mathbf{S}) &= -\frac{1}{c^{(2)}}(\bar{\mathbf{S}} - \bar{\mathbf{S}}^{(1)}) \otimes (\bar{\mathbf{S}} - \bar{\mathbf{S}}^{(1)}) \\ &\quad - \frac{1}{c^{(2)}} \left[(\mathbb{Q}^{(1)})^{-1}(\bar{\mathbf{S}} - \bar{\mathbf{S}}^{(1)}) \right] \cdot \frac{\partial \mathbb{Q}^{(1)}}{\partial \mathbb{M}^{(1)}} \left[(\mathbb{Q}^{(1)})^{-1}(\bar{\mathbf{S}} - \bar{\mathbf{S}}^{(1)}) \right], \end{aligned} \quad (47)$$

where the Cartesian components of $\partial \mathbb{Q}^{(1)} / \partial \mathbb{M}^{(1)}$ are given by

$$\begin{aligned} \frac{\partial Q_{ijkl}^{(1)}}{\partial M_{rsuv}^{(1)}} &= \frac{\partial L_{ijkl}^{(1)}}{\partial M_{rsuv}^{(1)}} - \frac{\partial L_{ijmn}^{(1)}}{\partial M_{rsuv}^{(1)}} P_{mnop}^{(1)} L_{opkl}^{(1)} \\ &\quad - L_{ijmn}^{(1)} \frac{\partial P_{mnop}^{(1)}}{\partial L_{efgh}^{(1)}} \frac{\partial L_{efgh}^{(1)}}{\partial M_{rsuv}^{(1)}} L_{opkl}^{(1)} - L_{ijmn}^{(1)} P_{mnop}^{(1)} \frac{\partial L_{opkl}^{(1)}}{\partial M_{rsuv}^{(1)}}. \end{aligned} \quad (48)$$

In the above expression, the Cartesian components of $\partial \mathbb{L}^{(1)} / \partial \mathbb{M}^{(1)}$ are $-\mathbb{L}^{(1)} \boxtimes_s \mathbb{L}^{(1)}$ are given by

$$\frac{\partial L_{ijkl}^{(1)}}{\partial M_{rsuv}^{(1)}} = -\frac{1}{2} \left(L_{ijrs}^{(1)} L_{kluv}^{(1)} + L_{ijuv}^{(1)} L_{klrs}^{(1)} \right), \quad (49)$$

which also serves to define the symmetrical box product \boxtimes_s . The covariance of the matrix stress fluctuations can be alternatively obtained using $\mathbb{C}^{(1)}(\mathbf{S}) = \mathbb{L}^{(1)}\mathbb{C}^{(1)}(\mathbf{F})\mathbb{L}^{(1)}$ (see Eq. (28)).

2.3.3. Two-phase isotropic composites

The expressions derived for composites with anisotropic phases and microstructure can be greatly simplified for isotropic elastic composites, where isotropic spherical inclusions are distributed isotropically ($\mathbf{Z} = \mathbf{I}$) in an isotropic matrix phase. In this case, we can present analytical expressions for the overall response and field statistics explicitly in terms of the material parameters.

We consider two-phase isotropic composites with the local constitutive relation given by

$$\mathbf{S} = \mathbb{L}^{(r)} \mathbf{F}, \quad (50)$$

where the stiffness tensor $\mathbb{L}^{(r)}$ is expressed as (Guo, 1983a,b; Jog, 2006):

$$\mathbb{L}^{(r)} = 3k^{(r)} \mathbb{J} + 2\mu^{(r)} \mathbb{K} + 2\gamma^{(r)} \mathbb{I}^a, \quad (51)$$

where \mathbb{I}^s and \mathbb{I}^a , such that $\mathbb{I}^s + \mathbb{I}^a = \mathbb{I}$, are the fourth-order identity tensors projecting second-order tensors in the subspaces of symmetric and anti-symmetric second-order tensors, respectively, while \mathbb{J} and \mathbb{K} , such that $\mathbb{J} + \mathbb{K} = \mathbb{I}^s$, are fourth-order tensors projecting into the hydrostatic and deviatoric subspaces of symmetric second-order tensors. In terms of the Kronecker delta, δ_{ij} , the Cartesian components of \mathbb{I}^s and \mathbb{I}^a are given by

$$I_{ijkl}^s = \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}), \quad I_{ijkl}^a = \frac{1}{2}(\delta_{ik}\delta_{jl} - \delta_{il}\delta_{jk}), \quad (52)$$

while the Cartesian components of \mathbb{J} and \mathbb{K} are given by

$$J_{ijkl} = \frac{1}{3}\delta_{ij}\delta_{kl}, \quad K_{ijkl} = \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) - \frac{1}{3}\delta_{ij}\delta_{kl}. \quad (53)$$

For the above composite with matrix modulus $\mathbb{L}^{(1)}$, as given by (51), and spherical inclusions and distribution, as given by $\mathbf{Z} = \mathbf{I}$, the microstructural tensor $\mathbb{P}^{(1)}$ is obtained by means of (32), and can be shown to reduce to

$$\mathbb{P}^{(1)} = \frac{1}{3k_p} \mathbb{J} + \frac{1}{2\mu_p} \mathbb{K} + \frac{1}{2\gamma_p} \mathbb{I}^a, \quad (54)$$

where

$$3k_p = 3k^{(1)} + 2\mu^{(1)}, \quad 2\mu_p = \frac{5(3k^{(1)} + 4\mu^{(1)})(\mu^{(1)} + \gamma^{(1)})}{3k^{(1)} + 2\mu^{(1)} + \frac{2}{3}\gamma^{(1)}} \quad \text{and} \quad (55)$$

$$2\gamma_p = 3(\mu^{(1)} + \gamma^{(1)}).$$

Then, making use of Eq. (40)₁, together with (54), the overall stiffness of the two-phase isotropic composite is given by

$$\begin{aligned} \tilde{\mathbb{L}} = & 3 \left[k^{(1)} + \frac{c^{(2)}k_p \Delta k}{k_p + c^{(1)}\Delta k} \right] \mathbb{J} + 2 \left[\mu^{(1)} + \frac{c^{(2)}\mu_p \Delta \mu}{\mu_p + c^{(1)}\Delta \mu} \right] \mathbb{K} \\ & + 2 \left[\gamma^{(1)} + \frac{c^{(2)}\gamma_p \Delta \gamma}{\gamma_p + c^{(1)}\Delta \gamma} \right] \mathbb{I}^a, \end{aligned} \quad (56)$$

where

$$\Delta k = k^{(2)} - k^{(1)}, \quad \Delta \mu = \mu^{(2)} - \mu^{(1)} \quad \text{and} \quad \Delta \gamma = \gamma^{(2)} - \gamma^{(1)}. \quad (57)$$

Next, using Eqs. (39), the average deformation gradient in the inclusions can be shown to reduce to

$$\bar{\mathbf{F}}^{(2)} = \left[\frac{k_p}{k_p + c^{(1)}\Delta k} \mathbb{J} + \frac{\mu_p}{\mu_p + c^{(1)}\Delta \mu} \mathbb{K} + \frac{\gamma_p}{\gamma_p + c^{(1)}\Delta \gamma} \mathbb{I}^a \right] \bar{\mathbf{F}}, \quad (58)$$

while the corresponding average deformation gradient in the matrix phase is provided by the average condition.

The second moments of the deformation gradient in the matrix phase can be obtained by means of expression (42). This calculation would require the effective modulus tensor $\tilde{\mathbb{L}}$ for general matrix elasticity tensors $\mathbb{L}^{(1)}$. However, it can be shown that the hydrostatic, deviatoric and skew-symmetric projections

can be obtained, respectively, from the expressions

$$\begin{aligned} \langle \mathbf{F} \otimes \mathbf{F} \rangle^{(1)} \cdot \mathbb{J} &= \frac{1}{3c^{(1)}} \bar{\mathbf{F}} \cdot \frac{\partial \tilde{\mathbb{L}}}{\partial k^{(1)}} \bar{\mathbf{F}}, \\ \langle \mathbf{F} \otimes \mathbf{F} \rangle^{(1)} \cdot \mathbb{K} &= \frac{1}{2c^{(1)}} \bar{\mathbf{F}} \cdot \frac{\partial \tilde{\mathbb{L}}}{\partial \mu^{(1)}} \bar{\mathbf{F}}, \\ \langle \mathbf{F} \otimes \mathbf{F} \rangle^{(1)} \cdot \mathbb{I}^a &= \frac{1}{2c^{(1)}} \bar{\mathbf{F}} \cdot \frac{\partial \tilde{\mathbb{L}}}{\partial \gamma^{(1)}} \bar{\mathbf{F}}. \end{aligned} \quad (59)$$

The corresponding projections of the fluctuation covariance tensor $\mathbb{C}^{(1)}(\mathbf{F})$ are respectively given by

$$\mathbb{C}^{(1)}(\mathbf{F}) \cdot \mathbb{J} = \frac{8}{5} c^{(2)} \left[\frac{\mu_p \Delta \mu}{(\mu_p + c^{(1)}\Delta \mu)(3k^{(1)} + 4\mu^{(1)})} \right]^2 \bar{\mathbf{F}} \cdot \mathbb{K} \bar{\mathbf{F}}, \quad (60)$$

$$\begin{aligned} \mathbb{C}^{(1)}(\mathbf{F}) \cdot \mathbb{K} &= 2c^{(2)} \left[\frac{\Delta k}{k_p + c^{(1)}\Delta k} \right]^2 \bar{\mathbf{F}} \cdot \mathbb{J} \bar{\mathbf{F}} \\ &+ \frac{3}{2} c^{(2)} \left[\frac{\Delta \gamma}{\gamma_p + c^{(1)}\Delta \gamma} \right]^2 \bar{\mathbf{F}} \cdot \mathbb{I}^a \bar{\mathbf{F}} \\ &+ c^{(2)} \left[-1 + \frac{6}{5} \frac{\alpha(k^{(1)}, \mu^{(1)}, \gamma^{(1)})}{(3k^{(1)} + 4\mu^{(1)})^2} \left(\frac{\mu_p}{\mu^{(1)} + \gamma^{(1)}} \right)^2 \right] \\ &\times \left[\frac{\Delta \mu}{\mu_p + c^{(1)}\Delta \mu} \right]^2 \bar{\mathbf{F}} \cdot \mathbb{K} \bar{\mathbf{F}}, \end{aligned} \quad (61)$$

and

$$\begin{aligned} \mathbb{C}^{(1)}(\mathbf{F}) \cdot \mathbb{I}^a &= \frac{2c^{(2)}}{5} \left[\frac{\mu_p \Delta \mu}{(\mu_p + c^{(1)}\Delta \mu)(\mu^{(1)} + \gamma^{(1)})} \right]^2 \bar{\mathbf{F}} \cdot \mathbb{K} \bar{\mathbf{F}} \\ &+ \frac{c^{(2)}}{2} \left[\frac{\Delta \gamma}{\gamma_p + c^{(1)}\Delta \gamma} \right]^2 \bar{\mathbf{F}} \cdot \mathbb{I}^a \bar{\mathbf{F}}, \end{aligned} \quad (62)$$

where $\alpha(k^{(1)}, \mu^{(1)}, \gamma^{(1)})$ is given in terms of the matrix phase moduli by

$$\begin{aligned} \alpha(k^{(1)}, \mu^{(1)}, \gamma^{(1)}) &= 3k^{(1)2} + 8\mu^{(1)2} + \frac{2}{3}\gamma^{(1)2} + 8k^{(1)}\mu^{(1)} \\ &- 3k^{(1)}\gamma^{(1)} - \frac{2}{3}\mu^{(1)}\gamma^{(1)}. \end{aligned} \quad (63)$$

As already mentioned, the corresponding fluctuations in the inclusion phase vanish identically, $\mathbb{C}^{(2)}(\mathbf{F}) = \mathbb{O}$.

The above expressions can be further simplified for isotropic incompressible composites, when $k^{(1)} \rightarrow \infty$ and $k^{(2)} \rightarrow \infty$. In this case, the allowed deformations are isochoric ($\text{tr}(\mathbf{F}) = 0$), and the deviatoric and skew-symmetric projections of the covariance tensor $\mathbb{C}^{(1)}(\mathbf{F})$ reduce to:

$$\begin{aligned} \mathbb{C}^{(1)}(\mathbf{F}) \cdot \mathbb{K} &= \frac{3}{2} c^{(2)} \left[\frac{2\Delta \mu}{5(\mu^{(1)} + \gamma^{(1)}) + 2c^{(1)}\Delta \mu} \right]^2 \bar{\mathbf{F}} \cdot \mathbb{K} \bar{\mathbf{F}} \\ &+ \frac{3}{2} c^{(2)} \left[\frac{2\Delta \gamma}{3(\mu^{(1)} + \gamma^{(1)}) + 2c^{(1)}\Delta \gamma} \right]^2 \bar{\mathbf{F}} \cdot \mathbb{I}^a \bar{\mathbf{F}}, \end{aligned} \quad (64)$$

and

$$\begin{aligned} \mathbb{C}^{(1)}(\mathbf{F}) \cdot \mathbb{I}^a &= \frac{2c^{(2)}}{5} \left[\frac{5\Delta \mu}{5(\mu^{(1)} + \gamma^{(1)}) + 2c^{(1)}\Delta \mu} \right]^2 \bar{\mathbf{F}} \cdot \mathbb{K} \bar{\mathbf{F}} \\ &+ \frac{c^{(2)}}{2} \left[\frac{2\Delta \gamma}{3(\mu^{(1)} + \gamma^{(1)}) + 2c^{(1)}\Delta \gamma} \right]^2 \bar{\mathbf{F}} \cdot \mathbb{I}^a \bar{\mathbf{F}}. \end{aligned} \quad (65)$$

2.4. Composites with granular microstructure: Self-consistent estimate

2.4.1. N -phase composites with granular microstructure

This section is concerned with N -phase composites with granular microstructures, where randomly distributed grains of each phase are surrounded by grains of the other phases in such a way that there is no matrix phase—as in polycrystals. The two-point correlation functions for all the phases are assumed to have ‘ellipsoidal’ symmetry and are all characterized by the same shape tensor \mathbf{Z} , which also correlates with the average shape and orientation of the grains.

The phase and macroscopic stored-energy functions are described by expressions (5) and (7), respectively, with the overall properties $\tilde{\mathbf{L}}$, $\tilde{\mathbf{T}}$ and \tilde{f} given in terms of the corresponding phase properties $\mathbb{L}^{(r)}$, $\mathbf{T}^{(r)}$, $f^{(r)}$ and concentration tensors by expressions (14). For the above-described composite with granular microstructure, the concentration tensors are obtained by generalizing the self-consistent (SC) estimate of Willis (1981); Bornert et al. (2001) for linear thermo-elastic composites to incremental linear-elastic composites (6), as was done by Ponte Castañeda and Tiberio (2000). In this case, the concentration tensors are given by

$$\mathbb{A}^{(r)} = [\mathbb{I} + \tilde{\mathbb{P}}\Delta\mathbb{L}^{(r)}]^{-1} \quad \text{and} \quad \mathbf{a}^{(r)} = -\tilde{\mathbb{P}}\mathbb{A}^{(r)T}\Delta\mathbf{T}^{(r)}, \quad (66)$$

where $\Delta\mathbb{L}^{(r)} = \mathbb{L}^{(r)} - \tilde{\mathbb{L}}$ and $\Delta\mathbf{T}^{(r)} = \mathbf{T}^{(r)} - \tilde{\mathbf{T}}$. The microstructural tensor $\tilde{\mathbb{P}}$ depends on the overall stiffness $\tilde{\mathbb{L}}$ and the shape tensor \mathbf{Z} , with the Cartesian components given by

$$\tilde{P}_{ijkl} = \frac{\det(\mathbf{Z})}{4\pi} \int_{|\zeta|=1} \frac{\tilde{N}_{ik}\zeta_j\zeta_l}{|\mathbf{Z}\zeta|^3} dS_\zeta, \quad (67)$$

where $\tilde{\mathbf{N}} = (\tilde{\mathbf{K}})^{-1}$ and $\tilde{K}_{ik} = \tilde{L}_{ijkl}\zeta_j\zeta_l$. Note that the microstructural tensor $\tilde{\mathbb{P}}$ exhibits only major symmetries.

By means of the above concentration tensors, together with expression (14)₁, the overall stiffness $\tilde{\mathbb{L}}$ can be obtained via the following implicit equation

$$(\tilde{\mathbb{L}} + \mathbb{L}^*)^{-1} = \sum_{r=1}^N c^{(r)} (\mathbb{L}^{(r)} + \mathbb{L}^*)^{-1}, \quad (68)$$

where $\mathbb{L}^* = \tilde{\mathbb{P}}^{-1} - \tilde{\mathbb{L}}$ is the Hill influence tensor. This equation can be alternatively recast as

$$\tilde{\mathbb{P}} = \sum_{s=1}^N c^{(s)} \mathbb{H}^{(s)}. \quad (69)$$

where

$$\mathbb{H}^{(s)} = \mathbb{A}^{(s)}\tilde{\mathbb{P}} = (\tilde{\mathbb{P}}^{-1} + \Delta\mathbb{L}^{(s)})^{-1}, \quad (70)$$

which possesses major symmetry.

Once the overall stiffness is obtained by solving the above-described implicit equation, the concentration tensors can be obtained by Eqs. (66). Then, the overall stress polarization $\tilde{\mathbf{T}}$ and zero deformation-gradient energy \tilde{f} are obtained by means of expressions (14)₂ and (14)₃, respectively.

The first moment of the deformation-gradient fields in the phases trivially follows from Eq. (9), i.e. $\bar{\mathbf{F}}^{(r)} = \mathbb{A}^{(r)}\bar{\mathbf{F}} + \mathbf{a}^{(r)}$. To obtain the second moments of the deformation gradient $(\mathbf{F} \otimes \mathbf{F})^{(r)}$, use of expression (23)₂ leads to an expression identical to (34), involving the derivatives of the overall moduli $\tilde{\mathbb{L}}$, $\tilde{\mathbf{T}}$ and \tilde{f} with respect to the phase stiffness $\mathbb{L}^{(r)}$. Next, we briefly describe the procedure to obtain these derivatives.

First, the derivative of the overall stiffness $\tilde{\mathbb{L}}$ with respect to the phase moduli $\mathbb{L}^{(r)}$ is obtained by term-by-term differentiation

of Eq. (69), leading to

$$\frac{\partial \tilde{\mathbb{P}}}{\partial \mathbb{L}^{(r)}} = \sum_{s=1}^N c^{(s)} \frac{\partial \mathbb{H}^{(s)}}{\partial \mathbb{L}^{(r)}}. \quad (71)$$

By taking the derivative of expression (70) for $\mathbb{H}^{(s)}$, it is shown in Appendix A that

$$\begin{aligned} \frac{\partial \mathbb{H}^{(s)}}{\partial \mathbb{L}^{(r)}} &= (\mathbb{H}^{(s)}\tilde{\mathbb{P}}^{-1}) \frac{\partial \tilde{\mathbb{P}}}{\partial \mathbb{L}^{(r)}} (\tilde{\mathbb{P}}^{-1}\mathbb{H}^{(s)}) - \delta_{(s)}^{(r)} \mathbb{H}^{(s)} \boxtimes_s \mathbb{H}^{(s)} \\ &\quad + [\mathbb{H}^{(s)} \boxtimes_s \mathbb{H}^{(s)}] \frac{\partial \tilde{\mathbb{L}}}{\partial \mathbb{L}^{(r)}}, \end{aligned} \quad (72)$$

where the symmetrical box product \boxtimes_s has been defined by Eq. (49). It follows from the chain rule that the derivative of $\partial \mathbb{H}^{(s)}/\partial \mathbb{L}^{(r)}$ can be expressed in terms of $\partial \tilde{\mathbb{L}}/\partial \mathbb{L}^{(r)}$ by means of

$$\frac{\partial \tilde{\mathbb{P}}}{\partial \mathbb{L}^{(r)}} = \frac{\partial \tilde{\mathbb{P}}}{\partial \tilde{\mathbb{L}}} \frac{\partial \tilde{\mathbb{L}}}{\partial \mathbb{L}^{(r)}}, \quad (73)$$

so that Cartesian components of the eight-order tensor $\partial \mathbb{H}^{(s)}/\partial \mathbb{L}^{(r)}$ can be written more explicitly as

$$\begin{aligned} \frac{\partial H_{ijkl}^{(s)}}{\partial L_{rsuv}^{(r)}} &= H_{ijab}^{(s)} \tilde{P}_{abmn}^{-1} \frac{\partial \tilde{P}_{mnop}}{\partial \tilde{L}_{efgh}} \tilde{P}_{opcd}^{-1} H_{cdkl}^{(s)} \frac{\partial \tilde{L}_{efgh}}{\partial L_{rsuv}^{(r)}} \\ &\quad - \delta_{(s)}^{(r)} \frac{1}{2} \left(H_{ijrs}^{(s)} H_{kluv}^{(s)} + H_{juvu}^{(s)} H_{klrs}^{(s)} \right) \\ &\quad + \frac{1}{2} \left(H_{ijef}^{(s)} H_{klgh}^{(s)} + H_{ijgh}^{(s)} H_{klfe}^{(s)} \right) \frac{\partial \tilde{L}_{efgh}}{\partial L_{rsuv}^{(r)}}. \end{aligned} \quad (74)$$

In this expression, \tilde{P}_{abmn}^{-1} are the Cartesian components of $\tilde{\mathbb{P}}^{-1}$, while the Cartesian components of $\partial \tilde{\mathbb{P}}/\partial \tilde{\mathbb{L}}$ are given by

$$\begin{aligned} \frac{\partial \tilde{P}_{ijkl}}{\partial \tilde{L}_{rsuv}} &= -\frac{\det(\mathbf{Z})}{8\pi} \int_{|\zeta|=1} \frac{1}{|\mathbf{Z}\zeta|^3} \left[(\tilde{N}_{ir}\zeta_s)(\tilde{N}_{ku}\zeta_v) \right. \\ &\quad \left. + (\tilde{N}_{kr}\zeta_s)(\tilde{N}_{iu}\zeta_v) \right] \zeta_j\zeta_l dS_\zeta. \end{aligned} \quad (75)$$

Now, substituting the above expression for $\partial \mathbb{H}^{(s)}/\partial \mathbb{L}^{(r)}$ into Eq. (71), the following linear system is obtained for the Cartesian components of the eight-order tensor $\partial \tilde{\mathbb{L}}/\partial \mathbb{L}^{(r)}$:

$$\begin{aligned} \sum_{s=1}^N c^{(s)} \left[\frac{1}{2} \left(H_{ijef}^{(s)} H_{klgh}^{(s)} + H_{ijgh}^{(s)} H_{klfe}^{(s)} \right) - \frac{\partial \tilde{P}_{ijkl}}{\partial \tilde{L}_{efgh}} \right. \\ \left. + H_{ijab}^{(s)} \tilde{P}_{abmn}^{-1} \frac{\partial \tilde{P}_{mnop}}{\partial \tilde{L}_{efgh}} \tilde{P}_{opcd}^{-1} H_{cdkl}^{(s)} \right] \frac{\partial \tilde{L}_{efgh}}{\partial L_{rsuv}^{(r)}} \\ = \frac{c^{(r)}}{2} \left(H_{ijrs}^{(r)} H_{kluv}^{(r)} + H_{juvu}^{(r)} H_{klrs}^{(r)} \right). \end{aligned} \quad (76)$$

Next, the derivative of the overall stress polarization $\partial \tilde{\mathbf{T}}/\partial \mathbb{L}^{(r)}$ is obtained by term-by-term differentiation of (14)₂. The Cartesian components of the sixth-order tensor $\partial \tilde{\mathbf{T}}/\partial \mathbb{L}^{(r)}$ can be written as

$$\frac{\partial \tilde{T}_{ij}}{\partial L_{rsuv}^{(r)}} = \sum_{s=1}^N c^{(s)} T_{pq}^{(s)} \frac{\partial A^{(s)}_{pqij}}{\partial L_{rsuv}^{(r)}}. \quad (77)$$

In this expression, the derivatives of the concentration tensor $\mathbb{A}^{(s)} = \mathbb{H}^{(s)}\tilde{\mathbb{P}}^{-1}$ are obtained by making use of the product-rule of differentiation:

$$\frac{\partial A^{(s)}_{ijkl}}{\partial L_{rsuv}^{(r)}} = \frac{\partial H^{(s)}_{ijpq}}{\partial L_{rsuv}^{(r)}} \tilde{P}_{pqkl}^{-1} - H_{ijab}^{(s)} \tilde{P}_{abmn}^{-1} \frac{\partial \tilde{P}_{mnop}}{\partial \tilde{L}_{efgh}} \tilde{P}_{opcd}^{-1} \frac{\partial \tilde{L}_{efgh}}{\partial L_{rsuv}^{(r)}}, \quad (78)$$

where $\partial \mathbb{H}^{(s)}/\partial \mathbb{L}^{(r)}$, $\partial \tilde{\mathbb{P}}/\partial \tilde{\mathbb{L}}$ and $\partial \tilde{\mathbb{L}}/\partial \mathbb{L}^{(r)}$ are given by (74), (75) and (76), respectively.

Lastly, the derivative of the zero deformation-gradient energy \tilde{f} , which, for the SC estimate, is given by

$$\tilde{f} = \sum_{s=1}^N c^{(s)} \mathbf{T}^{(s)} \cdot [\mathbb{H}^{(s)}(\tilde{\mathbf{T}} - \mathbf{T}^{(s)})], \quad (79)$$

can be obtained as

$$\frac{\partial \tilde{f}}{\partial L_{rsuv}^{(r)}} = \sum_{s=1}^N c^{(s)} T_{ij}^{(s)} \left[\frac{\partial H_{ijkl}^{(s)}}{\partial L_{rsuv}^{(r)}} (\tilde{T}_{kl} - T_{kl}^{(s)}) + H_{ijkl}^{(s)} \frac{\partial \tilde{T}_{kl}}{\partial L_{rsuv}^{(r)}} \right], \quad (80)$$

where $\partial \mathbb{H}^{(s)} / \partial L^{(r)}$ and $\partial \tilde{\mathbf{T}} / \partial L^{(r)}$ are given by (74) and (77), respectively. Finally, the second moments of deformation gradient in the phases are obtained by substituting Eqs. (76), (77) and (80) in Eq. (34). The intragranular fluctuation covariance tensors $\mathbb{C}^{(r)}(\mathbf{F}) = \langle \mathbf{F} \otimes \mathbf{F} \rangle^{(r)} - \bar{\mathbf{F}} \otimes \bar{\mathbf{F}}^{(r)}$ then follow straightforwardly. It should be emphasized here that the SC estimates for the tensors $\mathbb{C}^{(r)}(\mathbf{F})$ turn out to be non-zero in all the phases, indicating that the fields are heterogeneous in all the phases for composites with granular microstructures.

While the above expressions for the field statistics for general N -phase composites with granular microstructures do not simplify significantly for two-phase composites with anisotropic phases and microstructures, simpler expressions can be derived when the two phases are isotropic and incompressible and the microstructures are also isotropic, and will be provided next.

2.4.2. Two-phase isotropic incompressible composites

In this subsection, the results of the previous subsection are specialized for two-phase elastic composites with isotropic, incompressible phases and isotropic granular microstructures, such that $\mathbf{Z} = \mathbf{I}$. Thus, the local constitutive response of the phases is given by $\mathbf{S} = \mathbb{L}^{(r)} \mathbf{F}$ with

$$\mathbb{L}^{(r)} = 2\mu^{(r)} \mathbb{K} + 2\gamma^{(r)} \mathbb{I}^a, \quad r = 1, 2, \quad (81)$$

where the Cartesian components of the fourth-order tensors \mathbb{K} and \mathbb{I}^a are given by expressions (53)₂ and (52), respectively. For this class of composites, the overall stiffness tensor is also isotropic and is given by

$$\tilde{\mathbb{L}} = 2\tilde{\mu} \mathbb{K} + 2\tilde{\gamma} \mathbb{I}^a, \quad (82)$$

where $\tilde{\mu}$ and $\tilde{\gamma}$ are the appropriate overall moduli, while the expression for the microstructural tensor $\tilde{\mathbb{P}}$ is obtained from (67) by means of expression (82) for $\tilde{\mathbb{L}}$ and $\mathbf{Z} = \mathbf{I}$, with the result that

$$\tilde{\mathbb{P}} = \frac{1}{5(\tilde{\mu} + \tilde{\gamma})} \mathbb{K} + \frac{1}{3(\tilde{\mu} + \tilde{\gamma})} \mathbb{I}^a. \quad (83)$$

Then, making use of Eq. (69), the following set of coupled equations for the overall moduli $\tilde{\mu}$ and $\tilde{\gamma}$ is obtained:

$$\begin{aligned} 6\tilde{\mu}^2 + \tilde{\mu}[10(c^{(1)}\mu^{(2)} + c^{(2)}\mu^{(1)}) - 6(\mu^{(1)} + \mu^{(2)})] - 4\mu^{(1)}\mu^{(2)} \\ + 10\tilde{\gamma}[\tilde{\mu} - (c^{(1)}\mu^{(1)} + c^{(2)}\mu^{(2)})] = 0, \\ 2\tilde{\gamma}^2 + \tilde{\gamma}[6(c^{(1)}\gamma^{(2)} + c^{(2)}\gamma^{(1)}) - 2(\gamma^{(1)} + \gamma^{(2)})] - 4\gamma^{(1)}\gamma^{(2)} \\ + 6\tilde{\mu}[\tilde{\gamma} - (c^{(1)}\gamma^{(1)} + c^{(2)}\gamma^{(2)})] = 0. \end{aligned} \quad (84)$$

The phase averages of the deformation gradient can be obtained in terms of the above overall moduli, by specializing expressions (66) for the concentration tensors, to obtain the result

$$\bar{\mathbf{F}}^{(r)} = \left[\frac{5(\tilde{\mu} + \tilde{\gamma})}{3\tilde{\mu} + 5\tilde{\gamma} + 2\mu^{(r)}} \mathbb{K} + \frac{3(\tilde{\mu} + \tilde{\gamma})}{3\tilde{\mu} + \tilde{\gamma} + 2\gamma^{(r)}} \mathbb{I}^a \right] \bar{\mathbf{F}}. \quad (85)$$

The corresponding second moments of the deformation gradient in the phases may be obtained by following the procedure

outlined in the previous subsection; however, this would require obtaining results for general phase moduli $\mathbb{L}^{(r)}$ in order to take the required derivatives. Much simpler and explicit expressions can be derived for the isotropic projections, as given by the deviatoric $\langle \mathbf{F} \otimes \mathbf{F} \rangle^{(r)} \cdot \mathbb{K}$ and anti-symmetric $\langle \mathbf{F} \otimes \mathbf{F} \rangle^{(r)} \cdot \mathbb{I}^a$ projections, by making use of Eqs. (81) and (82) for the isotropic phase and overall stiffness tensor, respectively, and taking derivatives with respect to $\mu^{(r)}$ and $\gamma^{(r)}$, to obtain the results

$$\begin{aligned} \langle \mathbf{F} \otimes \mathbf{F} \rangle^{(r)} \cdot \mathbb{K} &= \frac{1}{2c^{(r)}} \bar{\mathbf{F}} \cdot \frac{\partial \tilde{\mathbb{L}}}{\partial \mu^{(r)}} \bar{\mathbf{F}} \\ &= \frac{1}{c^{(r)}} \left[\frac{\partial \tilde{\mu}}{\partial \mu^{(r)}} \bar{\mathbf{F}} \cdot \mathbb{K} \bar{\mathbf{F}} + \frac{\partial \tilde{\gamma}}{\partial \mu^{(r)}} \bar{\mathbf{F}} \cdot \mathbb{I}^a \bar{\mathbf{F}} \right], \\ \langle \mathbf{F} \otimes \mathbf{F} \rangle^{(r)} \cdot \mathbb{I}^a &= \frac{1}{2c^{(r)}} \bar{\mathbf{F}} \cdot \frac{\partial \tilde{\mathbb{L}}}{\partial \gamma^{(r)}} \bar{\mathbf{F}} \\ &= \frac{1}{c^{(r)}} \left[\frac{\partial \tilde{\mu}}{\partial \gamma^{(r)}} \bar{\mathbf{F}} \cdot \mathbb{K} \bar{\mathbf{F}} + \frac{\partial \tilde{\gamma}}{\partial \gamma^{(r)}} \bar{\mathbf{F}} \cdot \mathbb{I}^a \bar{\mathbf{F}} \right]. \end{aligned} \quad (86)$$

In these expressions, the partial derivatives of the overall moduli $\tilde{\mu}$ and $\tilde{\gamma}$ can be obtained by term-by-term differentiation of Eqs. (84) with respect to the phase moduli $\mu^{(r)}$ and $\gamma^{(r)}$. The results for the derivatives with respect to the moduli of phase 1 can be obtained from the solution of the linear system

$$\begin{aligned} A \begin{bmatrix} \frac{\partial \tilde{\mu}}{\partial \mu^{(1)}} & \frac{\partial \tilde{\mu}}{\partial \gamma^{(1)}} \\ \frac{\partial \tilde{\gamma}}{\partial \mu^{(1)}} & \frac{\partial \tilde{\gamma}}{\partial \gamma^{(1)}} \end{bmatrix} \\ = \begin{bmatrix} 10c^{(1)}\tilde{\gamma} + 4\mu^{(2)} + 6\tilde{\mu}(6 - 10c^{(2)}) & 0 \\ 0 & 6c^{(1)}\tilde{\mu} + 4\gamma^{(2)} + \tilde{\gamma}(2 - 6c^{(2)}) \end{bmatrix}, \end{aligned} \quad (87)$$

with A given by Eq. (88) in Box I.

The corresponding derivatives of the overall moduli with respect to the moduli of phase 2 are obtained by interchanging phases 1 and 2 in the above expressions.

3. Composites with linearly viscous constitutive response

This section is concerned with linearly viscous composites with constitutive behavior of the type

$$\boldsymbol{\sigma} = \mathbb{L}^s(\mathbf{x}) \mathbf{D} + \boldsymbol{\tau}(\mathbf{x}), \quad (89)$$

where $\boldsymbol{\sigma}$ is the Cauchy stress and \mathbf{D} is the Eulerian strain rate – both symmetric second-order tensors – while $\boldsymbol{\tau}$ is the stress polarization, also a symmetric second-order tensor, and \mathbb{L}^s is the viscosity (fourth-order) tensor exhibiting both major ($L_{ijkl}^s = L_{klij}^s$) and minor ($L_{ijkl}^s = L_{jikl}^s = L_{ijlk}^s$) symmetries. In addition, consistent with what was done in the previous section, $\boldsymbol{\tau}$ and \mathbb{L}^s will be assumed to be the uniform in each phase of the composites.

When such composites are subjected to affine velocity fields of the form $\mathbf{v} = \bar{\mathbf{L}}\mathbf{x}$ on their boundaries, non-uniform velocity-gradient fields $\mathbf{L}(\mathbf{x}) = \nabla v(\mathbf{x})$ develop within their interiors. The local strain rate and local spin are then given by $\mathbf{D} = (\mathbf{L} + \mathbf{L}^T)/2$ and $\mathbf{W} = (\mathbf{L} - \mathbf{L}^T)/2$, respectively. Note that $\bar{\mathbf{D}}, \bar{\mathbf{W}}$ and $\bar{\mathbf{L}} = \bar{\mathbf{D}} + \bar{\mathbf{W}}$ are respectively the macroscopic strain rate, macroscopic spin and macroscopic velocity gradient. In this section, we provide expressions for the overall response and field statistics of the stress, strain-rate and spin fields for linear composites and polycrystals with local constitutive response given by (89).

By comparing relation (89) with (3), it is evident that the constitutive relations for the linearly viscous composites are special cases of the constitutive relations for the linearized hyperelastic composites, after replacement of the deformation gradient

$$A = \begin{bmatrix} 12\tilde{\mu} + 10(c^{(1)}\mu^{(2)} + c^{(2)}\mu^{(1)}) - 6(\mu^{(1)} + \mu^{(2)}) + 10\tilde{\gamma} & 10[\tilde{\mu} - (c^{(1)}\mu^{(1)} + c^{(2)}\mu^{(2)})] \\ 6[\tilde{\gamma} - (c^{(1)}\gamma^{(1)} + c^{(2)}\gamma^{(2)})] & 4\tilde{\gamma} + 6(c^{(1)}\gamma^{(2)} + c^{(2)}\gamma^{(1)}) - 2(\gamma^{(1)} + \gamma^{(2)}) + 6\tilde{\mu} \end{bmatrix}. \quad (88)$$

Box I.

field \mathbf{F} by the velocity-gradient field \mathbf{L} , the first Piola–Kirchhoff (PK) stress \mathbf{S} by the Cauchy stress $\boldsymbol{\sigma}$, the incremental elasticity tensor \mathbb{L} by the symmetric viscosity tensor \mathbb{L}^s , and the non-symmetric stress polarization \mathbf{T} by the symmetric polarization $\boldsymbol{\tau}$. This mathematical analogy implies that the overall response and field statistics for the linearly viscous composites can be obtained by specializing the formulation in the previous section for linearized hyperelastic composites to symmetric modulus tensors and making the appropriate conversions. For instance, this procedure will allow us to generate expressions for the first and second moments of the velocity-gradient fields in the phases ($\bar{\mathbf{L}}^{(r)}$ and $\langle \mathbf{L} \otimes \mathbf{L} \rangle^{(r)}$) from the corresponding expressions for the statistics of the deformation-gradient fields ($\bar{\mathbf{F}}^{(r)}$ and $\langle \mathbf{F} \otimes \mathbf{F} \rangle^{(r)}$). The velocity-gradient statistics will then be projected onto appropriate subspaces to extract the corresponding moments of the strain-rate and spin fields in the phases.

3.1. Local constitutive relation and overall response

We consider composite materials with N randomly distributed phases. The microstructure of the composites is described by the indicator functions $\chi^{(r)}(\mathbf{x})$, as defined in Section 2.1 (although referred here to the position vector \mathbf{x} in the current configuration), such that the phase volume fractions are $c^{(r)}$ and the two-point probabilities are given by $p^{(rs)}(\mathbf{x} - \mathbf{x}')$.

As already mentioned, the phase potentials are obtained by making the substitutions $\mathbf{F} \rightarrow \mathbf{L}$, $\mathbb{L}^{(r)} \rightarrow \mathbb{L}^{s(r)}$, $\mathbf{T}^{(r)} \rightarrow \boldsymbol{\tau}^{(r)}$ in expression (5), where the phase viscosity tensors $\mathbb{L}^{s(r)}$ and stress polarizations $\boldsymbol{\tau}^{(r)}$ are fully symmetric. Note that the viscosity tensors $\mathbb{L}^{s(r)}$ are not positive definite in the ‘full’ space, in the sense that $\mathbf{W} \cdot \mathbb{L}^{s(r)} \mathbf{W} = 0$ for all skew-symmetric tensors \mathbf{W} , but are positive definite in the appropriate symmetric subspace, such that $\mathbf{D} \cdot \mathbb{L}^{s(r)} \mathbf{D} > 0$ for all non-zero symmetric second-order tensors \mathbf{D} . As a consequence, the results of the previous section must be appropriately re-interpreted in this section as holding in the subspace of positive definite viscosity tensors with minor and major symmetries (of the full space of positive definite viscosity tensors with major, but no minor symmetries).

The phase dissipation potentials then become

$$W^{(r)}(\mathbf{L}) = W^{(r)}(\mathbf{D}) = \frac{1}{2} \mathbf{D} \cdot \mathbb{L}^{s(r)} \mathbf{D} + \boldsymbol{\tau}^{(r)} \cdot \mathbf{D} + \frac{1}{2} f^{(r)}, \quad (90)$$

where the minor symmetries of $\mathbb{L}^{s(r)}$ and the symmetry of $\boldsymbol{\tau}^{(r)}$ are exploited to eliminate dependence of the phase potential on local spin field. The corresponding local constitutive relation (6) then reduces to

$$\boldsymbol{\sigma} = \mathbb{L}^{s(r)} \mathbf{D} + \boldsymbol{\tau}^{(r)}. \quad (91)$$

Now, by specializing Eq. (9) from the results of the previous section for linearized elastic composites, we obtain the following equation for the average velocity gradient in the linearly viscous composites

$$\bar{\mathbf{L}}^{(r)} = \langle \mathbf{L} \rangle^{(r)} = \mathbb{A}^{(r)} \bar{\mathbf{L}} + \mathbf{a}^{(r)}, \quad (92)$$

in terms of the velocity-gradient concentration tensors $\mathbb{A}^{(r)}$ and $\mathbf{a}^{(r)}$, which possess no symmetries.

Then, specializing expression (12) from the results of the previous section, the overall potential of the linearly viscous composites is given by

$$\tilde{W}(\bar{\mathbf{L}}) = \min_{\mathbf{L} \in \mathcal{K}(\bar{\mathbf{L}})} \sum_{r=1}^N c^{(r)} \langle W^{(r)}(\mathbf{L}) \rangle^{(r)} = \frac{1}{2} \bar{\mathbf{L}} \cdot \tilde{\mathbb{L}} \bar{\mathbf{L}} + \tilde{\mathbf{T}} \cdot \bar{\mathbf{L}} + \frac{1}{2} \tilde{f}, \quad (93)$$

where $\mathcal{K}(\bar{\mathbf{L}}) = \{\mathbf{L} | \mathbf{L} = \nabla v(\mathbf{x}) \text{ in } \Omega \text{ and } \mathbf{v} = \bar{\mathbf{L}} \mathbf{x} \text{ on } \partial\Omega\}$, and where the effective properties $\tilde{\mathbb{L}}$, $\tilde{\mathbf{T}}$ and \tilde{f} are given by relations completely analogous to expressions (14) in terms of the concentration tensors defined by expressions (92) and the properties of the phases $\mathbb{L}^{s(r)}$, $\boldsymbol{\tau}^{(r)}$ and $f^{(r)}$. Note that, at this stage, the overall viscosity tensor $\tilde{\mathbb{L}}$ exhibits major symmetry but not necessarily minor symmetries, and the overall stress polarization $\tilde{\mathbf{T}}$ does not necessarily exhibit any symmetries. However, as detailed below, the symmetries of the phase property tensors and the major symmetry of the overall viscosity implies that the overall property tensors are fully symmetric. This will then allow us to recover the classical relation for the overall potential in terms of the overall strain rate.

For this purpose, we will need the following identities. First, it is recalled that a general second-order tensor \mathbf{A} can be decomposed as

$$\mathbf{A} = (\mathbb{I}^s + \mathbb{I}^a) \mathbf{A} = \mathbf{A}^s + \mathbf{A}^a, \quad (94)$$

where $\mathbf{A}^s = \mathbb{I}^s \mathbf{A}$ is symmetric and $\mathbf{A}^a = \mathbb{I}^a \mathbf{A}$ is anti-symmetric. Similarly, a general fourth-order tensor \mathbb{A} can be decomposed into four components by means of the identity

$$\mathbb{A} = (\mathbb{I}^s + \mathbb{I}^a) \mathbb{A} (\mathbb{I}^s + \mathbb{I}^a) = \mathbb{A}^s + \mathbb{E} + \mathbb{F} + \mathbb{A}^a, \quad (95)$$

where $\mathbb{A}^s = \mathbb{I}^s \mathbb{A} \mathbb{I}^s$ exhibits minor symmetries ($A_{ijkl}^s = A_{jikl}^s = A_{ijlk}^s$), $\mathbb{E} = \mathbb{I}^a \mathbb{A} \mathbb{I}^s$ is anti-symmetric in its left two indices ($E_{ijkl} = -E_{jikl}$) and symmetric in its right two indices ($E_{ijkl} = E_{ijlk}$), $\mathbb{F} = \mathbb{I}^s \mathbb{A} \mathbb{I}^a$ is symmetric in its left two indices ($F_{ijkl} = F_{jikl}$) and anti-symmetric in its right two indices ($F_{ijkl} = -F_{ijlk}$), and finally $\mathbb{A}^a = \mathbb{I}^a \mathbb{A} \mathbb{I}^a$ is anti-symmetric in minor indices ($A_{ijkl}^a = -A_{jikl}^a = -A_{ijlk}^a$). Note that, if \mathbb{A} also possesses major symmetry, \mathbb{A}^s and \mathbb{A}^a share that symmetry, and it also follows, in this case (but not more generally), that $\mathbb{F} = \mathbb{E}^T$.

In particular, the concentration tensor $\mathbb{A}^{(r)}$ can be written in the form (95), so that

$$\mathbb{A}^{(r)} = \mathbb{A}^{s(r)} + \mathbb{E}^{(r)} + \mathbb{F}^{(r)} + \mathbb{A}^{a(r)}. \quad (96)$$

Making use of these results and of the symmetries of the material properties for the linearly viscous composites, it is shown in Appendix B that $\mathbb{F}^{(r)} = \mathbb{O}$, so that

$$\mathbb{A}^{(r)} = \mathbb{A}^{s(r)} + \mathbb{E}^{(r)} + \mathbb{A}^{a(r)}, \quad (97)$$

while the expressions for overall properties of the linearly viscous composites can be shown to reduce to

$$\tilde{\mathbb{L}} = \sum_{r=1}^N c^{(r)} \mathbb{L}^{s(r)} \mathbb{A}^{s(r)} \doteq \tilde{\mathbb{L}}^s, \quad \tilde{\mathbf{T}} = \sum_{r=1}^N c^{(r)} \mathbb{A}^{s(r)T} \boldsymbol{\tau}^{(r)} \doteq \tilde{\boldsymbol{\tau}}, \quad (98)$$

$$\tilde{f} = \sum_{r=1}^N c^{(r)} (f^{(r)} + \boldsymbol{\tau}^{(r)} \cdot \mathbf{a}^{s(r)}).$$

Note that the expressions for the overall viscosity and stress polarization in terms of the phase property tensors and concentration tensors are identical to the classical results in the literature (Laws, 1973; Willis, 1977) and exhibit all the usual symmetries.

Exploiting the symmetries of the overall viscosity and stress polarization, the dependence of $\tilde{W}(\bar{\mathbf{L}})$ on $\bar{\mathbf{W}}$ can be eliminated, and therefore the overall potential (93) simplifies to

$$\tilde{W}(\bar{\mathbf{L}}) = \frac{1}{2} \bar{\mathbf{D}} \cdot \tilde{\mathbb{L}}^s \bar{\mathbf{D}} + \tilde{\boldsymbol{\tau}} \cdot \bar{\mathbf{D}} + \frac{1}{2} \tilde{f} \doteq \tilde{W}(\bar{\mathbf{D}}), \quad (99)$$

as expected. The corresponding overall constitutive response relating the overall symmetric stress $\bar{\boldsymbol{\sigma}}$ with the overall symmetric strain rate $\bar{\mathbf{D}}$ is provided by

$$\bar{\boldsymbol{\sigma}} = \tilde{\mathbb{L}}^s \bar{\mathbf{D}} + \tilde{\boldsymbol{\tau}}. \quad (100)$$

Next, making use of expression (97) for the concentration tensors $\mathbb{A}^{(r)}$, and of the corresponding result for the concentration tensors $\mathbf{a}^{(r)}$, as given by

$$\mathbf{a}^{(r)} = \mathbf{a}^{s(r)} + \mathbf{a}^{a(r)}, \quad (101)$$

where $\mathbf{a}^{s(r)} = \mathbb{I}^s \mathbf{a}^{(r)}$ is symmetric and $\mathbf{a}^{a(r)} = \mathbb{I}^a \mathbf{a}^{(r)}$ is anti-symmetric, it is possible to obtain expressions for the average strain rate $\bar{\mathbf{D}}^{(r)} = \mathbb{I}^s \bar{\mathbf{L}}^{(r)}$ and spin $\bar{\mathbf{W}}^{(r)} = \mathbb{I}^a \bar{\mathbf{L}}^{(r)}$, such that $\bar{\mathbf{L}}^{(r)} = \bar{\mathbf{D}}^{(r)} + \bar{\mathbf{W}}^{(r)}$, directly from Eq. (92). The details are provided in Appendix C, but the results for the average strain rate and for the average spin are respectively given by

$$\bar{\mathbf{D}}^{(r)} = \mathbb{A}^{s(r)} \bar{\mathbf{D}} + \mathbf{a}^{s(r)}, \quad (102)$$

and

$$\bar{\mathbf{W}}^{(r)} = \mathbb{A}^{a(r)} \bar{\mathbf{W}} + \mathbb{E}^{(r)} \bar{\mathbf{D}} + \mathbf{a}^{a(r)}, \quad (103)$$

which are in agreement with the standard expressions (Laws, 1973; Willis, 1977).

Similarly, by making the substitutions $\mathbf{S} \rightarrow \boldsymbol{\sigma}$ and $\mathbb{M}^{(r)} \rightarrow \mathbb{M}^{s(r)}$, $\mathbf{G}^{(r)} \rightarrow \boldsymbol{\gamma}^{(r)}$, where the compliance tensor $\mathbb{M}^{s(r)}$, strain-rate polarization $\boldsymbol{\gamma}^{(r)}$ and zero-stress energy $h^{(r)}$ are given by

$$\begin{aligned} \mathbb{M}^{s(r)} &= (\mathbb{L}^{s(r)})^{-1}, \quad \boldsymbol{\gamma}^{(r)} = -(\mathbb{L}^{s(r)})^{-1} \boldsymbol{\tau}^{(r)}, \\ h^{(r)} &= -f^{(r)} + \boldsymbol{\tau}^{(r)} \cdot (\mathbb{L}^{s(r)})^{-1} \boldsymbol{\tau}^{(r)}, \end{aligned} \quad (104)$$

in the expressions (15) for the stress potentials, we find that

$$U^{(r)}(\boldsymbol{\sigma}) = \frac{1}{2} \boldsymbol{\sigma} \cdot \mathbb{M}^{s(r)} \boldsymbol{\sigma} + \boldsymbol{\gamma}^{(r)} \cdot \boldsymbol{\sigma} + \frac{1}{2} h^{(r)}. \quad (105)$$

On the other hand, we find that the corresponding overall stress potential is given by

$$\tilde{U}(\bar{\boldsymbol{\sigma}}) = \inf_{\boldsymbol{\sigma} \in \mathcal{T}(\bar{\boldsymbol{\sigma}})} \sum_{r=1}^N c^{(r)} \langle U^{(r)}(\boldsymbol{\sigma}) \rangle^{(r)} = \frac{1}{2} \bar{\boldsymbol{\sigma}} \cdot \tilde{\mathbb{M}}^s \bar{\boldsymbol{\sigma}} + \tilde{\boldsymbol{\gamma}} \cdot \bar{\boldsymbol{\sigma}} + \frac{1}{2} \tilde{h}, \quad (106)$$

where $\mathcal{T}(\bar{\boldsymbol{\sigma}}) = \{\boldsymbol{\sigma} | \nabla \cdot \boldsymbol{\sigma} = \mathbf{0} \text{ in } \Omega \text{ and } \boldsymbol{\sigma} \mathbf{n} = \bar{\boldsymbol{\sigma}} \mathbf{n} \text{ on } \partial\Omega\}$ and

$$\tilde{\mathbb{M}}^s = (\tilde{\mathbb{L}}^s)^{-1}, \quad \tilde{\boldsymbol{\gamma}} = -(\tilde{\mathbb{L}}^s)^{-1} \tilde{\boldsymbol{\tau}}, \quad \tilde{h} = -\tilde{f} + \tilde{\boldsymbol{\tau}} \cdot (\tilde{\mathbb{L}}^s)^{-1} \tilde{\boldsymbol{\tau}}. \quad (107)$$

The symmetries of the overall viscous compliance and strain-rate polarization follow from the corresponding symmetries of the overall viscosity tensor and stress polarization.

The stress concentration tensors can also be obtained by specializing Eq. (18) for the linearized elastic composites and then projecting on the symmetric subspace, making use of the identities (94) and (95). It follows that the phase averages of the stresses are given by

$$\bar{\boldsymbol{\sigma}}^{(r)} = \mathbb{B}^{s(r)} \bar{\boldsymbol{\sigma}} + \mathbf{b}^{s(r)}, \quad (108)$$

while expressions (20) lead to the expected relations

$$\begin{aligned} \tilde{\mathbb{M}}^s &= \sum_{r=1}^N c^{(r)} \mathbb{M}^{s(r)} \mathbb{B}^{s(r)}, \quad \tilde{\boldsymbol{\gamma}} = \sum_{r=1}^N c^{(r)} (\mathbb{B}^{s(r)})^T \boldsymbol{\gamma}^{(r)} \quad \text{and} \\ \tilde{h} &= \sum_{r=1}^N c^{(r)} (h^{(r)} + \boldsymbol{\gamma}^{(r)} \cdot \mathbf{b}^{s(r)}). \end{aligned} \quad (109)$$

3.2. Identities for the field statistics

In this section, we obtain expressions for the moments of the strain rate and spin in the phases of linearly viscous composites. To that end, we first determine the velocity-gradient moments by evaluating the expressions (23) in Section 2.2 for the linearized elastic composites with the substitution $\mathbf{F} \rightarrow \mathbf{L}$. Thus, we have

$$\begin{aligned} \bar{\mathbf{L}}^{(r)} &= \frac{1}{c^{(r)}} \frac{\partial \tilde{W}}{\partial \mathbf{T}^{(r)}}(\bar{\mathbf{L}}) \Big|_{\substack{\mathbb{L}^{(r)} = \mathbb{L}^{s(r)} \\ \mathbf{T}^{(r)} = \boldsymbol{\tau}^{(r)}}} \quad \text{and} \\ \langle \mathbf{L} \otimes \mathbf{L} \rangle^{(r)} &= \frac{2}{c^{(r)}} \frac{\partial \tilde{W}}{\partial \mathbb{L}^{(r)}}(\bar{\mathbf{L}}) \Big|_{\substack{\mathbb{L}^{(r)} = \mathbb{L}^{s(r)} \\ \mathbf{T}^{(r)} = \boldsymbol{\tau}^{(r)}}}, \end{aligned} \quad (110)$$

where the notation $(\cdot) \Big|_{\substack{\mathbb{L}^{(r)} = \mathbb{L}^{s(r)} \\ \mathbf{T}^{(r)} = \boldsymbol{\tau}^{(r)}}}$ means that we first carry out the differentiation with respect to the variables $\mathbb{L}^{(r)}$ and $\mathbf{T}^{(r)}$, and then evaluate the resulting expressions by letting $\mathbb{L}^{(r)} = \mathbb{L}^{s(r)}$ and $\mathbf{T}^{(r)} = \boldsymbol{\tau}^{(r)}$. Thus, the moments of the velocity-gradient for linearly viscous composites can be obtained by means of the overall potentials. Later, more explicit expressions for these moments will be derived making use of the appropriate phase property tensors and concentration tensors for the given microstructures of interest.

First, it is noted that, by means of expression (99) for the overall potential \tilde{W} , together with expressions (98) for the overall properties, it is easy to show that the average velocity gradient given by Eq. (110)₁ is consistent with expression (92), from which the average strain rate and spin have been determined, as given by in Eqs. (102) and (103), respectively.

To obtain the corresponding expressions for the second moments of the strain-rate and spin fields, we use the identity (95) for the symmetric fourth-order tensor $\langle \mathbf{L} \otimes \mathbf{L} \rangle^{(r)}$ and obtain

$$\begin{aligned} \langle \mathbf{D} \otimes \mathbf{D} \rangle^{(r)} &= \mathbb{I}^s \langle \mathbf{L} \otimes \mathbf{L} \rangle^{(r)} \mathbb{I}^s, \quad \langle \mathbf{W} \otimes \mathbf{W} \rangle^{(r)} = \mathbb{I}^a \langle \mathbf{L} \otimes \mathbf{L} \rangle^{(r)} \mathbb{I}^a, \\ \langle \mathbf{W} \otimes \mathbf{D} \rangle^{(r)} &= \mathbb{I}^a \langle \mathbf{L} \otimes \mathbf{L} \rangle^{(r)} \mathbb{I}^s, \quad \langle \mathbf{D} \otimes \mathbf{W} \rangle^{(r)} = \mathbb{I}^s \langle \mathbf{L} \otimes \mathbf{L} \rangle^{(r)} \mathbb{I}^a, \end{aligned} \quad (111)$$

with $\langle \mathbf{D} \otimes \mathbf{W} \rangle^{(r)} = [\langle \mathbf{W} \otimes \mathbf{D} \rangle^{(r)}]^T$.

In addition, similar to Eq. (26), expressions for the stress moments can be obtained in straightforward fashion from the strain-rate moments by means of

$$\begin{aligned} \bar{\boldsymbol{\sigma}}^{(r)} &= \mathbb{L}^{s(r)} \bar{\mathbf{D}}^{(r)} + \boldsymbol{\tau}^{(r)}, \\ \langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)} &= \mathbb{L}^{s(r)} \langle \mathbf{D} \otimes \mathbf{D} \rangle^{(r)} \mathbb{L}^{s(r)} + (\mathbb{L}^{s(r)} \bar{\mathbf{D}}^{(r)}) \otimes \boldsymbol{\tau}^{(r)} \\ &\quad + \boldsymbol{\tau}^{(r)} \otimes (\mathbb{L}^{s(r)} \bar{\mathbf{D}}^{(r)}) + \boldsymbol{\tau}^{(r)} \otimes \boldsymbol{\tau}^{(r)}. \end{aligned} \quad (112)$$

These relations imply that

$$\mathbb{C}^{(r)}(\boldsymbol{\sigma}) = \mathbb{L}^{s(r)} \mathbb{C}^{(r)}(\mathbf{D}) \mathbb{L}^{s(r)}, \quad (113)$$

where the phase fluctuation covariance tensors of the stress $\mathbb{C}^{(r)}(\boldsymbol{\sigma})$ and strain rate $\mathbb{C}^{(r)}(\mathbf{D})$ are given by

$$\begin{aligned} \mathbb{C}^{(r)}(\boldsymbol{\sigma}) &= \langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)} - \bar{\boldsymbol{\sigma}}^{(r)} \otimes \bar{\boldsymbol{\sigma}}^{(r)} \quad \text{and} \\ \mathbb{C}^{(r)}(\mathbf{D}) &= \langle \mathbf{D} \otimes \mathbf{D} \rangle^{(r)} - \bar{\mathbf{D}}^{(r)} \otimes \bar{\mathbf{D}}^{(r)}. \end{aligned} \quad (114)$$

We can also define in similar fashion the phase fluctuation covariance tensor of the spin $\mathbb{C}^{(r)}(\mathbf{W})$. In addition, we can define, similarly to (29), the overall fluctuation covariance tensors $\mathbb{C}(\sigma)$, $\mathbb{C}(\mathbf{D})$ and $\mathbb{C}(\mathbf{W})$ by means of the expressions of the form

$$\mathbb{C}(\sigma) = \sum_{r=1}^N c^{(r)} \langle \sigma \otimes \sigma \rangle^{(r)} - \bar{\sigma} \otimes \bar{\sigma}, \quad (115)$$

$$\mathbb{C}(\mathbf{D}) = \sum_{r=1}^N c^{(r)} \langle \mathbf{D} \otimes \mathbf{D} \rangle^{(r)} - \bar{\mathbf{D}} \otimes \bar{\mathbf{D}},$$

etc. For later reference, we also define the equivalent average stress $\bar{\sigma}_e^{(r)}$ and average strain rate $\bar{D}_e^{(r)}$ in the phases via

$$\bar{\sigma}_e^{(r)} = \sqrt{\frac{3}{2} \bar{\sigma}^{(r)} \cdot \mathbb{K} \bar{\sigma}^{(r)}} \quad \text{and} \quad \bar{D}_e^{(r)} = \sqrt{\frac{2}{3} \bar{\mathbf{D}}^{(r)} \cdot \mathbb{K} \bar{\mathbf{D}}^{(r)}}, \quad (116)$$

where \mathbb{K} is the identity tensor in the subspace of deviatoric fourth-order symmetric tensors, defined by (53)₂. The overall stress $\bar{\sigma}_e$ and strain rate \bar{D}_e can be defined similarly. In addition, we define the standard deviations of the phase fluctuations for the equivalent stress $SD^{(r)}(\sigma_e)$, strain rate $SD^{(r)}(D_e)$ and spin $SD^{(r)}(W)$ in the phases via

$$SD^{(r)}(\sigma_e) = \sqrt{\frac{3}{2} \mathbb{C}^{(r)}(\sigma) \cdot \mathbb{K}}, \quad SD^{(r)}(D_e) = \sqrt{\frac{2}{3} \mathbb{C}^{(r)}(\mathbf{D}) \cdot \mathbb{K}}, \quad (117)$$

etc., and similarly for the corresponding overall fluctuations $SD(\sigma_e)$, $SD(D_e)$ and $SD(W)$.

In the following subsections, we will provide more explicit expressions for the velocity-gradient, strain-rate and spin moments for particulate and granular microstructures. This is achieved by appropriately specializing the expressions in Sections 2.3.2 and 2.4.1, respectively.

3.3. Two-phase composites with particulate microstructure

We consider two-phase composites with particulate microstructures, where aligned ellipsoidal inclusions with phase properties $\mathbb{L}^{s(2)}$, $\boldsymbol{\tau}^{(2)}$, $f^{(2)}$ and volume fraction $c^{(2)}$ are randomly distributed in the matrix phase with phase properties $\mathbb{L}^{s(1)}$, $\boldsymbol{\tau}^{(1)}$, $f^{(1)}$ and volume fraction $c^{(1)} = 1 - c^{(2)}$. The shape of particles and particle-center distribution are assumed to be identical and are characterized by the shape tensor \mathbf{Z} , similar to Section 2.3.2. In this section, the expressions of Section 2.3.2 for linearized hyperelastic composites are specialized to linearly viscous composites, with the local potential given by (90), to obtain their overall response and field statistics.

For the above-described linearly viscous composites, the PCW estimates for the velocity-gradient concentration tensors for the inclusions, provided for linearized hyperelastic composites by Eqs. (39), reduce to

$$\mathbb{A}^{(2)} = [\mathbb{I} + c^{(1)} \mathbb{P}^{s(1)} \Delta \mathbb{L}^s]^{-1} \quad \text{and} \quad \mathbf{a}^{(2)} = -c^{(1)} \mathbb{P}^{s(1)} \mathbb{A}^{(2)T} \Delta \boldsymbol{\tau}, \quad (118)$$

after substituting $\mathbb{L}^{(r)} \rightarrow \mathbb{L}^{s(r)}$ and $\mathbb{T}^{(r)} \rightarrow \boldsymbol{\tau}^{(r)}$. Here $\Delta \mathbb{L}^s = \mathbb{L}^{s(2)} - \mathbb{L}^{s(1)}$, $\Delta \boldsymbol{\tau} = \boldsymbol{\tau}^{(2)} - \boldsymbol{\tau}^{(1)}$, and $\mathbb{P}^{s(1)}$ is obtained by substituting $\mathbb{L}^{s(1)}$ for $\mathbb{L}^{(1)}$ in Eq. (32). The microstructural tensor $\mathbb{P}^{s(1)}$ can be decomposed, by means of the identity (95) for a fourth-order tensor with major symmetry, into

$$\mathbb{P}^{s(1)} = \mathbb{P}^{s(1)} + \mathbb{R}^{(1)} + \mathbb{R}^{(1)T} + \mathbb{P}^{a(1)}, \quad (119)$$

where fully symmetric $\mathbb{P}^{s(1)} = \mathbb{I}^s \mathbb{P}^{s(1)} \mathbb{I}^s$, with the corresponding Cartesian components obtained by projecting expression (32)

onto the appropriate symmetric subspaces, i.e.

$$P_{ijkl}^{s(1)} = \frac{\det(\mathbf{Z})}{4\pi} \int_{|\zeta|=1} \frac{1}{4|\mathbf{Z}\zeta|^3} (N_{ik}^{(1)} \zeta_j \zeta_l + N_{jk}^{(1)} \zeta_i \zeta_l + N_{il}^{(1)} \zeta_j \zeta_k + N_{jl}^{(1)} \zeta_i \zeta_k) dS_\zeta. \quad (120)$$

Recall that $N_{ik}^{(1)}$ are the Cartesian components of the inverse of the acoustic tensor, which is $\mathbf{N}^{(1)} = (\mathbf{K}^{(1)})^{-1}$. Similarly, $\mathbb{R}^{(1)} = \mathbb{I}^a \mathbb{P}^{s(1)} \mathbb{I}^s$, with its Cartesian components given by

$$R_{ijkl}^{(1)} = \frac{\det(\mathbf{Z})}{4\pi} \int_{|\zeta|=1} \frac{1}{4|\mathbf{Z}\zeta|^3} (N_{ik}^{(1)} \zeta_j \zeta_l - N_{jk}^{(1)} \zeta_i \zeta_l + N_{il}^{(1)} \zeta_j \zeta_k - N_{jl}^{(1)} \zeta_i \zeta_k) dS_\zeta. \quad (121)$$

The other projections, $\mathbb{R}^{(1)T}$ and $\mathbb{P}^{a(1)}$, can also be similarly expressed in terms $\mathbf{N}^{(1)}$ by appropriately projecting expression (32) onto symmetric and anti-symmetric subspaces.

Making use of these results and of the identity (97), it is shown in Appendix D that the inclusion concentration tensor $\mathbb{A}^{(2)}$, as given by (118)₁, can be re-expressed in the form

$$\mathbb{A}^{(2)} = \mathbb{A}^{s(2)} + \mathbb{I}^a - c^{(1)} \mathbb{R}^{(1)} \Delta \mathbb{L}^s \mathbb{A}^{s(2)}, \quad (122)$$

where the symmetrized concentration tensor $\mathbb{A}^{s(2)} = \mathbb{I}^s \mathbb{A}^{(2)} \mathbb{I}^s$ is given by

$$\mathbb{A}^{s(2)} = [\mathbb{I}^s + c^{(1)} \mathbb{P}^{s(1)} \Delta \mathbb{L}^s]^{-1}, \quad (123)$$

where it should be emphasized that the inverse is now taken in the subspace of fourth-order tensors with minor symmetries.

Then, evaluating the transpose of expression (122) and using it in expression (118)₂ for the thermal concentration tensor $\mathbf{a}^{(2)}$, we obtain the result

$$\mathbf{a}^{(2)} = -c^{(1)} (\mathbb{P}^{s(1)} + \mathbb{R}^{(1)}) \mathbb{A}^{s(2)T} \Delta \boldsymbol{\tau}, \quad (124)$$

where the symmetries of the stress-polarization tensors have been exploited to show that $\mathbb{A}^{(2)T} \Delta \boldsymbol{\tau} = \mathbb{A}^{s(2)T} \Delta \boldsymbol{\tau}$. The stress polarization $\mathbf{a}^{(2)}$ can then be decomposed into its symmetric and anti-symmetric parts by means of (101), to get the results

$$\mathbf{a}^{s(2)} = \mathbb{I}^s \mathbf{a}^{(2)} = -c^{(1)} \mathbb{P}^{s(1)} \mathbb{A}^{s(2)T} \Delta \boldsymbol{\tau} \quad \text{and} \quad (125)$$

$$\mathbf{a}^{a(2)} = \mathbb{I}^a \mathbf{a}^{(2)} = -c^{(1)} \mathbb{R}^{(1)} \mathbb{A}^{s(2)T} \Delta \boldsymbol{\tau}.$$

The anti-symmetric part $\mathbf{a}^{a(2)}$ can be alternatively expressed as

$$\mathbf{a}^{a(2)} = -c^{(1)} \mathbb{R}^{(1)} (\Delta \mathbb{L}^s \mathbf{a}^{s(2)} + \Delta \boldsymbol{\tau}), \quad (126)$$

where we have used the identity $\mathbb{A}^{s(2)} - \mathbb{I}^s = -c^{(1)} \mathbb{A}^{s(2)} \mathbb{P}^{s(1)} \Delta \mathbb{L}^s$.

Making use of the symmetrized concentration tensors in Eqs. (98), the PCW estimates for the overall properties of the linearly viscous composites can be shown to be given by

$$\begin{aligned} \tilde{\mathbb{L}}^s &= \mathbb{L}^{s(1)} + c^{(2)} [(\mathbb{L}^{s(2)} - \mathbb{L}^{s(1)})^{-1} + c^{(1)} \mathbb{P}^{s(1)}]^{-1}, \\ \tilde{\boldsymbol{\tau}} &= c^{(1)} \boldsymbol{\tau}^{(1)} + c^{(2)} \boldsymbol{\tau}^{(2)} + (\tilde{\mathbb{L}}^s - \langle \mathbb{L}^s \rangle) (\Delta \mathbb{L}^s)^{-1} \Delta \boldsymbol{\tau}, \end{aligned} \quad (127)$$

$$\tilde{f} = \bar{f} + \Delta \boldsymbol{\tau} \cdot [(\Delta \mathbb{L}^s)^{-1} (\tilde{\mathbb{L}}^s - \langle \mathbb{L}^s \rangle) (\Delta \mathbb{L}^s)^{-1}] \Delta \boldsymbol{\tau},$$

where $\bar{f} = c^{(1)} f^{(1)} + c^{(2)} f^{(2)}$. Note that these expressions are identical to the standard expressions in the literature (Willis, 1977; Ponte Castañeda and Willis, 1995; Ponte Castañeda, 2005).

The velocity-gradient averages in the inclusions can be obtained in terms of the concentration tensors by means of $\bar{\mathbf{L}}^{(2)} = \mathbb{A}^{(2)} \bar{\mathbf{L}} + \mathbf{a}^{(2)}$. It then follows from the above-derived concentration tensors, together with Eqs. (102) and (103), that the average strain rate and spin in the inclusions are given by

$$\bar{\mathbf{D}}^{(2)} = \mathbb{A}^{s(2)} \bar{\mathbf{D}} + \mathbf{a}^{s(2)}, \quad (128)$$

$$\bar{\mathbf{W}}^{(2)} = \bar{\mathbf{W}} - c^{(1)} \mathbb{R}^{(1)} \Delta \mathbb{L}^s \mathbb{A}^{s(2)} \bar{\mathbf{D}} - c^{(1)} \mathbb{R}^{(1)} (\Delta \mathbb{L}^s \mathbf{a}^{s(2)} + \Delta \boldsymbol{\tau}),$$

which are, again, identical to the corresponding results in the literature (see Ponte Castañeda (2005) as well as appendix of Song and Ponte Castañeda (2017a, 2018b) for $\bar{\mathbf{D}}^{(2)}$, and of Song and Ponte Castañeda (2018b) for $\bar{\mathbf{W}}^{(2)}$). The average velocity gradient in the matrix phase is $\bar{\mathbf{L}}^{(1)} = (\bar{\mathbf{L}} - c^{(2)}\bar{\mathbf{L}}^{(2)})/c^{(1)}$, from which the corresponding strain-rate and spin averages can be analogously retrieved by projecting on symmetric and anti-symmetric subspaces, respectively.

On the other hand, the velocity-gradient fluctuations are obtained by specializing the corresponding expressions in Section 2.3.2. The velocity-gradient fields in the inclusions are found to be uniform, i.e., $\mathbb{C}^{(2)}(\mathbf{L}) = \mathbb{O}$, which follows from Eq. (41). This means that the strain-rate and spin fields are uniform as well, $\mathbb{C}^{(2)}(\mathbf{D}) = \mathbb{O}$ and $\mathbb{C}^{(2)}(\mathbf{W}) = \mathbb{O}$. However, the fields in the matrix phase are non-uniform and the corresponding velocity-gradient fluctuation covariances can be obtained by specializing Eq. (42) to linearly viscous composites. Using the fact, shown in Appendix E, that

$$(\mathbb{P}^{(1)})^{-1}(\bar{\mathbf{L}} - \bar{\mathbf{L}}^{(1)}) = (\mathbb{P}^{s(1)})^{-1}(\bar{\mathbf{D}} - \bar{\mathbf{D}}^{(1)}), \quad (129)$$

it can be shown that

$$\begin{aligned} \mathbb{C}^{(1)}(\mathbf{L}) = & -\frac{1}{c^{(2)}}(\bar{\mathbf{L}} - \bar{\mathbf{L}}^{(1)}) \otimes (\bar{\mathbf{L}} - \bar{\mathbf{L}}^{(1)}) \\ & - \frac{1}{c^{(2)}} \left[(\mathbb{P}^{s(1)})^{-1}(\bar{\mathbf{D}} - \bar{\mathbf{D}}^{(1)}) \right] \\ & \cdot \frac{\partial \mathbb{P}^{(1)}}{\partial \mathbb{L}^{(1)}} \bigg|_{\mathbb{L}^{(1)} = \mathbb{L}^{s(1)}} \left[(\mathbb{P}^{s(1)})^{-1}(\bar{\mathbf{D}} - \bar{\mathbf{D}}^{(1)}) \right], \end{aligned} \quad (130)$$

where $\partial \mathbb{P}^{(1)}/\partial \mathbb{L}^{(1)}|_{\mathbb{L}^{(1)} = \mathbb{L}^{s(1)}}$ is determined by substituting $\mathbb{L}^{s(1)}$ for $\mathbb{L}^{(1)}$ in Eq. (43). It is recalled that the free indices of $\mathbb{C}_{rsuv}^{(1)}(\mathbf{L})$ correspond to those associated with $\partial L_{rsuv}^{(1)}$ in the second term in the right hand side. Then, by projecting the velocity-gradient fluctuations in the matrix phase on symmetric subspaces, it can be shown that the corresponding strain-rate fluctuation covariances are given by

$$\begin{aligned} \mathbb{C}^{(1)}(\mathbf{D}) = & \mathbb{I}^s \mathbb{C}^{(1)}(\mathbf{L}) \mathbb{I}^s \\ = & -\frac{1}{c^{(2)}}(\bar{\mathbf{D}} - \bar{\mathbf{D}}^{(1)}) \otimes (\bar{\mathbf{D}} - \bar{\mathbf{D}}^{(1)}) \\ & - \frac{1}{c^{(2)}} \left[(\mathbb{P}^{s(1)})^{-1}(\bar{\mathbf{D}} - \bar{\mathbf{D}}^{(1)}) \right] \\ & \cdot \frac{\partial \mathbb{P}^{s(1)}}{\partial \mathbb{L}^{s(1)}} \left[(\mathbb{P}^{s(1)})^{-1}(\bar{\mathbf{D}} - \bar{\mathbf{D}}^{(1)}) \right], \end{aligned} \quad (131)$$

which is consistent with known results from the literature (Furer and Ponte Castañeda, 2018). Here, $\partial \mathbb{P}^{s(1)}/\partial \mathbb{L}^{s(1)}$ is obtained by substituting $\mathbb{L}^{s(1)}$ for $\mathbb{L}^{(1)}$ in Eq. (43) and subsequently symmetrizing minor indices, i.e.,

$$\frac{\partial P_{ijkl}^{s(1)}}{\partial L_{rsuv}^{s(1)}} = I_{ijmn}^s I_{klpq}^s I_{rsef}^s I_{uvgh}^s \frac{\partial P_{mnop}^{(1)}}{\partial L_{efgh}^{(1)}} \bigg|_{\mathbb{L}^{(1)} = \mathbb{L}^{s(1)}}, \quad (132)$$

where we use the chain rule $\partial(\cdot)/\partial \mathbb{L}^{(1)} = \sum_{p=1}^4 \partial(\cdot)/\partial \mathbb{L}^{p(1)} \partial \mathbb{L}^{p(1)}/\partial \mathbb{L}^{(1)}$, with $\mathbb{L}^{p(1)}$ ($p = 1, \dots, 4$) the projections on different subspaces as determined by Eq. (95). The derivatives $\partial \mathbb{L}^{p(1)}/\partial \mathbb{L}^{(1)}$ are easily obtained by appropriate expressions for the projections; for example, $\mathbb{L}^{s(1)} = \mathbb{I}^s \mathbb{L}^{(1)} \mathbb{I}^s$, leading to $\partial L_{rsuv}^{s(1)}/\partial L_{efgh}^{(1)} = I_{rsef}^s I_{uvgh}^s$. In any event, the final result is that relation (132) gives the same result as the direct computation of the derivative of $\mathbb{P}^{s(1)}$ with respect to $\mathbb{L}^{s(1)}$.

The spin fluctuation covariance in the matrix phase is obtained by projecting $\mathbb{C}^{(1)}(\mathbf{L})$ on the anti-symmetric subspaces, $\mathbb{C}^{(1)}(\mathbf{W}) = \mathbb{I}^a \mathbb{C}^{(1)}(\mathbf{L}) \mathbb{I}^a$, with the corresponding Cartesian components given

by

$$\begin{aligned} \mathbb{C}_{rsuv}^{(1)}(\mathbf{W}) = & -\frac{1}{c^{(2)}}(\bar{W}_{rs} - \bar{W}_{rs}^{(1)})(\bar{W}_{uv} - \bar{W}_{uv}^{(1)}) \\ & - \frac{1}{c^{(2)}} I_{rsef}^a I_{uvgh}^a \frac{\partial P_{ijkl}^{(1)}}{\partial L_{efgh}^{(1)}} \bigg|_{\mathbb{L}^{(1)} = \mathbb{L}^{s(1)}} \\ & \times [P_{ijmn}^{s(1)-1}(\bar{D}_{mn} - \bar{D}_{mn}^{(1)})][P_{klpq}^{s(1)-1}(\bar{D}_{pq} - \bar{D}_{pq}^{(1)})], \end{aligned} \quad (133)$$

where the $P_{ijmn}^{s(1)-1}$ are the Cartesian components of $\mathbb{P}^{s(1)-1}$. It should be emphasized here that the derivative of the microstructural tensor $\mathbb{P}^{(1)}$ with respect to $\mathbb{L}^{(1)}$, evaluated at $\mathbb{L}^{(1)} = \mathbb{L}^{s(1)}$, is computed in the same way as in the context of expression (130).

Similarly, the covariance $\mathbb{C}^{(1)}(\mathbf{W}, \mathbf{D})$ is obtained from the expression $\mathbb{C}^{(1)}(\mathbf{W}, \mathbf{D}) = \mathbb{I}^a \mathbb{C}^{(1)}(\mathbf{L}) \mathbb{I}^s$; its Cartesian components are given by

$$\begin{aligned} \mathbb{C}_{rsuv}^{(1)}(\mathbf{W}, \mathbf{D}) = & -\frac{1}{c^{(2)}}(\bar{W}_{rs} - \bar{W}_{rs}^{(1)})(\bar{D}_{uv} - \bar{D}_{uv}^{(1)}) \\ & - \frac{1}{c^{(2)}} I_{rsef}^a I_{uvgh}^s \frac{\partial P_{ijkl}^{(1)}}{\partial L_{efgh}^{(1)}} \bigg|_{\mathbb{L}^{(1)} = \mathbb{L}^{s(1)}} \\ & \times [P_{ijmn}^{s(1)-1}(\bar{D}_{mn} - \bar{D}_{mn}^{(1)})][P_{klpq}^{s(1)-1}(\bar{D}_{pq} - \bar{D}_{pq}^{(1)})]. \end{aligned} \quad (134)$$

Lastly, the covariance $\mathbb{C}^{(1)}(\mathbf{D}, \mathbf{W})$ is readily given by

$$\mathbb{C}^{(1)}(\mathbf{D}, \mathbf{W}) = [\mathbb{C}^{(1)}(\mathbf{W}, \mathbf{D})]^T. \quad (135)$$

Thus, the strain-rate and spin fluctuation covariances and their cross covariances in the matrix phase are obtained by projecting the velocity-gradient fluctuations on appropriate symmetric and anti-symmetric subspaces.

Following a similar procedure, the stress-field statistics are obtained by specializing Eqs. (44)–(49) for linearly viscous composites. The average stresses in the phases are given by

$$\bar{\boldsymbol{\sigma}}^{(2)} = \mathbb{B}^{s(2)} \bar{\boldsymbol{\sigma}} + \mathbf{b}^{s(2)} \quad \text{and} \quad \bar{\boldsymbol{\sigma}}^{(1)} = (\bar{\boldsymbol{\sigma}} - c^{(2)} \bar{\boldsymbol{\sigma}}^{(2)})/c^{(1)}, \quad (136)$$

where the symmetrized inclusion concentration tensors are obtained by first specializing expressions (44) to linearly viscous composites and then projecting them on the symmetric subspaces, so that

$$\begin{aligned} \mathbb{B}^{s(2)} = & \mathbb{I}^s \mathbb{B}^{(2)} \mathbb{I}^s = [\mathbb{I}^s + c^{(1)} \mathbb{Q}^{s(1)} \Delta \mathbb{M}^s]^{-1} \quad \text{and} \\ \mathbf{b}^{s(2)} = & \mathbb{I}^s \mathbf{b}^{(2)} = -c^{(1)} \mathbb{Q}^{s(1)} \mathbb{B}^{s(2)T} \Delta \boldsymbol{\gamma}, \end{aligned} \quad (137)$$

with $\Delta \mathbb{M}^s = \mathbb{M}^{s(2)} - \mathbb{M}^{s(1)}$, $\Delta \boldsymbol{\gamma} = \boldsymbol{\gamma}^{(2)} - \boldsymbol{\gamma}^{(1)}$ and $\mathbb{Q}^{s(1)} = \mathbb{L}^{s(1)} - \mathbb{L}^{s(1)} \mathbb{P}^{s(1)} \mathbb{L}^{s(1)}$.

The stress fields in the inclusions are uniform, resulting the corresponding stress fluctuations to be null,

$$\mathbb{C}^{(2)}(\boldsymbol{\sigma}) = \mathbb{O}, \quad (138)$$

while the stress fluctuation in the matrix phase can be derived from expression (47) after substituting $\mathbb{M}^{s(r)}$ for $\mathbb{M}^{(r)}$ and $\boldsymbol{\gamma}^{(r)}$ for $\mathbf{G}^{(r)}$, and can be shown to be given by the result

$$\begin{aligned} \mathbb{C}^{(1)}(\boldsymbol{\sigma}) = & -\frac{1}{c^{(2)}}(\bar{\boldsymbol{\sigma}} - \bar{\boldsymbol{\sigma}}^{(1)}) \otimes (\bar{\boldsymbol{\sigma}} - \bar{\boldsymbol{\sigma}}^{(1)}) \\ & - \frac{1}{c^{(2)}} \left[(\mathbb{Q}^{s(1)})^{-1}(\bar{\boldsymbol{\sigma}} - \bar{\boldsymbol{\sigma}}^{(1)}) \right] \\ & \cdot \frac{\partial \mathbb{Q}^{s(1)}}{\partial \mathbb{M}^{s(1)}} \left[(\mathbb{Q}^{s(1)})^{-1}(\bar{\boldsymbol{\sigma}} - \bar{\boldsymbol{\sigma}}^{(1)}) \right], \end{aligned} \quad (139)$$

which is identical to the one available in the literature (Song and Ponte Castañeda, 2017a; Furer and Ponte Castañeda, 2018). The above relations for the stress moments can also be derived

by directly treating linearly viscous composites in symmetric subspaces, as is usually carried out in literature. Because of the duality between the stress and strain-rate potentials, the stress statistics can equivalently be obtained by means of the strain-rate statistics using Eqs. (112).

3.3.1. Two-phase isotropic composites

The previous section treats two-phase anisotropic composites, where the anisotropy of phases and microstructure significantly affect the field fluctuations induced by phase heterogeneity. Here, we demonstrate explicitly the effects of the phase heterogeneity, and thus consider two-phase isotropic composites, where isotropic spherical inclusions are distributed isotropically ($\mathbf{Z} = \mathbf{I}$) in an isotropic matrix. The overall response and field statistics can be presented explicitly in terms of material parameters.

Then, the local constitutive relation is given by

$$\boldsymbol{\sigma} = \mathbb{L}^{s(r)} \mathbf{D}, \quad \mathbb{L}^{s(r)} = 3k^{(r)} \mathbb{J} + 2\mu^{(r)} \mathbb{K}, \quad \text{for } r = 1, 2. \quad (140)$$

The overall response and field statistics for this composite can be obtained by specializing the corresponding expressions for general $\mathbb{L}^{s(r)}$ in Section 3.3 for the isotropic composites of (140). Alternatively, a much simpler approach would be to directly specialize the expressions for isotropic composites in Section 2.3.3 for $\mathbf{F} \rightarrow \mathbf{L}$, and $\gamma^{(1)} = 0$, $\gamma^{(2)} = 0$. These two procedures would lead to identical tensorial expressions for the overall response and phase averages. However, for the fluctuation tensors, the second procedure provides a direct means to obtain explicit expressions for the isotropic projections of the covariance tensors in terms of material properties.

The microstructural tensor $\mathbb{P}^{(1)}$ can be obtained by evaluating expression (54) for $\gamma^{(1)} = 0$, $\gamma^{(2)} = 0$ and is given by

$$\mathbb{P}^{(1)} = \frac{1}{3k_p} \mathbb{J} + \frac{1}{2\mu_p} \mathbb{K} + \frac{1}{2\gamma_p} \mathbb{I}^a, \quad (141)$$

with

$$3k_p = 3k^{(1)} + 2\mu^{(1)}, \quad 2\mu_p = \frac{5\mu^{(1)}(3k^{(1)} + 4\mu^{(1)})}{3} \quad \text{and} \quad (142)$$

$$2\gamma_p = 3\mu^{(1)}.$$

Thus, for isotropic composites $\mathbb{P}^{s(1)} = 1/3k_p \mathbb{J} + 1/2\mu_p \mathbb{K}$, $\mathbb{P}^{a(1)} = 1/2\gamma_p \mathbb{I}^a$ and $\mathbb{R}^{(1)} = \mathbb{O}$.

Similarly, the overall viscosity tensor can be derived by evaluating (56) for $\gamma^{(1)} = 0$, $\gamma^{(2)} = 0$:

$$\tilde{\mathbb{L}}^s = 3 \left[k^{(1)} + \frac{c^{(2)} k_p \Delta k}{k_p + c^{(1)} \Delta k} \right] \mathbb{J} + 2 \left[\mu^{(1)} + \frac{c^{(2)} \mu_p \Delta \mu}{\mu_p + c^{(1)} \Delta \mu} \right] \mathbb{K}, \quad (143)$$

with $\Delta k = k^{(2)} - k^{(1)}$ and $\Delta \mu = \mu^{(2)} - \mu^{(1)}$.

The average velocity gradient in the inclusions are obtained by specializing Eq. (58) for the above-described composites and is given by

$$\bar{\mathbf{L}}^{(2)} = \left[\frac{k_p}{k_p + c^{(1)} \Delta k} \mathbb{J} + \frac{\mu_p}{\mu_p + c^{(1)} \Delta \mu} \mathbb{K} + \mathbb{I}^a \right] \bar{\mathbf{L}}, \quad (144)$$

from which the average strain rate and spin in the inclusions are retrieved as

$$\bar{\mathbf{D}}^{(2)} = \left[\frac{k_p}{k_p + c^{(1)} \Delta k} \mathbb{J} + \frac{\mu_p}{\mu_p + c^{(1)} \Delta \mu} \mathbb{K} \right] \bar{\mathbf{D}}, \quad \bar{\mathbf{W}}^{(2)} = \bar{\mathbf{W}}. \quad (145)$$

This implies that for isotropic composites, unlike the strain-rate fields, the spin fields exhibit no interphase fluctuations.

However, all the fields in the matrix phase are strongly heterogeneous, although they are estimated to be homogeneous in the inclusion phases. The isotropic projections of the velocity-gradient fluctuations in the matrix phase can be obtained by

setting $\gamma^{(1)} = \gamma^{(2)} = 0$ in Eqs. (60)–(62). In particular, the hydrostatic projection $\mathbb{C}^{(1)}(\mathbf{D}_m) = \mathbb{C}^{(1)}(\mathbf{L}) \cdot \mathbb{J}$ is given by

$$\mathbb{C}^{(1)}(\mathbf{D}_m) = \frac{12}{5} c^{(2)} \left[\frac{\mu_p \Delta \mu}{(\mu_p + c^{(1)} \Delta \mu)(3k^{(1)} + 4\mu^{(1)})} \right]^2 \bar{\mathbf{D}}_e^2. \quad (146)$$

Similarly, the deviatoric projection $\mathbb{C}^{(1)}(\mathbf{D}_e) = 2/3 \mathbb{C}^{(1)}(\mathbf{L}) \cdot \mathbb{K}$ is given by

$$\begin{aligned} \mathbb{C}^{(1)}(\mathbf{D}_e) &= 4c^{(2)} \left[\frac{\Delta k}{k_p + c^{(1)} \Delta k} \right]^2 \bar{\mathbf{D}}_m^2 \\ &+ c^{(2)} \left[\frac{\Delta \mu}{\mu_p + c^{(1)} \Delta \mu} \right]^2 \left[-1 + \frac{5}{6} \frac{3k^{(1)2} + 8\mu^{(1)2} + 8k^{(1)}\mu^{(1)}}{(k^{(1)} + 2\mu^{(1)})^2} \right] \bar{\mathbf{D}}_e^2, \end{aligned} \quad (147)$$

and the anti-symmetric projection $\mathbb{C}^{(1)}(\mathbf{W}) = 2/3 \mathbb{C}^{(1)}(\mathbf{L}) \cdot \mathbb{I}^a$ is given by

$$\mathbb{C}^{(1)}(\mathbf{W}) = \frac{2}{5} c^{(2)} \left[\frac{\mu_p \Delta \mu}{(\mu_p + c^{(1)} \Delta \mu)\mu^{(1)}} \right]^2 \bar{\mathbf{D}}_e^2. \quad (148)$$

Interestingly, $\mathbb{C}^{(1)}(\mathbf{W})$ depends only on the overall strain rate $\bar{\mathbf{D}}_e$ (and not on $\bar{\mathbf{W}}$), implying that the spin fluctuations can be significant even when the average spins are null. Note that the field fluctuations become null for homogeneous materials and increase with increasing phase heterogeneity (characterized by Δk and $\Delta \mu$), and tend to saturate for large heterogeneity.

The expressions provided above can be simplified further for incompressible composites, by taking the limits as $k^{(1)}, k^{(2)} \rightarrow \infty$. In this case, the isotropic projections become

$$\mathbb{C}^{(1)}(\mathbf{D}_e) = \frac{3}{2} c^{(2)} \left[\frac{2(\mu^{(2)} - \mu^{(1)})}{5\mu^{(1)} + 2c^{(1)}(\mu^{(2)} - \mu^{(1)})} \right]^2 \bar{\mathbf{D}}_e^2 \quad (149)$$

and

$$\mathbb{C}^{(1)}(\mathbf{W}) = \frac{2}{5} c^{(2)} \left[\frac{5(\mu^{(2)} - \mu^{(1)})}{5\mu^{(1)} + 2c^{(1)}(\mu^{(2)} - \mu^{(1)})} \right]^2 \bar{\mathbf{D}}_e^2. \quad (150)$$

Finally, the stress fluctuation in the matrix phase $\mathbb{C}^{(1)}(\boldsymbol{\sigma}_e)$ can be obtained from the strain-rate fluctuation $\mathbb{C}^{(1)}(\mathbf{D}_e)$ as

$$\mathbb{C}^{(1)}(\boldsymbol{\sigma}_e) = \frac{3}{2} \mathbb{C}^{(1)}(\boldsymbol{\sigma}) \cdot \mathbb{K} = 9\mu^{(1)2} \mathbb{C}^{(1)}(\mathbf{D}_e), \quad (151)$$

which leads to the following relation

$$\frac{\mathbb{C}^{(1)}(\boldsymbol{\sigma}_e)}{\bar{\boldsymbol{\sigma}}_e^{(1)2}} = \frac{\mathbb{C}^{(1)}(\mathbf{D}_e)}{\bar{\mathbf{D}}_e^{(1)2}}. \quad (152)$$

3.4. N-phase composites with granular microstructure

This section is concerned with the overall response and field statistics for N -phase composites with granular microstructures, where, similar to Section 2.4.1, each phase is surrounded by the other phases, in such a way that there is no matrix phase. The two-point correlation function, which characterizes the average grain shape and orientation, is assumed to be 'ellipsoidal' with shape tensor \mathbf{Z} . We then make use of the results of Section 2.4.1 for linearized hyperelastic composites and specialize them for linearly viscous composites, with local potentials given by (90), to estimate the overall response and field statistics for these composites.

For the linearly viscous composites, the SC estimates for the velocity-gradient concentration tensors, provided for linearized hyperelastic composites by Eqs. (66), reduce to

$$\mathbb{A}^{(r)} = [\mathbb{I} + \tilde{\mathbb{P}} \mathbb{L}^{s(r)}]^{-1} \quad \text{and} \quad \mathbf{a}^{(r)} = -\tilde{\mathbb{P}} \mathbb{A}^{(r)T} \Delta \boldsymbol{\tau}^{(r)}, \quad (153)$$

after substituting $\mathbb{L}^{(r)} \rightarrow \mathbb{L}^{s(r)}$ and $\mathbf{T}^{(r)} \rightarrow \boldsymbol{\tau}^{(r)}$. Here $\Delta \mathbb{L}^{s(r)} = \mathbb{L}^{s(r)} - \tilde{\mathbb{L}}^s$, $\Delta \boldsymbol{\tau}^{(r)} = \boldsymbol{\tau}^{(r)} - \tilde{\boldsymbol{\tau}}$, and $\tilde{\mathbb{P}}$ is obtained by substituting $\tilde{\mathbb{L}}^s$ for \mathbb{L} in Eq. (67). The microstructural tensor $\tilde{\mathbb{P}}$ can be decomposed, by means of identity (95) for a fourth-order tensor with major symmetry, into

$$\tilde{\mathbb{P}} = \tilde{\mathbb{P}}^s + \tilde{\mathbb{R}} + \tilde{\mathbb{R}}^T + \tilde{\mathbb{P}}^a. \quad (154)$$

The fully symmetric $\tilde{\mathbb{P}}^s = \mathbb{I}^s \tilde{\mathbb{P}} \mathbb{I}^s$, with the corresponding Cartesian components obtained by projecting expression (67) onto symmetric subspaces, i.e.,

$$\tilde{P}_{ijkl}^s = \frac{\det(\mathbf{Z})}{16\pi} \int_{|\zeta|=1} \frac{1}{|\mathbf{Z}\zeta|^3} (\tilde{N}_{ik}\zeta_j\zeta_l + \tilde{N}_{jk}\zeta_i\zeta_l + \tilde{N}_{il}\zeta_j\zeta_k + \tilde{N}_{jl}\zeta_i\zeta_k) dS_\zeta, \quad (155)$$

where \tilde{N}_{ik} are the Cartesian components of the inverse of the acoustic tensor, $\tilde{\mathbf{N}}$. Similarly, $\tilde{\mathbb{R}} = \mathbb{I}^a \tilde{\mathbb{P}} \mathbb{I}^s$, with the Cartesian components given by

$$\tilde{R}_{ijkl} = \frac{\det(\mathbf{Z})}{16\pi} \int_{|\zeta|=1} \frac{1}{|\mathbf{Z}\zeta|^3} (\tilde{N}_{ik}\zeta_j\zeta_l - \tilde{N}_{jk}\zeta_i\zeta_l + \tilde{N}_{il}\zeta_j\zeta_k - \tilde{N}_{jl}\zeta_i\zeta_k) dS_\zeta. \quad (156)$$

The other projections, $\tilde{\mathbb{R}}^T$ and $\tilde{\mathbb{P}}^a$, can also be expressed in terms of $\tilde{\mathbf{N}}$ by appropriately projecting expression (67) onto symmetric and anti-symmetric subspaces.

Now, following the procedure discussed in Section 3.3, the velocity-gradient concentration tensors can be expressed in the form of Eqs. (101) and (97). Thus, by means of an identity analogous to (D.5), the concentration tensor $\mathbb{A}^{(r)}$ can be expressed

$$\mathbb{A}^{(r)} = \mathbb{A}^{s(r)} + \mathbb{I}^a - \tilde{\mathbb{R}} \Delta \mathbb{L}^{s(r)} \mathbb{A}^{s(r)}, \quad (157)$$

where the symmetrized concentration tensors is

$$\mathbb{A}^{s(r)} = \mathbb{I}^s \mathbb{A}^{(r)} \mathbb{I}^s = [\mathbb{I}^s + \tilde{\mathbb{P}}^s \Delta \mathbb{L}^{s(r)}]^{-1} \quad (158)$$

and the other projections are provided by $\mathbb{I}^a \mathbb{A}^{(r)} \mathbb{I}^s = -\tilde{\mathbb{R}} \Delta \mathbb{L}^{s(r)} \mathbb{A}^{s(r)}$ and $\mathbb{I}^a \mathbb{A}^{(r)} \mathbb{I}^a = \mathbb{I}^a$.

On the other hand, the second-order concentration tensor $\mathbf{a}^{(r)}$ in Eq. (153)₂ becomes

$$\mathbf{a}^{(r)} = -(\tilde{\mathbb{P}}^s + \tilde{\mathbb{R}}) \mathbb{A}^{s(r)T} \Delta \boldsymbol{\tau}^{(r)}, \quad (159)$$

with symmetric and anti-symmetric parts given by

$$\begin{aligned} \mathbf{a}^{s(r)} &= \mathbb{I}^s \mathbf{a}^{(r)} = -\tilde{\mathbb{P}}^s \mathbb{A}^{s(r)T} \Delta \boldsymbol{\tau}^{(r)} \quad \text{and} \\ \mathbf{a}^{a(r)} &= \mathbb{I}^a \mathbf{a}^{(r)} = -\tilde{\mathbb{R}} \mathbb{A}^{s(r)T} \Delta \boldsymbol{\tau}^{(r)}. \end{aligned} \quad (160)$$

By substituting these expressions for the concentration tensors into Eqs. (98)₁, we obtain the following implicit equation for the SC estimate for the overall stiffness $\tilde{\mathbb{L}}^s$

$$(\tilde{\mathbb{L}}^s + \mathbb{L}^{s*})^{-1} = \sum_{r=1}^N c^{(r)} (\mathbb{L}^{s(r)} + \mathbb{L}^{s*})^{-1}, \quad (161)$$

where $\mathbb{L}^{s*} = (\tilde{\mathbb{P}}^s)^{-1} - \tilde{\mathbb{L}}^s$. The corresponding expressions for the overall stress polarization $\tilde{\boldsymbol{\tau}}$ and zero strain-rate energy \tilde{f} are obtained similarly obtained by means of expressions (98)₂ and (98)₃, respectively.

The velocity-gradient averages in the phases are given in terms of the concentration tensors by $\bar{\mathbb{L}}^{(r)} = \mathbb{A}^{(r)} \bar{\mathbb{L}} + \mathbf{a}^{(r)}$. The phase averages of the strain rate and spin can then be retrieved using

Eqs. (102) and (103), and are given by

$$\begin{aligned} \bar{\mathbf{D}}^{(r)} &= \mathbb{A}^{s(r)} \bar{\mathbf{D}} + \mathbf{a}^{s(r)}, \\ \bar{\mathbf{W}}^{(r)} &= \bar{\mathbf{W}} - \tilde{\mathbb{R}} (\tilde{\mathbb{P}}^s)^{-1} (\bar{\mathbf{D}} - \bar{\mathbf{D}}^{(r)}). \end{aligned} \quad (162)$$

In the last expression, we have used $\bar{\mathbf{D}}^{(r)} - \bar{\mathbf{D}} = (\mathbb{A}^{s(r)} - \mathbb{I}^s) \bar{\mathbf{D}} + \mathbf{a}^{s(r)} = -\tilde{\mathbb{P}}^s \mathbb{A}^{s(r)T} \Delta \mathbb{L}^s \bar{\mathbf{D}} - \tilde{\mathbb{P}}^s \mathbb{A}^{s(r)T} \Delta \boldsymbol{\tau}^{(r)}$. These expressions for the phase averages are of course identical to the corresponding expressions in the literature (Liu et al., 2005; Song and Ponte Castañeda, 2018a).

Next, the second moments of the velocity-gradient fields in the phases, $\langle \mathbf{L} \otimes \mathbf{L} \rangle^{(r)}$, are obtained by specializing the corresponding expressions from Section 2.4.1. This involves specializing the derivatives of the overall properties with respect to the phase elasticity tensor for linearized hyperelastic composites to linearly viscous composites, i.e., $\partial \tilde{\mathbb{L}} / \partial \mathbb{L}^{(r)}|_{\mathbb{L}^{(r)} = \mathbb{L}^{s(r)}, \mathbf{T}^{(r)} = \boldsymbol{\tau}^{(r)}}$ and $\partial \tilde{\mathbf{T}} / \partial \mathbb{L}^{(r)}|_{\mathbb{L}^{(r)} = \mathbb{L}^{s(r)}, \mathbf{T}^{(r)} = \boldsymbol{\tau}^{(r)}}$ and

$\partial \tilde{f} / \partial \mathbb{L}^{(r)}|_{\mathbb{L}^{(r)} = \mathbb{L}^{s(r)}, \mathbf{T}^{(r)} = \boldsymbol{\tau}^{(r)}}$. The derivative $\partial \tilde{\mathbb{L}} / \partial \mathbb{L}^{(r)}|_{\mathbb{L}^{(r)} = \mathbb{L}^{s(r)}}$ is obtained by evaluating Eq. (76) for linearly viscous composites and can be expressed in terms of Cartesian components as

$$\begin{aligned} &\sum_{s=1}^N c^{(s)} \left[\frac{1}{2} \left(H_{ijef}^{(s)} H_{klgh}^{(s)} + H_{ijgh}^{(s)} H_{klfe}^{(s)} \right) - \frac{\partial \tilde{P}_{ijkl}}{\partial \tilde{L}_{efgh}} \right]_{\tilde{\mathbb{L}} = \tilde{\mathbb{L}}^s} \\ &+ (H_{ijab}^{(s)} \tilde{P}_{abmn}^{-1}) \frac{\partial \tilde{P}_{mnop}}{\partial \tilde{L}_{efgh}} \bigg|_{\tilde{\mathbb{L}} = \tilde{\mathbb{L}}^s} (\tilde{P}_{opcd}^{-1} H_{cdkl}^{(s)}) \bigg] \frac{\partial \tilde{L}_{efgh}}{\partial L_{rsuv}^{(r)}} \bigg|_{\mathbb{L}^{(r)} = \mathbb{L}^{s(r)}} \\ &= \frac{c^{(r)}}{2} \left(H_{ijrs}^{(r)} H_{kluv}^{(r)} + H_{ijuv}^{(r)} H_{klrs}^{(r)} \right), \end{aligned} \quad (163)$$

which is a linear tensorial equation in the full space. Here, $\partial \tilde{\mathbb{P}} / \partial \tilde{\mathbb{L}}|_{\tilde{\mathbb{L}} = \tilde{\mathbb{L}}^s}$ is evaluated from Eqs. (75) after substituting $\tilde{\mathbb{L}}^s$ for \mathbb{L} , and $\mathbb{H}^{(r)} = (\tilde{\mathbb{P}}^{-1} + \Delta \mathbb{L}^{s(r)})^{-1}$, which is obtained by specializing Eq. (70) for linearly viscous composites. Similarly, the other two derivatives, $\partial \tilde{\mathbf{T}} / \partial \mathbb{L}^{(r)}|_{\mathbb{L}^{(r)} = \mathbb{L}^{s(r)}, \mathbf{T}^{(r)} = \boldsymbol{\tau}^{(r)}}$ and $\partial \tilde{f} / \partial \mathbb{L}^{(r)}|_{\mathbb{L}^{(r)} = \mathbb{L}^{s(r)}, \mathbf{T}^{(r)} = \boldsymbol{\tau}^{(r)}}$, are obtained by specializing Eqs. (77)–(78) and (80), respectively, for linearly viscous composites. This, in turn, involves the quantities $\partial \tilde{\mathbb{L}} / \partial \mathbb{L}^{(r)}|_{\mathbb{L}^{(r)} = \mathbb{L}^{s(r)}}$, which are given by Eq. (163).

Once we have obtained the second moments of the velocity gradient $\langle \mathbf{L} \otimes \mathbf{L} \rangle^{(r)}$, the second moments of the strain rate $\langle \mathbf{D} \otimes \mathbf{D} \rangle^{(r)}$ and spin $\langle \mathbf{W} \otimes \mathbf{W} \rangle^{(r)}$, together with the cross-moments $\langle \mathbf{W} \otimes \mathbf{D} \rangle^{(r)}$ and $\langle \mathbf{D} \otimes \mathbf{W} \rangle^{(r)}$, are obtained by the means of the projections (111). It is important to point out that the expressions for the second moments $\langle \mathbf{D} \otimes \mathbf{D} \rangle^{(r)}$ that are obtained by the above-described procedure are identical to those obtained earlier using standard methodologies making use of derivatives of the type $\partial \tilde{\mathbb{P}}^s / \partial \mathbb{L}^s$, $\partial \tilde{\mathbb{L}}^s / \partial \mathbb{L}^{s(r)}$, $\partial \tilde{\boldsymbol{\tau}} / \partial \mathbb{L}^{s(r)}$, etc., computed in the appropriately symmetrized subspaces (Liu, 2003; Brenner et al., 2004; Liu et al., 2005; Lebensohn et al., 2007). However, by evaluating the derivatives in the full space to obtain expressions for $\langle \mathbf{L} \otimes \mathbf{L} \rangle^{(r)}$, allows us to extract not only the strain-rate second moments, but also the spin second moments and the cross moments of the strain rate and spin in the phases of composites with granular microstructures, which have not been previously determined.

3.4.1. Two-phase isotropic incompressible composites

Similar to Subsection 3.3.1, this section is concerned with isotropic composites, thus allowing us to obtain explicit expressions for the overall response and field statistics in terms of the material properties. In particular, we consider two-phase incompressible composites with isotropic granular microstructures. The phases are also isotropic with phase viscosity tensors

$$\mathbb{L}^{s(r)} = 2\mu^{(r)} \mathbb{K}, \quad \text{for } r = 1, 2, \quad (164)$$

which corresponds to those given in Eq. (81) with $\gamma^{(1)} = \gamma^{(2)} = 0$.

The overall response and field statistics for this composite can be obtained by specializing the corresponding expressions for general $\mathbb{L}^{(r)}$ in Section 3.4 for the isotropic composites (164). However, this would not be able to provide explicit expressions for the field fluctuations in terms of material properties. This problem could be alleviated by specializing the expressions for isotropic composites in Section 2.4.2 by setting $\gamma^{(r)} = 0$. Although this approach would not allow us to access the full fluctuation tensor, this enables us to derive analytical expressions for the isotropic projections of field fluctuations, as well as the overall responses and phase averages, directly in terms of material properties.

For the above-described composites, the microstructural tensor $\tilde{\mathbb{P}}$ can be obtained by evaluating expression (83) for $\gamma^{(1)} = \gamma^{(2)} = 0$, and is given by

$$\tilde{\mathbb{P}} = \frac{1}{5\tilde{\mu}}\mathbb{K} + \frac{1}{3\tilde{\mu}}\mathbb{I}^a, \quad (165)$$

where $\tilde{\mu}$ is the overall viscosity, with the overall viscosity tensor $\tilde{\mathbb{L}} = 2\tilde{\mu}\mathbb{K}$, and is determined by

$$6\tilde{\mu}^2 + \tilde{\mu}[10(c^{(1)}\mu^{(2)} + c^{(2)}\mu^{(1)}) - 6(\mu^{(1)} + \mu^{(2)})] - 4\mu^{(1)}\mu^{(2)} = 0. \quad (166)$$

Thus, $\tilde{\mathbb{P}}^s = (1/5\tilde{\mu})\mathbb{K}$, $\tilde{\mathbb{P}}^a = (1/3\tilde{\mu})\mathbb{I}^a$ and $\tilde{\mathbb{R}} = \mathbb{O}$.

The average velocity gradient in the phases is then obtained from expression (85) by setting $\gamma^{(r)} = 0$:

$$\bar{\mathbf{L}}^{(r)} = \left[\frac{5\tilde{\mu}}{3\tilde{\mu} + 2\mu^{(r)}}\mathbb{K} + \mathbb{I}^a \right] \bar{\mathbf{L}}, \quad (167)$$

from which the average strain rate and spin can be retrieved as

$$\bar{\mathbf{D}}^{(r)} = \left[\frac{5\tilde{\mu}}{3\tilde{\mu} + 2\mu^{(r)}}\mathbb{K} \right] \bar{\mathbf{D}}, \quad \bar{\mathbf{W}}^{(r)} = \bar{\mathbf{W}}. \quad (168)$$

Similarly, the isotropic projections of the second moments in the phases are obtained by setting $\gamma^{(r)} = 0$ in expressions (86)–(88) of Section 2.4.2, i.e.,

$$\begin{aligned} \langle \mathbf{D} \otimes \mathbf{D} \rangle^{(r)} \cdot \mathbb{K} &= \frac{1}{2c^{(r)}} \bar{\mathbf{L}} \cdot \frac{\partial \tilde{\mathbb{L}}}{\partial \mu^{(r)}} \bar{\mathbf{L}} \bigg|_{\substack{\gamma^{(1)}=0 \\ \gamma^{(2)}=0}} \quad \text{and} \\ \langle \mathbf{W} \otimes \mathbf{W} \rangle^{(r)} \cdot \mathbb{I}^a &= \frac{1}{2c^{(r)}} \bar{\mathbf{L}} \cdot \frac{\partial \tilde{\mathbb{L}}}{\partial \gamma^{(r)}} \bar{\mathbf{L}} \bigg|_{\substack{\gamma^{(1)}=0 \\ \gamma^{(2)}=0}} \end{aligned} \quad (169)$$

Then, using Eq. (87), the fluctuation covariance of the equivalent strain rate in phase 1 is found to be given by

$$\begin{aligned} C^{(1)}(D_e) &= \frac{2}{3} C^{(1)}(\mathbf{D}) \cdot \mathbb{K} \\ &= \left[\frac{\tilde{\mu}}{c^{(1)}} \frac{4\mu^{(2)} + \tilde{\mu}(10c^{(1)} - 4)}{6\tilde{\mu}^2 + 4\mu^{(1)}\mu^{(2)}} - \frac{25\tilde{\mu}^2}{(3\tilde{\mu} + 2\mu^{(1)})^2} \right] \bar{D}_e^2. \end{aligned} \quad (170)$$

The corresponding fluctuation covariance in phase 2 is obtained by interchanging phases 1 and 2 in the above relation. These fluctuation covariances become identical for both phases when normalized by the corresponding phase averages of equivalent strain rate, as given by

$$\frac{C^{(r)}(D_e)}{\bar{D}_e^{(r)2}} = \frac{2}{5} \tilde{\mu} \frac{15\tilde{\mu} + 10(c^{(1)}\mu^{(2)} + c^{(2)}\mu^{(1)})}{6\tilde{\mu}^2 + 4\mu^{(1)}\mu^{(2)}} - 1, \quad (171)$$

demonstrating that the phases play similar roles, when the homogenized response of the granular composites is estimated using the (symmetric) SC approximation.

The determination of the spin fluctuation also requires the use of Eq. (87), which reduce to

$$\frac{\partial \tilde{\gamma}}{\partial \gamma^{(r)}} \bigg|_{\substack{\gamma^{(1)}=0 \\ \gamma^{(2)}=0}} = c^{(r)} \quad (172)$$

and

$$\frac{\partial \tilde{\mu}}{\partial \gamma^{(r)}} \bigg|_{\substack{\gamma^{(1)}=0 \\ \gamma^{(2)}=0}} = 10c^{(r)} \frac{[(c^{(1)}\mu^{(1)} + c^{(2)}\mu^{(2)}) - \tilde{\mu}]\tilde{\mu}}{6\tilde{\mu}^2 + 4\mu^{(1)}\mu^{(2)}}. \quad (173)$$

It then follows that the isotropic projections of the spin covariance tensor in the phases $C^{(r)}(\mathbf{W})$ are given by

$$C^{(r)}(\mathbf{W}) = \frac{2}{3} C^{(r)}(\mathbf{W}) \cdot \mathbb{I}^a = 10\bar{D}_e^2 \frac{[(c^{(1)}\mu^{(1)} + c^{(2)}\mu^{(2)}) - \tilde{\mu}]\tilde{\mu}}{6\tilde{\mu}^2 + 4\mu^{(1)}\mu^{(2)}}. \quad (174)$$

It is interesting to note that these quantities are identical in both phases of two-phase isotropic incompressible composites with granular microstructure. Moreover, the spin fluctuations depend only on the overall strain rate (and not on $\bar{\mathbf{W}}$), indicating that $C^{(r)}(\mathbf{W})$ can be large—even when the average spins are null.

The isotropic projections of the overall fluctuation covariance tensors (115) for the strain rate and spin, which are respectively given by

$$C(D_e) = \frac{2}{3} C(\mathbf{D}) \cdot \mathbb{K} \quad \text{and} \quad C(\mathbf{W}) = \frac{2}{3} C(\mathbf{W}) \cdot \mathbb{I}^a, \quad (175)$$

are found to be identical as well, and given by

$$C(\mathbf{W}) = C(D_e) = 10\bar{D}_e^2 \frac{[(c^{(1)}\mu^{(1)} + c^{(2)}\mu^{(2)}) - \tilde{\mu}]\tilde{\mu}}{[6\tilde{\mu}^2 + 4\mu^{(1)}\mu^{(2)}]}. \quad (176)$$

This is in spite of the fact that the phase fluctuations for the strain rate and spin fields are themselves different ($C^{(r)}(\mathbf{W}) \neq C^{(r)}(D_e)$). Finally, the stress fluctuation in the phases are easily obtained from the corresponding strain-rate fluctuations using a relation analogous to expression (151), which leads to the result

$$\frac{C^{(r)}(\sigma_e)}{\bar{\sigma}_e^{(r)2}} = \frac{C^{(r)}(D_e)}{\bar{D}_e^{(r)2}}. \quad (177)$$

4. Numerical applications for linearly viscous composites and polycrystals

This section is concerned with the overall response and field statistics for linearly viscous composites with two different types of isotropic microstructures: (i) particulate microstructures, consisting of inclusions that are embedded in a well-defined matrix, and which are homogenized using the Ponte Castañeda–Willis (PCW) method; and (ii) granular microstructures, with no distinct matrix phase, and which are homogenized using the self-consistent (SC) approximation. In this section, we will consider composites with isotropic and anisotropic phases, as well as polycrystalline aggregates.

4.1. Two-phase isotropic incompressible composites

First, we investigate simple examples of two-phase composites consisting of isotropic and incompressible phases with contrast or viscosity ratio $\mu^{(2)}/\mu^{(1)}$. We consider two subclasses of particulate composites, which are modeled using the results of Section 3.3.1 for the PCW estimate: one with hard inclusions

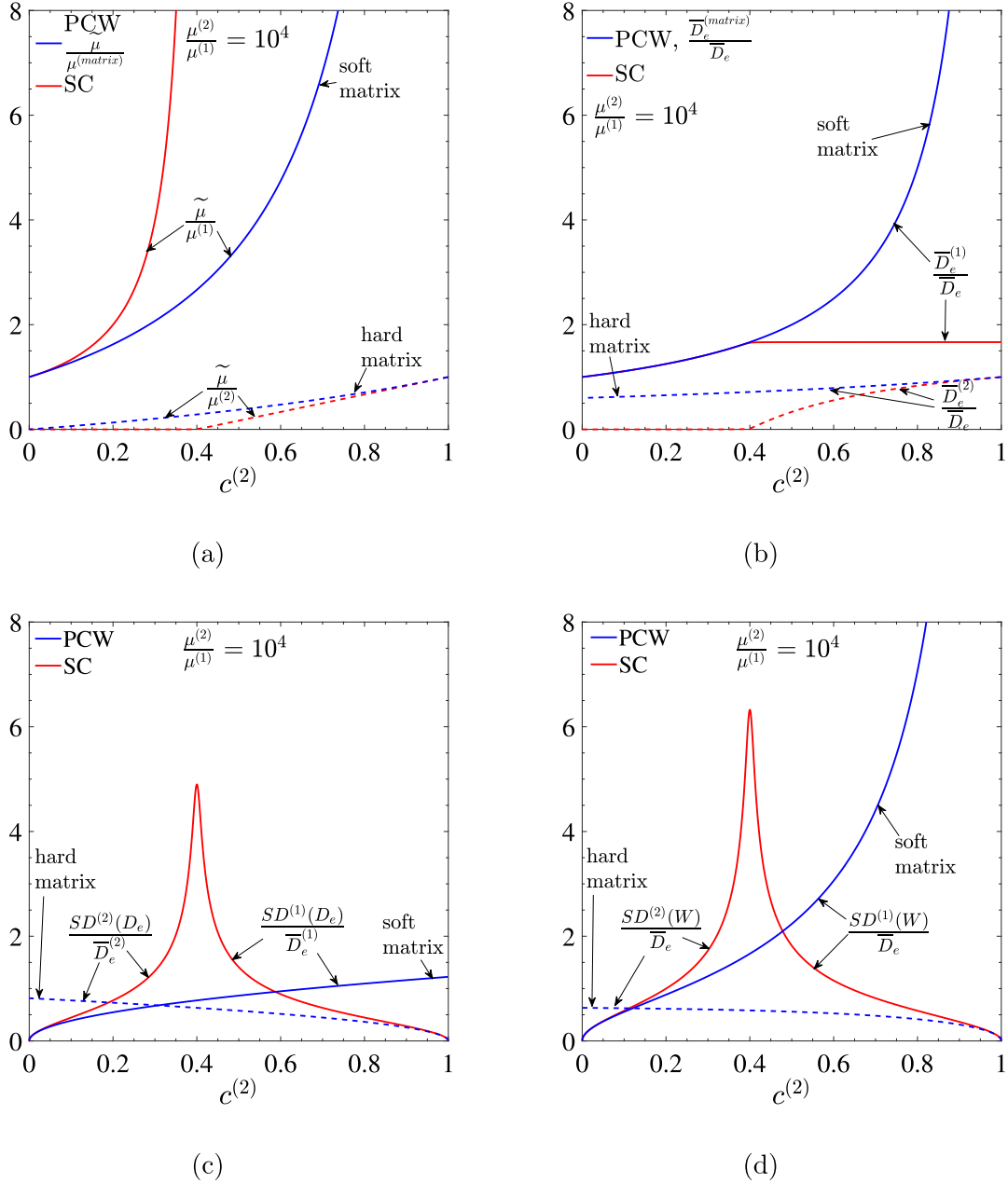


Fig. 1. Results for two-phase isotropic incompressible composites with phase contrast $\mu^{(2)}/\mu^{(1)} = 10^4$, where phases 1 and 2 are labeled 'soft' and 'hard,' respectively. Two types of microstructures are considered: particulate, homogenized using the PCW estimate (blue lines), and granular, homogenized using the SC estimate (red lines). For the particulate composites, two cases are distinguished: 'soft matrix' (continuous lines) and 'hard matrix' (dashed lines). (a) Normalized overall viscosity $\tilde{\mu}$. (b) Normalized average strain rate in the phases $\bar{D}_e^{(r)}/\bar{D}_e$. (c) Normalized strain-rate standard deviations $SD^{(r)}(D_e)/\bar{D}_e^{(r)}$. (d) Normalized spin standard deviations $SD^{(r)}(W)/\bar{D}_e$. All the results are shown as functions of the hard phase volume fraction $c^{(2)}$.

embedded in a 'soft matrix' phase and the other with soft inclusions in a 'hard matrix.' In addition, we consider composites with granular microstructures, where both phases play 'symmetric' roles, and are modeled using the results given in Section 3.4.1 for the SC estimate. Results are shown in Fig. 1 as function of the volume fraction $c^{(2)}$ for a fixed value of $\mu^{(2)}/\mu^{(1)} = 10^4$ and in Fig. 2 as functions of the viscosity ratio $\mu^{(2)}/\mu^{(1)}$ for two different values of $c^{(2)}$.

Fig. 1(a) shows results for the overall viscosity $\tilde{\mu}$. The PCW estimates for the 'soft matrix' and 'hard matrix' cases are normalized by the viscosities of the soft phase $\mu^{(1)}$ and hard phase $\mu^{(2)}$,

respectively, while the SC estimate for the granular microstructures is normalized by both the viscosities of the soft phase $\mu^{(1)}$ and hard phase $\mu^{(2)}$ (i.e., the same SC estimate normalized in two different ways). These normalizations are chosen to highlight the well-known fact that the SC estimate has the same dilute limit as the 'soft matrix' PCW estimate for $c^{(2)} \rightarrow 0$, while it has the same dilute limit as the 'hard matrix' PCW estimate for $c^{(2)} \rightarrow 1$. For this reason, we refer to these two different normalizations of the SC estimate as 'soft matrix' and 'hard matrix' SC estimates, respectively. In addition, for this large value of the viscosity ratio $\mu^{(2)}/\mu^{(1)}$, the 'soft matrix' SC estimate

is seen to 'percolate' with $\tilde{\mu}/\mu^{(1)}$ tending to increase dramatically (from the left) near $c^{(2)} \approx 0.4$, while the 'hard matrix' SC estimate for $\tilde{\mu}/\mu^{(2)}$ tends to vanish (from the right) near $c^{(1)} \approx 0.6$ (or $c^{(2)} \approx 0.4$). (Of course, it can be checked that the 'soft matrix' SC estimate percolates exactly as $c^{(2)} \rightarrow 0.4$ for rigid particles and that the 'hard matrix' SC estimate percolates exactly as $c^{(1)} \rightarrow 0.6$ for vacuous particles.) The behavior is quite different for the 'soft matrix' and 'hard matrix' PCW estimates, which do not 'percolate' until $c^{(2)} \rightarrow 1$ and $c^{(1)} \rightarrow 1$, respectively.

Fig. 1(b) presents corresponding results for the normalized strain rates $\bar{D}_e^{(r)}/\bar{D}_e$, as functions of the hard phase volume fraction $c^{(2)}$. For the particulate composites, results are shown only for the matrix phase, while for the granular composites results are shown for both phases. In the 'soft matrix' particulate composites, the deformation is accommodated primarily by the soft matrix phase ($\bar{D}_e^{(1)} \approx \bar{D}_e/(1 - c^{(2)})$ and $\bar{D}_e^{(2)} \approx 0$), while for the 'hard matrix' particulate composites both phases deform. For the granular composite, where results are shown for both phases, it is seen that, for $c^{(2)} \lesssim 0.4$, the hard phase hardly deforms at all ($\bar{D}_e^{(1)} \approx 0$), while the strain rate in the soft phase is given by $\bar{D}_e^{(2)} \approx \bar{D}_e/(1 - c^{(2)})$. However, for $c^{(2)} \gtrsim 0.4$, the hard phases are expected to start forming clusters of long-range extent and thus are forced to deform. As a consequence, in this regime of $c^{(2)}$ ($0.4 \lesssim c^{(2)} < 1.0$), the strain rate in the soft phases (of the granular composites) no longer continues to increase with the volume fraction, remaining constant with $\bar{D}_e^{(1)} \approx (5/3)\bar{D}_e$. On the other hand, the strain rate in the hard phase $\bar{D}_e^{(2)}$ is seen to slowly decrease (from the right) until it becomes nearly zero at $c^{(1)} \approx 0.6$ (and beyond). These SC results for the 'soft' and 'hard' phases should be respectively compared with the 'soft matrix' and 'hard matrix' PCW estimates for particulate composites. Again, it is seen that the results agree in the appropriate dilute limits, but exhibit very different behaviors near and beyond percolation.

Fig. 1(c) shows the normalized standard deviations of the strain-rate fluctuations $SD^{(r)}(D_e)/\bar{D}_e^{(r)}$, characterizing the field heterogeneity in phase r . Again, for the particulate composites, results are shown only for the matrix phase (recall that, according to the PCW estimates, the fields are uniform in the inclusions), while for the granular composites results are shown for both phases. At dilute volume fractions of inclusions, the field heterogeneity in the matrix phase of the particulate composites is similar to that of the corresponding phase in the granular composite. For the particulate composites, the field in the matrix phase becomes progressively more heterogeneous with increasing inclusion volume fraction until reaching 100%. On the other hand, for the granular composites, the heterogeneity is a non-monotonic function of $c^{(2)}$, being zero at $c^{(2)} = 0$ or 1 and attaining a maximum near the 'percolation' limit at around $c^{(2)} = 0.4$. In fact, for rigidly-reinforced granular composites ($\mu^{(2)}/\mu^{(1)} \rightarrow \infty$), we can show by means of expression (171) that at the percolation limit ($c^{(2)} = 0.4$), where $\tilde{\mu}$ blows up, $SD^{(r)}(D_e)/\bar{D}_e^{(r)} = \sqrt{c^{(1)}\tilde{\mu}/\mu^{(1)} - 1}$ also blows up. Therefore, the strain rate fields become strongly heterogeneous near the percolation limit, as expected. Moreover, it is interesting to note that the SC estimates for the suitably normalized standard deviations are identical in both phases, as already noted in the context of Eq. (171), i.e., $SD^{(1)}(D_e)/\bar{D}_e^{(1)} = SD^{(2)}(D_e)/\bar{D}_e^{(2)}$, reflecting the similar roles played by both phases in composites with granular microstructures.

Fig. 1(d) presents results for the spin standard deviations $SD^{(r)}(W)$, normalized by the overall strain rate \bar{D}_e . Once again, for the particulate composites, results are shown only for the matrix phase, while for the granular composites results are shown for both phases. The first thing to note is that the spin fields are

strongly heterogeneous, even though their phase averages are null (as can be shown from Eqs. (168)₂ and (145)₂ for the PCW and SC estimates, respectively). Thus, it can be seen that the spin fields become progressively more heterogeneous in the matrix phase of the particulate composites as the inclusion volume fractions increase, while the fluctuations in the phases of the granular composite exhibit non-monotonic trends with $c^{(2)}$, attaining their maximum values at the 'percolation' volume fraction ($c^{(2)} \approx 0.4$). It is also interesting to remark that the spin matrix fluctuations in the 'soft matrix' particulate composites tend to blow up as the volume fraction of the hard phase approaches 100%, while they remain finite for the 'hard matrix' particulate composites, even as $c^{(1)} \rightarrow 1$. Moreover, the spin fluctuations for the granular composites are identical in both phases, i.e., $SD^{(1)}(W) = SD^{(2)}(W)$, consistent with Eq. (174).

Next, to better understand the effect of phase contrast on the fields, Fig. 2 presents results for the field statistics as functions of the viscosity ratio $\mu^{(2)}/\mu^{(1)}$. In this figure, for the PCW estimates phases 1 and 2 correspond to the matrix and inclusions, respectively, while for the SC estimates, where both phases play symmetric roles (since there is no matrix phase), the result is the same in both phases.

Figs. 2(a) and 2(b) show the normalized strain-rate standard deviations $SD^{(r)}(D_e)/\bar{D}_e^{(r)}$, as functions of the viscosity ratio $\mu^{(2)}/\mu^{(1)}$, at $c^{(2)} = 0.4$ and $c^{(2)} = 0.6$, respectively. The normalized standard deviations, which are measures for the field heterogeneity, are seen to increase with the phase heterogeneity both as $\mu^{(2)}/\mu^{(1)}$ increases from the value 1, or decreases from 1. For the PCW estimates for the particulate microstructures, it is seen that the normalized fluctuations $SD^{(1)}(D_e)/\bar{D}_e^{(1)}$ generally tend to level off to a finite value, which depends on the volume fraction $c^{(2)}$. On the other hand, the SC estimates for the granular microstructures shown in Fig. 2(a) are seen to exhibit generally larger normalized fluctuations, which tend to blow up as $SD^{(1)}(D_e)/\bar{D}_e^{(1)} \sim (\mu^{(2)}/\mu^{(1)})^{1/4}$ with increasing values of $\mu^{(2)}/\mu^{(1)}$, consistent with what was observed in the context of Fig. 1(c) for $c^{(2)} = 0.4$. It should also be noted that the results for the SC estimate at $c^{(2)} = 0.6$ in Fig. 2(b) is the same as that obtained by 'flipping' phase 1 and 2 in the results at $c^{(2)} = 0.4$ in Fig. 2(a). This 'duality' between phases is absent in particulate composites (which have well-defined matrix phases) and explains the different behavior observed in Fig. 2(b) for $c^{(2)} = 0.6$.

Figs. 2(c) and 2(d) present the normalized spin standard deviations $SD^{(1)}(W)/\bar{D}_e$ as functions of the viscosity ratio $\mu^{(2)}/\mu^{(1)}$ at $c^{(2)} = 0.4$ and $c^{(2)} = 0.6$, respectively. Similar to the strain-rate field, the spin field becomes progressively more heterogeneous with the phase heterogeneity (increasing or decreasing values of $\mu^{(2)}/\mu^{(1)}$ from 1). For the particulate composite, the matrix-field heterogeneity increases with the phase contrast until saturation, with the saturation levels increasing with $c^{(2)}$. On the other hand, as shown in Fig. 2(c) for the granular composite with $c^{(2)} = 0.4$, the spin heterogeneity intensifies with the phase contrast, growing like $SD^{(1)}(W)/\bar{D}_e \sim (\mu^{(2)}/\mu^{(1)})^{1/4}$ for large $\mu^{(2)}/\mu^{(1)}$. As before, the SC estimate for $c^{(2)} = 0.6$ shown in Fig. 2(d) can be obtained by flipping phase 1 and 2 in Fig. 2(c)—this duality is absent for particulate composites.

4.2. Composites with anisotropic crystalline phases

In the last section, we demonstrated the dependence of the overall fluctuation on phase heterogeneity for composites with isotropic phases. However, the phase anisotropy, which introduces preferential orientations for the deformation patterns in the phases, influences the field fluctuations. In this section, we seek to elucidate the effect of the anisotropy of the phases by

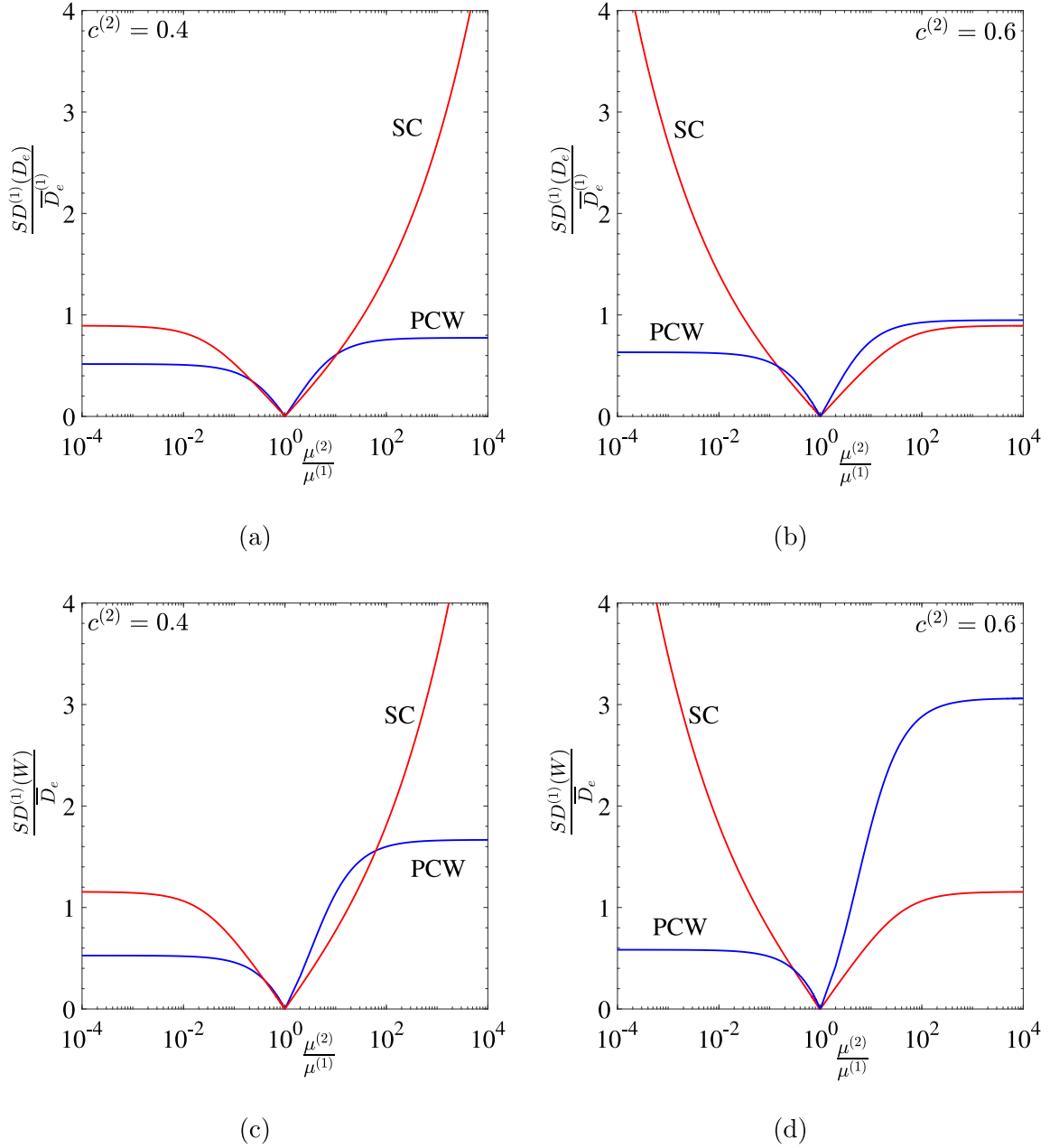


Fig. 2. Results for two-phase isotropic incompressible composites with particulate and granular microstructures homogenized by means of the PCW estimates (blue lines) and SC estimates (red lines), respectively. For the particulate composites, the matrix and inclusion phases are labeled 1 and 2, respectively. Normalized strain-rate standard deviations $SD^{(1)}(D_e)/\bar{D}_e^{(1)}$, as functions of the phase contrast $\mu^{(2)}/\mu^{(1)}$, for (a) $c^{(2)} = 0.4$ and (b) $c^{(2)} = 0.6$. Normalized spin standard deviations $SD^{(1)}(W)/\bar{D}_e$, as functions of $\mu^{(2)}/\mu^{(1)}$, for (c) $c^{(2)} = 0.4$ and (d) $c^{(2)} = 0.6$.

considering applications to porous hexagonal-close-packed (HCP) single crystals, as well as solid HCP polycrystals. In particular, we consider ice-like HCP crystalline phases with three soft $\{0001\}\langle 1\bar{2}10 \rangle$ basal ('ba') slip systems, three hard $\{10\bar{1}0\}\langle 1\bar{2}10 \rangle$ prismatic ('pr') systems and six hard $\{11\bar{2}2\}\langle 11\bar{2}3 \rangle$ pyramidal ('py') systems. The basal and prismatic systems contains only four linearly independent slip systems, allowing no deformation along the $\langle c \rangle$ -axis ($\{0001\}$), the symmetry axis perpendicular to the basal plane. However, the pyramidal systems provides five linearly independent slip systems, thus allowing arbitrary deformation.

While ice single crystals and polycrystals exhibit nonlinear constitutive response, as already mentioned, results for the overall behavior and field statistics in nonlinear composites and polycrystals can be obtained directly from corresponding results for suitably optimized linear composites and polycrystals by means of the linear comparison variational approximations (Ponte Castañeda, 1991, 2016). For this reason, we focus our attention here on linear composites and polycrystals, leaving further consideration of nonlinear composites and polycrystals for future work. Thus, the local stress potential for the single-crystal phases is

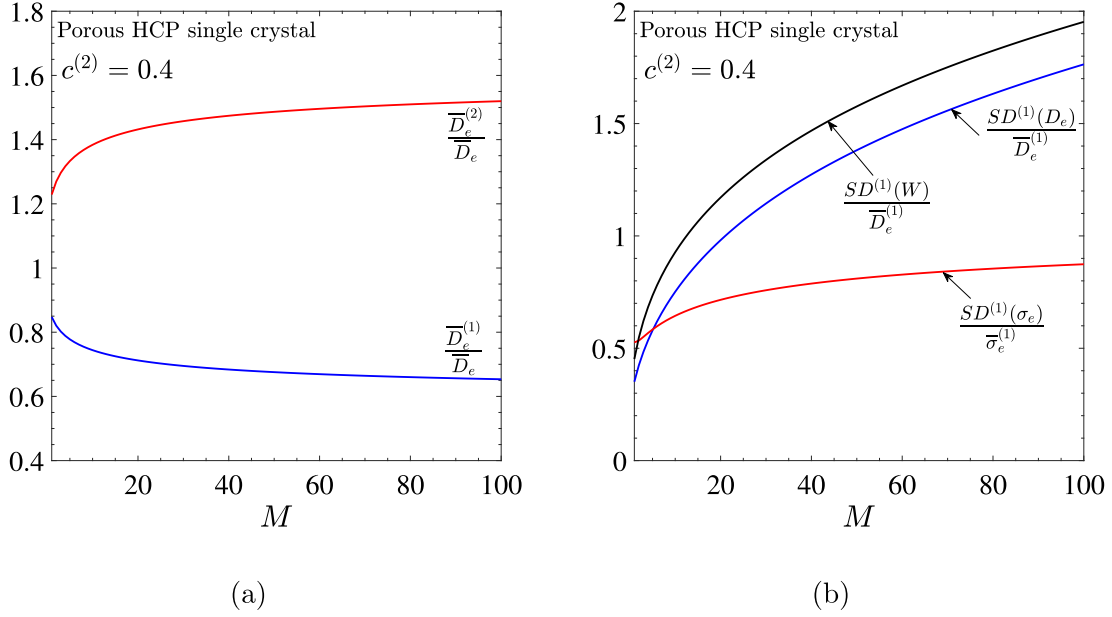


Fig. 3. PCW estimates for the field statistics in porous HCP single crystals with spherical pores of volume fraction $c^{(2)} = 0.4$. (a) Normalized average strain rate in the matrix phase $\bar{D}_e^{(1)}/\bar{D}_e$ and the pores $\bar{D}_e^{(2)}/\bar{D}_e$, as functions of the matrix anisotropy M . (b) Normalized matrix standard deviations of the equivalent stress $SD^{(1)}(\sigma_e)/\bar{\sigma}_e^{(1)}$, strain rate $SD^{(1)}(D_e)/\bar{D}_e^{(1)}$ and spin $SD^{(1)}(W)/\bar{D}_e^{(1)}$, all as functions of M .

taken to be of the form

$$U^{(r)}(\sigma) = \sum_{k=1}^K \frac{1}{2\tau_{(k)}} \left(\tau_{(k)}^{(r)} \right)^2 = \frac{1}{2} \sigma \cdot \mathbb{M}^{s(r)} \sigma$$

$$\text{with } \mathbb{M}^{s(r)} = \sum_{k=1}^K \frac{1}{\tau_{(k)}} \mu_{(k)}^{(r)} \otimes \mu_{(k)}^{(r)}, \quad (178)$$

where K is the number of slip systems ($K = 12$ for ice-like HCP single crystals), $\tau_{(k)}$ is the flow stress of k th slip system and $\tau_{(k)}^{(r)}$ is the resolved shear stress that is defined as

$$\tau_{(k)}^{(r)} = \sigma \cdot \mu_{(k)}^{(r)}, \quad \text{with } \mu_{(k)}^{(r)} = \frac{1}{2} (\mathbf{m}_{(k)}^{(r)} \otimes \mathbf{n}_{(k)}^{(r)} + \mathbf{n}_{(k)}^{(r)} \otimes \mathbf{m}_{(k)}^{(r)}). \quad (179)$$

The Schmidt tensor $\mu_{(k)}^{(r)}$ is obtained from the unit vectors $\mathbf{m}_{(k)}^{(r)}$, along the slip direction, and $\mathbf{n}_{(k)}^{(r)}$, normal to the slip plane, of the k th slip system in phase r . The flow stresses for each family of slip systems are taken to be identical, but could be different for different slip systems. In particular, the basal systems are taken to be ‘soft,’ while the prismatic and pyramidal systems are taken to be hard, with identical flow stresses $\tau_{pr} = \tau_{py}$. Then, the anisotropy of the HCP grains is characterized by $M = \tau_{py}/\tau_{ba} = \tau_{pr}/\tau_{ba} \geq 1$.

4.2.1. Porous HCP single crystals

Here we consider porous single crystals consisting of randomly distributed spherical pores in volume fraction $c^{(2)} = 0.4$, embedded in ice-like HCP crystals. Due to the anisotropy of the matrix phase, the overall response of the porous crystals is anisotropic. For brevity, we restrict our attention to the axisymmetric loading condition

$$\bar{\mathbf{D}} = \mathbf{e}_3 \otimes \mathbf{e}_3 - \frac{1}{2} (\mathbf{e}_1 \otimes \mathbf{e}_1 + \mathbf{e}_2 \otimes \mathbf{e}_2), \quad (180)$$

where the unit vector \mathbf{e}_3 is aligned with the $\langle c \rangle$ -axis. Under this loading condition, the hard non-basal systems are required to deform, leading to highly heterogeneous fields in the anisotropic

matrix phase. However, it follows from Eq. (128)₂ that, like the macroscopic spin, the averages of the spin in the phases are null.

Fig. 3(a) shows the average strain rates in the matrix phase $\bar{D}_e^{(1)}$ and the pores $\bar{D}_e^{(2)}$, both normalized by the overall strain rate \bar{D}_e . It is seen that the average strain rate in the matrix decreases with increasing matrix anisotropy M , as a consequence of the increasing matrix viscosity, which makes the matrix harder to deform. In turn, the deformation of the pores increases with M in order to accommodate the prescribed overall strain rate (180). Note that the pores cannot support any stress ($\bar{\sigma}_e^{(2)} = 0$) and thus the stress in the matrix phase is $\bar{\sigma}_e^{(1)}/\bar{\sigma}_e = 1/(1 - c^{(2)})$.

Fig. 3(b) presents the normalized matrix standard deviations of the stress $SD^{(1)}(\sigma_e)/\bar{\sigma}_e^{(1)}$, strain rate $SD^{(1)}(D_e)/\bar{D}_e^{(1)}$, and spin $SD^{(1)}(W)/\bar{D}_e^{(1)}$, all as functions of the grain anisotropy M . It can be seen that all the fields become progressively more heterogeneous with increasing values of M . However, the fluctuations of the stress tend to level off for large values of M , while the fluctuations of the strain rate and spin continue to grow with increasing values of M . Thus, it can be seen that the anisotropy of the single-crystal matrix phase in these porous materials plays a roughly similar role to that of the phase contrast in the previous example, and adds to the fluctuations already present as a consequence of the relatively large contrast between the viscosity of the matrix and vacuous inclusions.

4.2.2. HCP polycrystals

Here we consider untextured HCP polycrystals, containing 400 randomly oriented equiaxed ice-like HCP single crystals. The phases (also loosely referred to as grains) correspond to all the single crystals with given orientation tensors $\mathbf{Q}^{(r)}$ ($r = 1, \dots, 400$), such that their lattice vectors $\mathbf{l}_i^{(r)}$ are related to those of a reference single crystal \mathbf{l}_i by $\mathbf{l}_i^{(r)} = \mathbf{Q}^{(r)} \mathbf{l}_i$ ($i = 1, 2, 3$). The volume fractions of the grains are assumed to be identical with $c^{(r)} = 1/400$. Because of the random distribution of equiaxed single crystals, the overall behavior of the polycrystal is isotropic. The polycrystals are subjected to the loading condition (180),

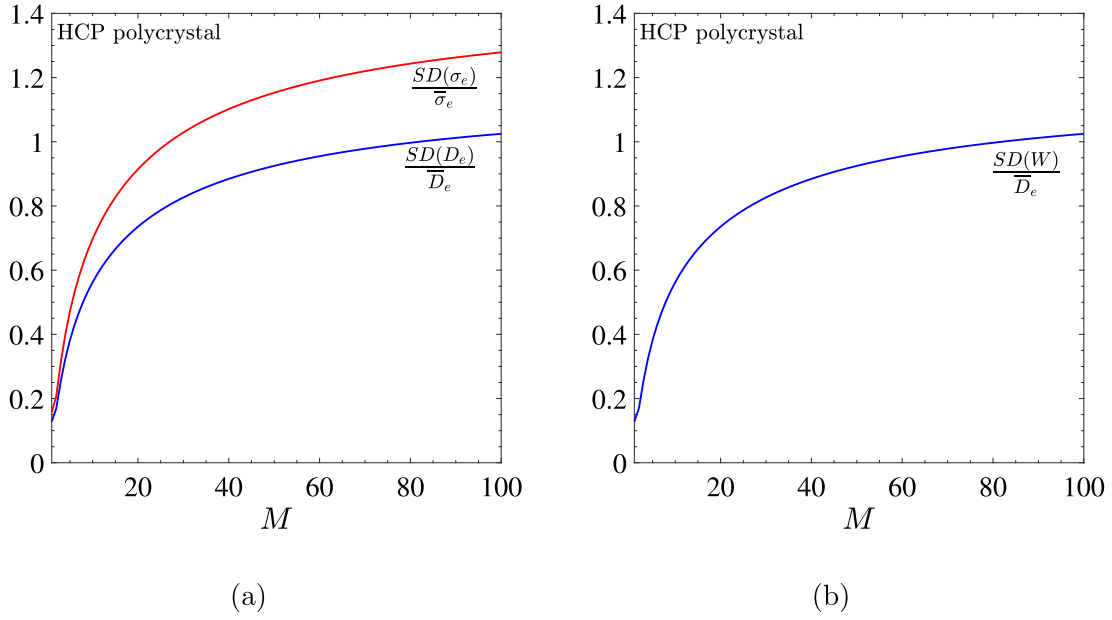


Fig. 4. SC estimates for the overall field fluctuations in untextured HCP polycrystals, as functions of the grain viscoplastic anisotropy M . (a) Normalized overall standard deviations for the stress $SD(\sigma_e)/\bar{\sigma}_e$ and strain rate $SD(D_e)/\bar{D}_e$. (b) Normalized overall standard deviations for the spin $SD(W)/\bar{D}_e$.

for which the overall spin is null. As already noted, the grain anisotropy is characterized by the parameter $M = \tau_{py}/\tau_{ba} = \tau_{pr}/\tau_{ba} \geq 1$, so that the hard non-basal systems become more difficult to deform with increasing values of M . The macroscopic behavior of this type of polycrystal has already been investigated by Song and Ponte Castañeda (2018a), where it was observed that the polycrystals tends to become rigid as $M \rightarrow \infty$. For brevity, such results are not detailed here.

Fig. 4 shows results for the overall field fluctuations, which accounts for both the field heterogeneity within the grains (intragranular fluctuations), as well as the field heterogeneity for different grains (intergranular fluctuations). In particular, Fig. 4(a) shows the normalized overall standard deviations of the equivalent stress and strain-rate fields, $SD(\sigma_e)/\bar{\sigma}_e$ and $SD(D_e)/\bar{D}_e$, while Fig. 4(b) provides the corresponding results for the overall standard deviations of the spin fields $SD(W)/\bar{D}_e$, all as functions of the grain anisotropy M . It should be noted, first of all, that the HCP single crystals are (slightly) anisotropic for $M = 1$ and, as a consequence, the field fluctuations in the polycrystals are small but do not vanish. As the grain anisotropy M increases beyond 1 and the grains become increasingly more difficult to deform, all the fields – including the spin field – become progressively more heterogeneous. In this respect, it is interesting to remark that the overall fluctuations of the strain rate and spin are (numerically) equal–similar to what has been observed for isotropic incompressible granular composites, as can be seen in Eq. (176).

Fig. 5 presents results for the standard deviations of the intragranular fluctuations in the grains for $M = 20$ and 100, as functions of the corresponding normalized equivalent average strain rate in the grains. In particular, Fig. 5(a) shows the normalized standard deviations for the equivalent stress $SD^{(r)}(\sigma_e)/\bar{\sigma}_e$ and strain rate $SD^{(r)}(D_e)/\bar{D}_e$, as functions of $\bar{D}_e^{(r)}/\bar{D}_e$. Consistent with Fig. 4, the standard deviations are much larger for $M = 100$ than for $M = 20$. However, the magnitude of these fluctuations is roughly the same for all grains, unlike the average equivalent strain rates, which vary significantly from grain to grain. On the other hand, Fig. 5(b) shows the normalized intragranular spin fluctuations $SD^{(r)}(W)/\bar{D}_e$, again as functions of $\bar{D}_e^{(r)}/\bar{D}_e$. Similar to the strain-rate fields, the intragranular spin fields are significantly

more heterogeneous for $M = 100$ than for $M = 20$. More importantly, the intragranular fluctuations of the spin are found to be quite large, in spite of the fact that the average spins in the grains are null. Finally, It should be noted that despite the fact that the isotropic projections of all the field fluctuations for different grain orientations are quite similar in magnitude, the actual phase fluctuation covariance tensors, which are anisotropic, do depend strongly on the grain orientation.

5. Concluding remarks

In this work, we have developed a methodology to estimate the overall response and field statistics in linearized elastic and viscous composites and polycrystals. The central idea is to make use of generalized linear-elastic (i.e., linearized hyperelastic) composites with non-symmetric stress and strain fields, as well as elastic modulus tensors that exhibit major symmetry but no minor symmetries. By means of a well-know mathematical analogy, these results can then be applied to linearly viscous composites with symmetric stress and strain-rate fields, as well as fully symmetric viscosity tensors. In addition, it is possible to generate estimates for the first and second moments of the stress and strain-rate tensors, as well as for the spin tensor (including coupled moments with the strain rate), by projections onto the appropriate symmetric and non-symmetric subspaces.

The generalized framework that has been developed in this paper for the overall response and field statistics for linearized composites can also be used, together with the fully optimized second-order linear comparison homogenization method (Ponte Castañeda, 2015, 2016), to generate corresponding estimates for the overall response and field statistics in nonlinear composites, including hyperelastic and viscoplastic composites and polycrystals, although this was not pursued in this work. In addition, it is envisaged that the methodology could be extended for elasto-viscoplastic composites (Lahellec and Suquet, 2007; Kowalczyk-Gajewska and Petryk, 2011; Lahellec and Suquet, 2013; Kurska et al., 2018; Cotelto et al., 2020).

We have also obtained analytical expressions and numerical results for the overall response and field statistics of the strain-rate and spin fields in the phases of two-phase isotropic

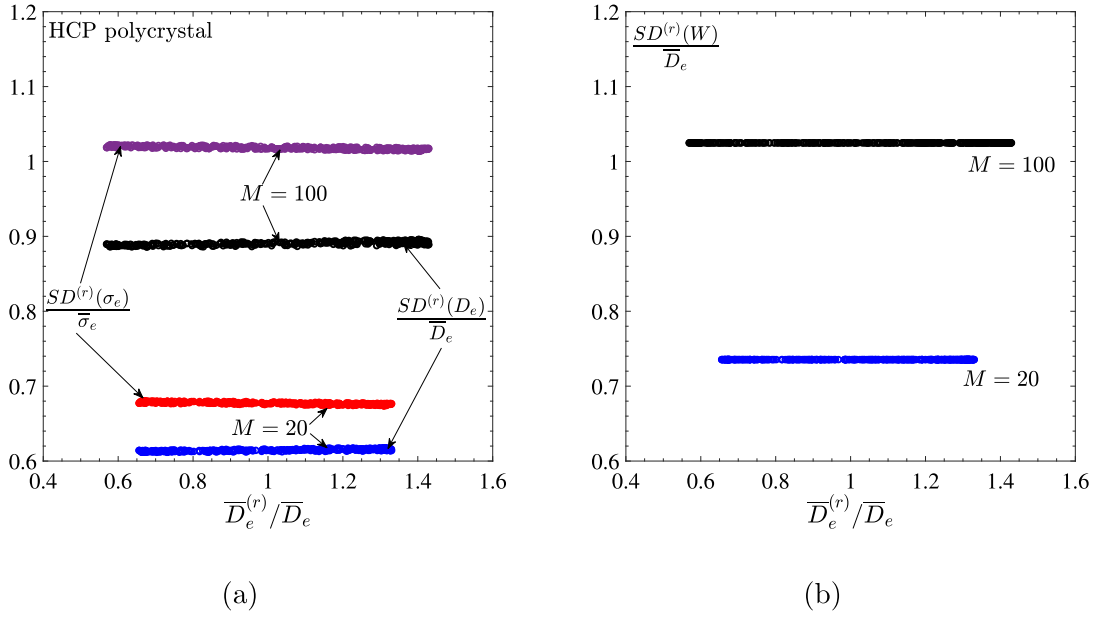


Fig. 5. SC estimates for the intragranular field fluctuations vs. the normalized strain-rate phase averages $\bar{D}_e^{(r)}/\bar{D}_e$, for grain anisotropies $M = 20, 100$. (a) Normalized standard deviations of the intragranular stress fluctuations $SD^{(r)}(\sigma_e)/\bar{\sigma}_e$ and of the strain rate fluctuations $SD^{(r)}(D_e)/\bar{D}_e$. (b) Normalized standard deviations of the intragranular spin fluctuations $SD^{(r)}(W)/\bar{D}_e$.

composites with both particulate and granular microstructures, by making use of the PCW and SC estimates, respectively. It was found that the fluctuations generally increase with increasing heterogeneity contrast and become significant for both particulate and granular composites. Indeed, the field fluctuations tend to blow up near percolation limits for granular composites. Interestingly, the spin fluctuations in the phases were found to depend only on the overall strain rate and not on the overall spin, leading to highly heterogeneous spin fields even when their phase averages are null. Moreover, while the appropriately normalized spin fluctuations are identical in both phases for granular composites, the strain-rate fluctuations are different; however, the overall spin and strain-rate fluctuations are identical. For particulate composites, the fields in the inclusions are uniform while strong field heterogeneities (including in the spin field) can develop in the matrix phase.

The fields also become progressively more heterogeneous with increasing matrix anisotropy, as demonstrated for porous composites comprised of spherical pores embedded in hexagonal closed-packed (HCP) single crystals. Under axisymmetric loading aligned with the symmetry axis of the crystals, the hard non-basal slip systems of ice-like HCP single crystals are required to deform, resulting in highly heterogeneous fields. The spin fluctuations in the matrix phase were found to be of the same order as the strain-rate fluctuations, even though the corresponding phase averages of the spin are null. We have also provided results for the overall and intragranular field fluctuations in linear ice-like HCP untextured polycrystals, which show that the fields can become strongly heterogeneous, because the deformation has to be accommodated primarily by the softer basal slip systems. Indeed, the fields become progressively more heterogeneous with increasing grain anisotropy until saturation. Interestingly, the overall strain-rate and spin fluctuations are approximately equal, while the intragranular spin fluctuations are larger than the corresponding strain-rate fluctuations. Finally, it is noted that an application of these results to (nonlinear) viscoplastic polycrystals is given in a separate publication (Das and Ponte Castañeda, 2021).

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. The derivatives of $\mathbb{H}^{(s)}$ with respect to $\mathbb{L}^{(r)}$

In Section 2.4.1, we obtained the linear tensorial equation (76) for the derivative $\partial \tilde{\mathbb{L}}/\partial \mathbb{L}^{(r)}$ by taking the derivative of Eq. (69) with respect to $\mathbb{L}^{(r)}$. This, in turn, requires evaluating the derivative of $\mathbb{H}^{(s)}$, as given by (70), with respect to $\mathbb{L}^{(r)}$, as an intermediate step. Here, we provide a detailed derivation of expression (74) for $\partial \mathbb{H}^{(s)}/\partial \mathbb{L}^{(r)}$.

Performing term-by-term differentiation of the expression $\mathbb{H}^{(s)} = (\tilde{\mathbb{P}}^{-1} + \Delta \mathbb{L}^{(s)})^{-1}$, we arrive at

$$\frac{\partial \mathbb{H}^{(s)}}{\partial \mathbb{L}^{(r)}} = -(\tilde{\mathbb{P}}^{-1} + \Delta \mathbb{L}^{(s)})^{-1} \frac{\partial [\tilde{\mathbb{P}}^{-1} + \Delta \mathbb{L}^{(s)}]}{\partial \mathbb{L}^{(r)}} (\tilde{\mathbb{P}}^{-1} + \Delta \mathbb{L}^{(s)})^{-1}, \quad (\text{A.1})$$

which can be written alternatively as

$$\begin{aligned} \frac{\partial \mathbb{H}^{(s)}}{\partial \mathbb{L}^{(r)}} &= -\mathbb{H}^{(s)} \frac{\partial [\tilde{\mathbb{P}}^{-1} + \Delta \mathbb{L}^{(s)}]}{\partial \mathbb{L}^{(r)}} \mathbb{H}^{(s)}, \\ &= -\mathbb{H}^{(s)} \frac{\partial \tilde{\mathbb{P}}^{-1}}{\partial \mathbb{L}^{(r)}} \mathbb{H}^{(s)} - \mathbb{H}^{(s)} \frac{\partial \Delta \mathbb{L}^{(s)}}{\partial \mathbb{L}^{(r)}} \mathbb{H}^{(s)} + \mathbb{H}^{(s)} \frac{\partial \tilde{\mathbb{L}}}{\partial \mathbb{L}^{(r)}} \mathbb{H}^{(s)}. \end{aligned} \quad (\text{A.2})$$

Now using the chain rule

$$\frac{\partial \tilde{\mathbb{P}}^{-1}}{\partial \mathbb{L}^{(r)}} = -\tilde{\mathbb{P}}^{-1} \frac{\partial \tilde{\mathbb{P}}}{\partial \mathbb{L}^{(r)}} \tilde{\mathbb{P}}^{-1}, \quad (\text{A.3})$$

we arrive at

$$\frac{\partial \mathbb{H}^{(s)}}{\partial \mathbb{L}^{(r)}} = (\mathbb{H}^{(s)} \tilde{\mathbb{P}}^{-1}) \frac{\partial \tilde{\mathbb{P}}}{\partial \mathbb{L}^{(r)}} (\tilde{\mathbb{P}}^{-1} \mathbb{H}^{(s)}) - \mathbb{H}^{(s)} \frac{\partial \Delta \mathbb{L}^{(s)}}{\partial \mathbb{L}^{(r)}} \mathbb{H}^{(s)} + \mathbb{H}^{(s)} \frac{\partial \tilde{\mathbb{L}}}{\partial \mathbb{L}^{(r)}} \mathbb{H}^{(s)}. \quad (\text{A.4})$$

Appendix B. The symmetries of the overall tensors for linearly viscous composites

In Section 3.1, we stated that the symmetries of phase viscosity tensors $\mathbb{L}^{(r)}$ and the major symmetry of the overall viscosity tensor $\tilde{\mathbb{L}}$ implies that $\tilde{\mathbb{L}}$ is fully symmetric. This, in turn, implies that the overall stress polarization is also symmetric. In this appendix, we provide detailed proofs for these statements.

If the overall stiffness for linearized hyperelastic composites, given by expression (14)₁, is specialized to linearly viscous composites, it is found that

$$\begin{aligned}\tilde{\mathbb{L}} &= \sum_{r=1}^N c^{(r)} \mathbb{L}^{s(r)} \mathbb{A}^{(r)} \\ &= \sum_{r=1}^N c^{(r)} \mathbb{L}^{s(r)} \mathbb{A}^{s(r)} + \sum_{r=1}^N c^{(r)} \mathbb{L}^{s(r)} \mathbb{E}^{(r)} \\ &\quad + \sum_{r=1}^N c^{(r)} \mathbb{L}^{s(r)} \mathbb{F}^{(r)} + \sum_{r=1}^N c^{(r)} \mathbb{L}^{s(r)} \mathbb{A}^{a(r)} \\ &= \sum_{r=1}^N c^{(r)} \mathbb{L}^{s(r)} \mathbb{A}^{s(r)} + \sum_{r=1}^N c^{(r)} \mathbb{L}^{s(r)} \mathbb{F}^{(r)},\end{aligned}\quad (\text{B.1})$$

where the decomposition (96) is used for $\mathbb{A}^{(r)}$, and the minor symmetry of $\mathbb{L}^{s(r)}$ with respect to its right two indices is exploited to reduce $\mathbb{L}^{s(r)} \mathbb{E}^{(r)} = \mathbb{O}$ and $\mathbb{L}^{s(r)} \mathbb{A}^{a(r)} = \mathbb{O}$. The minor symmetry of $\mathbb{L}^{s(r)}$ with respect to its left two indices implies the corresponding minor symmetry of $\tilde{\mathbb{L}}$. Thus, we have that

$$\tilde{\mathbb{L}} = \mathbb{I}^s \tilde{\mathbb{L}}, \quad (\text{B.2})$$

and, by taking its transpose, that

$$\tilde{\mathbb{L}}^T = \tilde{\mathbb{L}}^T \mathbb{I}^s. \quad (\text{B.3})$$

Since $\tilde{\mathbb{L}}$ exhibits major symmetry, $\tilde{\mathbb{L}}^T = \tilde{\mathbb{L}}$, it readily follows that

$$\tilde{\mathbb{L}} = \tilde{\mathbb{L}} \mathbb{I}^s, \quad (\text{B.4})$$

implying that $\tilde{\mathbb{L}}$ possesses minor symmetry with respect to its right two indices. Thus, major symmetry and one minor symmetry imply the other minor symmetry (Jog, 2015). In turn, this leads to

$$\mathbb{F}^{(r)} = \mathbb{O}, \quad (\text{B.5})$$

resulting in the decomposition (97) for $\mathbb{A}^{(r)}$. Then, the standard expression for the overall tensor is recovered as

$$\tilde{\mathbb{L}}^s = \sum_{r=1}^N c^{(r)} \mathbb{L}^{s(r)} \mathbb{A}^{s(r)}. \quad (\text{B.6})$$

On the other hand, the overall stress polarization for linearized hyperelastic composites, given by expression (14)₂, is specialized to linearly viscous composites to obtain the result that

$$\tilde{\boldsymbol{\tau}} = \sum_{r=1}^N c^{(r)} (\mathbb{A}^{s(r)T} + \mathbb{E}^{(r)T} + \mathbb{A}^{a(r)T}) \boldsymbol{\tau}^{(r)}. \quad (\text{B.7})$$

The anti-symmetry of $\mathbb{E}^{(r)T}$ and $\mathbb{A}^{a(r)T}$ with respect to its right two indices and the symmetry of $\boldsymbol{\tau}^{(r)}$ imply that $\mathbb{E}^{(r)T} \boldsymbol{\tau}^{(r)} = \mathbf{0}$ and $\mathbb{A}^{a(r)T} \boldsymbol{\tau}^{(r)} = \mathbf{0}$. This then implies the symmetry of the overall stress polarization tensor, which is then given by

$$\tilde{\boldsymbol{\tau}} = \sum_{r=1}^N c^{(r)} \mathbb{A}^{s(r)T} \boldsymbol{\tau}^{(r)}, \quad (\text{B.8})$$

thus recovering the usual relation for the stress polarization tensor.

Appendix C. The average strain rate and spin in terms of the concentration tensors

Section 3.1 provides expressions (102) and (103) for the average strain rate and spin in the phases, respectively, in terms of the appropriate projections of the concentration tensors $\mathbb{A}^{(r)}$ and $\mathbf{a}^{(r)}$. This appendix is concerned with a detailed derivation of those expressions.

The velocity-gradient average $\bar{\mathbb{L}}^{(r)}$ is given by expression (92), which together with expression (97) for $\mathbb{A}^{(r)}$ implies that the average strain rate in the phases can be written as

$$\begin{aligned}\bar{\mathbb{D}}^{(r)} &= \mathbb{I}^s \bar{\mathbb{L}}^{(r)} \\ &= \mathbb{I}^s \mathbb{A}^{(r)} \bar{\mathbb{L}} + \mathbb{I}^s \mathbf{a}^{(r)} \\ &= \mathbb{I}^s \mathbb{A}^{(r)} (\mathbb{I}^s + \mathbb{I}^a) \bar{\mathbb{L}} + \mathbf{a}^{s(r)} \\ &= \mathbb{A}^{s(r)} \bar{\mathbb{D}} + \mathbf{a}^{s(r)},\end{aligned}\quad (\text{C.1})$$

where in the last step, we use the fact that $\mathbb{F}^{(r)} = \mathbb{I}^s \mathbb{A}^{(r)} \mathbb{I}^a = \mathbb{O}$ and $\mathbb{A}^{s(r)} \bar{\mathbb{L}} = \mathbb{A}^{s(r)} \bar{\mathbb{D}}$, as a result of the right minor symmetry of $\mathbb{A}^{s(r)}$.

Similarly, the average spin in the phases can be written as

$$\begin{aligned}\bar{\mathbb{W}}^{(r)} &= \mathbb{I}^a \bar{\mathbb{L}}^{(r)} \\ &= \mathbb{I}^a \mathbb{A}^{(r)} \bar{\mathbb{L}} + \mathbb{I}^a \mathbf{a}^{(r)} \\ &= \mathbb{I}^a \mathbb{A}^{(r)} (\mathbb{I}^s + \mathbb{I}^a) \bar{\mathbb{L}} + \mathbf{a}^{a(r)} \\ &= \mathbb{E}^{(r)} \bar{\mathbb{L}} + \mathbb{A}^{a(r)} \bar{\mathbb{L}} + \mathbf{a}^{a(r)} \\ &= \mathbb{E}^{(r)} \bar{\mathbb{D}} + \mathbb{A}^{a(r)} \bar{\mathbb{W}} + \mathbf{a}^{a(r)},\end{aligned}\quad (\text{C.2})$$

where in the last step, we exploit the right minor symmetry of $\mathbb{E}^{(r)} = \mathbb{I}^a \mathbb{A}^{(r)} \mathbb{I}^s$ to reduce $\mathbb{E}^{(r)} \bar{\mathbb{L}} = \mathbb{E}^{(r)} \bar{\mathbb{D}}$, and the right anti-symmetry of $\mathbb{A}^{a(r)} = \mathbb{I}^a \mathbb{A}^{(r)} \mathbb{I}^a$ to reduce $\mathbb{A}^{a(r)} \bar{\mathbb{L}} = \mathbb{A}^{a(r)} \bar{\mathbb{W}}$.

Appendix D. Derivation of expression (122) for the concentration tensors $\mathbb{A}^{(r)}$

First, we note that

$$\begin{aligned}\mathbb{A}^{(2)-1} [\mathbb{A}^{s(2)} + \mathbb{I}^a - c^{(1)} \mathbb{R}^{(1)} \Delta \mathbb{L}^s \mathbb{A}^{s(2)}] \\ = [\mathbb{I} + c^{(1)} \mathbb{P}^{(1)} \Delta \mathbb{L}^s] [\mathbb{A}^{s(2)} + \mathbb{I}^a - c^{(1)} \mathbb{R}^{(1)} \Delta \mathbb{L}^s \mathbb{A}^{s(2)}] \\ = \mathbb{A}^{s(2)} + \mathbb{I}^a - c^{(1)} \mathbb{R}^{(1)} \Delta \mathbb{L}^s \mathbb{A}^{s(2)} + c^{(1)} \mathbb{P}^{(1)} \Delta \mathbb{L}^s \mathbb{A}^{s(2)},\end{aligned}\quad (\text{D.1})$$

where $\mathbb{A}^{s(2)}$ is provided by expression (123), and we use the fact $\Delta \mathbb{L}^s \mathbb{I}^a = \mathbb{O}$ and $\Delta \mathbb{L}^s \mathbb{R}^{(1)} = \mathbb{O}$,

which follows from minor symmetries of $\Delta \mathbb{L}^s$ and anti-symmetries of \mathbb{I}^a and $\mathbb{R}^{(1)}$. By means of the decomposition (119) for $\mathbb{P}^{(1)}$, together with the symmetries of its projections, Eq. (D.1) can be simplified further as

$$\begin{aligned}\mathbb{A}^{(2)-1} [\mathbb{A}^{s(2)} + \mathbb{I}^a - c^{(1)} \mathbb{R}^{(1)} \Delta \mathbb{L}^s \mathbb{A}^{s(2)}] \\ = \mathbb{A}^{s(2)} + \mathbb{I}^a + c^{(1)} \mathbb{P}^{s(1)} \Delta \mathbb{L}^s \mathbb{A}^{s(2)}.\end{aligned}\quad (\text{D.3})$$

Now, expression (123) for $\mathbb{A}^{s(2)}$ can be rewritten as

$$\mathbb{A}^{s(2)} = \mathbb{I}^s - c^{(1)} \mathbb{P}^{s(1)} \Delta \mathbb{L}^s \mathbb{A}^{s(2)}, \quad (\text{D.4})$$

which allows us to obtain

$$\mathbb{A}^{(2)-1} [\mathbb{A}^{s(2)} + \mathbb{I}^a - c^{(1)} \mathbb{R}^{(1)} \Delta \mathbb{L}^s \mathbb{A}^{s(2)}] = \mathbb{I}^s + \mathbb{I}^a = \mathbb{I}. \quad (\text{D.5})$$

Similarly, it can be shown that

$$[\mathbb{A}^{s(2)} + \mathbb{I}^a - c^{(1)}\mathbb{R}^{(1)}\Delta\mathbb{L}^s\mathbb{A}^{s(2)}]\mathbb{A}^{(2)-1} = \mathbb{I}. \quad (\text{D.6})$$

Thus, we arrive at the identity (122):

$$\mathbb{A}^{(2)} = \mathbb{A}^{s(2)} + \mathbb{I}^a - c^{(1)}\mathbb{R}^{(1)}\Delta\mathbb{L}^s\mathbb{A}^{s(2)}. \quad (\text{D.7})$$

Appendix E. Derivation of Eq. (129) in Section 3.3

This appendix is concerned with a detailed derivation of Eq. (129) in Section 3.3. The average velocity gradients in the inclusions are given by

$$\bar{\mathbf{L}}^{(2)} = \mathbb{A}^{(2)}\bar{\mathbf{L}} + \mathbf{a}^{(2)}, \quad (\text{E.1})$$

with the concentration tensors provided by expressions (118). This could be used to rewrite the velocity-gradient difference

$$\begin{aligned} \bar{\mathbf{L}} - \bar{\mathbf{L}}^{(1)} &= \frac{c^{(2)}}{c^{(1)}}(\bar{\mathbf{L}}^{(2)} - \bar{\mathbf{L}}) \\ &= \frac{c^{(2)}}{c^{(1)}}((\mathbb{A}^{(2)} - \mathbb{I})\bar{\mathbf{L}} + \mathbf{a}^{(2)}). \end{aligned} \quad (\text{E.2})$$

Now, the expression (118) for the concentration tensor $\mathbb{A}^{(2)}$ can be rewritten as

$$\mathbb{A}^{(2)} = \mathbb{I} - c^{(1)}\mathbb{P}^{(1)}\mathbb{A}^{(2)T}\Delta\mathbb{L}^s. \quad (\text{E.3})$$

This expression, together with (118)₂ for the thermal concentration tensor $\mathbf{a}^{(2)}$, leads to

$$\begin{aligned} \bar{\mathbf{L}} - \bar{\mathbf{L}}^{(1)} &= -c^{(2)}\mathbb{P}^{(1)}\mathbb{A}^{(2)T}(\Delta\mathbb{L}^s\bar{\mathbf{L}} + \Delta\boldsymbol{\tau}) \\ &= -c^{(2)}\mathbb{P}^{(1)}\mathbb{A}^{s(2)T}(\Delta\mathbb{L}^s\bar{\mathbf{D}} + \Delta\boldsymbol{\tau}), \end{aligned} \quad (\text{E.4})$$

where we have exploited minor symmetries of $\Delta\mathbb{L}^s$ to reduce $\Delta\mathbb{L}^s\bar{\mathbf{L}} = \mathbb{L}^s\bar{\mathbf{D}}$, and expression (122) for $\mathbb{A}^{(2)}$ to reduce $\mathbb{A}^{(2)T}(\Delta\mathbb{L}^s\bar{\mathbf{D}} + \Delta\boldsymbol{\tau}) = \mathbb{A}^{s(2)T}(\Delta\mathbb{L}^s\bar{\mathbf{D}} + \Delta\boldsymbol{\tau})$. The decomposition (119) allows us to further simplify the above expression such that

$$\bar{\mathbf{L}} - \bar{\mathbf{L}}^{(1)} = -c^{(2)}(\mathbb{P}^{s(1)} + \mathbb{R}^{(1)})\mathbb{A}^{s(2)T}(\Delta\mathbb{L}^s\bar{\mathbf{D}} + \Delta\boldsymbol{\tau}). \quad (\text{E.5})$$

By projecting on the symmetric subspace, the strain-rate difference is obtained as

$$\bar{\mathbf{D}} - \bar{\mathbf{D}}^{(1)} = -c^{(2)}\mathbb{P}^{s(1)}\mathbb{A}^{s(2)T}(\Delta\mathbb{L}^s\bar{\mathbf{D}} + \Delta\boldsymbol{\tau}). \quad (\text{E.6})$$

Thus, expression (E.5) can be rewritten as

$$\bar{\mathbf{L}} - \bar{\mathbf{L}}^{(1)} = (\mathbb{P}^{s(1)} + \mathbb{R}^{(1)})(\mathbb{P}^{s(1)})^{-1}(\bar{\mathbf{D}} - \bar{\mathbf{D}}^{(1)}). \quad (\text{E.7})$$

Multiplying the above equation by $(\mathbb{P}^{(1)})^{-1}$, we obtain

$$(\mathbb{P}^{(1)})^{-1}(\bar{\mathbf{L}} - \bar{\mathbf{L}}^{(1)}) = (\mathbb{P}^{(1)})^{-1}(\mathbb{P}^{s(1)} + \mathbb{R}^{(1)})(\mathbb{P}^{s(1)})^{-1}(\bar{\mathbf{D}} - \bar{\mathbf{D}}^{(1)}). \quad (\text{E.8})$$

Now, from the decomposition (119) for $\mathbb{P}^{(1)}$, we know that

$$\mathbb{P}^{(1)}\mathbb{I}^s = \mathbb{P}^{s(1)} + \mathbb{R}^{(1)}, \quad (\text{E.9})$$

leading to

$$\mathbb{I}^s = (\mathbb{P}^{(1)})^{-1}(\mathbb{P}^{s(1)} + \mathbb{R}^{(1)}) \quad (\text{E.10})$$

Thus, we arrive at Eq. (129):

$$(\mathbb{P}^{(1)})^{-1}(\bar{\mathbf{L}} - \bar{\mathbf{L}}^{(1)}) = (\mathbb{P}^{s(1)})^{-1}(\bar{\mathbf{D}} - \bar{\mathbf{D}}^{(1)}). \quad (\text{E.11})$$

References

Avazmohammadi, R., Ponte Castañeda, P., 2014. On the macroscopic response, microstructure evolution, and macroscopic stability of short-fibre-reinforced elastomers at finite strains: I—Analytical results. *Phil. Mag.* 94 (10), 1031–1067.

Avazmohammadi, R., Ponte Castañeda, P., 2016. Macroscopic constitutive relations for elastomers reinforced with short aligned fibers: Instabilities and post-bifurcation response. *J. Mech. Phys. Solids* 97, 37–67.

Ball, J.M., 1977. Convexity conditions and existence theorems in nonlinear elasticity. *Arch. Ration. Mech. Anal.* 63, 337–403.

Bobeth, M., Diener, G., 1986. Field fluctuations in multicomponent mixtures. *J. Mech. Phys. Solids* 34 (1), 1–17.

Bornert, M., Masson, R., Ponte Castañeda, P., Zaoui, A., 2001. Second-order estimates for the effective behaviour of viscoplastic polycrystalline materials. *J. Mech. Phys. Solids* 49 (11), 2737–2764.

Brenner, R., Castelnau, O., Badea, L., 2004. Mechanical field fluctuations in polycrystals estimated by homogenization techniques. *Proc. R. Soc. Lond. Ser. A Math. Phys. Eng. Sci.* 460 (2052), 3589–3612.

Cotelo, J., Das, S., Ponte Castañeda, P., 2020. A differential homogenization method for estimating the macroscopic response and field statistics of particulate viscoelastic composites. *Int. J. Solids Struct.* 204–205, 199–219.

Danas, K., Idiart, M.I., Ponte Castañeda, P., 2008. A homogenization-based constitutive model for isotropic viscoplastic porous media. *Int. J. Solids Struct.* 45 (11–12), 3392–3409.

Das, S., Ponte Castañeda, P., 2021. Statistics of the stress, strain-rate and spin fields in viscoplastic polycrystals. *Int. J. Solids Struct.* 217–218, 193–214.

Finlay, H.M., Whittaker, P., Canham, P.B., 1998. Collagen organization in the branching region of human brain arteries. *Stroke* 29 (8), 1595–1601.

Furer, J., Ponte Castañeda, P., 2018. A symmetric fully optimized second-order method for nonlinear homogenization. *ZAMM-J. Appl. Math. Mech./Z. Angew. Math. Mech.* 98 (2), 222–254.

Geymonat, G., Müller, S., Triantafyllidis, N., 1993. Homogenization of nonlinearly elastic materials, microscopic bifurcation and macroscopic loss of rank-one convexity. *Arch. Ration. Mech. Math.* 122, 231–290.

Guo, Z.-H., 1983a. The representation theorem for isotropic, linear asymmetric stress-strain relations. *J. Elasticity* 13 (2), 121–124.

Guo, Z.H., 1983b. An alternative proof of the representation theorem for isotropic, linear asymmetric stress-strain relations. *Quart. Appl. Math.* 41 (1), 119–123.

Hershey, A., 1954. The elasticity of an isotropic aggregate of anisotropic cubic crystals. *J. Appl. Mech.-Trans. ASME* 21 (3), 236–240.

Hill, R., 1972. On constitutive macro-variables for heterogeneous solids at finite strain. *Proc. R. Soc. Lond. Ser. A Math. Phys. Eng. Sci.* 326 (1565), 131–147.

Honeker, C.C., Thomas, E.L., 1996. Impact of morphological orientation in determining mechanical properties in triblock copolymer systems. *Chem. Mater.* 8 (8), 1702–1714.

Honeker, C.C., Thomas, E.L., Albalak, R.J., Hajduk, D.A., Gruner, S.M., Capel, M.C., 2000. Perpendicular deformation of a near-single-crystal triblock copolymer with a cylindrical morphology. 1. Synchrotron SAXS. *Macromolecules* 33 (25), 9395–9406.

Idiart, M.I., Ponte Castañeda, P., 2007a. Field statistics in nonlinear composites. I. Theory. *Proc. R. Soc. A* 463 (2077), 183–202.

Idiart, M.I., Ponte Castañeda, P., 2007b. Field statistics in nonlinear composites. II. Applications. *Proc. R. Soc. A* 463 (2077), 203–222.

Jog, C., 2006. A concise proof of the representation theorem for fourth-order isotropic tensors. *J. Elasticity* 85 (2), 119–124.

Jog, C.S., 2015. *Continuum Mechanics*, Vol. 1. Cambridge University Press.

Kowalczyk-Gajewska, K., Petryk, H., 2011. Sequential linearization method for viscous/elastic heterogeneous materials. *Eur. J. Mech. A* 30 (5), 650–664.

Kreher, W., 1990. Residual stresses and stored elastic energy of composites and polycrystals. *J. Mech. Phys. Solids* 38 (1), 115–128.

Kröner, E., 1958. Berechnung der elastischen Konstanten des Vielkristalls aus den Konstanten des Einkristalls. *Z. Phys.* 151 (4), 504–518.

Kursa, M., Kowalczyk-Gajewska, K., Lewandowski, M.J., Petryk, H., 2018. Elastic-plastic properties of metal matrix composites: Validation of mean-field approaches. *Eur. J. Mech. A* 68, 53–66.

Lahellec, N., Suquet, P., 2007. On the effective behavior of nonlinear inelastic composites: I. Incremental variational principles. *J. Mech. Phys. Solids* 55 (9), 1932–1963.

Lahellec, N., Suquet, P., 2013. Effective response and field statistics in elasto-plastic and elasto-viscoplastic composites under radial and non-radial loadings. *Int. J. Plast.* 42, 1–30.

Laws, N., 1973. On the thermostatics of composite materials. *J. Mech. Phys. Solids* 21 (1), 9–17.

Lebensohn, R., Liu, Y., Ponte Castañeda, P., 2004. On the accuracy of the self-consistent approximation for polycrystals: comparison with full-field numerical simulations. *Acta Mater.* 52 (18), 5347–5361.

Lebensohn, R., Tomé, C., Ponte Castañeda, P., 2007. Self-consistent modelling of the mechanical behaviour of viscoplastic polycrystals incorporating intragranular field fluctuations. *Phil. Mag.* 87 (28), 4287–4322.

Lebensohn, R.A., Zecevic, M., Knezevic, M., McCabe, R.J., 2016. Average intragranular misorientation trends in polycrystalline materials predicted by a viscoplastic self-consistent approach. *Acta Mater.* 104, 228–236.

Leroy, Y., Ponte Castañeda, P., 2001. Bounds on the self-consistent approximation for nonlinear media and implications for the second-order method. *C. R. Acad. Sci. IIB* 329 (8), 571–577.

- Levin, V., 1992. Thermal expansion coefficients of heterogeneous materials. 2, SAGE Publications, pp. 58–61.
- Liu, Y., 2003. Macroscopic Behavior, Field Fluctuations and Texture Evolution in Viscoplastic Polycrystals (Ph.D. Thesis). University of Pennsylvania.
- Liu, Y., Gilormini, P., Ponte Castañeda, P., 2005. Homogenization estimates for texture evolution in halite. *Tectonophysics* 406 (3–4), 179–195.
- Liu, Y., Ponte Castañeda, P., 2004. Second-order theory for the effective behavior and field fluctuations in viscoplastic polycrystals. *J. Mech. Phys. Solids* 52 (2), 467–495.
- Lopez-Pamies, O., Ponte Castañeda, P., 2004. Second-order homogenization estimates incorporating field fluctuations in finite elasticity. *Math. Mech. Solids* 9 (3), 243–270.
- Lopez-Pamies, O., Ponte Castañeda, P., 2006a. On the overall behavior, microstructure evolution, and macroscopic stability in reinforced rubbers at large deformations: I—Theory. *J. Mech. Phys. Solids* 54 (4), 807–830.
- Moulinec, H., Suquet, P., 2003. Intraphase strain heterogeneity in nonlinear composites: a computational approach. *Eur. J. Mech. A Solids* 22 (5), 751–770.
- Nebozhyn, M., Ponte Castañeda, P., 1998. Second-order estimates for the effective behavior of nonlinear porous materials. In: *IUTAM Symposium on Transformation Problems in Composite and Active Materials*. Springer, pp. 73–88.
- Ogden, R.W., 1997. *Non-linear Elastic Deformations*. Courier Corporation.
- Parton, V., Buryachenko, V., 1990. Stress fluctuations in elastic composites. *Sov. Phys. Dokl.* 310 (5), 1075–1078.
- Petryk, H., Stupkiewicz, S., Kuziak, R., 2008. Grain refinement and strain hardening in IF steel during multi-axis compression: Experiment and modelling. *J. Mater. Process. Technol.* 204 (1–3), 255–263.
- Ponte Castañeda, P., 1991. The effective mechanical properties of nonlinear isotropic composites. *J. Mech. Phys. Solids* 39 (1), 45–71.
- Ponte Castañeda, P., 1996. Exact second-order estimates for the effective mechanical properties of nonlinear composite materials. *J. Mech. Phys. Solids* 44 (6), 827–862.
- Ponte Castañeda, P., 2002. Second-order homogenization estimates for nonlinear composites incorporating field fluctuations: I – Theory. *J. Mech. Phys. Solids* 50 (4), 737–757.
- Ponte Castañeda, P., 2005. *Heterogeneous Materials. Lecture Notes*, Ecole Polytechnique.
- Ponte Castañeda, P., 2015. Fully optimized second-order variational estimates for the macroscopic response and field statistics in viscoplastic crystalline composites. *Proc. R. Soc. A* 471 (2184), 20150665.
- Ponte Castañeda, P., 2016. Stationary variational estimates for the effective response and field fluctuations in nonlinear composites. *J. Mech. Phys. Solids* 96, 660–682.
- Ponte Castañeda, P., Suquet, P., 1998. Nonlinear composites. In: *Advances in Applied Mechanics*, Vol. 34. Elsevier, pp. 171–302.
- Ponte Castañeda, P., Tiberio, E., 2000. A second-order homogenization method in finite elasticity and applications to black-filled elastomers. *J. Mech. Phys. Solids* 48 (6–7), 1389–1411.
- Ponte Castañeda, P., Willis, J.R., 1995. The effect of spatial distribution on the effective behavior of composite materials and cracked media. *J. Mech. Phys. Solids* 43 (12), 1919–1951.
- Ponte Castañeda, P., Willis, J.R., 1999. Variational second-order estimates for nonlinear composites. *Proc. R. Soc. Lond. Ser. A* 455, 1799–1811.
- Quapp, K., Weiss, J., 1998. Material characterization of human medial collateral ligament. *J. Biomech. Eng.*
- Racherla, V., Lopez-Pamies, O., Ponte Castañeda, P., 2010. Macroscopic response and stability in lamellar nanostructured elastomers with “oriented” and “unoriented” polydomain microstructures. *Mech. Mater.* 42 (4), 451–468.
- Siboni, M.H., Ponte Castañeda, P., 2016. Macroscopic response of particle-reinforced elastomers subjected to prescribed torques or rotations on the particles. *J. Mech. Phys. Solids* 91, 240–264.
- Song, D., Ponte Castañeda, P., 2017a. A finite-strain homogenization model for viscoplastic porous single crystals: I—Theory. *J. Mech. Phys. Solids* 107, 560–579.
- Song, D., Ponte Castañeda, P., 2017b. A finite-strain homogenization model for viscoplastic porous single crystals: II—Applications. *J. Mech. Phys. Solids* 107, 580–602.
- Song, D., Ponte Castañeda, P., 2018a. Fully optimized second-order homogenization estimates for the macroscopic response and texture evolution of low-symmetry viscoplastic polycrystals. *Int. J. Plast.* 110, 272–293.
- Song, D., Ponte Castañeda, P., 2018b. A multi-scale homogenization model for fine-grained porous viscoplastic polycrystals: I—Finite-strain theory. *J. Mech. Phys. Solids* 115, 102–122.
- Suquet, P., 1995. Overall properties of nonlinear composites: a modified secant moduli theory and its link with Ponte Castañeda's nonlinear variational procedure. *C. R. Acad. Sci. II* 320 (11), 563–571.
- Willis, J., 1977. Bounds and self-consistent estimates for the overall properties of anisotropic composites. *J. Mech. Phys. Solids* 25 (3), 185–202.
- Willis, J., 1978. Variational principles and bounds for the overall properties of composites. *Continuum Models Discret. Syst.* 185–215.
- Willis, J.R., 1981. Variational and related methods for the overall properties of composites. *Adv. Appl. Mech.* 21, 1–78.
- Zecevic, M., Lebensohn, R.A., McCabe, R.J., Knezevic, M., 2019. Modelling recrystallization textures driven by intragranular fluctuations implemented in the viscoplastic self-consistent formulation. *Acta Mater.* 164, 530–546.
- Zecevic, M., Pantleon, W., Lebensohn, R.A., McCabe, R.J., Knezevic, M., 2017. Predicting intragranular misorientation distributions in polycrystalline metals using the viscoplastic self-consistent formulation. *Acta Mater.* 140, 398–410.