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This work presents a means for extracting the statistics of the local fields in nonlinear composites from the effective potential of suitably perturbed composites. The idea is to introduce a parameter in the local potentials, generally a tensor, such that differentiation of the corresponding effective potential with respect to the parameter yields the volume average of the desired quantity. In particular, this provides a generalization to the nonlinear case of well-known formulas in the context of linear composites, which express phase averages and second moments of the local fields in terms of derivatives of the effective potential. Such expressions are useful since they allow the generation of estimates for the field statistics in nonlinear composites, directly from homogenization estimates for appropriately defined effective potentials. Here, use is made of these expressions in the context of the 'variational', 'tangent second-order' and 'second-order' homogenization methods, to obtain rigorous estimates for the first and second moments of the fields in nonlinear composites. While the variational estimates for these quantities are found to be identical to those proposed in previous works, the tangent second-order and second-order estimates are found be different. In particular, the new estimates for the first moments given in this work are found to be entirely consistent with the corresponding estimates for the macroscopic behaviour. Sample results for two-phase, power-law composites are provided in part II of this work.

Keywords: nonlinear homogenization; variational methods; field fluctuations

1. Introduction

The main objective of homogenization is to predict the macroscopic behaviour of composite materials in terms of the behaviour of their constituents and prescribed statistical information about their microstructure. Recent work in this area include bounds on the overall stress–strain relation for composites by Talbot & Willis (2004). However, in many circumstances, it is also of interest to estimate the statistics of the spatial distribution of the local fields within the composite. For instance, in viscoplastic composites and polycrystals undergoing finite deformations, a certain knowledge about the distribution (e.g. the phase

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averages) of the strain-rate field is necessary to be able to account for the evolution of the microstructure, which, in turn, can strongly affect the macroscopic behaviour. Also, information on the stress distribution can be useful for developing theories of damage nucleation and evolution in heterogeneous material systems. In the context of *linear* composites, there are already well-known exact formulas expressing the first and second moments of the local fields in the phases, in terms of the effective potentials (e.g. Bobeth & Diener 1986; Kreher 1990; Parton & Buryachenko 1990; Ponte Castañeda & Suquet 1998; Lipton 2005). Such formulas are useful as they allow the extraction of estimates for the statistics of the local fields from homogenization estimates for the effective potentials. In this work, we present a means for generalizing those formulas to the case of *nonlinear* composites, and we make use of them in the context of nonlinear homogenization methods based on linear comparison composites (LCCs; Ponte Castañeda 1991, 1996, 2002*a*). Some illustrative results for two-phase, power-law composites are provided in part II of this work.

2. Effective behaviour

We consider composite materials made of N different homogeneous constituents, or *phases*, which are assumed to be *randomly* distributed in a specimen occupying a volume Ω, at a length scale that is much smaller than the size of Ω and the scale of variation of the loading conditions. The constitutive behaviour of each phase is characterized by *convex* potential functions $w^{(r)}$ ($r=1, \ldots, N$), such that the stress $\boldsymbol{\sigma}$ and strain $\boldsymbol{\varepsilon}$ tensors are related by

$$\boldsymbol{\sigma} = \partial_{\boldsymbol{\varepsilon}} w(\boldsymbol{x}, \boldsymbol{\varepsilon}), \quad w(\boldsymbol{x}, \boldsymbol{\varepsilon}) = \sum_{r=1}^{N} \chi^{(r)}(\boldsymbol{x}) w^{(r)}(\boldsymbol{\varepsilon}), \tag{2.1}$$

where $\partial_{\boldsymbol{\varepsilon}}$ denotes differentiation with respect to $\boldsymbol{\varepsilon}$, and the characteristic functions $\chi^{(r)}$ serve to describe the microstructure, being 1 if the position vector $\boldsymbol{x}$ is in phase r, and 0 otherwise. This constitutive relation can be used within the context of the deformation theory of plasticity, where $\boldsymbol{\sigma}$ and $\boldsymbol{\varepsilon}$ represent the infinitesimal stress and strain, respectively. Equation (2.1) applies equally well to viscoplastic materials, in which case $\boldsymbol{\sigma}$ and $\boldsymbol{\varepsilon}$ are the Cauchy stress and Eulerian strain rate, respectively.

Let $\langle\cdot\rangle$ and $\langle\cdot\rangle^{(r)}$ denote the volume averages over the composite (Ω) and over phase r ($\Omega^{(r)}$), respectively. The effective behaviour of the composite, which is defined as the relation between the average stress $\bar{\boldsymbol{\sigma}} = \langle\boldsymbol{\sigma}\rangle$ and the average strain $\bar{\boldsymbol{\varepsilon}} = \langle\boldsymbol{\varepsilon}\rangle$, can be characterized by an *effective strain potential* $\tilde{W}$, such that

$$\bar{\boldsymbol{\sigma}} = \partial_{\bar{\boldsymbol{\varepsilon}}} \tilde{W}(\bar{\boldsymbol{\varepsilon}}), \quad \tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \inf_{\boldsymbol{\varepsilon} \in \mathcal{K}(\bar{\boldsymbol{\varepsilon}})} \langle w(\boldsymbol{x}, \boldsymbol{\varepsilon}) \rangle = \inf_{\boldsymbol{\varepsilon} \in \mathcal{K}(\bar{\boldsymbol{\varepsilon}})} \sum_{r=1}^{N} c^{(r)} \langle w^{(r)}(\boldsymbol{\varepsilon}) \rangle^{(r)}, \tag{2.2}$$

where $c^{(r)}$ denotes the volume fraction of phase r, and $\mathcal{K}(\bar{\boldsymbol{\varepsilon}}) = \{\boldsymbol{\varepsilon} |$ there is $\boldsymbol{u}$ such that $\boldsymbol{\varepsilon} = (1/2)[\nabla \boldsymbol{u} + (\nabla \boldsymbol{u})^{\mathrm{T}}]$ in Ω, $u = \bar{\boldsymbol{\varepsilon}}\boldsymbol{x}$ on $\partial\Omega\}$ is the set of kinematically admissible strain fields consistent with an average strain $\bar{\boldsymbol{\varepsilon}}$.

Alternatively, the behaviour of the phases can be characterized by *convex* stress potentials $u^{(r)}$, which are the Legendre transforms of $w^{(r)}$, in other words

$$u^{(r)}(\boldsymbol{\sigma}) = (w^{(r)})^*(\boldsymbol{\sigma}) = \sup_{\boldsymbol{\varepsilon}}[\boldsymbol{\sigma}\cdot\boldsymbol{\varepsilon} - w^{(r)}(\boldsymbol{\varepsilon})]. \tag{2.3}$$

Then, the local stress and strain are related by $\boldsymbol{\varepsilon} = \partial_{\boldsymbol{\sigma}} u^{(r)}(\boldsymbol{\sigma})$, and the effective behaviour can be described in terms of the *effective stress potential* $\tilde{U}$, such that

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

where $\mathcal{S}(\bar{\boldsymbol{\sigma}})$ is the set of divergence-free stresses such that $\bar{\boldsymbol{\sigma}} = \langle\boldsymbol{\sigma}\rangle$. The variational formulations (2.2) and (2.4) can be shown to be completely equivalent, in the sense that the functions $\tilde{W}$ and $\tilde{U}$ are Legendre duals of each other, i.e. $\tilde{U} = \tilde{W}^*$.

In §3, use will be made of the following lemma. Its proof has been given by Ponte Castañeda & Suquet (1998) (see appendix B) for a scalar parameter t, but the proof applies *mutatis mutandis* when t is a tensor. It is a simple consequence of the chain rule, plus the fact that the effective potentials $\tilde{W}_t$ and $\tilde{U}_t$ are stationary with respect to $\boldsymbol{\varepsilon}_t$ and $\boldsymbol{\sigma}_t$, respectively.

Lemma 2.1. *Consider convex local potentials w_t and u_t depending on a parameter t. Then, the corresponding effective potentials $\tilde{W}_t$ and $\tilde{U}_t$ also depend on t, and their derivatives with respect to this parameter are given by*

$$\frac{\partial\tilde{W}_t}{\partial t}(\bar{\boldsymbol{\varepsilon}}) = \left\langle\frac{\partial w_t}{\partial t}(\boldsymbol{x},\boldsymbol{\varepsilon}_t)\right\rangle, \quad \frac{\partial\tilde{U}_t}{\partial t}(\bar{\boldsymbol{\sigma}}) = \left\langle\frac{\partial u_t}{\partial t}(\boldsymbol{x},\boldsymbol{\sigma}_t)\right\rangle, \tag{2.5}$$

where the local fields $\boldsymbol{\varepsilon}_t$ and $\boldsymbol{\sigma}_t$ are the solutions to the minimization problems $(2.2)_2$ and $(2.4)_2$, respectively, with w and u given by w_t and u_t. (The derivatives are taken with $\bar{\boldsymbol{\varepsilon}}$ and $\bar{\boldsymbol{\sigma}}$ held fixed.)

3. Exact relations for the statistics of the local fields

In this section, a methodology is provided for extracting, at least theoretically, the statistics of the strain and stress fields, from the knowledge of the effective potentials of suitably perturbed problems. In general, the statistical information of interest corresponds to the first, second and higher moments of the fields in each phase. This is accomplished through the following propositions.

Proposition 3.1. *Consider a composite with local potential (2.1). The first moment, or phase average, of the strain in phase r is given by*

$$\bar{\boldsymbol{\varepsilon}}^{(r)} = \langle\boldsymbol{\varepsilon}\rangle^{(r)} = \frac{1}{c^{(r)}}\partial_{\boldsymbol{\tau}^{(r)}}\tilde{W}_{\boldsymbol{\tau}}\Big|_{\boldsymbol{\tau}^{(r)}=\mathbf{0}}, \tag{3.1}$$

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

$$w_{\tau}(\boldsymbol{x},\boldsymbol{\varepsilon}) = \sum_{s=1}^{N}\chi^{(s)}(\boldsymbol{x})w^{(s)}(\boldsymbol{\varepsilon}) + \chi^{(r)}(\boldsymbol{x})\boldsymbol{\tau}^{(r)}\cdot\boldsymbol{\varepsilon}. \tag{3.2}$$

Proof. The local potential (3.2) is convex for any value of the parameter $\boldsymbol{\tau}^{(r)}$, and so the corresponding effective potential $\tilde{W}_{\tau}$, which depends on $\boldsymbol{\tau}^{(r)}$, is well

defined. It then follows from lemma 2.1 that

$$\partial_{\boldsymbol{\tau}^{(r)}} \tilde{W}_\tau(\bar{\boldsymbol{\varepsilon}}) = \langle \partial_{\boldsymbol{\tau}^{(r)}} w_\tau(\boldsymbol{x}, \boldsymbol{\varepsilon}_\tau) \rangle = \langle \chi^{(r)}(\boldsymbol{x}) \boldsymbol{\varepsilon}_\tau \rangle = c^{(r)} \langle \boldsymbol{\varepsilon}_\tau \rangle^{(r)}, \tag{3.3}$$

where $\boldsymbol{\varepsilon}_\tau$ is the solution to the minimization problem $(2.2)_2$ with a local potential given by equation (3.2), and the subscript $\boldsymbol{\tau}$ has been used to emphasize that it depends on the parameter $\boldsymbol{\tau}^{(r)}$. In particular, for $\boldsymbol{\tau}^{(r)}=\mathbf{0}$, $\boldsymbol{\varepsilon}_\tau$ reduces to the strain field in a composite with local potential $(2.1)_2$, and so relation (3.1) follows. ■

Proposition 3.2. *Consider a composite with local potential (2.1). The even moments of order $2K$ ($K=1, 2, 3, \dots$) of the strain field in phase r are given by*

$$\langle \underbrace{\boldsymbol{\varepsilon} \otimes \boldsymbol{\varepsilon} \otimes \dots \otimes \boldsymbol{\varepsilon}}_{2K \text{ times}} \rangle^{(r)} = \frac{2K}{c^{(r)}} \partial_{\boldsymbol{t}^{(r)}} \tilde{W}_t|_{\boldsymbol{t}^{(r)}=0}, \tag{3.4}$$

where $\boldsymbol{t}^{(r)}$ is a constant, completely symmetric, positive semi-definite tensor of order $4K$ (from the space of $2K$th tensors to the space of $2K$th tensors), and $\tilde{W}_t$ is the effective strain potential of a composite with (perturbed) local potential

$$w_t(\boldsymbol{x}, \boldsymbol{\varepsilon}) = \sum_{s=1}^{N} \chi^{(s)}(\boldsymbol{x}) w^{(s)}(\boldsymbol{\varepsilon}) + \chi^{(r)}(\boldsymbol{x}) \frac{1}{2K} t^{(r)}_{ijkl\dots} \underbrace{(\varepsilon_{ij}\varepsilon_{kl}\dots)}_{2K \text{ times}}. \tag{3.5}$$

Proof. Since $\boldsymbol{t}^{(r)}$ is a positive semi-definite tensor, the local potential (3.5) is convex, and so the corresponding effective potential $\tilde{W}_t$ is well defined. Then, it follows from lemma 2.1 that

$$\begin{aligned} \partial_{\boldsymbol{t}^{(r)}} \tilde{W}_t(\bar{\boldsymbol{\varepsilon}}) = \langle \partial_{\boldsymbol{t}^{(r)}} w_t(\boldsymbol{x}, \boldsymbol{\varepsilon}_t) \rangle &= \frac{1}{2K} \langle \chi^{(r)}(\boldsymbol{x}) \underbrace{\boldsymbol{\varepsilon}_t \otimes \boldsymbol{\varepsilon}_t \otimes \dots \otimes \boldsymbol{\varepsilon}_t}_{2K \text{ times}} \rangle \\ &= \frac{c^{(r)}}{2K} \langle \underbrace{\boldsymbol{\varepsilon}_t \otimes \boldsymbol{\varepsilon}_t \otimes \dots \otimes \boldsymbol{\varepsilon}_t}_{2K \text{ times}} \rangle^{(r)}, \end{aligned} \tag{3.6}$$

where $\boldsymbol{\varepsilon}_t$ is the solution to the minimization problem $(2.2)_2$ with a local potential given by equation (3.5). For $\boldsymbol{t}^{(r)}=\mathbf{0}$, $\boldsymbol{\varepsilon}_t$ reduces to the strain field in a composite with local potential $(2.1)_2$, and so relation (3.4) follows. ■

Thus, we have obtained identities expressing the phase averages and even moments of the strain field in terms of suitably perturbed effective potentials. It is noted that analogous relations for the odd moments (higher than one) of the strain could be obtained in a similar manner, but this is mathematically more involved, since the perturbed potentials would be non-convex and unbounded from below.

In §4, the focus will be on moments up to second order, and it is then useful to consider the following corollary of proposition 3.2 ($K=1$).

Corollary 3.3. *Consider a composite with local potential (2.1). The second moments of the strain in phase r are given by*

$$\langle \boldsymbol{\varepsilon} \otimes \boldsymbol{\varepsilon} \rangle^{(r)} = \frac{2}{c^{(r)}} \partial_{\boldsymbol{\lambda}^{(r)}} \tilde{W}_\lambda|_{\boldsymbol{\lambda}^{(r)}=\mathbf{0}}, \tag{3.7}$$

where $\boldsymbol{\lambda}^{(r)}$ is a constant, symmetric, positive semi-definite, fourth-order tensor, and $\tilde{W}_\lambda$ denotes the effective potential of a composite with (perturbed) local potential

$$w_\lambda(\boldsymbol{\varepsilon}) = \sum_{s=1}^{N} \chi^{(s)}(\boldsymbol{x}) w^{(s)}(\boldsymbol{\varepsilon}) + \chi^{(r)}(\boldsymbol{x}) \frac{1}{2} \boldsymbol{\varepsilon} \cdot \boldsymbol{\lambda}^{(r)} \boldsymbol{\varepsilon}. \tag{3.8}$$

It is noted that the phase covariance tensors $\mathsf{C}_{\boldsymbol{\varepsilon}}^{(r)}$, which provide a measure of the intraphase field fluctuations, can be written in terms of (3.1) and (3.7),

$$\mathsf{C}_{\boldsymbol{\varepsilon}}^{(r)} \doteq \langle (\boldsymbol{\varepsilon} - \bar{\boldsymbol{\varepsilon}}^{(r)}) \otimes (\boldsymbol{\varepsilon} - \bar{\boldsymbol{\varepsilon}}^{(r)}) \rangle^{(r)} = \langle \boldsymbol{\varepsilon} \otimes \boldsymbol{\varepsilon} \rangle^{(r)} - \bar{\boldsymbol{\varepsilon}}^{(r)} \otimes \bar{\boldsymbol{\varepsilon}}^{(r)}. \tag{3.9}$$

When the potentials $w^{(r)}$ are quadratic, expressions (3.1) and (3.7) reduce to the well-known formulas for linear composites (Kreher 1990; Parton & Buryachenko 1990; Ponte Castañeda & Suquet 1998). Of course, it is possible to obtain, by completely analogous means, corresponding expressions for the statistics of the stress field $\boldsymbol{\sigma}$ in terms of suitably perturbed *stress* potentials. For brevity, such analogous expressions will not be spelled out here. Instead, it is shown next how the statistics of the stress field may be obtained in terms of suitably perturbed *strain* potentials, which may be more useful in some cases.

Proposition 3.4. *Consider a composite with local potential (2.1). The first moment, or phase average, of the stress in phase r is given by*

$$\bar{\boldsymbol{\sigma}}^{(r)} = \langle \boldsymbol{\sigma} \rangle^{(r)} = -\frac{1}{c^{(r)}} \partial_{\boldsymbol{\eta}^{(r)}} \tilde{W}_\eta|_{\boldsymbol{\eta}^{(r)} = \mathbf{0}}, \tag{3.10}$$

where $\boldsymbol{\eta}^{(r)}$ is a constant, symmetric, second-order tensor, and $\tilde{W}_\eta$ denotes the effective potential of a composite with (perturbed) local potential w_η given by the Legendre transform of u_η, $w_\eta = u_\eta^*$, where

$$u_\eta(\boldsymbol{x}, \boldsymbol{\sigma}) = \sum_{s=1}^{N} \chi^{(s)}(\boldsymbol{x}) u^{(s)}(\boldsymbol{\sigma}) + \chi^{(r)}(\boldsymbol{x}) \boldsymbol{\eta}^{(r)} \cdot \boldsymbol{\sigma}. \tag{3.11}$$

Remark 3.5. It is noted (van Tiel 1984) that w_η can be written in terms of the (unperturbed) potentials $w^{(r)}$ as

$$w_\eta(\boldsymbol{x}, \boldsymbol{\varepsilon}) = \sum_{\substack{s=1 \\ s \neq r}}^{N} \chi^{(s)}(\boldsymbol{x}) w^{(s)}(\boldsymbol{\varepsilon}) + \chi^{(r)}(\boldsymbol{x}) w^{(r)}(\boldsymbol{\varepsilon} - \boldsymbol{\eta}^{(r)}). \tag{3.12}$$

Proof. The local potential (3.12) is convex for any value of the parameter $\boldsymbol{\eta}^{(r)}$, and so the corresponding effective potential $\tilde{W}_\eta$, which depends on $\boldsymbol{\eta}^{(r)}$, is well defined. Then, it follows from lemma 2.1 that

$$\begin{aligned} \partial_{\boldsymbol{\eta}^{(r)}} \tilde{W}_\eta(\bar{\boldsymbol{\varepsilon}}) &= \langle \partial_{\boldsymbol{\eta}^{(r)}} w_\eta(\boldsymbol{x}, \boldsymbol{\varepsilon}_\eta) \rangle \\ &= -\langle \chi^{(r)}(\boldsymbol{x}) \partial_{\boldsymbol{\varepsilon}} w^{(r)}(\boldsymbol{\varepsilon}_\eta - \boldsymbol{\eta}^{(r)}) \rangle = -c^{(r)} \langle \boldsymbol{\sigma}_\eta \rangle^{(r)}, \end{aligned} \tag{3.13}$$

where $\boldsymbol{\varepsilon}_\eta$ is the solution to the minimization problem $(2.2)_2$ with a local potential given by (3.12), and $\boldsymbol{\sigma}_\eta = \partial_{\boldsymbol{\varepsilon}} w^{(r)}(\boldsymbol{\varepsilon}_\eta - \boldsymbol{\eta}^{(r)})$ is the corresponding stress. Relation (3.13) is valid for any value of $\boldsymbol{\eta}^{(r)}$. In particular, for $\boldsymbol{\eta}^{(r)} = 0$, $\boldsymbol{\varepsilon}_\eta$ and $\boldsymbol{\sigma}_\eta$ reduce to the strain and stress fields in a composite with local potential $(2.1)_2$, and so relation (3.10) follows. ∎

Proposition 3.6. *Consider a composite with local potential (2.1). The even moments of order $2K$ ($K=1, 2, 3, \dots$) of the stress field in phase r are given by*

$$\langle \underbrace{\boldsymbol{\sigma}\otimes\boldsymbol{\sigma}\otimes\dots\otimes\boldsymbol{\sigma}}_{2K \text{ times}} \rangle^{(r)} = -\frac{2K}{c^{(r)}}\, \partial_{\boldsymbol{t}^{(r)}}\, \tilde{W}_t \Big|_{\boldsymbol{t}^{(r)}=\boldsymbol{0}}, \tag{3.14}$$

where $\boldsymbol{t}^{(r)}$ is a constant, completely symmetric, positive semi-definite tensor of order $4K$, and $\tilde{W}_t$ is the effective strain potential of a composite with a local potential w_t given by the Legendre transform of u_t, $w_t = u_t^*$, where

$$u_t(\boldsymbol{x},\boldsymbol{\sigma}) = \sum_{s=1}^{N} \chi^{(s)}(\boldsymbol{x}) u^{(s)}(\boldsymbol{\sigma}) + \chi^{(r)}(\boldsymbol{x}) \frac{1}{2K}\, t^{(r)}_{ijkl\dots} (\underbrace{\sigma_{ij}\sigma_{kl}\dots}_{K \text{ times}}). \tag{3.15}$$

Proof. Let $\tilde{U}_t$ denote the effective stress potential of a composite with local potential (3.15). Since $\boldsymbol{t}^{(r)}$ is positive semi-definite, the potential (3.15) is convex, and so $\tilde{U}_t$ is well defined. Let the strain potential w_t be the Legendre dual of (3.15), i.e. $w_t = u_t^*$, and let $\tilde{W}_t$ be the corresponding effective strain potential. Then, $\tilde{W}_t = \tilde{U}_t^*$, or

$$\tilde{W}_t(\bar{\boldsymbol{\varepsilon}}) = \sup_{\bar{\boldsymbol{\sigma}}} [\bar{\boldsymbol{\sigma}}\cdot\bar{\boldsymbol{\varepsilon}} - \tilde{U}_t(\bar{\boldsymbol{\sigma}})]. \tag{3.16}$$

Assuming the supremum over $\bar{\boldsymbol{\sigma}}$ in (3.16) is attained at a stationary point, and differentiating with respect to $\boldsymbol{t}^{(r)}$, we obtain

$$\partial_{\boldsymbol{t}^{(r)}}\, \tilde{W}_t|_{\boldsymbol{t}(r)=\boldsymbol{0}} = -\partial_{\boldsymbol{t}^{(r)}}\, \tilde{U}_t \Big|_{\boldsymbol{t}^{(r)}=\boldsymbol{0}} = -\frac{c^{(r)}}{2K} \langle \underbrace{\boldsymbol{\sigma}\otimes\boldsymbol{\sigma}\otimes\dots\otimes\boldsymbol{\sigma}}_{2K \text{ times}} \rangle^{(r)}, \tag{3.17}$$

where the last identity follows from the dual version of proposition 3.2. Relation (3.14) follows immediately. ∎

In §4, use is made of the following corollary of proposition 3.6 ($K=1$), for the second moments of the stress.

Corollary 3.7. *Consider a composite with local potential (2.1). The second moments of the stress in phase r are given by*

$$\langle \boldsymbol{\sigma}\otimes\boldsymbol{\sigma} \rangle^{(r)} = -\frac{2}{c^{(r)}}\, \partial_{\boldsymbol{\mu}^{(r)}}\, \tilde{W}_\mu \Big|_{\boldsymbol{\mu}^{(r)}=\boldsymbol{0}}, \tag{3.18}$$

where $\boldsymbol{\mu}^{(r)}$ is a constant, symmetric, positive semi-definite, fourth-order tensor, and $\tilde{W}_\mu$ denotes the effective potential of a composite with (perturbed) local potential w_μ given by the Legendre transform of u_μ, $w_\mu = u_\mu^*$, where

$$u_\mu(\boldsymbol{\sigma}) = \sum_{s=1}^{N} \chi^{(s)}(\boldsymbol{x}) u^{(s)}(\boldsymbol{\sigma}) + \chi^{(r)}(\boldsymbol{x}) \frac{1}{2}\, \boldsymbol{\sigma}\cdot\boldsymbol{\mu}^{(r)}\boldsymbol{\sigma}. \tag{3.19}$$

It is straightforward to verify that relations (3.1) and (3.10) for the phase averages are consistent with the macroscopic averages $(2.2)_1$ and $(2.4)_1$, so that

$$\bar{\boldsymbol{\varepsilon}} = \sum_{r=1}^{N} c^{(r)} \bar{\boldsymbol{\varepsilon}}^{(r)}, \quad \bar{\boldsymbol{\sigma}} = \sum_{r=1}^{N} c^{(r)} \bar{\boldsymbol{\sigma}}^{(r)}. \tag{3.20}$$

Once again, exactly analogous arguments can be used to derive expressions relating the statistics of the local strain field and the effective stress potential $\widetilde{U}$,

but are not given here for brevity. In §4, we make use of these identities in the context of nonlinear homogenization methods based on LCCs, to obtain estimates for the first and second moments of the strain and stress fields.

4. Homogenization estimates based on linear composites

The relations derived in §3 allow us to extract statistics of the local fields from perturbed effective potentials. In general, these potentials are very difficult to compute exactly, and so we need to resort to approximate homogenization methods to estimate them. A fairly general class of nonlinear homogenization methods has been introduced by Ponte Castañeda (1991, 1996, 2002*a*). These methods make use of a LCC with the same microstructure as the nonlinear composite, but with phase potentials that correspond to appropriate linearizations of the nonlinear ones, as determined by suitably designed variational principles. Use can then be made of the various estimates available for linear composites to estimate the effective behaviour of the LCC, to generate corresponding estimates for the effective behaviour of the nonlinear composites.

These LCC-based homogenization methods deliver estimates that are rigorous *only* for the effective potentials of nonlinear composites. However, in the context of the so-called 'variational' method (Ponte Castañeda 1991), Ponte Castañeda & Zaidman (1994) conjectured that the first moment of the local field in the LCC constitutes a reasonable approximation for the corresponding first moment in the nonlinear composite. Later, Kailasam & Ponte Castañeda (1998) demonstrated that this approximation was indeed consistent with the *exact* version of the variational method (Ponte Castañeda 1992). The conjecture was also used in the context of the 'tangent second-order' (Ponte Castañeda 1996) and 'second-order' methods (Ponte Castañeda 2002*a*), as well as extended to consider higher-order moments. The relations derived in §4 make it possible to assess the validity of these approximations. In this section, use is made of those relations to obtain rigorous estimates for the first and second moments of the nonlinear fields in terms of the corresponding quantities in the LCC, for the variational, tangent second-order and second-order methods.

In the following, for simplicity, we restrict the analysis to composites made of *isotropic* phases, characterized by potentials of the form

$$w^{(r)}(\boldsymbol{\varepsilon}) = \frac{9}{2}\kappa^{(r)}\varepsilon_{\rm m}^2 + \phi^{(r)}(\varepsilon_{\rm e}), \quad u^{(r)}(\boldsymbol{\sigma}) = \frac{1}{2\kappa^{(r)}}\sigma_{\rm m}^2 + \psi^{(r)}(\sigma_{\rm e}), \tag{4.1}$$

where $\varepsilon_{\rm m}$ and $\sigma_{\rm m}$ denote the hydrostatic components of the strain and stress tensors, and the von Mises equivalent strain and stress are given in terms of the deviatoric strain and stress tensors by $\varepsilon_{\rm e} = \sqrt{(2/3)\boldsymbol{\varepsilon}_{\rm d}\cdot\boldsymbol{\varepsilon}_{\rm d}}$ and $\sigma_{\rm e} = \sqrt{(3/2)\boldsymbol{\sigma}_{\rm d}\cdot\boldsymbol{\sigma}_{\rm d}}$.

(*a*) *'Variational' estimates*

The variational method is based on the identity (Ponte Castañeda 1992)

$$w^{(r)}(\boldsymbol{\varepsilon}) = \inf_{\mu_0^{(r)}>0}\left\{ w_L^{(r)}(\boldsymbol{\varepsilon};\mu_0^{(r)}) + V^{(r)}(\mu_0^{(r)})\right\}, \tag{4.2}$$

which assumes that the isotropic potentials $(4.1)_1$ are concave in $\boldsymbol{\varepsilon}_{\mathrm{e}}^2$. Also in this relation, $w_L^{(r)}$ denotes the phase potential of an isotropic linear material given by

$$w_L^{(r)}(\boldsymbol{\varepsilon};\mu_0^{(r)}) = \frac{1}{2}\boldsymbol{\varepsilon}\cdot\mathsf{L}_0^{(r)}\boldsymbol{\varepsilon}, \quad \mathsf{L}_0^{(r)} = 3\kappa^{(r)}\mathsf{J} + 2\mu_0^{(r)}\mathsf{K}, \tag{4.3}$$

and the function $V^{(r)}$ is defined by

$$V^{(r)}(\mu_0^{(r)}) = \sup_{\boldsymbol{\varepsilon}^{(r)}} \left\{ w^{(r)}(\boldsymbol{\varepsilon}^{(r)}) - w_L^{(r)}(\boldsymbol{\varepsilon}^{(r)};\mu_0^{(r)}) \right\}. \tag{4.4}$$

In expression (4.3), J and K denote the standard fourth-order, isotropic, hydrostatic and shear projection tensors, and $\kappa^{(r)}$ are the same as those appearing in (4.1). The optimality condition in (4.4) is given by the 'secant' condition

$$\partial_{\boldsymbol{\varepsilon}} w^{(r)}(\hat{\boldsymbol{\varepsilon}}^{(r)}) = \mathsf{L}_0^{(r)}\hat{\boldsymbol{\varepsilon}}^{(r)}, \tag{4.5}$$

where $\hat{\boldsymbol{\varepsilon}}^{(r)}$ denotes the optimal value of $\boldsymbol{\varepsilon}^{(r)}$ in (4.4). Then, an estimate for the effective potential $\tilde{W}$ may be obtained by introducing (4.2) into (2.2), interchanging the optimization conditions over $\boldsymbol{\varepsilon}(\boldsymbol{x})$ and $\mu_0^{(r)}$, and restricting the latter to be constant per phase. The estimate is a rigorous upper bound for $\tilde{W}$,

$$\tilde{W}(\bar{\boldsymbol{\varepsilon}}) \leq \inf_{\mu_0^{(s)}>0} \left\{ \tilde{W}_L(\bar{\boldsymbol{\varepsilon}};\mu_0^{(s)}) + \sum_{r=1}^{N} c^{(r)}\, V^{(r)}(\mu_0^{(r)}) \right\}, \tag{4.6}$$

where $\tilde{W}_L = (1/2)\bar{\boldsymbol{\varepsilon}}\cdot\tilde{\mathsf{L}}_0\bar{\boldsymbol{\varepsilon}}$ is the effective strain potential of an LCC with phase potentials (4.3). Thus, a linear homogenization estimate is required for the effective elastic tensor $\tilde{\mathsf{L}}_0$ of the LCC to compute $\tilde{W}_L$. Then, the optimality conditions in (4.6) generate a system of algebraic nonlinear equations for the optimal values $\hat{\mu}_0^{(r)}$ of the variables $\mu_0^{(r)}$, which can be written as

$$\frac{1}{2c^{(r)}}\bar{\boldsymbol{\varepsilon}}\cdot\frac{\partial\tilde{\mathsf{L}}_0}{\partial\mu_0^{(r)}}\left(\hat{\mu}_0^{(s)}\right)\bar{\boldsymbol{\varepsilon}} + \frac{\partial V^{(r)}}{\partial\mu_0^{(r)}}\left(\hat{\mu}_0^{(r)}\right) = 0. \tag{4.7}$$

The variational estimate for the effective behaviour of a nonlinear composite is obtained by differentiation of (4.6) with respect to $\bar{\boldsymbol{\varepsilon}}$. This is made more explicit in the following result, due to deBotton & Ponte Castañeda (1993).

Result 4.1. *Since the estimate (4.6) is stationary with respect to the variables* $\mu_0^{(r)}$, *it follows that the variational estimate for the effective behaviour is given by*

$$\bar{\boldsymbol{\sigma}} = \partial_{\bar{\boldsymbol{\varepsilon}}}\tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \partial_{\bar{\boldsymbol{\varepsilon}}}\tilde{W}_L\left(\bar{\boldsymbol{\varepsilon}};\hat{\mu}_0^{(s)}\right) = \tilde{\mathsf{L}}_0\left(\hat{\mu}_0^{(s)}\right)\bar{\boldsymbol{\varepsilon}} = \bar{\boldsymbol{\sigma}}_L, \tag{4.8}$$

where $\tilde{\mathsf{L}}_0$ *is evaluated at the optimal values* $\hat{\mu}_0^{(r)}$ *from equation (4.7), and the notation* $\bar{\boldsymbol{\sigma}}_L$ *has been used to emphasize that it corresponds to the macroscopic stress in the LCC.*

Thus, the variational estimate for the macroscopic stress in the nonlinear composite *coincides* with that in the LCC evaluated at the $\hat{\mu}_0^{(r)}$. It is emphasized, however, that the stress–strain relation (4.8) is nonlinear, as it should be, since the moduli $\hat{\mu}_0^{(r)}$, and therefore $\tilde{\mathsf{L}}_0$, depend on $\bar{\boldsymbol{\varepsilon}}$. Suquet (1995) remarked that the

first term in expression (4.7) is nothing more than the second moment of the equivalent strain $\langle \varepsilon_{\rm e}^2 \rangle^{(r)}$ over phase r in the LCC, while it can be deduced from (4.4) that the second term is precisely $(\hat{\varepsilon}_{\rm e}^{(r)})^2$ in the secant condition (4.5), and so it follows that $(\hat{\varepsilon}_{\rm e}^{(r)})^2 = \langle \varepsilon_{\rm e}^2 \rangle^{(r)}$, and that the variables $\hat{\boldsymbol{\mu}}_0^{(s)}$ in the effective stress–strain relation (4.8) for the nonlinear composite can be given the interpretation of secant moduli evaluated at the second moments of the strain field. This provides a 'modified secant' interpretation of the variational estimate (4.8) for the effective stress–strain relation of the nonlinear composite. It also follows that the estimate (4.6) for the effective potential can be written as

$$\tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \sum_{r=1}^{N} c^{(r)} \boldsymbol{\phi}^{(r)}(\hat{\boldsymbol{\varepsilon}}^{(r)}). \tag{4.9}$$

In order to obtain corresponding variational estimates for the phase averages of the strain, we consider a composite with (perturbed) phase potentials $w_{\boldsymbol{\tau}}^{(r)}$ given by (3.2), where $w^{(r)}$ is given by $(4.1)_1$, and we evaluate the derivative (3.1) with $\tilde{W}_{\boldsymbol{\tau}}$ given by the variational procedure described above. Similarly, variational estimates for the second moments of the strain, as well as the first and second moments of the stress, can be obtained by considering (perturbed) potentials (3.8), (3.12) and (3.19), and differentiating the variational estimates for the corresponding effective potentials with respect to the perturbation parameters. These estimates are spelled out in the following result.

Result 4.2. *The variational estimates for the first and second moments of the local fields are given by*

$$\bar{\boldsymbol{\varepsilon}}^{(r)} = \bar{\boldsymbol{\varepsilon}}_L^{(r)}, \quad \langle \boldsymbol{\varepsilon} \otimes \boldsymbol{\varepsilon} \rangle^{(r)} = \langle \boldsymbol{\varepsilon}_L \otimes \boldsymbol{\varepsilon}_L \rangle^{(r)}, \tag{4.10}$$

$$\bar{\boldsymbol{\sigma}}^{(r)} = \bar{\boldsymbol{\sigma}}_L^{(r)}, \quad \langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)} = \langle \boldsymbol{\sigma}_L \otimes \boldsymbol{\sigma}_L \rangle^{(r)}. \tag{4.11}$$

where again, the subscript L has been used to denote quantities in the LCC associated with the variational estimate (4.6)*.*

Proof. We begin by proving the identity $(4.10)_1$ for the phase averages of the strain. In order to make use of proposition 3.1, we consider a composite with perturbed local potential (3.2), where the unperturbed phase potentials $w^{(s)}$ are given by $(4.1)_1$. Thus, phase r in this composite is characterized by

$$w_{\boldsymbol{\tau}}^{(r)}(\boldsymbol{\varepsilon}) = w^{(r)}(\boldsymbol{\varepsilon}) + \boldsymbol{\tau}^{(r)} \cdot \boldsymbol{\varepsilon}. \tag{4.12}$$

Making use of the identity (4.2) for $w^{(r)}$, this potential can be written as

$$w_{\boldsymbol{\tau}}^{(r)}(\boldsymbol{\varepsilon}) = \inf_{\boldsymbol{\mu}_0^{(r)} > 0} \Big\{ w_{L\boldsymbol{\tau}}^{(r)}(\boldsymbol{\varepsilon}; \boldsymbol{\mu}_0^{(r)}) + V^{(r)}(\boldsymbol{\mu}_0^{(r)}) \Big\}, \tag{4.13}$$

where $w_{L\boldsymbol{\tau}}^{(r)}$ denotes the phase potential of a perturbed (anisotropic) LCC, given in terms of (4.3) by

$$w_{L\boldsymbol{\tau}}^{(r)}(\boldsymbol{\varepsilon}; \boldsymbol{\mu}_0^{(r)}) = w_L^{(r)}(\boldsymbol{\varepsilon}; \boldsymbol{\mu}_0^{(r)}) + \boldsymbol{\tau}^{(r)} \cdot \boldsymbol{\varepsilon}. \tag{4.14}$$

The variational estimate for the perturbed effective potential $\tilde{W}_{\boldsymbol{\tau}}$ is then obtained by following the procedure described in the context of expression (4.6).

The resulting expression for $\tilde{W}_{\boldsymbol{\tau}}$ is in fact (4.6), but with $\tilde{W}_L$ replaced by $\tilde{W}_{L\boldsymbol{\tau}}$, the effective potential of the perturbed LCC with phase r characterized by (4.14). Then, recalling that the variational estimate for $\tilde{W}_{\boldsymbol{\tau}}$ is stationary with respect to the variables $\boldsymbol{\mu}_0^{(r)}$, and that the functions $V^{(r)}$ do not depend explicitly on the parameters $\boldsymbol{\tau}^{(r)}$, it follows from proposition 3.1 that the variational estimate for the average of the strain in phase r is given by

$$\bar{\boldsymbol{\varepsilon}}^{(r)} = \frac{1}{c^{(r)}} \partial_{\boldsymbol{\tau}^{(r)}} \tilde{W}_{\boldsymbol{\tau}} \Big|_{\boldsymbol{\tau}^{(r)}=\mathbf{0}} = \frac{1}{c^{(r)}} \partial_{\boldsymbol{\tau}^{(r)}} \tilde{W}_{L\boldsymbol{\tau}} \Big|_{\boldsymbol{\tau}^{(r)}=\mathbf{0}} = \bar{\boldsymbol{\varepsilon}}_L^{(r)}, \tag{4.15}$$

where $\bar{\boldsymbol{\varepsilon}}_L^{(r)}$ denotes the average strain in phase r in the LCC associated with the estimate (4.6) for the unperturbed effective potential $\tilde{W}$, and the last identity follows also from proposition 3.1.

The remaining estimates in result 4.2 can be derived in a completely analogous fashion, by making use of proposition 3.4 and corollaries 3.3 and 3.7, and identities for the relevant perturbed phase potentials analogous to (4.13) with perturbed (anisotropic) LCC phase potentials given in terms of (4.3) by

$$w_{L\lambda}^{(r)}(\boldsymbol{\varepsilon}; \boldsymbol{\mu}_0^{(r)}) = w_L^{(r)}(\boldsymbol{\varepsilon}; \boldsymbol{\mu}_0^{(r)}) + \frac{1}{2} \boldsymbol{\varepsilon} \cdot \boldsymbol{\lambda}^{(r)} \boldsymbol{\varepsilon}, \tag{4.16}$$

$$w_{L\eta}^{(r)}\Big(\boldsymbol{\varepsilon}; \boldsymbol{\mu}_0^{(r)}\Big) = w_L^{(r)}(\boldsymbol{\varepsilon} - \boldsymbol{\eta}^{(r)}; \boldsymbol{\mu}_0^{(r)}), \tag{4.17}$$

$$w_{L\mu}^{(r)}(\boldsymbol{\varepsilon}; \boldsymbol{\mu}_0^{(r)}) = \inf_{\boldsymbol{\varepsilon}_1 + \boldsymbol{\varepsilon}_2 = \boldsymbol{\varepsilon}} \left[w_L^{(r)}(\boldsymbol{\varepsilon}_1; \boldsymbol{\mu}_0^{(r)}) + \frac{1}{2} \boldsymbol{\varepsilon}_2 \cdot (\boldsymbol{\mu}^{(r)})^{-1} \boldsymbol{\varepsilon}_2 \right]. \tag{4.18}$$

■

Thus, the variational estimates for the first and second moments of the local fields *coincide* with those in the LCC. This result is in exact agreement with the conjecture of Ponte Castañeda & Zaidman (1994). It is also worth noting that the nonlinear estimates for the phase averages are consistent with the corresponding estimates for the macroscopic behaviour (4.8), in the sense that they satisfy relation (3.20). (This is so provided the linear estimates used in the context of the LCC are themselves consistent.) It is useful to recall here that the phase averages and second moments of the strain and stress fields in the LCC can be computed from (e.g. Willis 1981, Parton & Buryachenko 1990)

$$\bar{\boldsymbol{\varepsilon}}_L^{(r)} = \mathsf{A}_0^{(r)} \bar{\boldsymbol{\varepsilon}}, \qquad \langle \boldsymbol{\varepsilon}_L \otimes \boldsymbol{\varepsilon}_L \rangle^{(r)} = \frac{2}{c^{(r)}} \partial_{\mathsf{L}_0^{(r)}} \tilde{W}_L, \tag{4.19}$$

$$\bar{\boldsymbol{\sigma}}_L^{(r)} = \mathsf{L}_0^{(r)} \bar{\boldsymbol{\varepsilon}}_L^{(r)}, \quad \langle \boldsymbol{\sigma}_L \otimes \boldsymbol{\sigma}_L \rangle^{(r)} = \mathsf{L}_0^{(r)} \langle \boldsymbol{\varepsilon}_L \otimes \boldsymbol{\varepsilon}_L \rangle^{(r)} \mathsf{L}_0^{(r)}, \tag{4.20}$$

where the $\mathsf{A}_0^{(r)}$ are the strain concentration tensors, which are related to the effective modulus tensor by $\tilde{\mathsf{L}}_0 = \sum_{r=1}^{N} c^{(r)} \mathsf{L}_0^{(r)} \mathsf{A}_0^{(r)}$, and which depend on the linear homogenization method utilized.

'Variational' estimates for the dual potential $\tilde{U}$ follow from exactly analogous expressions in terms of stress potentials $u^{(r)}$ and $u_L^{(r)}$. Such estimates can be shown to be exactly equivalent to the variational estimates (4.6) for $\tilde{W}$, in the

sense that they are Legendre duals of each other. In addition, the LCCs associated with each of these estimates are also equivalent to each other, i.e. $u_L^{(r)} = (w_L^{(r)})^*$. In other words, the variational estimates exhibit no *duality gap*. Following exactly similar arguments, it can be shown that the field statistics arising from the variational estimates for the stress potential $\tilde{U}$ coincide with those in the associated LCC. Thus, the identities (4.10) and (4.11) also hold for the dual version of the method, and are, of course, entirely consistent with those resulting from the potential $\tilde{W}$.

Finally, it is noted that a similar procedure can be used with proposition 3.2 to generalize the above results for higher-order moments.

Remark 4.3. The variational estimates for the $2K$ moments of the local fields in the nonlinear composite are given by

$$\langle \underbrace{\boldsymbol{\varepsilon}\otimes\boldsymbol{\varepsilon}\otimes\ldots\otimes\boldsymbol{\varepsilon}}_{2K \text{ times}} \rangle^{(r)} = \langle \underbrace{\boldsymbol{\varepsilon}_L\otimes\boldsymbol{\varepsilon}_L\otimes\ldots\otimes\boldsymbol{\varepsilon}_L}_{2K \text{ times}} \rangle^{(r)}, \tag{4.21}$$

$$\langle \underbrace{\boldsymbol{\sigma}\otimes\boldsymbol{\sigma}\otimes\ldots\otimes\boldsymbol{\sigma}}_{2K \text{ times}} \rangle^{(r)} = \langle \underbrace{\boldsymbol{\sigma}_L\otimes\boldsymbol{\sigma}_L\otimes\ldots\otimes\boldsymbol{\sigma}_L}_{2K \text{ times}} \rangle^{(r)}. \tag{4.22}$$

Unfortunately, these results are not very useful, because there are no simple formulas to extract the moments of the order higher than 2 in linear composites. This suggests making use of the variational method itself to estimate these higher order moments, which can be shown to yield estimates for the higher-order moments of the nonlinear composite depending only on the second-order moments of the field in the LCC. While such estimates would be easily computed, it is unlikely that they would be very accurate.

(*b*) *'Tangent second-order' estimates*

The variational method considered in §4*b* delivers estimates for the effective potentials that are exact only to first order in the heterogeneity contrast. In this section we consider the so-called tangent second-order method introduced by Ponte Castañeda (1996), which delivers estimates for the effective potentials that are exact to second order in the heterogeneity contrast, and are therefore expected to be more accurate in general. In this case, the following identity (Ponte Castañeda & Willis 1999) is used for the phase potentials $w^{(r)}$:

$$w^{(r)}(\boldsymbol{\varepsilon}) = \underset{\check{\boldsymbol{\varepsilon}}^{(r)}}{\operatorname{stat}} \left\{ w_L^{(r)}\left(\boldsymbol{\varepsilon}; \check{\boldsymbol{\varepsilon}}^{(r)}, \mathsf{L}_0^{(r)}\right) \right\}, \tag{4.23}$$

where the *stationary* operation consists in setting the partial derivative of the argument with respect to the variable equal to zero, and $w_L^{(r)}$ is the potential of a linear *thermoelastic* comparison composite defined in terms of a reference strain tensor $\check{\boldsymbol{\varepsilon}}^{(r)}$ and a tensor of moduli $\mathsf{L}_0^{(r)}$ by

$$w_L^{(r)}(\boldsymbol{\varepsilon}; \check{\boldsymbol{\varepsilon}}^{(r)}, \mathsf{L}_0^{(r)}) = w^{(r)}(\check{\boldsymbol{\varepsilon}}^{(r)}) + \partial_{\boldsymbol{\varepsilon}} w^{(r)}(\check{\boldsymbol{\varepsilon}}^{(r)}) \cdot (\boldsymbol{\varepsilon} - \check{\boldsymbol{\varepsilon}}^{(r)}) + \frac{1}{2}(\boldsymbol{\varepsilon} - \check{\boldsymbol{\varepsilon}}^{(r)}) \cdot \mathsf{L}_0^{(r)} (\boldsymbol{\varepsilon} - \check{\boldsymbol{\varepsilon}}^{(r)}). \tag{4.24}$$

Note that the identity (4.23) is valid for any $\mathsf{L}_0^{(r)}$. The tangent second-order estimates for the effective potential $\tilde{W}$ are then obtained by introducing (4.23) into (2.2), interchanging the optimization operations over $\boldsymbol{\varepsilon}(\boldsymbol{x})$ and $\check{\boldsymbol{\varepsilon}}^{(r)}$, and restricting the latter to be constant per phase. The result of this calculation is the approximation

$$\tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \underset{\check{\boldsymbol{\varepsilon}}^{(s)}}{\operatorname{stat}} \Big\{ \tilde{W}_L(\bar{\boldsymbol{\varepsilon}}; \check{\boldsymbol{\varepsilon}}^{(s)}, \mathsf{L}_0^{(s)}) \Big\}, \tag{4.25}$$

where $\tilde{W}_L$ is the effective strain potential of the thermoelastic LCC with phase potentials (4.24). The stationary operation in (4.25) leads to the conditions

$$\check{\boldsymbol{\varepsilon}}^{(r)} = \bar{\boldsymbol{\varepsilon}}_L^{(r)}, \tag{4.26}$$

where the $\bar{\boldsymbol{\varepsilon}}_L^{(r)}$ denote the averages of the strain in phase r of the associated LCC, which depends on the $\check{\boldsymbol{\varepsilon}}^{(s)}$ and $\mathsf{L}_0^{(s)}$ according to the homogenization procedure utilized. Given this choice for the variables $\check{\boldsymbol{\varepsilon}}^{(r)}$, it is not possible to make the resulting estimate stationary with respect to the tensor $\mathsf{L}_0^{(r)}$. For this reason, the following physically motivated choice was proposed (Ponte Castañeda 1996) for these tensors:

$$\mathsf{L}_0^{(r)} = \mathsf{L}_t^{(r)}(\check{\boldsymbol{\varepsilon}}^{(r)}) \doteq \partial^2_{\boldsymbol{\varepsilon}\boldsymbol{\varepsilon}} w^{(r)}(\check{\boldsymbol{\varepsilon}}^{(r)}). \tag{4.27}$$

The relations (4.26) and (4.27) thus serve to specify the variables $\check{\boldsymbol{\varepsilon}}^{(r)}$ and $\mathsf{L}_0^{(r)}$ defining the phase potentials (4.24) of the LCC in terms of the phase averages of the strain field in the phases of the LCC, itself.

The stationarity condition (4.26) for the reference strains $\check{\boldsymbol{\varepsilon}}^{(r)}$ can be used to simplify the expression for the effective potential (4.25) to

$$\tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \sum_{r=1}^{N} c^{(r)} \left[w^{(r)}(\bar{\boldsymbol{\varepsilon}}_L^{(r)}) + \frac{1}{2} \partial_{\boldsymbol{\varepsilon}} w^{(r)} \left(\bar{\boldsymbol{\varepsilon}}_L^{(r)} \right) \cdot \left(\bar{\boldsymbol{\varepsilon}} - \bar{\boldsymbol{\varepsilon}}_L^{(r)} \right) \right]. \tag{4.28}$$

On the other hand, because of the lack of stationarity of the estimates (4.25) with respect to the moduli tensors $\mathsf{L}_0^{(r)}$, the corresponding estimates for the effective stress–strain relation are given (Ponte Castañeda & Suquet 1998) by those in the LCC, plus some 'correction' terms, as described next.

Result 4.4. *The tangent second-order estimate for the effective stress–strain relation is given by*

$$\bar{\boldsymbol{\sigma}} = \partial_{\bar{\boldsymbol{\varepsilon}}} \tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \bar{\boldsymbol{\sigma}}_L + \sum_{r=1}^{N} c^{(r)} \boldsymbol{\rho}^{(r)} \cdot \partial_{\bar{\boldsymbol{\varepsilon}}} \bar{\boldsymbol{\varepsilon}}_L^{(r)}, \tag{4.29}$$

where $\bar{\boldsymbol{\sigma}}_L$ denotes the macroscopic stress in the LCC, and the tensors $\boldsymbol{\rho}^{(r)}$ are defined in terms of the phase covariance tensors $\mathsf{C}_{\boldsymbol{\varepsilon}_L}^{(r)}$ of the strain in the LCC via

$$\boldsymbol{\rho}^{(r)} = \frac{1}{2} \mathsf{C}_{\boldsymbol{\varepsilon}_L}^{(r)} \cdot \partial_{\boldsymbol{\varepsilon}} \mathsf{L}_t^{(r)}(\bar{\boldsymbol{\varepsilon}}_L^{(r)}). \tag{4.30}$$

In expressions (4.29) and (4.30), the notation $\mathsf{a} \cdot \partial_{\mathsf{c}} \mathsf{b}$ and $\mathsf{A} \cdot \partial_{\mathsf{c}} \mathsf{B}$ has been used to denote second-order tensors with ijth components $a_{kl} \partial b_{kl} / \partial c_{ij}$ and $A_{klmn} \partial B_{klmn} / \partial c_{ij}$.

Thus, it is seen that the determination of the effective potential and effective stress–strain relation for the nonlinear composite requires the computation of the phase averages of the strain and stress fields, as well as the phase covariance tensors in the 'thermoelastic' LCC defined by the phase potentials (4.24). These phase potentials may be rewritten in the form

$$w_L^{(r)}(\boldsymbol{\varepsilon};\check{\boldsymbol{\varepsilon}}^{(r)},\mathsf{L}_0^{(r)}) = \frac{1}{2}\boldsymbol{\varepsilon}\cdot\mathsf{L}_0^{(r)}\boldsymbol{\varepsilon} + \boldsymbol{\tau}_0^{(r)}\cdot\boldsymbol{\varepsilon} + f_0^{(r)}, \tag{4.31}$$

where the 'thermal stress' $\boldsymbol{\tau}_0^{(r)}$ and 'specific heat' $f_0^{(r)}$ are defined in terms of $\check{\boldsymbol{\varepsilon}}^{(r)}$ and $\mathsf{L}_0^{(r)}$ via

$$\boldsymbol{\tau}_0^{(r)} = \partial_{\boldsymbol{\varepsilon}} w^{(r)}(\check{\boldsymbol{\varepsilon}}^{(r)}) - \mathsf{L}_0^{(r)}\check{\boldsymbol{\varepsilon}}^{(r)}, \quad f_0^{(r)} = w^{(r)}(\check{\boldsymbol{\varepsilon}}^{(r)}) - \boldsymbol{\tau}_0^{(r)}\cdot\check{\boldsymbol{\varepsilon}}^{(r)} - \frac{1}{2}\check{\boldsymbol{\varepsilon}}^{(r)}\cdot\mathsf{L}_0^{(r)}\check{\boldsymbol{\varepsilon}}^{(r)}. \tag{4.32}$$

The effective potential of this LCC can thus be written as (Willis 1981)

$$\tilde{W}_L(\bar{\boldsymbol{\varepsilon}}) = \frac{1}{2}\bar{\boldsymbol{\varepsilon}}\cdot\tilde{\mathsf{L}}_0\bar{\boldsymbol{\varepsilon}} + \tilde{\boldsymbol{\tau}}_0\cdot\bar{\boldsymbol{\varepsilon}} + \tilde{f}_0, \tag{4.33}$$

where $\tilde{\mathsf{L}}_0$, $\tilde{\boldsymbol{\tau}}_0$ and $\tilde{f}_0$ depend on the linear homogenization estimates utilized. The phase averages and second moments of the local fields in the LCC can then be extracted from the relations

$$\bar{\boldsymbol{\varepsilon}}_L^{(r)} = \mathsf{A}_0^{(r)}\bar{\boldsymbol{\varepsilon}} + \mathsf{a}_0^{(r)}, \qquad \langle\boldsymbol{\varepsilon}_L\otimes\boldsymbol{\varepsilon}_L\rangle^{(r)} = \mathsf{C}_{\boldsymbol{\varepsilon}_L}^{(r)} + \bar{\boldsymbol{\varepsilon}}_L^{(r)}\otimes\bar{\boldsymbol{\varepsilon}}_L^{(r)}, \tag{4.34}$$

$$\bar{\boldsymbol{\sigma}}_L^{(r)} = \mathsf{L}_0^{(r)}\bar{\boldsymbol{\varepsilon}}_L^{(r)} + \boldsymbol{\tau}_0^{(r)}, \quad \langle\boldsymbol{\sigma}_L\otimes\boldsymbol{\sigma}_L\rangle^{(r)} = \bar{\boldsymbol{\sigma}}_L^{(r)}\otimes\bar{\boldsymbol{\sigma}}_L^{(r)} + \mathsf{L}_0^{(r)}\mathsf{C}_{\boldsymbol{\varepsilon}_L}^{(r)}\mathsf{L}_0^{(r)}, \tag{4.35}$$

where the $\mathsf{A}_0^{(r)}$ and $\mathsf{a}_0^{(r)}$ are strain concentration tensors, and $\mathsf{C}_{\boldsymbol{\varepsilon}_L}^{(r)}$ denotes the phase covariance tensors of the strain in the LCC, as given by

$$\mathsf{C}_{\boldsymbol{\varepsilon}_L}^{(r)} \doteq \langle(\boldsymbol{\varepsilon}_L - \bar{\boldsymbol{\varepsilon}}_L^{(r)})\otimes(\boldsymbol{\varepsilon}_L - \bar{\boldsymbol{\varepsilon}}_L^{(r)})\rangle^{(r)} = \frac{2}{c^{(r)}}\partial_{\mathsf{L}_0^{(r)}}\tilde{W}_L. \tag{4.36}$$

In this last relation, the derivatives should be taken with the $\check{\boldsymbol{\varepsilon}}^{(r)}$ held fixed.

As in the context of the variational estimates, tangent second-order estimates for the first and second moments of the local fields are obtained by considering composites with perturbed phase potentials (3.2), (3.8), (3.12) and (3.19), and evaluating the derivatives (3.1), (3.7), (3.10) and (3.18), with $\tilde{W}_\tau$, $\tilde{W}_\lambda$, $\tilde{W}_\eta$ and $\tilde{W}_\mu$ given by the tangent second-order procedure. The latter are given by expression (4.25), with $\tilde{W}_L$ replaced by the effective potential of the relevant perturbed LCC, respectively, $\tilde{W}_{L\tau}$, $\tilde{W}_{L\lambda}$, $\tilde{W}_{L\eta}$ and $\tilde{W}_{L\mu}$. The potentials in phase r of these perturbed LCCs are given by expressions analogous to (4.14), (4.16), (4.17) and (4.18), with $w_L^{(r)}$ given by (4.24). In addition, the modulus tensors $\mathsf{L}_0^{(r)}$ are still given by (4.27), and the reference tensors $\check{\boldsymbol{\varepsilon}}^{(r)}$ follow from the appropriate stationarity condition (with $\mathsf{L}_0^{(r)}$ held fixed). In the results below, the symbols $\bar{\boldsymbol{\varepsilon}}_{L\tau}^{(r)}$, $\bar{\boldsymbol{\varepsilon}}_{L\eta}^{(r)}$, $\bar{\boldsymbol{\varepsilon}}_{L\lambda}^{(r)}$ and $\bar{\boldsymbol{\varepsilon}}_{L\mu}^{(r)}$ denote the phase averages in the perturbed LCCs.

Result 4.5. *The tangent second-order estimates for the first and second moments of the local fields are given by*

$$\bar{\boldsymbol{\varepsilon}}^{(r)} = \bar{\boldsymbol{\varepsilon}}_L^{(r)} + \sum_{s=1}^{N} \frac{c^{(s)}}{c^{(r)}} \boldsymbol{\rho}^{(s)} \cdot \partial_{\boldsymbol{\tau}^{(r)}} \bar{\boldsymbol{\varepsilon}}_{L\tau}^{(s)} \Big|_{\boldsymbol{\tau}^{(r)}=\mathbf{0}}, \tag{4.37}$$

$$\langle \boldsymbol{\varepsilon} \otimes \boldsymbol{\varepsilon} \rangle^{(r)} = \langle \boldsymbol{\varepsilon}_L \otimes \boldsymbol{\varepsilon}_L \rangle^{(r)} + 2 \sum_{s=1}^{N} \frac{c^{(s)}}{c^{(r)}} \boldsymbol{\rho}^{(s)} \cdot \partial_{\boldsymbol{\lambda}^{(r)}} \bar{\boldsymbol{\varepsilon}}_{L\lambda}^{(s)} \Big|_{\boldsymbol{\lambda}^{(r)}=\mathbf{0}}, \tag{4.38}$$

$$\bar{\boldsymbol{\sigma}}^{(r)} = \bar{\boldsymbol{\sigma}}_L^{(r)} + \boldsymbol{\rho}^{(r)} - \sum_{s=1}^{N} \frac{c^{(s)}}{c^{(r)}} \boldsymbol{\rho}^{(s)} \cdot \partial_{\boldsymbol{\eta}^{(r)}} \bar{\boldsymbol{\varepsilon}}_{L\eta}^{(s)} \Big|_{\boldsymbol{\eta}^{(r)}=\mathbf{0}}, \tag{4.39}$$

$$\langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)} = \langle \boldsymbol{\sigma}_L \otimes \boldsymbol{\sigma}_L \rangle^{(r)} + 2 \bar{\boldsymbol{\sigma}}_L^{(r)} \otimes_s \boldsymbol{\rho}^{(r)} - 2 \sum_{s=1}^{N} \frac{c^{(s)}}{c^{(r)}} \boldsymbol{\rho}^{(s)} \cdot \partial_{\boldsymbol{\mu}^{(r)}} \bar{\boldsymbol{\varepsilon}}_{L\mu}^{(s)} \Big|_{\boldsymbol{\mu}^{(r)}=\mathbf{0}}, \tag{4.40}$$

where the symbol $\otimes_s$ *denotes symmetrized tensor product, and the subscript L has been used to denote, once again, quantities in the LCC.*

Proof. We begin by proving the identity (4.37) for the phase averages of the strain. In order to make use of proposition 3.1, we consider a composite with perturbed local potential (3.2). Thus, phase r in this composite is characterized by (4.12). Making use of the identity (4.23) for the unperturbed potential $w^{(r)}$, we can write the perturbed potential as

$$w_{\boldsymbol{\tau}}^{(r)}(\boldsymbol{\varepsilon}) = \operatorname*{stat}_{\check{\boldsymbol{\varepsilon}}^{(r)}} \Big\{ w_{L\tau}^{(r)}(\boldsymbol{\varepsilon}; \check{\boldsymbol{\varepsilon}}^{(r)}, \mathsf{L}_0^{(r)}) \Big\}, \tag{4.41}$$

where $w_{L\tau}^{(r)}$ denotes the phase potential of the perturbed LCC, given in terms of (4.24) by (4.14). The tangent second-order estimate for the perturbed effective potential $\tilde{W}_{\boldsymbol{\tau}}$ is thus given by (4.25), with $\tilde{W}_L$ replaced by $\tilde{W}_{L\tau}$, the effective potential of the perturbed LCC with phase r characterized by $w_{L\tau}^{(r)}$. The optimal tensors $\check{\boldsymbol{\varepsilon}}^{(r)}$ and $\mathsf{L}_0^{(r)}$ in the perturbed problem are given by (4.26) and (4.27), with $\bar{\boldsymbol{\varepsilon}}_L^{(r)}$ replaced by $\bar{\boldsymbol{\varepsilon}}_{L\tau}^{(r)}$, the phase averages of the strain in the perturbed LCC. Then, recalling that the tangent second-order estimate for $\tilde{W}_{\boldsymbol{\tau}}$ is stationary with respect to the variables $\check{\boldsymbol{\varepsilon}}^{(r)}$, but not with respect to the variables $\mathsf{L}_0^{(r)}$, and noting that the latter depends on $\boldsymbol{\tau}^{(r)}$ only through the tensors $\check{\boldsymbol{\varepsilon}}^{(r)}$, we have

$$\begin{aligned} \partial_{\boldsymbol{\tau}^{(r)}} \tilde{W}_{\tau}|_{\boldsymbol{\tau}^{(r)}=\mathbf{0}} &= \partial_{\boldsymbol{\tau}^{(r)}} \tilde{W}_{L\tau}|_{\boldsymbol{\tau}^{(r)}=\mathbf{0}} + \sum_{s=1}^{N} \Big(\partial_{\check{\boldsymbol{\varepsilon}}^{(s)}} \mathsf{L}_0^{(s)} \cdot \partial_{\boldsymbol{L}_0^{(s)}} \tilde{W}_L \Big) \cdot \partial_{\boldsymbol{\tau}^{(r)}} \check{\boldsymbol{\varepsilon}}^{(s)}|_{\boldsymbol{\tau}^{(r)}=0} \\ &= c^{(r)} \bar{\boldsymbol{\varepsilon}}_L^{(r)} + \sum_{s=1}^{N} c^{(s)} \boldsymbol{\rho}^{(s)} \cdot \partial_{\boldsymbol{\tau}^{(r)}} \bar{\boldsymbol{\varepsilon}}_{L\tau}^{(s)}|_{\boldsymbol{\tau}^{(r)}=\mathbf{0}}, \end{aligned} \tag{4.42}$$

where the derivative of $\tilde{W}_{L\tau}$ is taken with the $\mathsf{L}_0^{(r)}$ held fixed. In the last equality, use has been made of proposition 3.1 in the first term, and (4.27), (4.30) and (4.36) in the second term. Finally, the identity (4.37) follows from proposition 3.1 with (4.42). The identity (4.38) can be derived in an analogous manner, making use of corollary 3.3.

Next, we prove the identity (4.39). In order to make use of proposition 3.4, we consider a composite with perturbed local potential (3.11). Thus, phase r in this composite is characterized by $w_\eta^{(r)} = (u_\eta^{(r)})^*$, where

$$u_\eta^{(r)}(\boldsymbol{\sigma}) = u^{(r)}(\boldsymbol{\sigma}) + \boldsymbol{\eta}^{(r)} \cdot \boldsymbol{\sigma}, \tag{4.43}$$

with $u^{(r)} = (w^{(r)})^*$. Making use of the dual version of the identity (4.23) for $u^{(r)}$, we can write (4.43) as

$$u_\eta^{(r)}(\boldsymbol{\sigma}) = \underset{\check{\boldsymbol{\sigma}}^{(r)}}{\operatorname{stat}} \Big\{ u_L^{(r)}(\boldsymbol{\sigma}; \check{\boldsymbol{\sigma}}^{(r)}, \mathsf{M}_0^{(r)}) + \boldsymbol{\eta}^{(r)} \cdot \boldsymbol{\sigma} \Big\}. \tag{4.44}$$

Performing the change of variables (see Ponte Castañeda 2002*a*)

$$\mathsf{L}_0^{(r)} = \Big(\mathsf{M}_0^{(r)}\Big)^{-1}, \quad \check{\boldsymbol{\varepsilon}}^{(r)} = \frac{\partial u^{(r)}}{\partial \boldsymbol{\sigma}}(\check{\boldsymbol{\sigma}}^{(r)}), \tag{4.45}$$

the Legendre transform of (4.44) can be written as

$$w_\eta^{(r)}(\boldsymbol{\varepsilon}) = \underset{\check{\boldsymbol{\varepsilon}}^{(r)}}{\operatorname{stat}} \Big\{ w_{L\eta}^{(r)}(\boldsymbol{\varepsilon}; \check{\boldsymbol{\varepsilon}}^{(r)}, \mathsf{L}_0^{(r)}) \Big\}, \tag{4.46}$$

where $w_{L\eta}^{(r)}$ is given by (4.17) with $w_L^{(r)}$ given by (4.24). The tangent second-order estimate for the perturbed effective potential $\tilde{W}_\eta$ is thus given by (4.25) with $\tilde{W}_L$ replaced by $\tilde{W}_{L\eta}$, the effective potential of a perturbed LCC with phase r characterized by (4.26). The optimal tensors $\check{\boldsymbol{\varepsilon}}^{(r)}$ in the perturbed problem are then

$$\check{\boldsymbol{\varepsilon}}^{(r)} = \bar{\boldsymbol{\varepsilon}}_{L\eta}^{(r)} - \boldsymbol{\eta}^{(r)}, \tag{4.47}$$

and the tensors $\mathsf{L}_0^{(r)}$ are related to $\check{\boldsymbol{\varepsilon}}^{(r)}$ by (4.27). Then, recalling that the tangent second-order estimate for $\tilde{W}_\eta$ is stationary with respect to the variables $\check{\boldsymbol{\varepsilon}}^{(r)}$ but not with respect to the variables $\mathsf{L}_0^{(r)}$, and noting that the latter depend on $\boldsymbol{\eta}^{(r)}$ only through the tensors $\check{\boldsymbol{\varepsilon}}^{(r)}$, we have

$$\begin{aligned} \partial_{\boldsymbol{\eta}^{(r)}} \tilde{W}_\eta|_{\boldsymbol{\eta}^{(r)}=\mathbf{0}} &= \partial_{\boldsymbol{\eta}^{(r)}} \tilde{W}_{L\eta}|_{\boldsymbol{\eta}^{(r)}=\mathbf{0}} + \sum_{s=1}^{N} \Big(\partial_{\check{\boldsymbol{\varepsilon}}^{(s)}} \mathsf{L}_0^{(s)} \cdot \partial_{\mathsf{L}_0^{(s)}} \tilde{W}_L \Big) \cdot \partial_{\boldsymbol{\eta}^{(r)}} \check{\boldsymbol{\varepsilon}}^{(s)}|_{\boldsymbol{\eta}^{(r)}=\mathbf{0}} \\ &= -c^{(r)} \bar{\boldsymbol{\sigma}}_L^{(r)} - c^{(r)} \boldsymbol{\rho}^{(r)} + \sum_{s=1}^{N} c^{(s)} \boldsymbol{\rho}^{(s)} \cdot \partial_{\boldsymbol{\eta}^{(r)}} \bar{\boldsymbol{\varepsilon}}_{L\eta}^{(s)}|_{\boldsymbol{\eta}^{(r)}=\mathbf{0}}, \end{aligned} \tag{4.48}$$

where the derivative of $\tilde{W}_{L\eta}$ is taken with the $\mathsf{L}_0^{(r)}$ held fixed. In the last equality, use has been made of proposition 3.4 in the first term, and (4.27), (4.30), (4.36) and (4.47) in the second term. Finally, the identity (4.39) follows from proposition 3.4 with (4.48). The identity (4.40) can be derived in an analogous manner, making use of corollary 3.7. ■

Several observations are relevant in the context of result 4.5. First, it can be shown that the estimates (4.37) and (4.39) for the phase averages are consistent with the corresponding estimates (4.29) for the effective behaviour, in the sense that they satisfy relations (3.20). This implies that the derivatives appearing in

(4.37) and (4.39) satisfy the constraints

$$\sum_{r=1}^{N}\sum_{s=1}^{N} c^{(s)}\boldsymbol{\rho}^{(s)}\cdot\partial_{\boldsymbol{\tau}^{(r)}}\bar{\boldsymbol{\varepsilon}}^{(s)}_{L\tau}|_{\boldsymbol{\tau}^{(r)}=\mathbf{0}} = \mathbf{0}, \tag{4.49}$$

$$\sum_{r=1}^{N}\sum_{s=1}^{N} c^{(s)}\boldsymbol{\rho}^{(s)}\cdot\partial_{\boldsymbol{\eta}^{(r)}}\bar{\boldsymbol{\varepsilon}}^{(s)}_{L\eta}\bigg|_{\boldsymbol{\eta}^{(r)}=\mathbf{0}} = \sum_{r=1}^{N} c^{(r)}\Big[\boldsymbol{\rho}^{(r)} - \boldsymbol{\rho}^{(r)}\cdot\partial_{\bar{\boldsymbol{\varepsilon}}}\bar{\boldsymbol{\varepsilon}}^{(r)}_{L}\Big]. \tag{4.50}$$

It can also be shown that the estimates (4.37)–(4.40) are exact to first order in the heterogeneity contrast, which follows from the fact that the estimate (4.25) for $\tilde{W}$ is exact to second order. Third, it is interesting to note that the terms 'correcting' the LCC quantities (effective behaviour and field statistics) depend explicitly on the intraphase field fluctuations, through the tensors $\mathsf{C}^{(r)}_{\boldsymbol{\varepsilon}_L}$ and on the degree of nonlinearity of the local potential, through the tensors $\partial_{\boldsymbol{\varepsilon}}\mathsf{L}^{(r)}_{t}$ (which vanish in the linear case). It is also interesting to note that the 'correction' of a nonlinear quantity in phase r depends explicitly, in general, on the properties of all other phases. Finally, it is worth mentioning that the derivatives appearing in expressions (4.37)–(4.40) can be expressed in terms of the unperturbed phase averages $\bar{\boldsymbol{\varepsilon}}^{(r)}_{L}$, by differentiating the perturbed system of nonlinear equations $(4.34)_1$ with respect to the perturbation parameter, setting the parameter equal to zero, and inverting the resulting system of linear equations for the derivatives.

A dual formulation of this method is also available, but it is not given here for brevity. Of course, analogous expressions can also be derived from this formulation for the macroscopic stress–strain relation, and the first and second moments of the local fields. It should be mentioned, however, that, unlike the variational method, this method exhibits a duality gap, and consequently, such estimates will not be equivalent to those arising from the strain formulation.

(*c*) *'Second-order' estimates*

An improved version of the tangent second-order method has been introduced by Ponte Castañeda (2002*a*), which incorporates information about the field fluctuations in the linearization scheme, and has been found to deliver estimates that are, in general, more accurate than the ones given in §4*b* (e.g. Ponte Castañeda 2002*b*). These so-called second-order estimates also make use of a thermoelastic LCC with phase potentials $w^{(r)}_{L}$ given in terms of reference tensors $\check{\boldsymbol{\varepsilon}}^{(r)}$ and tensors of moduli $\mathsf{L}^{(r)}_{0}$ by (4.24). However, unlike for the tangent second-order method, in this case the tensors $\mathsf{L}^{(r)}_{0}$ are not identified with the tangent tensors of moduli (4.27). For composites with isotropic constituents characterized by potentials of the form (4.1), Ponte Castañeda (2002*a*) proposed the use of anisotropic tensors of the form

$$\mathsf{L}^{(r)}_{0} = 3\kappa^{(r)}\mathsf{J} + 2\lambda^{(r)}_{0}\mathsf{E}^{(r)} + 2\mu^{(r)}_{0}\mathsf{F}^{(r)}, \tag{4.51}$$

with

$$\mathsf{E}^{(r)} = \frac{2}{3}\,\frac{\check{\boldsymbol{\varepsilon}}^{(r)}_{\mathrm{d}}}{\check{\varepsilon}^{(r)}_{\mathrm{e}}}\otimes\frac{\check{\boldsymbol{\varepsilon}}^{(r)}_{\mathrm{d}}}{\check{\varepsilon}^{(r)}_{\mathrm{e}}}, \quad \mathsf{F}^{(r)} = \mathsf{K} - \mathsf{E}^{(r)}. \tag{4.52}$$

Then, the following identity for the local potentials holds:

$$w^{(r)}(\boldsymbol{\varepsilon}) = \underset{\lambda_0^{(r)},\,\boldsymbol{\mu}_0^{(r)}}{\operatorname{stat}} \Big\{ w_L^{(r)}(\boldsymbol{\varepsilon};\check{\boldsymbol{\varepsilon}}^{(r)},\mathsf{L}_0^{(r)}) + V^{(r)}(\check{\boldsymbol{\varepsilon}}^{(r)},\mathsf{L}_0^{(r)}) \Big\}, \tag{4.53}$$

where the function $V^{(r)}$ is defined as

$$V^{(r)}(\check{\boldsymbol{\varepsilon}}^{(r)},\mathsf{L}_0^{(r)}) = \underset{\boldsymbol{\varepsilon}^{(r)}}{\operatorname{stat}} \Big\{ w^{(r)}(\boldsymbol{\varepsilon}^{(r)}) - w_L^{(r)}(\boldsymbol{\varepsilon}^{(r)};\check{\boldsymbol{\varepsilon}}^{(r)},\mathsf{L}_0^{(r)}) \Big\}. \tag{4.54}$$

The stationary operation here leads to the 'generalized-secant' conditions

$$\partial_{\boldsymbol{\varepsilon}} w^{(r)}(\hat{\boldsymbol{\varepsilon}}^{(r)}) - \partial_{\boldsymbol{\varepsilon}} w^{(r)}(\check{\boldsymbol{\varepsilon}}^{(r)}) = \mathsf{L}_0^{(r)}(\hat{\boldsymbol{\varepsilon}}^{(r)} - \check{\boldsymbol{\varepsilon}}^{(r)}). \tag{4.55}$$

Note that the identity (4.53) is valid for any $\check{\boldsymbol{\varepsilon}}^{(r)}$. The second-order estimates for the effective strain potential are obtained by inserting (4.53) into (2.2), interchanging the optimization operations over $\boldsymbol{\varepsilon}(\boldsymbol{x})$ and the moduli $\lambda_0^{(r)}$ and $\mu_0^{(r)}$, and restricting the latter to be constant per phase. The result is the approximation

$$\tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \underset{\lambda_0^{(s)},\,\mu_0^{(s)}}{\operatorname{stat}} \left\{ \tilde{W}_L(\bar{\boldsymbol{\varepsilon}};\check{\boldsymbol{\varepsilon}}^{(s)},\mathsf{L}_0^{(s)}) + \sum_{r=1}^{N} c^{(r)}\, V^{(r)}(\check{\boldsymbol{\varepsilon}}^{(r)},\mathsf{L}_0^{(r)}) \right\}, \tag{4.56}$$

where $\tilde{W}_L$ is the effective potential of the LCC. The stationary operation in this expression leads to the following conditions for the phase moduli $\lambda_0^{(r)}$ and $\mu_0^{(r)}$:

$$\hat{\varepsilon}_{\parallel}^{(r)} - \check{\varepsilon}_{\mathrm{e}}^{(r)} = \pm\sqrt{\frac{2}{3}\,\frac{1}{c^{(r)}}\,\frac{\partial\,\tilde{W}_L}{\partial\lambda_0^{(r)}}} = \pm\sqrt{\frac{2}{3}\,\langle(\boldsymbol{\varepsilon}_L - \check{\boldsymbol{\varepsilon}}^{(r)})\cdot\mathsf{E}^{(r)}(\boldsymbol{\varepsilon}_L - \check{\boldsymbol{\varepsilon}}^{(r)})\rangle^{(r)}}, \tag{4.57}$$

$$\hat{\varepsilon}_{\perp}^{(r)} = \pm\sqrt{\frac{2}{3}\,\frac{1}{c^{(r)}}\,\frac{\partial\,\tilde{W}_L}{\partial\mu_0^{(r)}}} = \pm\sqrt{\frac{2}{3}\,\langle\boldsymbol{\varepsilon}_L\cdot\mathsf{F}^{(r)}\boldsymbol{\varepsilon}_L\rangle^{(r)}}, \tag{4.58}$$

where $\hat{\varepsilon}_{\parallel}^{(r)} = \sqrt{(2/3)\hat{\boldsymbol{\varepsilon}}^{(r)}\cdot\mathsf{E}^{(r)}\hat{\boldsymbol{\varepsilon}}^{(r)}}$ and $\hat{\varepsilon}_{\perp}^{(r)} = \sqrt{(2/3)\hat{\boldsymbol{\varepsilon}}^{(r)}\cdot\mathsf{F}^{(r)}\hat{\boldsymbol{\varepsilon}}^{(r)}}$ denote the components of the tensors $\hat{\boldsymbol{\varepsilon}}^{(r)}$ that are 'parallel' and 'perpendicular' to the corresponding reference tensor $\hat{\boldsymbol{\varepsilon}}^{(r)}$, respectively. For a given set of reference tensors $\check{\boldsymbol{\varepsilon}}^{(r)}$, the stationarity conditions (4.55), (4.57) and (4.58) completely specify the tensors $\hat{\boldsymbol{\varepsilon}}^{(r)}$ and $\mathsf{L}_0^{(r)}$. Unfortunately, the optimal choice of reference strains $\check{\boldsymbol{\varepsilon}}^{(r)}$ is still an open question. While they should depend on $\bar{\boldsymbol{\varepsilon}}$ and reduce to it to zeroth order in the weak-heterogeneity expansion, in general, they could depend on other parameters in the problem. Ponte Castañeda (2002*a*) initially proposed enforcing stationarity of (4.56) with respect to the $\check{\boldsymbol{\varepsilon}}^{(r)}$, but it has not been possible to find a satisfactory solution to the resulting equations. In what follows, expressions for the effective behaviour and field statistics will first be given for general $\check{\boldsymbol{\varepsilon}}^{(r)}$, and then specialized further below. In any case, the expression (4.56) can be shown to simplify to

$$\tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \sum_{r=1}^{N} c^{(r)} \Big[w^{(r)}(\hat{\boldsymbol{\varepsilon}}^{(r)}) - \partial_{\boldsymbol{\varepsilon}} w^{(r)}(\check{\boldsymbol{\varepsilon}}^{(r)})\cdot\Big(\hat{\boldsymbol{\varepsilon}}^{(r)} - \bar{\boldsymbol{\varepsilon}}_L^{(r)}\Big)\Big], \tag{4.59}$$

where $\bar{\boldsymbol{\varepsilon}}_L^{(r)}$ and $\hat{\boldsymbol{\varepsilon}}^{(r)}$ have been defined above in terms of the averages and second moments of the strain field in phase r of the thermoelastic LCC.

Since, unlike the variational estimates, the second-order estimates are not fully stationary, the corresponding estimates for the effective stress–strain relation and field statistics are given by those in the LCC plus certain 'correction' terms. In fact, their form is similar to that of the tangent second-order estimates given in §4b, as can be seen in the following results. The derivation of these results is analogous to that given in the context of result 4.5, and will be omitted here for brevity.

Result 4.6. *The second-order estimates for the effective behaviour are given by*

$$\bar{\boldsymbol{\sigma}} = \partial_{\bar{\boldsymbol{\varepsilon}}} \tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \bar{\boldsymbol{\sigma}}_L + \sum_{r=1}^{N} c^{(r)} \boldsymbol{\rho}^{(r)} \cdot \partial_{\bar{\boldsymbol{\varepsilon}}} \check{\boldsymbol{\varepsilon}}^{(r)}, \tag{4.60}$$

where $\bar{\boldsymbol{\sigma}}_L$ denotes the macroscopic stress in the LCC, and the tensors $\boldsymbol{\rho}^{(r)}$ are

$$\boldsymbol{\rho}^{(r)} = \left[\mathsf{L}_0^{(r)} - \mathsf{L}_t^{(r)}(\check{\boldsymbol{\varepsilon}}^{(r)})\right]\left(\hat{\boldsymbol{\varepsilon}}^{(r)} - \bar{\boldsymbol{\varepsilon}}_L^{(r)}\right) + \frac{4}{3}\frac{\lambda_0^{(r)} - \mu_0^{(r)}}{(\check{\varepsilon}_{\mathrm{e}}^{(r)})^2}$$

$$\times \left[\langle(\boldsymbol{\varepsilon}_{\mathrm{d}_L} - \check{\boldsymbol{\varepsilon}}_{\mathrm{d}}^{(r)}) \otimes (\boldsymbol{\varepsilon}_{\mathrm{d}_L} - \check{\boldsymbol{\varepsilon}}_{\mathrm{d}}^{(r)})\rangle^{(r)} - (\hat{\boldsymbol{\varepsilon}}_{\mathrm{d}}^{(r)} - \check{\boldsymbol{\varepsilon}}_{\mathrm{d}}^{(r)}) \otimes (\hat{\boldsymbol{\varepsilon}}_{\mathrm{d}}^{(r)} - \check{\boldsymbol{\varepsilon}}_{\mathrm{d}}^{(r)})\right] \check{\boldsymbol{\varepsilon}}_{\mathrm{d}}^{(r)}. \tag{4.61}$$

In (4.61), the subscript L denotes quantities in the LCC associated with the estimate (4.56), the subscript d denotes deviatoric parts, and $\mathsf{L}_t^{(r)}$ is the tangent modulus tensor defined by expression (4.27).

Result 4.7. *The second-order estimates for the first and second moments of the local fields are given by*

$$\bar{\boldsymbol{\varepsilon}}^{(r)} = \bar{\boldsymbol{\varepsilon}}_L^{(r)} + \sum_{s=1}^{N} \frac{c^{(s)}}{c^{(r)}} \boldsymbol{\rho}^{(s)} \cdot \partial_{\boldsymbol{\tau}^{(r)}} \check{\boldsymbol{\varepsilon}}_{\tau}^{(s)}\Big|_{\boldsymbol{\tau}^{(r)} = \mathbf{0}}, \tag{4.62}$$

$$\langle \boldsymbol{\varepsilon} \otimes \boldsymbol{\varepsilon} \rangle^{(r)} = \langle \boldsymbol{\varepsilon}_L \otimes \boldsymbol{\varepsilon}_L \rangle^{(r)} + 2 \sum_{s=1}^{N} \frac{c^{(s)}}{c^{(r)}} \boldsymbol{\rho}^{(s)} \cdot \partial_{\boldsymbol{\lambda}^{(r)}} \check{\boldsymbol{\varepsilon}}_{\lambda}^{(s)}\Big|_{\boldsymbol{\lambda}^{(r)} = \mathbf{0}}, \tag{4.63}$$

$$\bar{\boldsymbol{\sigma}}^{(r)} = \bar{\boldsymbol{\sigma}}_L^{(r)} + \boldsymbol{\rho}^{(r)} - \sum_{s=1}^{N} \frac{c^{(s)}}{c^{(r)}} \boldsymbol{\rho}^{(s)} \cdot \partial_{\boldsymbol{\eta}^{(r)}} \check{\boldsymbol{\varepsilon}}_{\eta}^{(s)}\Big|_{\boldsymbol{\eta}^{(r)} = \mathbf{0}}, \tag{4.64}$$

$$\langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)} = \langle \boldsymbol{\sigma}_L \otimes \boldsymbol{\sigma}_L \rangle^{(r)} + 2\check{\boldsymbol{\sigma}}^{(r)} \otimes_s \boldsymbol{\rho}^{(r)} - 2 \sum_{s=1}^{N} \frac{c^{(s)}}{c^{(r)}} \boldsymbol{\rho}^{(s)} \cdot \partial_{\boldsymbol{\mu}^{(r)}} \check{\boldsymbol{\varepsilon}}_{\mu}^{(s)}\Big|_{\boldsymbol{\mu}^{(r)} = \mathbf{0}}, \tag{4.65}$$

where $\check{\boldsymbol{\sigma}}^{(r)} = \partial_{\boldsymbol{\varepsilon}} w^{(r)}(\check{\boldsymbol{\varepsilon}}^{(r)})$, the symbol $\otimes_s$ denotes symmetrized tensor product, the subscript L denotes quantities in the LCC, and $\check{\boldsymbol{\varepsilon}}_{\tau}^{(s)}$, $\check{\boldsymbol{\varepsilon}}_{\lambda}^{(s)}$, $\check{\boldsymbol{\varepsilon}}_{\eta}^{(s)}$ and $\check{\boldsymbol{\varepsilon}}_{\mu}^{(s)}$ denote the relevant perturbed reference strains.

An important observation in the context of this result is that, like the tangent second-order estimates, the second-order estimates (4.62) and (4.64) for the phase averages can be shown to be consistent with the corresponding estimates for the effective stress–strain relation (4.60). Thus, this implies that the terms involving derivatives of the reference tensors in (4.62) and (4.64) satisfy constraints analogous to (4.49) and (4.50).

The results given above are valid for general reference tensors $\check{\boldsymbol{\varepsilon}}^{(r)}$. The simplest prescription for these tensors, which has been shown to be fully consistent in the sense described in the previous paragraph, and to give results that are physically reasonable at least in the context of two-phase composites with isotropic phases (see part II of this work), is given by

$$\check{\boldsymbol{\varepsilon}}^{(r)} = \bar{\boldsymbol{\varepsilon}}_{\mathrm{d}}, \quad \text{for all } r, \tag{4.66}$$

where $\bar{\boldsymbol{\varepsilon}}_{\mathrm{d}}$ denotes the deviatoric part of the macroscopic strain. Thus, prescription (4.66) together with the stationarity conditions mentioned above, constitute a system of algebraic nonlinear equations for the variables $\hat{\boldsymbol{\varepsilon}}^{(r)}$, $\lambda_0^{(r)}$ and $\mu_0^{(r)}$. When the simple prescription (4.66) is used for the reference tensors, the derivatives in expressions (4.60)–(4.65) become trivial. Thus, in the expression for the effective behaviour we have that $\partial_{\bar{\boldsymbol{\varepsilon}}} \check{\boldsymbol{\varepsilon}}^{(r)} = \mathsf{K}$, and all derivatives of the perturbed reference tensors with respect to the various perturbation parameters in (4.62)–(4.64) become zero, since they are taken with $\bar{\boldsymbol{\varepsilon}}$ held fixed. The simplified expressions are provided in the following result.

Result 4.8. *The second-order estimates for the effective behaviour, with prescription (4.66), are given by*

$$\bar{\boldsymbol{\sigma}} = \partial_{\bar{\boldsymbol{\varepsilon}}} \tilde{W}(\bar{\boldsymbol{\varepsilon}}) = \bar{\boldsymbol{\sigma}}_L + \sum_{r=1}^{N} c^{(r)} \boldsymbol{\rho}^{(r)}, \tag{4.67}$$

and the corresponding estimates for the field statistics are given by

$$\bar{\boldsymbol{\varepsilon}}^{(r)} = \bar{\boldsymbol{\varepsilon}}_L^{(r)}, \qquad \langle \boldsymbol{\varepsilon} \otimes \boldsymbol{\varepsilon} \rangle^{(r)} = \langle \boldsymbol{\varepsilon}_L \otimes \boldsymbol{\varepsilon}_L \rangle^{(r)}, \tag{4.68}$$

$$\bar{\boldsymbol{\sigma}}^{(r)} = \bar{\boldsymbol{\sigma}}_L^{(r)} + \boldsymbol{\rho}^{(r)}, \quad \langle \boldsymbol{\sigma} \otimes \boldsymbol{\sigma} \rangle^{(r)} = \langle \boldsymbol{\sigma}_L \otimes \boldsymbol{\sigma}_L \rangle^{(r)} + 2\check{\boldsymbol{\sigma}}^{(r)} \otimes_s \boldsymbol{\rho}^{(r)}, \tag{4.69}$$

where $\check{\boldsymbol{\sigma}}^{(r)} = \partial_{\boldsymbol{\varepsilon}} w^{(r)}(\bar{\boldsymbol{\varepsilon}})$, *and the tensors* $\boldsymbol{\rho}^{(r)}$ are defined by *(4.61) with (4.66).*

In this case, the second-order estimates for the phase averages and second moments of the strain (arising from the strain version) *coincide* with those in the LCC, while the corresponding estimates for the stress quantities still exhibit certain 'correction' terms. It is easy to see for prescription (4.66) that the constraints (4.49) and (4.50) are indeed satisfied.

Analogous expressions can be derived from the stress formulation of the second-order method, but are not given here for brevity. In general, the second-order estimates arising from the strain and stress formulations are *not* equivalent, i.e. they exhibit a duality gap, which depends on the prescriptions used for the reference tensors in both versions. It is worth mentioning, though, that the observations of the previous paragraph are also valid for the stress version if the reference stresses $\check{\boldsymbol{\sigma}}^{(r)}$ are set equal to the macroscopic stress $\bar{\boldsymbol{\sigma}}$.

5. Concluding remarks

Exact relations for nonlinear composites have been given which express the first and even moments of the spatial distribution of the local fields in terms of the effective potentials of suitably perturbed composites. Similar relations could also be obtained for other kinds of averaged quantities, following similar arguments. These relations were used in the context of the variational, tangent second-order and second-order nonlinear homogenization methods, to obtain rigorous estimates for

the first and second moments of the fields. Thus, it was shown that, while the variational estimates for these quantities coincide with those in the associated LCC, as had been previously conjectured, the tangent second-order and second-order estimates were found to incorporate certain 'correction' terms which depend explicitly on the nonlinearity of the local potentials and on the fluctuations of the fields in the LCC. These 'correction' terms are such that the estimates for the first moments (phase averages) of the fields are entirely consistent with the corresponding estimates for the macroscopic constitutive relation.

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