Second-Order Homogenization Estimates Incorporating Field Fluctuations in Finite Elasticity

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Dedicated to Ray Ogden on the occasion of his 60th birthday

Abstract: This paper presents the application of a recently proposed "second-order" homogenization method (Ponte Castañeda 2002; *J. Mech. Phys. Solids* **50**, 737) to the estimation of the effective behavior of hyperelastic composites subjected to *finite* deformations. The key idea is to introduce an optimally chosen "linear comparison composite" which can then be used to convert available homogenization estimates for linear composites directly into new estimates for the nonlinear hyperelastic composites. More precisely, the method makes use of "generalized" secant moduli that are intermediate between the standard "secant" and "tangent" moduli of the nonlinear phases, and that depend not only on the averages, or first moments of the fields in the phases, but also on the second moments of the field fluctuations, or phase covariance tensors. The use of the method is illustrated in the context of carbon-black-filled, and fiber-reinforced elastomers, and estimates analogous to the well-known Hashin=–Shtrikman and self-consistent estimates for linear–elastic composites are generated. The new estimates are compared with corresponding estimates using an earlier version of the method (Ponte Castañeda and Tiberio 2000; *J. Mech. Phys. Solids* **48**, 1389) neglecting the use of fluctuations, and the new results are found to be superior. In particular, the new estimates, unlike the earlier ones, are found to satisfy a rigorous bound, and to give more realistic predictions in the important limit of incompressible behavior.

Key Words: homogenization, finite strain, rubber material, variational calculus

1. HYPERELASTIC COMPOSITES AND EFFECTIVE BEHAVIOR

The objective of this paper is to develop estimates for the effective behavior of hyperelastic composite materials subjected to *finite deformations*. The materials are made up of N different (homogeneous) phases, which are assumed to be distributed randomly in a specimen occupying a volume Ω_0 in the reference configuration. Furthermore, the size of the typical inhomogeneity (e.g., particle, void, crystal) is much smaller than the size of the specimen and the scale of variation of the loading conditions. The constitutive behavior of the phases is characterized by stored energies $W^{(r)}$ $(r = 1, \ldots, N)$ that are *nonconvex* functions of the deformation gradient $\mathbf{F}$. The local energy function of the composite may be written as

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$$W(\mathbf{X},\mathbf{F}) = \sum_{r=1}^{N} \chi^{(r)}(\mathbf{X})\, W^{(r)}(\mathbf{F}), \tag{1}$$

where the functions $\chi^{(r)}$ are equal to 1 if the position vector $\mathbf{X}$ is inside phase r (i.e. $\mathbf{X} \in \Omega_0^{(r)}$) and zero otherwise. The stored-energy functions of the phases are, of course, assumed to be *objective* in the sense that $W^{(r)}(\mathbf{QF}) = W^{(r)}(\mathbf{F})$ for all proper orthogonal $\mathbf{Q}$ and arbitrary deformation gradients $\mathbf{F}$. Making use of the polar decomposition $\mathbf{F} = \mathbf{RU}$, where $\mathbf{U}$ is the right stretch tensor and $\mathbf{R}$ is the rotation tensor, it follows, in particular, that $W^{(r)}(\mathbf{F}) = W^{(r)}(\mathbf{U})$.

The local or microscopic constitutive relation for the material is given by

$$\mathbf{S} = \frac{\partial W}{\partial \mathbf{F}}(\mathbf{X},\mathbf{F}), \tag{2}$$

where $\mathbf{S}$ denotes the first Piola–Kirchhoff stress tensor. Note that sufficient smoothness has been assumed for W in $\mathbf{F}$ and that $\mathbf{F}$ is required to satisfy the *material impenetrability* condition: $\det \mathbf{F}(\mathbf{X}) > 0$ for $\mathbf{X}$ in Ω_0. For example, this condition would be satisfied for incompressible materials, where $\det \mathbf{F}$ is required to be exactly 1. For more details on hyperelastic materials, refer to the monograph by Ogden [1].

Following the works of Hill [2], Hill and Rice [3] and Ogden [4], the *effective stored-energy function* of the composite is defined by

$$\widetilde{W}(\overline{\mathbf{F}}) = \inf_{\mathbf{F}\in\mathcal{K}(\overline{\mathbf{F}})} \langle W(\mathbf{X},\mathbf{F})\rangle = \inf_{\mathbf{F}\in\mathcal{K}(\overline{\mathbf{F}})} \sum_{r=1}^{N} c^{(r)} \langle W^{(r)}(\mathbf{F})\rangle^{(r)}, \tag{3}$$

where $\mathcal{K}$ denotes the set of admissible deformation gradients:

$$\mathcal{K}(\overline{\mathbf{F}}) = \left\{ \mathbf{F} \mid \mathbf{x} = \boldsymbol{\chi}(\mathbf{X}) \text{ with } \mathbf{F} = \mathrm{Grad}\boldsymbol{\chi},\ \det \mathbf{F} > 0 \text{ in } \Omega_0,\ \mathbf{x} = \bar{\mathbf{F}}\mathbf{X} \text{ on } \partial\Omega_0 \right\}. \tag{4}$$

Above, the symbols $\langle . \rangle$ and $\langle . \rangle^{(r)}$ have been introduced to denote volume averages over the composite (Ω_0) and over phase r ($\Omega_0^{(r)}$), respectively, so that the scalars $c^{(r)} = \langle \chi^{(r)} \rangle$ serve to denote the volume fractions of the given phases.

It is noted that $\widetilde{W}$ physically corresponds to the average elastic energy that is stored in the composite when it is subjected to an affine displacement boundary condition with prescribed average deformation gradient $\langle \mathbf{F} \rangle = \overline{\mathbf{F}}$. Furthermore, it can be easily shown that $\widetilde{W}$ is objective, that is, $\widetilde{W}(\overline{\mathbf{F}}) = \widetilde{W}(\overline{\mathbf{U}})$, where $\overline{\mathbf{U}}$ is the macroscopic right stretch tensor in the polar decomposition of the macroscopic deformation gradient $\overline{\mathbf{F}} = \overline{\mathbf{R}}\,\overline{\mathbf{U}}$, with $\overline{\mathbf{R}}$ denoting the macroscopic rotation tensor.

The usefulness of the definition (3) derives from the fact that the *average stress*, defined by $\overline{\mathbf{S}} = \langle \mathbf{S} \rangle$, can be shown to be related to the average deformation gradient $\overline{\mathbf{F}}$ via the relation

$$\overline{\mathbf{S}} = \frac{\partial \widetilde{W}}{\partial \overline{\mathbf{F}}}, \tag{5}$$

where, again, sufficient smoothness must be assumed for $\widetilde{W}$. This is the effective or macroscopic constitutive relation for the nonlinear elastic composite. Of course, the average stress and deformation gradient must satisfy macroscopic equilibrium and compatibility. In particular, the macroscopic rotational balance equation $\overline{\mathbf{S}}\,\overline{\mathbf{F}}^T = \overline{\mathbf{F}}\,\overline{\mathbf{S}}^T$ must be satisfied (Hill [2]).

It is further recalled that under the hypotheses of *polyconvexity* of W, together with suitable growth conditions for W, the infimum in relation (3) defining $\widetilde{W}$ is known (Ball [5]) to be attained when the field $\mathbf{x}$ is assumed to be in a suitable functional space. Ogden [6] proposed alternative constitutive hypotheses on W ensuring the existence of extremum principles of potential and complementary energy in finite elasticity. More precise definitions of the effective energy $\widetilde{W}$ are available at least for *periodic* microstructures (Müller [7], Braides [8]). Such definitions generalize the classical definition of the effective energy for periodic media with convex energies (Marcellini [9]), by allowing for possible interactions between unit cells, essentially by taking an infimum over the set of all possible combinations of units cells. Physically, this corresponds to accounting for the possibility of the development of instabilities in the composite at sufficiently high deformation. In practice, however, the definition (3) should provide an adequate measure of the effective behavior up to the point at which instabilities develop (see Geymonat et al. [10]). Note that $\widetilde{W}$ is essentially the quasiconvexification (or relaxation) of W.

The focus here will be in the characterization of the effective behavior of composites made up of *rubber elastic* phases. Given objectivity, isotropy then implies that the stored-energy functions of the phases can be written as symmetric functions of the principal stretches $\lambda_1, \lambda_2, \lambda_3$ (i.e. the principal values of $\mathbf{U}$), so that $W^{(r)}(\mathbf{F}) = \Phi^{(r)}(\lambda_1, \lambda_2, \lambda_3) = \Phi^{(r)}(\lambda_2, \lambda_1, \lambda_3)$, etc. A fairly general class of such stored-energy functions, which has been found to give good agreement with experimental data for rubberlike materials, was proposed by Ogden [12]. Although the methods developed in this paper will apply more generally, for simplicity, the attention here will be focused on *polyconvex* energy functions of the type

$$W(\mathbf{F}) = f(\mathbf{F}) + g(J), \tag{6}$$

where $J = \det \mathbf{F} = \lambda_1\lambda_2\lambda_3$, and f and g are taken to be *convex* functions of the tensor $\mathbf{F}$ and the scalar variable $J > 0$, respectively. A simple, special case of this general class of materials, which will be considered in some detail below is given by the *compressible* neo-Hookean material with stored-energy function of the form:

$$W(\mathbf{F}) = \frac{\mu}{2}\left(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3\right) + \frac{\mu'}{2}(J-1)^2 - \mu \ln J. \tag{7}$$

where the parameters $\mu > 0$ and $\mu' > 0$ denote the standard Lamé moduli. Note that $W(\mathbf{F}) \sim (1/2)\mu'(\mathrm{tr}\boldsymbol{\varepsilon})^2 + \mu \mathrm{tr}\boldsymbol{\varepsilon}^2$, where $\boldsymbol{\varepsilon}$ is the infinitesimal strain tensor, as $\mathbf{F} \to \mathbf{I}$, so that the stored-energy function (7) linearizes properly. In addition, the limit as $\mu' \to \infty$ in relation (7) corresponds to incompressible behavior ($J \to 1$).

The objective of this work then becomes to obtain estimates for the effective stored-energy function $\widetilde{W}$ of hyperelastic composites subjected to finite deformations. This is an extremely difficult problem, because it amounts to solving a set of highly nonlinear partial differential equations with random coefficients. As a consequence, there are precious few *analytical* estimates for $\widetilde{W}$. Ogden [13] noted that use of the trial field $\mathbf{F} = \overline{\mathbf{F}}$ in the definition (3) for $\widetilde{W}$ leads to an upper bound analogous to the well-known Voigt upper bound in linear elasticity. Also, under appropriate hypotheses on W ensuring the existence of a principle of minimum complementary energy, Ogden [13] also proposed a generalization of the Reuss lower bound. However, the required constitutive hypothesis is too strong to include materials such as the compressible neo-Hookean material defined by relation (7). For this reason, Ponte Castañeda [14] proposed an alternative generalization of the Reuss lower bound, exploiting the polyconvexity hypothesis. For polyconvex materials of the type (6), this lower bound takes the form:

$$\widetilde{W}(\overline{\mathbf{F}}) \geq \widetilde{W}_{PC}\,(\overline{\mathbf{F}}) \doteq \left(\overline{f^*}\right)^*(\overline{\mathbf{F}}) + \left(\overline{g^*}\right)^*(\det \overline{\mathbf{F}}). \tag{8}$$

Thus, the bound $\widetilde{W}_{PC}$ reduces to the polyconvex envelope [11] of the function W, given by relation (6), when the special case of a homogeneous material is considered. Note that, due to the lack of convexity of the function W, this lower bound is much sharper (see [14]) than the bound that would be obtained by means of the standard Legendre–Fenchel transform applied directly to the function W, which would lead to a bound of the type $\left(\overline{W^*}\right)^*(\overline{\mathbf{F}})$.

There are also numerous empirically based, and *ad hoc* estimates for various special cases, including the case of rigidly reinforced rubbers (see [15, 16, 17, 18, 19]). Our aim here is to develop a general class of *analytical estimates* that are based on homogenization theory and that are applicable to large classes of composite systems, including rigidly reinforced rubbers, porous elastomers and other heterogeneous elastomeric systems, such as nematic elastomers and block copolymers. Such estimates should allow for the incorporation of statistical information beyond the phase volume fractions, thus allowing for a more precise characterization of the influence of microstructure on effective behavior. Some progress along these lines has been made recently [20, 21] with the extension of an earlier version of the "second-order" nonlinear homogenization technique [22] to finite elasticity.

2. THE SECOND-ORDER VARIATIONAL PROCEDURE

Our proposal for generating homogenization estimates in finite elasticity is based on an appropriate extension of the "second-order" homogenization procedure that has been recently developed by Ponte Castañeda [23, 24] in the context of nonlinear dielectrics and viscous composites with convex, nonlinear potentials. This new method is in turn a generalization of the "linear comparison" variational method of Ponte Castañeda [25] in a way that incorporates many of the desirable features of an earlier version of the second-order method [22, 26], including the fact that the estimates generated should be exact to second order in the heterogeneity contrast [27]. It is relevant to mention in this context that earlier works (e.g., Talbot and Willis [28], Ponte Castañeda [25]) delivered bounds that are exact *only* to *first* order in the contrast. Next we give a brief description of the proposed method.

Following [22], define a *comparison* linear "thermoelastic" composite with potential:

$$W_0(\mathbf{X},\mathbf{F}) = \sum_{r=1}^{N} \chi^{(r)}(\mathbf{X})\, W_0^{(r)}(\mathbf{F}), \tag{9}$$

where the quadratic functions $W_0^{(r)}$ correspond to second-order Taylor approximations of the nonlinear stored-energy functions $W^{(r)}$ about certain uniform reference deformations $\mathbf{F}^{(r)}$:

$$W_0^{(r)}(\mathbf{F}) = W^{(r)}(\mathbf{F}^{(r)}) + \mathbf{S}^{(r)}(\mathbf{F}^{(r)}) \cdot (\mathbf{F} - \mathbf{F}^{(r)}) + \frac{1}{2}(\mathbf{F} - \mathbf{F}^{(r)}) \cdot \mathbf{L}_0^{(r)}(\mathbf{F} - \mathbf{F}^{(r)}). \tag{10}$$

Here $\mathbf{S}^{(r)} = \partial W^{(r)}/\partial \mathbf{F}$, and $\mathbf{L}_0^{(r)}$ is a positive definite, constant tensor to be determined later. Then, $\mathbf{S} = \mathbf{S}^{(r)}(\mathbf{F}^{(r)}) + \mathbf{L}_0^{(r)}(\mathbf{F} - \mathbf{F}^{(r)})$ is the stress associated with $\mathbf{F}$ in phase *r* of the linear comparison composite. Note that the nonlinear stored-energy functions $W^{(r)}$ can then be *approximated* as:

$$W^{(r)}(\mathbf{F}) = W_0^{(r)}(\mathbf{F}) + V^{(r)}(\mathbf{F}^{(r)}, \mathbf{L}_0^{(r)}). \tag{11}$$

where the $V^{(r)}$ are "error" functions defined by:

$$V^{(r)}(\mathbf{F}^{(r)}, \mathbf{L}_0^{(r)}) = \operatorname*{stat}_{\hat{\mathbf{F}}^{(r)}} \left[W^{(r)}(\hat{\mathbf{F}}^{(r)}) - W_0^{(r)}(\hat{\mathbf{F}}^{(r)}) \right]. \tag{12}$$

In these expressions, the optimization operation *stat* with respect to a variable means differentiation with respect to that variable and setting the result equal to zero to generate an expression for the optimal value of the variable.

For later use, let

$$\widetilde{W}_0(\overline{\mathbf{F}}; \mathbf{F}^{(s)}, \mathbf{L}_0^{(s)}) = \min_{\mathbf{F}\in\mathcal{K}} \langle W_0(\mathbf{X},\mathbf{F}) \rangle = \min_{\mathbf{F}\in\mathcal{K}} \sum_{r=1}^{N} c^{(r)} \langle W_0^{(r)}(\mathbf{F}) \rangle^{(r)} \tag{13}$$

be the effective free-energy density associated with the *fictitious* linear thermoelastic composite, which has the same microstructure as the original nonlinear elastic composite. To see this more explicitly (see, for example, Willis [29]), note that the Euler-Lagrange equations of the variational problem for $\widetilde{W}_0$ are

$$\nabla \cdot (\mathbf{L}_0 \nabla \mathbf{x} - \mathbf{T}) = \mathbf{0} \quad \text{in } \Omega, \quad \mathbf{x} = \overline{\mathbf{F}}\mathbf{X} \quad \text{on } \partial\Omega, \tag{14}$$

where $\mathbf{L}_0$ is the elasticity tensor of the linear comparison composite with free energy (10), $\mathbf{T}$ denotes a suitably defined thermal stress tensor, and where the temperature and heat capacity at constant strain are taken to be unity and zero, respectively. Note that this fictitious linear thermoelastic problem is one involving, in general, non-symmetric "stress" and "strain" measures, so that suitable generalizations of the classical thermoelastic analyses are required [20]. In particular, estimates of the self-consistent and Hashin–Shtrikman types may be obtained by appropriate extension of the corresponding methods for linear–elastic composites [30, 29].

Using relations (11), averaging the resulting expression for W over Ω_0, minimizing over $\mathbf{F}$ in $\mathcal{K}$ and optimizing over tensors $\mathbf{F}^{(s)}$ and $\mathbf{L}_0^{(s)}$, it follows that [24]:

$$\widetilde{W}(\overline{\mathbf{F}}) = \operatorname*{stat}_{\mathbf{F}^{(s)},\,\mathbf{L}_0^{(s)}} \left\{ \widetilde{W}_0(\overline{\mathbf{F}}; \mathbf{F}^{(s)}, \mathbf{L}_0^{(s)}) + \sum_{r=1}^{N} c^{(r)}\, V^{(r)}(\mathbf{F}^{(r)}, \mathbf{L}_0^{(r)}) \right\}, \tag{15}$$

where the $V^{(r)}$ and $\widetilde{W}_0$ are defined by relations (12) and (13), respectively. Here it has been assumed that the resulting optimal values of $\mathbf{F}^{(s)}$ and $\mathbf{L}_0^{(s)}$ are such that the linear comparisons problem (13) is well posed.

It is easy to verify that formally setting the tensors $\mathbf{F}^{(s)}$ identically equal to zero leads to estimates of the type first proposed in [25] for materials with convex energy functions. In order to do better, it is necessary to consider the definition of the functions $V^{(r)}$ above in more detail. First note that optimizing with respect to the variables $\hat{\mathbf{F}}^{(r)}$ in (12) leads to the relations

$$\mathbf{S}^{(r)}(\hat{\mathbf{F}}^{(r)}) - \mathbf{S}^{(r)}(\mathbf{F}^{(r)}) = \mathbf{L}_0^{(r)}(\hat{\mathbf{F}}^{(r)} - \mathbf{F}^{(r)}), \tag{16}$$

where again sufficient smoothness has been assumed for the $W^{(r)}$. This condition has a nice physical interpretation as depicted in the one-dimensional sketch shown in Figure 1(a): it corresponds to a *linear approximation* to the nonlinear constitutive relation for the elastic material in phase r interpolating between the deformations $\mathbf{F}^{(r)}$ and $\hat{\mathbf{F}}^{(r)}$. Note that this condition does not have a unique solution, and appropriate choices must be made for the relevant variables. Indeed, as illustrated in Figure 1(a), there are three stationary points (the three points where the dashed, straight line intersects the continuous nonlinear curve), which lead to three different types of approximations. Thus, as illustrated in Figure 1(b), the "secant" approximation is obtained by setting $\mathbf{F}^{(r)} = \mathbf{0}$, while the "tangent" approximation is obtained by letting $\hat{\mathbf{F}}^{(r)}$ tend to $\mathbf{F}^{(r)}$. On the other hand, when $\hat{\mathbf{F}}^{(r)} \neq \mathbf{F}^{(r)}$ and $\mathbf{F}^{(r)} \neq \mathbf{0}$, a new type of approximation is obtained, which has been referred to [24] as a "generalized secant" approximation.

Under the assumption that $\hat{\mathbf{F}}^{(r)} \neq \mathbf{F}^{(r)}$, consideration of the optimality conditions with respect to the variables $\mathbf{F}^{(r)}$ and $\mathbf{L}_0^{(r)}$ in expression (15) formally leads to the following prescriptions:

$$\mathbf{F}^{(r)} = \langle \mathbf{F} \rangle^{(r)} \doteq \overline{\mathbf{F}}^{(r)}, \tag{17}$$

and

$$(\hat{\mathbf{F}}^{(r)} - \overline{\mathbf{F}}^{(r)}) \otimes (\hat{\mathbf{F}}^{(r)} - \overline{\mathbf{F}}^{(r)}) = \langle (\mathbf{F} - \overline{\mathbf{F}}^{(r)}) \otimes (\mathbf{F} - \overline{\mathbf{F}}^{(r)}) \rangle^{(r)} \doteq \mathbf{C}_{\mathbf{F}}^{(r)}, \tag{18}$$

where use has been made of the relation:

$$\mathbf{C}_{\mathbf{F}}^{(r)} = \frac{2}{c^{(r)}} \frac{\partial \widetilde{W}_0}{\partial \mathbf{L}_0^{(r)}}. \tag{19}$$

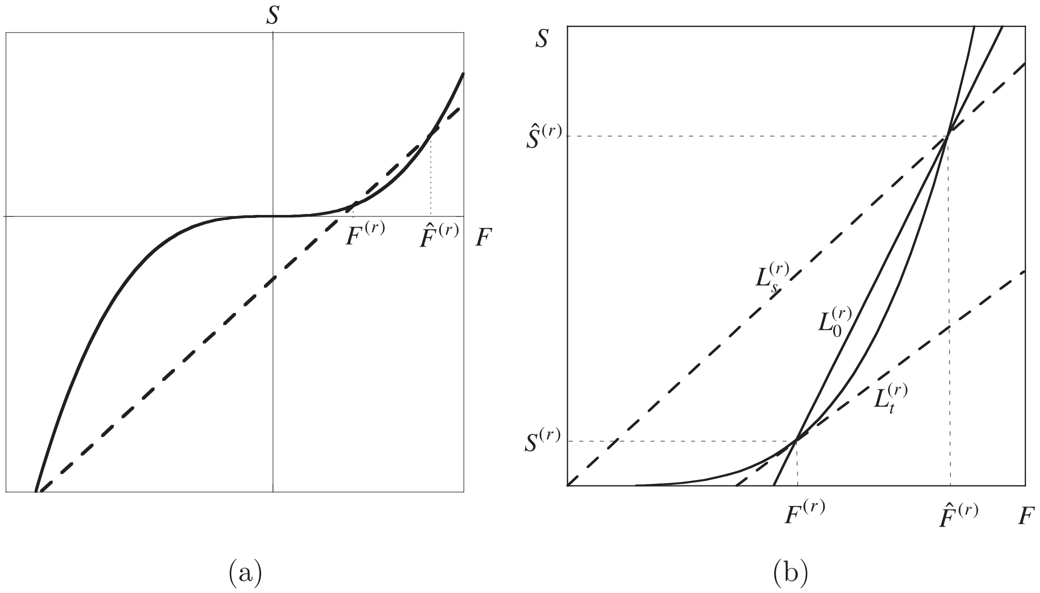


Figure 1. (a) Sketch of the nonlinear constitutive relation (continuous stress S vs. stretch F curve) and of its linear approximation (dashed line). (b) Schematic comparison of the "secant" ($\mathbf{L}_s^{(r)}$), "tangent" ($\mathbf{L}_t^{(r)}$) and new "generalized secant" ($\mathbf{L}_0^{(r)}$) approximations.

In the first relation, the symbol $\overline{\mathbf{F}}^{(r)}$ has been used to denote the phase averages of the deformation field $\langle\mathbf{F}\rangle^{(r)}$. Thus, the reference deformations $\mathbf{F}^{(r)}$ have been identified with the *phase averages* or *first moments* of the deformation field $\overline{\mathbf{F}}^{(r)}$. In the second relation, the symbol $\mathbf{C}_{\mathbf{F}}^{(r)}$ have been introduced to denote the *covariance tensor* of the *deformation fluctuations* in phase r (e.g., Bobeth & Diener [31]). Therefore, the variables $\hat{\mathbf{F}}^{(r)}$ have been associated with the second moments of the deformation field in the phases.

In connection with the above prescriptions, it is necessary to make the following clarifications. Concerning the prescription (17), it should be noted that relation (17) only makes stationary with respect to $\mathbf{F}^{(r)}$ the terms arising from the linear comparison energy $\widetilde{W}_0$. In other words, there are additional terms arising from the functions $V^{(r)}$, which have been neglected, for simplicity. Concerning the prescription (18), it needs to be emphasized that it is not possible to satisfy conditions (18) in full generality. This is due to the fact that the left-hand of relation (18) is a fourth-order tensor of rank 1, whereas the right-hand side is generally of full rank. This means that only certain components (or traces) of these expressions can be enforced. This point will be discussed in more detail in the context of the specific examples considered in the applications section. Generally speaking, the optimal choice of the variables $\mathbf{F}^{(r)}$ and $\mathbf{L}_0^{(r)}$ is still an open problem. However, it is known at least in the context of plasticity [32] that conditions (17) and appropriate traces of (18) lead to accurate estimates for the effective behavior. This suggests that even if the prescriptions (17) and (18) are not strictly optimal, they are still probably not far from optimal.

It follows from the above prescriptions that the secant-type condition (16) specializes to:

$$\frac{\partial W^{(r)}}{\partial \mathbf{F}}(\hat{\mathbf{F}}^{(r)}) - \frac{\partial W^{(r)}}{\partial \mathbf{F}}(\overline{\mathbf{F}}^{(r)}) = \mathbf{L}_0^{(r)}(\hat{\mathbf{F}}^{(r)} - \overline{\mathbf{F}}^{(r)}), \tag{20}$$

and that the expression (15) for the effective potential of the hyperelastic composite reduces to:

$$\widetilde{W}(\overline{\mathbf{F}}) = \sum_{r=1}^{N} c^{(r)} \left[W^{(r)}(\hat{\mathbf{F}}^{(r)}) - \frac{\partial W^{(r)}}{\partial \mathbf{F}}(\overline{\mathbf{F}}^{(r)}) \cdot (\hat{\mathbf{F}}^{(r)} - \overline{\mathbf{F}}^{(r)}) \right]. \tag{21}$$

In summary, the estimate (21) has been generated. Like the earlier "second-order" estimates [22], it depends on the phase averages $\overline{\mathbf{F}}^{(r)}$ of the deformation field in a suitably defined linear "thermoelastic" comparison composite, subject to the self-consistent prescription (17) on the reference variables $\mathbf{F}^{(r)}$. However, the new prescription (20) for the comparison moduli $\mathbf{L}_0^{(r)}$ is different from earlier *ad hoc* choices, being somewhat intermediate between the "secant" [25] and the "tangent" conditions [22]. In addition, the new estimate depends directly on the variables $\hat{\mathbf{F}}^{(r)}$, which, in turn, depend on (appropriate traces of) the "second moments" of the *fluctuations* $\mathbf{C}_{\mathbf{F}}^{(r)}$ of the deformation field in the phases of the linear comparison composite, as specified by the prescription (18). Furthermore, like the earlier "second-order" estimates, they are exact to second-order in the heterogeneity contrast [27].

It is remarked finally that the linear comparison problem (13) that needs to be considered for the determination of the phase averages $\overline{\mathbf{F}}^{(r)}$ and fluctuations $\mathbf{C}_{\mathbf{F}}^{(r)}$ needed in expression (21) for $\widetilde{W}$ is precisely the same that was considered by Ponte Castañeda and Tiberio [20] in the earlier version of the second-order method. These authors provided expressions of the Hashin–Shtrikman and self-consistent types [29] for the average deformations $\overline{\mathbf{F}}^{(r)}$ in the generalized N-phase "thermoelastic" composites (10), from which corresponding estimates may be generated for the corresponding effective stored-energy functions $\widetilde{W}_0$, and, in turn, for the fluctuations $\mathbf{C}_{\mathbf{F}}^{(r)}$ via (19). For brevity, the relevant general expressions will not be repeated here, and only the appropriate specialized versions of the results will be quoted in the applications sections for the special case of rigidly reinforced systems.

3. APPLICATION TO PARTICLE-REINFORCED ELASTOMERS

The second-order estimate (21) for the effective stored-energy function of hyperelastic composites applies for N-phase systems, including, with a suitable reinterpretation, polycrystalline aggregates of anisotropic phases. In this section, the special case of isotropic rigidly reinforced rubbers is considered. This case has already been considered using the earlier version of the second-order method [20] and these earlier results will be used here as a reference. Thus, the focus will be on two-phase composites consisting of rigid, spherical inclusions distributed isotropically with volume fraction $c^{(2)} = c$ in a hyperelastic matrix with energy function $W^{(1)} = W$, such that the composite is statistically isotropic in the undeformed configuration.

Because of the objectivity of $\widetilde{W}$, it suffices to consider macroscopic stretch loading histories (i.e. $\overline{\mathbf{F}} = \overline{\mathbf{U}}$; $\overline{\mathbf{R}} = \mathbf{I}$). Because of the spherical (isotropic) symmetry of the reinforcement and its distribution, it is expected [20] that the average rotation tensor of the rigid phase is the identity, so that the average deformation gradient in the inclusion phase is also equal to the identity (i.e. $\overline{\mathbf{F}}^{(2)} = \mathbf{I}$). It then follows trivially that the average deformation gradient in the hyperelastic phase is given by

$$\overline{\mathbf{F}}^{(1)} = \frac{1}{1-c}\left(\overline{\mathbf{U}} - c\,\mathbf{I}\right). \tag{22}$$

Note that $\overline{\mathbf{F}}^{(1)} = \overline{\mathbf{U}}^{(1)}$, so it is convenient to define the principal stretches associated with $\overline{\mathbf{U}}^{(1)}$ via $\overline{\lambda}_i^{(1)} = (\overline{\lambda}_i - c)/(1-c)$ $(i = 1,2,3)$, where $\overline{\lambda}_i$ $(i = 1,2,3)$ are the principal stretches associated with $\overline{\mathbf{U}}$. The above result would still be expected to apply, up to the onset of some possible instability, even if the shape or distribution (see Ponte Castañeda and Willis [33]) of the rigid phase were not spherical, provided that their symmetry axes were aligned with the principal directions of $\overline{\mathbf{U}}$. Otherwise, the reinforcement would undergo an average rigid rotation $\overline{\mathbf{R}}^{(2)}$, which would have to be determined from the full homogenization procedure described in the earlier sections of this paper. This would, of course, make the treatment of such cases more complicated. However, such more general analyses should lead to the result that $\overline{\mathbf{F}}^{(2)} = \overline{\mathbf{R}}^{(2)} = \mathbf{I}$ for the special case considered here where the reinforcement is isotropically distributed. Lahellec [21] has pursued this more general approach in the analogous context of a two-phase, periodic composite loaded symmetrically. (The case of rigid fibers was approximated by taking the contrast to be sufficiently large.)

3.1. Lower Bounds

Before proceeding with the second-order estimates, the above-mentioned "polyconvex" lower bounds of Ponte Castañeda [14] will be specialized here for later reference. Note that the classical Voigt upper bound is infinite in this case. It can be shown that specialization of the bound (8) to an isotropic rigidly reinforced elastomer with a compressible neo-Hookean matrix (7) leads to

$$\widetilde{W}_{PC}\left(\overline{\mathbf{U}}\right) = (1-c)\left[\frac{\mu}{2}\left(\overline{\mathbf{F}}^{(1)}\cdot\overline{\mathbf{F}}^{(1)} - 3\right) + \frac{\mu'}{2}\left(\frac{\bar{J}-1}{1-c}\right)^2 - \mu\ln\left(\frac{\bar{J}-c}{1-c}\right)\right], \tag{23}$$

where μ, μ' are the Lamé moduli of the elastic phase, $\overline{\mathbf{F}}^{(1)}$ is given by (22), and $\bar{J} = \overline{\lambda}_1\overline{\lambda}_2\overline{\lambda}_3$. In the derivation of this result, it has been assumed that the stored energy $W^{(2)}$ of the rigid phase is infinite unless $\overline{\mathbf{F}}^{(2)} = \mathbf{I}$, in which case it is zero. This assumption is consistent with the hypothesis that due to the isotropy of the rigid particles and their distribution, the particles do not rotate.

In the incompressible limit, $\mu' \to \infty$ the above lower bound reduces to $\widetilde{W}_{PC}^{I}\left(\overline{\mathbf{U}}\right) = \widetilde{\Phi}_{PC}^{I}\left(\overline{\lambda}_1, \overline{\lambda}_2, \overline{\lambda}_3\right)$, where

$$\widetilde{\Phi}^{I}_{PC}\left(\overline{\lambda}_1,\overline{\lambda}_2,\overline{\lambda}_3\right)=(1-c)\frac{\mu}{2}\left[\left(\frac{\overline{\lambda}_1-c}{1-c}\right)^2+\left(\frac{\overline{\lambda}_2-c}{1-c}\right)^2+\left(\frac{\overline{\lambda}_3-c}{1-c}\right)^2-3\right], \quad (24)$$

whenever $\bar{J}=\overline{\lambda}_1\overline{\lambda}_2\overline{\lambda}_3=1$ (and ∞ otherwise). Note that this bound is, therefore, consistent with the “exact” incompressibility constraint

$$g_E\left(\overline{\lambda}_1,\overline{\lambda}_2,\overline{\lambda}_3\right)=\overline{\lambda}_1\overline{\lambda}_2\overline{\lambda}_3-1=0, \quad (25)$$

expected on physical grounds (i.e., a composite with an incompressible matrix and rigid inclusions should be incompressible). However, this bound does not linearize properly, i.e., it does not reduce to the classical Reuss lower bound for infinitesimal deformations. In spite of this fact, this bound will prove to be useful below in checking the validity of the new second-order estimates to be developed next.

3.2. Second-Order Estimates

For the above-defined class of rigidly reinforced elastomers, the second-order estimate (21) reduces to

$$\widetilde{W}(\overline{\mathbf{U}})=(1-c)\left[W(\hat{\mathbf{F}}^{(1)})-\frac{\partial W}{\partial \mathbf{F}}(\overline{\mathbf{F}}^{(1)})\cdot(\hat{\mathbf{F}}^{(1)}-\overline{\mathbf{F}}^{(1)})\right], \quad (26)$$

where $\overline{\mathbf{F}}^{(1)}$ has already been specified in (22). It remains to determine the variable $\hat{\mathbf{F}}^{(1)}$, as well as the modulus tensor $\mathbf{L}_0$ of the matrix phase in the linear comparison composite, which can be achieved by means of the relation

$$\frac{\partial W}{\partial \mathbf{F}}(\hat{\mathbf{F}}^{(1)})-\frac{\partial W}{\partial \mathbf{F}}(\overline{\mathbf{F}}^{(1)})=\mathbf{L}_0(\hat{\mathbf{F}}^{(1)}-\overline{\mathbf{F}}^{(1)}), \quad (27)$$

together with suitably chosen traces of the relation

$$(\hat{\mathbf{F}}^{(1)}-\overline{\mathbf{F}}^{(1)})\otimes(\hat{\mathbf{F}}^{(1)}-\overline{\mathbf{F}}^{(1)})=\mathbf{C}^{(1)}_{\mathbf{F}}. \quad (28)$$

In this last relation,

$$\mathbf{C}^{(1)}_{\mathbf{F}}=\frac{2}{1-c}\,\frac{\partial \widetilde{W}_0}{\partial \mathbf{L}_0} \quad (29)$$

is the covariance of the fluctuations in the matrix phase of the linear comparison composite, with effective stored energy function given by

$$\widetilde{W}_0(\overline{\mathbf{U}})=(1-c)W(\overline{\mathbf{F}}^{(1)})+\frac{1}{2}(\overline{\mathbf{U}}-\mathbf{I})\cdot\left(\widetilde{\mathbf{L}}_0-\frac{1}{1-c}\mathbf{L}_0\right)(\overline{\mathbf{U}}-\mathbf{I}). \quad (30)$$

This last relation has been generated by making use of a generalization of Levin’s relation [34] for two-phase *thermoelastic* composites (see also [20]), letting phase two have the energy function

$$W^{(2)}(\mathbf{F}) = \frac{1}{2}\mu_0^{(2)}(\mathbf{F}-\mathbf{I})\cdot(\mathbf{F}-\mathbf{I}), \tag{31}$$

and taking the limit as $\mu_0^{(2)} \to \infty$ in the free energy expression (13). Again, note that the above form for $W^{(2)}$ is consistent with the requirement that $\overline{\mathbf{F}}^{(2)}$ should tend to $\mathbf{I}$ in the limit as $\mu_0^{(2)} \to \infty$. In expression (30), $\widetilde{\mathbf{L}}_0$ thus denotes the effective modulus tensor of a two-phase, *linear–elastic* comparison composite consisting of a distribution of rigid inclusions with volume fraction c in a matrix with elastic modulus $\mathbf{L}_0$ and the *same* microstructure as the nonlinear elastic composite (in its undeformed configuration).

It is emphasized that the above estimate for $\widetilde{W}$ is actually valid for *any* estimate for the effective modulus tensor $\widetilde{\mathbf{L}}_0$ of the linear comparison composite. For example, use can be made of the following *isotropic* Hashin–Shtrikman and self-consistent estimates for $\widetilde{\mathbf{L}}_0$:

$$\widetilde{\mathbf{L}}_0 = \begin{cases} L_0 + \frac{c}{1-c}\mathbf{P}^{-1} & HS, \\ L_0 + c\widetilde{\mathbf{P}}^{-1} & SC. \end{cases} \tag{32}$$

where $\mathbf{P}$ and $\widetilde{\mathbf{P}}$ are obtained by setting $\mathbf{L}^{(0)}$ equal to $\mathbf{L}_0$ and $\widetilde{\mathbf{L}}_0$, respectively, in the expression

$$\mathbf{P}^{(0)} = \frac{1}{4\pi}\int_{|\boldsymbol{\xi}|=1} \mathbf{H}^{(0)}(\boldsymbol{\xi})\,\mathrm{d}S, \tag{33}$$

with $K_{ik}^{(0)} = L_{ijkh}^{(0)}\xi_j\xi_h$, $\mathbf{N}^{(0)} = \mathbf{K}^{(0)^{-1}}$, $H_{ijkh}^{(0)}(\boldsymbol{\xi}) = N_{ik}^{(0)}\xi_j\xi_h$.

While fairly explicit, the above results, in general, require the computation of the tensor $\mathbf{P}$ (or $\tilde{\mathbf{P}}$), which depends on the anisotropy of $\mathbf{L}_0$ (or $\widetilde{\mathbf{L}}_0$). In turn, the anisotropy of these tensors depends on the functional form of the stored-energy function W and the loading configuration, as determined by $\overline{\mathbf{F}} = \overline{\mathbf{U}}$. In addition, the derivatives of the tensor $\mathbf{P}$ with respect to $\mathbf{L}_0$ are needed in the characterization of the fluctuations $\mathbf{C}_{\mathbf{F}}^{(1)}$, which requires further computations. In this work, which presents the first application of the (improved version of the) second-order method to finite elasticity, a simple, yet illustrative example, where the computation of the $\mathbf{P}$ tensor and its derivatives is simple, will be worked out in detail. Thus, estimates of the Hashin–Shtrikman type will be derived for *plane strain* loading of a *two-dimensional* fiber-reinforced composite. More general situations, including *uniaxial* and *shear* loading of *three-dimensional* particle-reinforced composites, and types of estimates will be presented elsewhere.

However, before specializing to the two-dimensional fiber-reinforced composite, it is noted here that when used together with the Reuss estimate for the effective modulus tensor ($\widetilde{\mathbf{L}}_0 = (1-c)^{-1}\mathbf{L}_0$), the second-order method yields the explicit result

$$\widetilde{W}(\overline{\mathbf{U}}) = (1-c)W\left[\frac{1}{1-c}\left(\overline{\mathbf{U}} - c\mathbf{I}\right)\right] \doteq \widetilde{W}_R(\overline{\mathbf{U}}). \tag{34}$$

This estimate agrees exactly with the corresponding estimate generated using the earlier version of the second-order method without fluctuations [20]. This is a direct consequence of the fact that the fluctuations in the Reuss theory are identically zero, so that there are no differences between the earlier and newer versions of the second-order theory. As already known [20], the Reuss estimate (34) is not necessarily a lower bound, except for small deformations, when the above result reduces (exactly to second order in the infinitesimal strain) to the classical Reuss lower bound. It is interesting to note that the estimate (34) was first obtained by Govindjee and Simo [18] by different means.

When the Reuss estimate (34) is specialized to a *compressible* neo-Hookean matrix phase with W given by the relation (7), it specializes to

$$\widetilde{W}_R(\overline{\mathbf{U}}) = (1-c)\left[\frac{\mu}{2}\left(\overline{\mathbf{F}}^{(1)}\cdot\overline{\mathbf{F}}^{(1)}-3\right)+\frac{\mu'}{2}\left(\bar{J}^{(1)}-1\right)^2-\mu\ln\left(\bar{J}^{(1)}\right)\right], \tag{35}$$

where μ, μ' are the Lamé moduli of the elastic phase, $\overline{\mathbf{F}}^{(1)}$ is given by (22), and $\bar{J}^{(1)} = \overline{\lambda}_1^{(1)}\overline{\lambda}_2^{(1)}\overline{\lambda}_3^{(1)}$ (with $\overline{\lambda}_i^{(1)} = (\overline{\lambda}_i - c)/(1-c)$).

It is interesting to remark that the Reuss estimate (35) differs from the lower bound (23) only through the terms that depend on the determinant. It can be shown that this estimate *violates* the bound (23) for certain loadings. For example, for loadings such that $\bar{J}^{(1)} = 1$, it can be verified that the Reuss estimate (35) actually lies below the bound (23).

From the result (35) it is possible to generate the corresponding result for an *incompressible* neo-Hookean matrix phase by considering the limit as μ' tends to infinity. The result may be written in the form $\widetilde{W}_R^I(\overline{\mathbf{U}}) = \widetilde{\Phi}_R^I(\overline{\lambda}_1,\overline{\lambda}_2,\overline{\lambda}_3)$, where

$$\widetilde{\Phi}_R^I(\overline{\lambda}_1,\overline{\lambda}_2,\overline{\lambda}_3) = (1-c)\frac{\mu}{2}\left[\left(\frac{\overline{\lambda}_1-c}{1-c}\right)^2+\left(\frac{\overline{\lambda}_2-c}{1-c}\right)^2+\left(\frac{\overline{\lambda}_3-c}{1-c}\right)^2-3\right]. \tag{36}$$

In this expression, the principal stretches $\overline{\lambda}_i$ are required to satisfy the "approximate" incompressibility constraint $\bar{J}^{(1)} = 1$, which can be written as

$$g_A(\overline{\lambda}_1,\overline{\lambda}_2,\overline{\lambda}_3) = \left(\frac{\overline{\lambda}_1-c}{1-c}\right)\left(\frac{\overline{\lambda}_2-c}{1-c}\right)\left(\frac{\overline{\lambda}_3-c}{1-c}\right)-1=0. \tag{37}$$

While exact to second-order in the infinitesimal strain, this "approximate" macroscopic incompressibility constraint is not identical to the "exact" constraint (25). Note that, because of this, the incompressible Reuss estimate (36) is really different from the incompressible polyconvex bound (24).

Thus, it appears that at least for the Reuss-type estimate, where the fluctuations are ignored, the second-order method yields predictions that are inaccurate in the incompressible limit and have been shown to violate a rigorous bound more generally. Within the context of the *earlier* second-order theory, it was found that even for estimates of the Hashin–Shtrikman type, the incompressible limit was troublesome, and direct application of the theory led to the same "approximate" macroscopic incompressibility constraint (37). For this reason, an "alternate" approach (see also [21] for yet a third approach) was proposed in reference [20] to avoid this limitation of the earlier version of the second-order theory. This approach consisted in evaluating the compressible term proportional to μ' directly. For later reference, the result of this "alternate" approach is recalled here:

$$\widetilde{\Phi}_R^A(\overline{\lambda}_1, \overline{\lambda}_2, \overline{\lambda}_3) = (1-c)\frac{\mu}{2}\left[\left(\frac{\overline{\lambda}_1 - c}{1-c}\right)^2 + \left(\frac{\overline{\lambda}_2 - c}{1-c}\right)^2 + \left(\frac{\overline{\lambda}_3 - c}{1-c}\right)^2 - 3 - 2\,\ln\left(\bar{J}^{(1)}\right)\right], \quad (38)$$

where the logarithmic term arises because the exact constraint (25) must be enforced (and therefore $\bar{J}^{(1)} = (\overline{\lambda}_1 - c)(\overline{\lambda}_2 - c)(\overline{\lambda}_3 - c)/(1-c)^3$ is not necessarily equal to 1).

Naturally, the hope is that the *new* version of the second-order theory, incorporating field fluctuations, should lead to better predictions, which should not only yield the "exact" incompressibility constraint in the limit of an incompressible matrix phase, but should also *not* violate any known bounds. This expectation will be explored in the next section in the context of the fiber-reinforced example mentioned earlier.

4. PLANE STRAIN LOADING OF TRANSVERSELY ISOTROPIC, FIBER-REINFORCED NEO-HOOKEAN COMPOSITES

4.1. Formulation

In this section, plane strain deformations of a fiber-reinforced composite are considered where the rigid fibers, which are perpendicular to the plane of the deformation, are aligned in the x_3 direction. The distribution of the reinforcement in the transverse plane is isotropic, so that the hypotheses that were made in the derivation of relation (26) for $\widetilde{W}$ carry over to this special case, with an appropriate (two-dimensional) modification of the relevant $\mathbf{P}$ tensor in the relevant expressions for $\widetilde{\mathbf{L}}_0$. Here, for simplicity, estimates of the Hashin–Shtrikman (HS) type $(32)_1$ will be determined for the special case of a neo-Hookean matrix phase with stored-energy function given by (7). The applied deformation $\overline{\mathbf{F}} = \overline{\mathbf{U}}$ in this case is entirely characterized by the two in-plane principal stretches $\overline{\lambda}_1$ and $\overline{\lambda}_2$, the out-of-plane principal stretch $\overline{\lambda}_3$ being identically 1.

Because of the transverse isotropy of the microstructure and the orthogonal symmetry of the loading condition, it is reasonable to assume that the linear comparison problem of relevance here will also exhibit orthotropic symmetry, with the symmetry axes aligned with the applied loading $\overline{\mathbf{F}} = \overline{\mathbf{U}}$. For plane strain conditions, it suffices to consider the in-plane components of a general deformation tensor $\mathbf{F}$ relative to the symmetry axes, which for convenience will be written as a vector in $\mathcal{R}^4$:

$$\left[\begin{array}{cccc} F_{11} & F_{22} & F_{12} & F_{21} \end{array}\right]^T. \quad (39)$$

The modulus tensor $\mathbf{L}_0$ of the linear comparison composite, which is expected to also exhibit orthotropic symmetry, will correspondingly be expressed as a matrix in $\mathcal{R}^{4\times4}$:

$$\begin{bmatrix} L_{1111} & L_{1122} & 0 & 0 \\ L_{1122} & L_{2222} & 0 & 0 \\ 0 & 0 & L_{1212} & L_{1221} \\ 0 & 0 & L_{1221} & L_{2121} \end{bmatrix}, \tag{40}$$

where the diagonal symmetry of the tensor $\mathbf{L}_0$ has been used (i.e. $L_{ijkl} = L_{klij}$).

Now, given the above assumptions, the tensor $\hat{\mathbf{F}}^{(1)}$ is seen to have at most four independent components ($\hat{F}_{11}^{(1)}$, $\hat{F}_{22}^{(1)}$, $\hat{F}_{12}^{(1)}$, $\hat{F}_{21}^{(1)}$,), which must be extracted from relation (28). This suggests that the tensor $\mathbf{L}_0$ should have at most four independent components, with respect to which $\widetilde{W}_0$ should be differentiated to generate four relations for the four components of $\hat{\mathbf{F}}^{(1)}$ using relation (29). Carrying this program out would, in effect, fix the traces of relation (28) to be considered. At the present time, it is not clear what the best choice for the components of $\mathbf{L}_0$ (and, therefore, for the traces of (28)) should be. Here, use will be made of the following prescriptions:

$$L_{1212} = L_{2121}, \quad \text{and} \quad L_{1221} + L_{1122} = \sqrt{(L_{1111} - L_{1212})\,(L_{2222} - L_{1212})}, \tag{41}$$

which reduce the components of the $\mathbf{L}_0$ to only four independent ones (L_{1111}, L_{2222}, L_{1122}, L_{1212}). The motivations for these choices are: (*i*) they are consistent with those satisfied by the components of the tangent modulus of a neo-Hookean material, expressed relative to the symmetry axes; and (*ii*) they simplify considerably the expressions for the tensor $\mathbf{P}$ (in fact, they lead to simple analytical results for all components, which are spelled out in Appendix A).

With these additional hypotheses, Equations (28)–(30), together with Equation $(32)_1$ for the Hashin–Shtrikman estimate for $\widetilde{\mathbf{L}}_0$, can be used to generate four equations for the four components of $\hat{\mathbf{F}}^{(1)}$, which are of the form

$$\begin{aligned} \left(\hat{F}_{11}^{(1)} - \bar{F}_{11}^{(1)}\right)^2 + 2f_1\hat{F}_{12}^{(1)}\hat{F}_{21}^{(1)} &= k_1 \\ \left(\hat{F}_{22}^{(1)} - \bar{F}_{22}^{(1)}\right)^2 + 2f_2\hat{F}_{12}^{(1)}\hat{F}_{21}^{(1)} &= k_2 \\ \left(\hat{F}_{12}^{(1)}\right)^2 + \left(\hat{F}_{21}^{(1)}\right)^2 + 2f_3\hat{F}_{12}^{(1)}\hat{F}_{21}^{(1)} &= k_3 \\ \left(\hat{F}_{11}^{(1)} - \bar{F}_{11}^{(1)}\right)\left(\hat{F}_{22}^{(1)} - \bar{F}_{22}^{(1)}\right) - \hat{F}_{12}^{(1)}\hat{F}_{21}^{(1)} &= k_4, \end{aligned} \tag{42}$$

where f_1, f_2, f_3, k_1, k_2, k_3, k_4 are functions of the components of $\mathbf{L}_0$ (or, more precisely, of the three ratios L_{1111}/L_{2222}, L_{1122}/L_{2222}, and L_{1212}/L_{2222}), as well as of the deformation $\overline{\mathbf{F}}$ and the fiber concentration c. These equations can be shown to have only two distinct solutions for $\hat{F}_{11}^{(1)}$ and $\hat{F}_{22}^{(1)}$, in terms of which $\hat{F}_{12}^{(1)}$ and $\hat{F}_{21}^{(1)}$ may be computed. Note that there are four possible solutions for these last two variables (this is because only the combinations $\hat{F}_{12}^{(1)}\hat{F}_{21}^{(1)}$

and $\left(\hat{F}_{12}^{(1)}\right)^2 + \left(\hat{F}_{21}^{(1)}\right)^2$ can be determined uniquely from these equations), but for a given root for $\hat{F}_{11}^{(1)}$ and $\hat{F}_{22}^{(1)}$, they all give the same predictions for the energy, so that they are all essentially identical. For completeness, it is noted that the two roots for $\hat{F}_{11}^{(1)}$ and $\hat{F}_{22}^{(1)}$ are given by

$$\begin{aligned} \hat{F}_{11}^{(1)} - \bar{F}_{11}^{(1)} &= \pm\frac{2f_1k_4 + k_1}{\sqrt{4f_1 2k_2 + 4f_1k_4 + k_1}}, \\ \hat{F}_{22}^{(1)} - \bar{F}_{22}^{(1)} &= \pm\frac{2f_1k_2 + k_4}{\sqrt{4f_1 2k_2 + 4f_1k_4 + k_1}}, \end{aligned} \tag{43}$$

where it is emphasized that the positive (and negative) signs in the roots for $\hat{F}_{11}^{(1)}$ and $\hat{F}_{22}^{(1)}$ go together.

Finally, for each of the two essentially distinct roots for the components of $\hat{\mathbf{F}}^{(1)}$ in terms of the four independent components of $\mathbf{L}_0$, two sets of four additional equations are generated for L_{1111}, L_{2222}, L_{1122}, and L_{1212} from the generalized secant conditions (27). Now, for the particular case of the neo-Hookean potential (7), one of these equations can be solved exactly for L_{1212}, giving the result $L_{1212} = \mu$. The remaining three equations must be solved numerically. Having computed the values of all the components of $\mathbf{L}_0$ for a given fiber volume fraction c, given material parameters (μ and μ'), and given loading ($\overline{\lambda}_1$ and $\overline{\lambda}_2$), the values of the components of $\hat{\mathbf{F}}^{(1)}$ can be computed using relations (43). Then, these results may be used together with the expression (22) for $\overline{\mathbf{F}}^{(1)}$ to compute the effective stored-energy function $\widetilde{W}$ for the rigidly reinforced composite using relation (26).

Some illustrative results will be presented in the next section and compared with earlier results and bounds. However, before doing this, the incompressible limit ($\mu' \to \infty$) of the effective stored energy is considered here. (Note that for actual rubbers $\mu'/\mu \approx 10^4$.) In this context, it is important to note that the above two distinct roots have very different asymptotic behaviors in the limit as μ' increases. The main distinguishing feature of the solutions associated with the two roots (43) of the equations (42) is that for one root, which is labeled the "positive" $(+)$ root, $\hat{J}^{(1)} = \det \hat{\mathbf{F}}^{(1)} \geq \bar{J}^{(1)} = \det \overline{\mathbf{F}}^{(1)}$, while for the other, labeled the "negative" $(-)$ root, the opposite is true.

For the negative-root solution, it can be shown that consideration of the incompressible limit of the energy for $\widetilde{W}$ leads to the "approximate" incompressibility constraint (37), in agreement with the corresponding limit obtained from the earlier version of the "second-order" theory (not incorporating field fluctuations). Because of this negative feature, and for reasons that will be detailed in the next section on results, this solution will not be pursued further here.

On the other hand, for the positive-root solution, it can be shown that the incompressible limit of $\widetilde{W}$ is consistent with the "exact" incompressibility constraint (25), and therefore consistent with the expected physics of the problem. The mathematical limit is a bit unusual in that some of the components of $\mathbf{L}_0$ (i.e. $L_{1111}, L_{1122}, L_{2222}$) become unbounded at *finite* values of μ', depending on μ, the loading level and the particle concentration. Further details are given in Appendix B, but the final result for the effective stored-energy function of the rigidly reinforced composite with a neo-Hookean matrix phase may be written as

$$\widetilde{\Phi}_{HS}^{I}\left(\overline{\lambda}_1,\overline{\lambda}_2\right) = \widetilde{\Phi}_{R}^{I}\left(\overline{\lambda}_1,\overline{\lambda}_2\right)+\frac{\mu}{2}\left(\frac{c}{1-c}\right)\left[\frac{\left(\overline{\lambda}_2-c\right)\left(\overline{\lambda}_1-1\right)^2}{\left(\overline{\lambda}_1-c\right)} + \frac{\left(\overline{\lambda}_1-c\right)\left(\overline{\lambda}_2-1\right)^2}{\left(\overline{\lambda}_2-c\right)}+\left(\overline{\lambda}_1-\overline{\lambda}_2\right)^2\right], \quad (44)$$

where $\widetilde{\Phi}_{R}^{I}$ is given by expression (36) with $\overline{\lambda}_3 = 1$, and it is emphasized that the exact incompressibility constraint (25) is satisfied. This result should be compared with the corresponding result (cf. (46) in 20) from the earlier version of the second-order procedure (without fluctuations), which, unlike (44), is inconsistent with the exact incompressibility constraint (25).

In the next section, comparisons will be made with the "alternate" version (see Appendix of [20]) of the old second-order estimate, which also leads to the exact incompressibility constraint, and is given by

$$\widetilde{\Phi}_{OHS}^{A}\left(\overline{\lambda}_1,\overline{\lambda}_2\right) = \widetilde{\Phi}_{R}^{A}\left(\overline{\lambda}_1,\overline{\lambda}_2\right)+\frac{\mu}{2}\left(\frac{c}{1-c}\right) \times\left[1+\frac{(1-c)^2}{(\overline{\lambda}_1-c)(\overline{\lambda}_2-c)}\right]\left[\left(\overline{\lambda}_1-1\right)^2+\left(\overline{\lambda}_2-1\right)^2\right]. \quad (45)$$

where $\widetilde{\Phi}_{R}^{A}$ is given by expression (38). Briefly, this estimate was generated by applying the old second-order method to only part of the energy, the additional terms being evaluated by other means. This required certain manipulations that were difficult to justify. The new estimate (44), on the other hand, is generated directly from the improved version of the second-order method, without the need of additional assumptions.

4.2. Results

Figure 2 provides a comparison of the new second-order estimates of the HS type with earlier estimates and bounds for a *compressible* neo-Hookean matrix phase with given moduli μ, and μ', reinforced with $c = 0.30$ rigid fibers. Results are shown as function of the applied stretch λ for two different types of loading: (*a*) pure shear with $\overline{\lambda}_1 = \lambda$ and $\overline{\lambda}_2 = 1/\lambda$, which satisfies the exact overall incompressibility constraint $\overline{J} = \det \overline{\mathbf{F}} = 1$; and (*b*) $\overline{\lambda}_1 = \lambda$ and $\overline{\lambda}_2$ chosen such that the "approximate" overall incompressibility constraint $\overline{J}^{(1)} = \det \overline{\mathbf{F}}^{(1)} = 1$ is satisfied. (It is emphasized that the composite is compressible and therefore should be able to accommodate both types of deformation conditions.) For completeness, both "roots" are shown for the new second-order estimates, respectively labelled $\widetilde{\Phi}_{HS}^{(+)}$ and $\widetilde{\Phi}_{HS}^{(-)}$ for the above-defined "positive-root" and "negative-root" solutions. They are compared against the polyconvex *lower* bound (23), denoted $\widetilde{\Phi}_{PC}$, the Reuss estimate (35), denoted $\widetilde{\Phi}_{R}$, and the old version [20] of the second-order HS estimates, labelled $\widetilde{\Phi}_{OHS}$. The energy function of the matrix phase is also shown in dashed lines for reference.

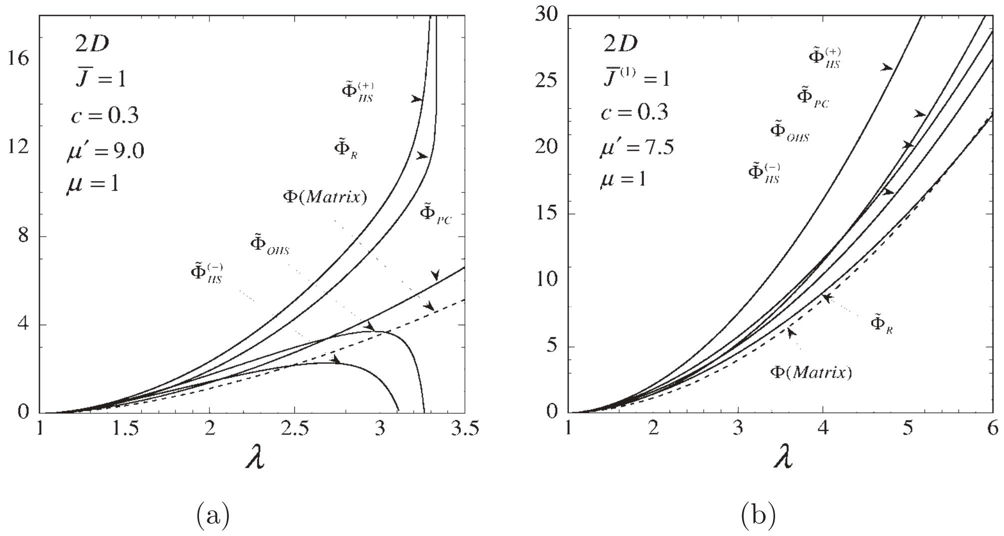
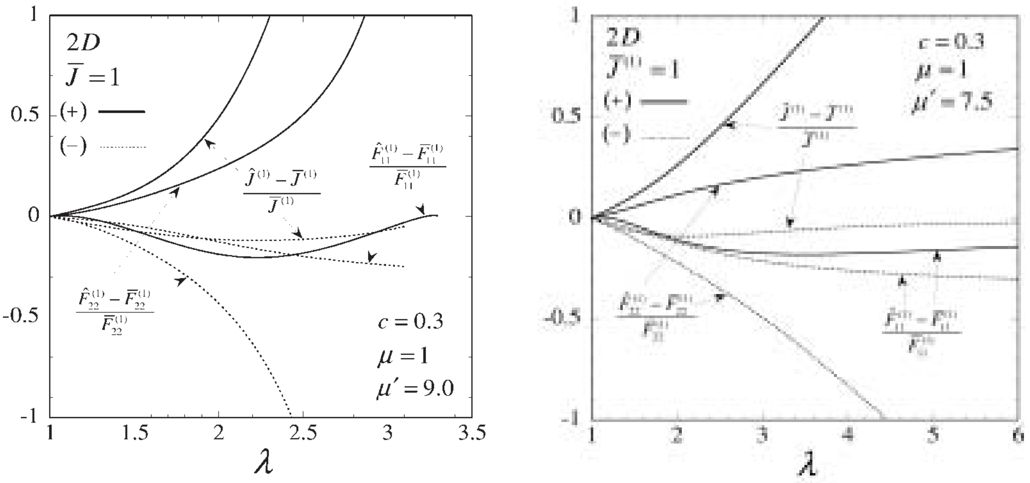


Figure 2. New second-order and other estimates and bounds for the effective stored energy of a compressible neo-Hookean rubber reinforced with a fixed concentration ($c = 0.3$) of aligned rigid fibers, as functions of the applied stretch λ. (a) Pure shear loading with $\overline{\lambda}_1 = \lambda$ and $\overline{\lambda}_2 = 1/\lambda$ and (b) loading with $\overline{\lambda}_1 = \lambda$ and $\overline{\lambda}_2$ such that $\bar{J}^{(1)} = 1$. The labels $\widetilde{\Phi}_{HS}^{(+)}$ and $\widetilde{\Phi}_{HS}^{(-)}$ correspond to the "positive" and "negative" roots of the new Hashin–Shtrikman second-order estimates, and the labels $\widetilde{\Phi}_{OHS}$, $\widetilde{\Phi}_R$, and $\widetilde{\Phi}_{PC}$ correspond to the old version [20] of the second-order HS estimates, the Reuss estimate and the polyconvex lower bound, respectively.

The main observation that can be made from Figure 2(a) is that while the new "positive-root" estimate $\widetilde{\Phi}_{HS}^{(+)}$ satisfies the polyconvex lower bound $\widetilde{\Phi}_{PC}$, both the "negative-root" estimate $\widetilde{\Phi}_{HS}^{(-)}$ and the old second-order estimate $\widetilde{\Phi}_{OHS}$ violate this bound at sufficiently large stretches λ. In fact, it can be seen from this figure that both $\widetilde{\Phi}_{HS}^{(-)}$ and $\widetilde{\Phi}_{OHS}$ have a seemingly unphysical behavior since they become lower than the matrix energy for sufficiently large stretches. This anomalous behavior for $\widetilde{\Phi}_{HS}^{(-)}$ and $\widetilde{\Phi}_{OHS}$ can be seen to be consistent with the above-mentioned observations that they both lead to overall incompressibility constraints that are inconsistent with the imposed deformation (i.e. pure shear). Even though the matrix phase is compressible, the value of μ' in this case ($\mu' = 9$) is sufficiently large to show the effect of the incompressible limit of these estimates, which again is inconsistent with the applied loading in this case. The implication of all of this is that the "positive-root" estimate $\widetilde{\Phi}_{HS}^{(+)}$ must be superior to both the "negative-root" estimate $\widetilde{\Phi}_{HS}^{(-)}$ and the old second-order estimate $\widetilde{\Phi}_{OHS}$, at least for nearly incompressible behavior for the matrix phase.

However, as shown in Figure 2(b), the "positive-root" estimate $\widetilde{\Phi}_{HS}^{(+)}$ also does better than the other two HS-type estimates when an overall loading condition is imposed that is consistent with the "approximate" incompressibility constraint ($\bar{J}^{(1)} = \det \overline{\mathbf{F}}^1 = 1$), so that the "negative-root" estimate $\widetilde{\Phi}_{HS}^{(-)}$ and the old second-order estimate $\widetilde{\Phi}_{OHS}$ would not be expected to blow up in the incompressible limit. (Note that $\mu' = 7.5$ in this case, which is



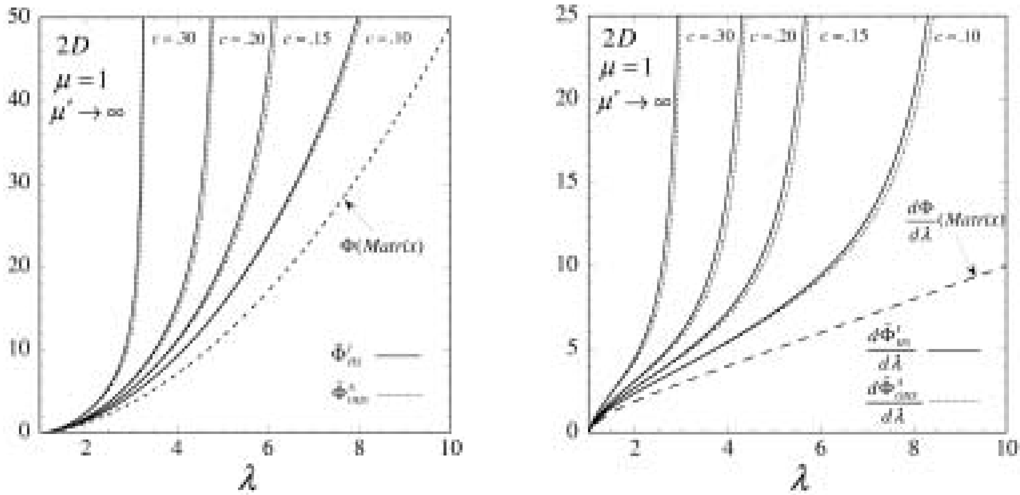
(a) (b)

Figure 3. Plots of the phase fluctuation measures $\hat{F}_{11}^{(1)} - \bar{F}_{11}^{(1)}$, $\hat{F}_{22}^{(1)} - \bar{F}_{22}^{(1)}$ and $\hat{J}^{(1)} - \bar{J}^{(1)}$, versus the stretch $\lambda$for the same reinforced, compressible, neo-Hookean rubbers considered in the context of Figure 2. (a) Pure shear loading with $\overline{\lambda}_1 = \lambda$ and $\overline{\lambda}_2 = 1/\lambda$; and (b) loading with $\overline{\lambda}_1 = \lambda$ and $\overline{\lambda}_2$ such that $\bar{J}^{(1)} = 1$.

also a relatively large value.) This figure also shows that the Reuss estimate (identical for the new and old second-order theories because of the lack of fluctuations) violates the bound in this case.

Putting these observations together with the earlier observations concerning the incompressible limits, the unescapable conclusion is that the "positive-root" estimate $\widetilde{\Phi}_{HS}^{(+)}$ must be superior to the "negative-root" estimate $\widetilde{\Phi}_{HS}^{(-)}$, as well as to the old second-order estimate $\widetilde{\Phi}_{OHS}$. Therefore, the new theory with fluctuations has been demonstrated to have the capability to give much improved predictions in finite elasticity, at least relative to the earlier theory [20].

In Figure 3, the new second-order predictions for the variables $\hat{F}_{11}^{(1)} - \bar{F}_{11}^{(1)}$ and $\hat{F}_{22}^{(1)} - \bar{F}_{22}^{(1)}$, as well as for the variable $\hat{J}^{(1)} - \bar{J}^{(1)}$, appropriately normalized by the corresponding phase averages, are given for compressible neo-Hookean rubbers reinforced by $c = 0.3$ of rigid particles. Both the predictions of the "positive" and "negative" roots are shown for completeness. Figure 3(a) and (b) give results for the two loadings identified in the context of Figure 2: (a) pure shear with $\overline{\lambda}_1 = \lambda$ and $\overline{\lambda}_2 = 1/\lambda$, and (b) $\overline{\lambda}_1 = \lambda$ and $\overline{\lambda}_2$ chosen such that $\bar{J}^{(1)} = 1$. Although the variables $\hat{F}_{11}^{(1)} - \bar{F}_{11}^{(1)}$ and $\hat{F}_{22}^{(1)} - \bar{F}_{22}^{(1)}$ cannot be identified exactly with the fluctuations of the deformation fields F_{11} and F_{22} over the matrix phase (because of the complex interactions among the various components of the deformation field arising from the selected traces of relation (28)), they do provide some measure of the fluctuations of the deformation field in the matrix. The main observation in the context of these plots is that $\hat{J}^{(1)} > \bar{J}^{(1)}$ for the positive root, which is physically more appealing than $\hat{J}^{(1)} < \bar{J}^{(1)}$

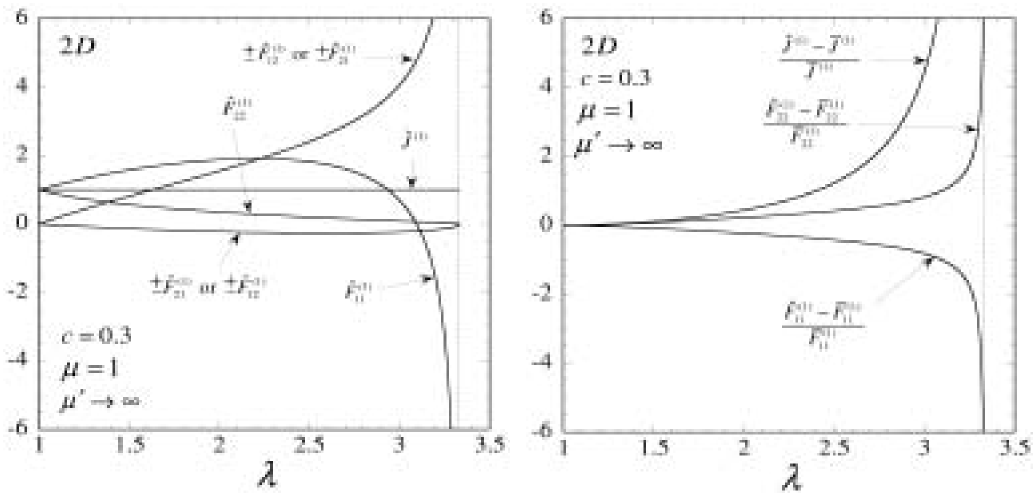


(a) (b)

Figure 4. Plots of the new (positive-root only) and old second-order predictions for the effective response of an incompressible neo-Hookean rubber reinforced with various concentrations c of aligned rigid fibers, and loaded in pure shear with $\overline{\lambda}_1 = \lambda$ and $\overline{\lambda}_2 = 1/\lambda$. (a) The stored-energy functions $\widetilde{\Phi}^I_{HS}$ and $\widetilde{\Phi}^A_{OHS}$; and (b) the corresponding stresses $S = \mathrm{d}\widetilde{\Phi}^I_{HS}/\,\mathrm{d}\lambda$ and $\mathrm{d}\widetilde{\Phi}^A_{OHS}/\,\mathrm{d}\lambda$, both as functions of the applied stretch λ.

for the negative root. In addition, it appears that there are considerable differences between the two roots, for a given loading, and between the predictions of the same root, for the two different loadings, which is consistent with the observations already made in the context of the energies.

In Figure 4, plots are given for the new second-order estimates ("positive root" only) for the stored-energy function $\widetilde{\Phi}^I_{HS}$ and corresponding stress $S = \mathrm{d}\widetilde{\Phi}^I_{HS}\,/\,\mathrm{d}\lambda$, as functions of the applied stretch λ, for an *incompressible* neo-Hookean material reinforced by rigid fibers at various concentrations c, subjected to pure shear $\overline{\lambda}_1 = \lambda$ and $\overline{\lambda}_2 = 1/\lambda$. These new second-order results were obtained by making use of the explicit expression (44) and are compared with the "alternate" form of the earlier version [20] of the second-order theory, as given by expression (45) and shown in dotted lines. First of all, note that the behavior of the composite is quite different from that of the neo-Hookean matrix phase in that it becomes much stiffer as the applied stretch $\overline{\lambda}$ tends to $1/c$, where the composite is found to lock up (i.e. the energy and the stress blow up). This is an interesting feature that was already predicted by the earlier version of the theory [20] and is confirmed by the more accurate results arising from the improved theory incorporating fluctuations. It is also interesting to remark that the predictions of the new second-order theory are in fact very close to the corresponding predictions of the "alternate" version of the old second-order theory. The fact that these two estimates, which have been generated by very different methods, are quite close may suggest that the predictions generated are fairly accurate in this case. In turn, this similarity in the predictions of the two theories is consistent with the expectation that the



(a) (b)

Figure 5. Plots of the phase fluctuations variable $\hat{\mathbf{F}}^{(1)}$ versus the stretch λ for an incompressible neo-Hookean rubber reinforced with $c = 0.3$ concentration of aligned rigid fibers, and loaded in pure shear with $\overline{\lambda}_1 = \lambda$ and $\overline{\lambda}_2 = 1/\lambda$. (a) The four non-zero components of $\hat{\mathbf{F}}^{(1)}$, as well as $\hat{J}^{(1)}$; and (b) the normalized field fluctuations variables $\hat{F}^{(1)}_{11} - \bar{F}^{(1)}_{11}$, $\hat{F}^{(1)}_{22} - \bar{F}^{(1)}_{22}$ and $\hat{J}^{(1)} - \bar{J}^{(1)}$.

most significant nonlinearities giving rise to large fluctuations are associated with the strongly nonlinear incompressibility constraint. The fact that the fluctuations are not used in the old second-order theory is compensated in its "alternate" version by the exact computation of the terms associated with the determinant constraint. The new second-order theory, using fluctuations, is robust enough to handle the incompressibility constraint directly (without the need of any special treatment for the determinant terms in the energy expression).

Finally, in Figure 5, plots are given for the variables $\hat{F}^{(1)}_{11}$, $\hat{F}^{(1)}_{22}$, $\hat{F}^{(1)}_{12}$ and $\hat{F}^{(1)}_{21}$ for the incompressible neo-Hookean rubber reinforced with $c = 0.3$ of rigid fibers. Figure $5(a)$ gives the raw values of these variables, which demonstrate that $\hat{J}^{(1)} = 1$, a feature that was critical in the asymptotic solution for nearly incompressible behavior, as shown in Appendix B. As can be deduced from Equations (42), the basic variables in the analysis are $\hat{F}^{(1)}_{11} - \bar{F}^{(1)}_{11}$ and $\hat{F}^{(1)}_{22} - \bar{F}^{(1)}_{22}$, and the combinations $\hat{F}^{(1)}_{12}\hat{F}^{(1)}_{21}$ and $\left(\hat{F}^{(1)}_{12}\right)^2 + \left(\hat{F}^{(1)}_{21}\right)^2$. These combinations of variables allow the determination of the effective energy and associated stress in a unique manner, but do not allow the unique determination of the variables $\hat{F}^{(1)}_{12}$ and $\hat{F}^{(1)}_{21}$ themselves. This is the reason for the multiple labels on these curves. Figure $5(b)$ gives plots of the variables $\hat{F}^{(1)}_{11} - \bar{F}^{(1)}_{11}$ and $\hat{F}^{(1)}_{22} - \bar{F}^{(1)}_{22}$, normalized by the corresponding phase averages. Although these variables cannot be identified exactly with the fluctuations of the deformation fields F_{11} and F_{22} over the matrix phase (because of the complex interactions among the various components of the deformation field arising from the selected traces of relation (28)), they do provide some measure of the fluctuations of the deformation field in the matrix. With the given normalizations, it is seen that these fluctuation variables preserve the symmetry of the loading (pure shear) and increase with increasing stretch, blowing up at $\overline{\lambda} = 1/c$. A scalar measure of the fluctuations, which also incorporates dependence on the variables $\hat{F}^{(1)}_{12}$ and $\hat{F}^{(1)}_{21}$, is provided by $\hat{J}^{(1)} - \bar{J}^{(1)}$. This variable also suggests that the fluctuations, suitably normalized by $\bar{J}^{(1)}$, increase with λ from zero at $\lambda = 1$ to $\lambda = 1/c$, when it blows up.

5. CONCLUDING REMARKS

In this paper, the recently proposed "second-order" homogenization method [24] has been extended to finite elasticity and applied to estimate the macroscopic response of particle- and fiber-reinforced elastomers subjected to large deformations. The resulting predictions appear to be quite good, exhibiting two qualitative features of special note.

First, the predicted constitutive behavior for rigidly reinforced composites with *incompressible* neo-Hookean matrix phases turn out to be incompressible in an overall sense. Simple as this requirement may be from the physical point of view, it is a non-trivial mathematical result due to the strong nonlinearities associated with the incompressibility of the matrix phase ($\det \mathbf{F} = 1$). In fact, it is known that an earlier version of the method [20], which neglected the use of the field fluctuations, led to predictions for the overall response of such rigidly reinforced incompressible elastomers that were not consistent with the overall incompressibility constraint, except in the limit of small deformations. The fact that the new second-order estimates are consistent with the overall incompressibility constraint for arbitrarily large deformations is an accomplishment for the theory, and serves to provide further evidence that the fluctuations are essential in generating accurate estimates for the effective behavior of nonlinear composites, in general, especially when such fluctuations are expected to be significant.

Second, the predictions for the effective response of such incompressible elastomers is found to exhibit a curious "lock-up" phenomenon at a finite stretch, even when the matrix behavior is assumed to allow arbitrarily large stretches. This interesting feature, which had already been observed in the context of the earlier version of the theory [20], can only be explained in terms of the evolution of the microstructure produced by finite changes in geometry, and serves to provide further evidence of the strength of the second-order homogenization methods in terms of capturing the effects of these additional nonlinearities in the field equations. A curious consequence of this lock-up phenomenon is the fact that the overall stress–strain relations for these materials exhibit a familiar "S" shape, even for relatively small concentrations of the rigid particles. This is in spite of the fact that the matrix phase, taken to be neo-Hookean, does not have such a shape.

APPENDIX A

In this appendix, the in-plane components of the tensor $\mathbf{P}$ associated with the orthotropic modulus tensor $\mathbf{L}_0$, given by expression (40) and subjected to constraints (41), are computed for the special case of cylindrical inclusions with circular cross section. In this case, the general expression (33) for $\mathbf{P}$ reduces to

$$\mathbf{P} = \frac{1}{2\pi}\int_{\xi_1^2+\xi_2^2=1} H_{ijkl}\ (\xi_1, \xi_2, \xi_3 = 0)\,\mathrm{d}S, \tag{46}$$

where it has been assumed that the fibers are aligned in the x_3 direction (note that the "surface" integral is evaluated over the unit circle).

Now, using the change of variables defined by $\xi_1 = \cos(\theta)$ and $\xi_2 = \sin(\theta)$, it follows that the non-vanishing (in-plane) components of $\mathbf{P}$ can be expressed as

$$\begin{aligned}
P_{1111} &= \frac{1}{2\pi}\int_0^{2\pi} \frac{K_{22}\cos^2(\theta)}{\det\mathbf{K}}\,\mathrm{d}\theta = \frac{1+\frac{L_{2222}}{L_{1212}}+2\sqrt{\frac{L_{2222}}{L_{1111}}}}{2L_{1111}\left(1+\sqrt{\frac{L_{2222}}{L_{1111}}}\right)^2}\\
P_{2222} &= \frac{1}{2\pi}\int_0^{2\pi} \frac{K_{11}\sin^2(\theta)}{\det\mathbf{K}}\,\mathrm{d}\theta = \frac{1+\frac{L_{1111}}{L_{1212}}+2\sqrt{\frac{L_{1111}}{L_{2222}}}}{2L_{1111}\left(1+\sqrt{\frac{L_{2222}}{L_{1111}}}\right)^2}\\
P_{1122} &= \frac{1}{2\pi}\int_0^{2\pi} \frac{-K_{12}\cos(\theta)\sin(\theta)}{\det\mathbf{K}}\,\mathrm{d}\theta = \frac{-\left(L_{1221}+L_{1122}\right)}{2L_{1111}L_{1212}\left(1+\sqrt{\frac{L_{2222}}{L_{1111}}}\right)^2}\\
P_{1212} &= \frac{1}{2\pi}\int_0^{2\pi} \frac{K_{22}\sin^2(\theta)}{\det\mathbf{K}}\,\mathrm{d}\theta = \frac{1+\frac{L_{2222}}{L_{1212}}+2\frac{L_{2222}}{L_{1212}}\sqrt{\frac{L_{1111}}{L_{2222}}}}{2L_{1111}\left(1+\sqrt{\frac{L_{2222}}{L_{1111}}}\right)^2}\\
P_{2121} &= \frac{1}{2\pi}\int_0^{2\pi} \frac{K_{11}\cos^2(\theta)}{\det\mathbf{K}}\,\mathrm{d}\theta = \frac{1+\frac{L_{1111}}{L_{1212}}+2\frac{L_{1111}}{L_{1212}}\sqrt{\frac{L_{2222}}{L_{1111}}}}{2L_{1111}\left(1+\sqrt{\frac{L_{2222}}{L_{1111}}}\right)^2}\\
P_{1221} &= P_{1122}.
\end{aligned} \tag{47}$$

Note that the tensor $\mathbf{P}$ exhibits both major symmetry, $P_{ijkl} = P_{klij}$, as well as orthotropic symmetry, consistent with similar requirements for $\mathbf{L}_0$. Also note that, due to the symmetry of the modulus tensor $\mathbf{L}_0$ and the type of loading considered in this paper (i.e. $\overline{\mathbf{F}} = \overline{\mathbf{U}}$; $\overline{\mathbf{R}} = \mathbf{I}$), the only relevant components that enter in the homogenization process are P_{1111}, P_{2222} and P_{1122}. The other components have been included in this appendix for completeness.

APPENDIX B

In this appendix, additional details are presented on the incompressible limit associated with the "positive" root of the new second-order method applied to a neo-Hookean-type material reinforced with rigid fibers. The asymptotic solution resulting from this heuristic derivation has been checked to give good agreement with the full numerical solution. This limit is a bit unusual in the sense that some of the components of the modulus tensor $\mathbf{L}_0$ become unbounded at a finite value, μ^*, of the Lamé modulus μ' of the elastomeric matrix, which depends on the loading, material parameters, and fiber concentration. Thus, motivated by the numerical solution for general μ', an expansion is attempted in the limit as $\mu' \to \mu^*$ of the following form:

$$\begin{aligned} L_{2222} &= \frac{1}{\Delta} \\ L_{1111} &= \frac{a_1}{\Delta} + a_2 + \mathrm{O}(\Delta) \\ L_{1122} &= \frac{b_1}{\Delta} + b_2 + \mathrm{O}(\Delta) \\ L_{1212} &= \frac{c_1}{\Delta} + c_2 + \mathrm{O}(\Delta) \\ L_{1221} &= \frac{d_1}{\Delta} + d_2 + \mathrm{O}(\Delta), \end{aligned} \tag{48}$$

where, by definition, $\Delta = 1/L_{2222}$ is a small parameter and $a_1, b_1, c_1, d_1, a_2, b_2, c_2$, and d_2 are unknown coefficients (more precisely, they are ratios between the components of $\mathbf{L}_0$) that ultimately depend on the applied loading, the material parameters, and the fiber concentration.

Now, as already mentioned earlier, the constraint $(41)_1$, together with the generalized secant conditions (27), can be shown to lead to the exact result that $L_{1212} = \mu$. In turn, the constraint $(41)_2$ can be used to solve for L_{1221} in terms of the other components of $\mathbf{L}$. Because of these facts, the following simplifications are obtained:

$$c_1 = 0, \quad c_2 = \mu, \quad d_1 = \sqrt{a_1} - b_1 \quad \textit{and} \quad d_2 = \frac{a_2 - \mu\,(a_1 + 1)}{2\sqrt{a_1}} - b_2. \tag{49}$$

Next, using the relations (48) and (49), the general expressions (43) for the ("positive" root) components of $\hat{\mathbf{F}}^{(1)} - \overline{\mathbf{F}}^{(1)}$ lead to expansions of the type

$$\begin{aligned} \hat{F}^{(1)}_{11} - \overline{F}^{(1)}_{11} &= x_1(a_1) + x_2(a_1, a_2)\Delta + \mathrm{O}(\Delta^2) \\ \hat{F}^{(1)}_{22} - \overline{F}^{(1)}_{22} &= y_1(a_1) + y_2(a_1, a_2)\Delta + \mathrm{O}(\Delta^2) \\ \hat{F}^{(1)}_{12} &= u_1(a_1) + u_2(a_1, a_2)\Delta + \mathrm{O}(\Delta^2) \\ \hat{F}^{(1)}_{21} &= v_1(a_1) + v_2(a_1, a_2)\Delta + \mathrm{O}(\Delta^2). \end{aligned} \tag{50}$$

The explicit form of the coefficients in these expansions is too cumbersome to be included here. Instead, only the unknown arguments upon which they depend have been specified. For instance, note that the leading term of all the components of $\hat{\mathbf{F}}^{(1)} - \overline{\mathbf{F}}^{(1)}$ depend solely on a_1.

At this point, expressions (48) and (50) can be introduced into the three reduced (i.e. using $L_{1212} = \mu$) generalized secant conditions to obtain a hierarchical system of equations. The leading order terms $O\left(\Delta^{-1}\right)$ of these equations are given by

$$a_1 x_1 + b_1 y_1 = 0, \quad b_1 x_1 + y_1 = 0, \quad \sqrt{a_1} - b_1 = 0, \tag{51}$$

where the arguments of x_1 and y_1 have been omitted for brevity. Note that setting the determinant associated with expressions $(51)_1$ and $(51)_2$ to zero implies $(51)_3$. Furthermore, it can be shown that the equation system (51) can be trivially satisfied by x_1 and y_1 (i.e. $y_1 = -\sqrt{a_1} x_1$). In summary, the terms of order $O\left(\Delta^{-1}\right)$ yield only one new condition, namely that $b_1 = \sqrt{a_1}$.

After some simplification, the terms of order $O\left(\Delta^{0}\right)$ in the generalized secant equations can be shown to reduce to

$$\begin{aligned}
\frac{\sqrt{c}\mu\left(1+b_1\right)\left|\left(\overline{\lambda}_2 - 1\right) - b_1\left(\overline{\lambda}_1 - 1\right)\right|}{\sqrt{2}(1-c)a_1^{1/4}} &= -\frac{\overline{\lambda}_2^{(1)}}{\hat{j}^{(1)}}\mu + \frac{\mu}{\overline{\lambda}_1^{(1)}} + \mu^* \overline{\lambda}_2^{(1)} (\hat{j}^{(1)} - \overline{J}^{(1)}) \\
\frac{\sqrt{c}\mu\left(1+b_1\right)\left|\left(\overline{\lambda}_2 - 1\right) - b_1\left(\overline{\lambda}_1 - 1\right)\right|}{\sqrt{2}(1-c)a_1^{3/4}} &= -\frac{\overline{\lambda}_1^{(1)}}{\hat{j}^{(1)}}\mu + \frac{\mu}{\overline{\lambda}_2^{(1)}} + \mu^* \overline{\lambda}_1^{(1)} (\hat{j}^{(1)} - \overline{J}^{(1)}) \\
b_2 = \frac{a_2 - \mu(a_1+1)}{2\sqrt{a_1}} + \mu^*(\hat{j}^{(1)} - 1) &- \frac{\mu}{\hat{j}^{(1)}},
\end{aligned} \tag{52}$$

where $\hat{j}^{(1)} = (x_1 + \overline{\lambda}_1^{(1)})(y_1 + \overline{\lambda}_2^{(1)}) - u_1 v_1$, which depends exclusively on a_1, is the first term in the expansion of $\hat{J}^{(1)}$ $(i.e.\, \hat{J}^{(1)} = \hat{j}^{(1)} + O\left(\Delta\right))$. It is noted now that $(51)_1$ and $(52)_2$ constitute a system of two equations for the two unknowns a_1 and μ^* (with b_1 being known in terms of a_1). Furthermore, Equation $(53)_3$ establishes a relationship between a_2 and b_2 in an analogous manner to the relationship established between a_1 and b_1 by $(51)_3$. This structure suggests that the coefficients a_i and b_i ($i = 1, 2, 3, \ldots,$), along with the corrections to μ^*, could be determined from the generalized secant equations of $O\left(\Delta^{i-1}\right)$ (although this will not be pursued here).

Returning to the problem involving a_1 and μ^*, it is easy to show from $(52)_1$ and $(52)_2$ that

$$a_1 = \left(\frac{\overline{\lambda}_2^{(1)}}{\overline{\lambda}_1^{(1)}} \right)^2 \quad \text{and} \quad \mu^* = \frac{\frac{\sqrt{c}\mu\,(1+\sqrt{a_1})\left|\left(\overline{\lambda}_2-1\right)-\sqrt{a_1}\left(\overline{\lambda}_1-1\right)\right|}{\sqrt{2}(1-c)a_1^{1/4}} + \frac{\overline{\lambda}_2^{(1)}}{\hat{j}^{(1)}}\mu - \frac{\mu}{\overline{\lambda}_1^{(1)}}}{\overline{\lambda}_2^{(1)}\left(\hat{j}^{(1)} - \bar{J}^{(1)}\right)}. \tag{53}$$

The results given by expressions (49), (51) and (53) suffice to characterize – through (48) and (50) – the leading order terms of the components of $\mathbf{L}_0$ and $\hat{\mathbf{F}}^{(1)} - \overline{\mathbf{F}}^{(1)}$, as well as the value of μ^*.

Next, the limit $\mu' \to \infty$ is considered in the context of expression (26) for the effective stored energy function of the rigidly reinforced, neo-Hookean, elastomer. In this connection, it is important to note that the required expression for $\hat{\mathbf{F}}^{(1)}$ does not change for values of μ' greater than μ^*. ($\overline{\mathbf{F}}^{(1)}$ is, of course, also independent of μ'.) This means that for sufficiently large values of μ', the effective stored energy function takes the form

$$\widetilde{W}(\overline{\mathbf{U}}) = \mu' g\left(\overline{\lambda}_1, \overline{\lambda}_2\right) + \tilde{\Phi}\left(\overline{\lambda}_1, \overline{\lambda}_2\right), \tag{54}$$

where it is emphasized that $\tilde{\Phi}$ is independent of μ', and

$$g\left(\overline{\lambda}_1, \overline{\lambda}_2\right) = \frac{1}{2}(\hat{j}^{(1)} - 1)^2 - \left(\bar{J}^{(1)} - 1\right)\left[\hat{j}^{(1)} - \frac{\overline{\lambda}_1\overline{\lambda}_2 - c}{1-c}\right] = 0. \tag{55}$$

As already discussed in the body of the text, consideration of the limit as $\mu' \to \infty$ leads to the overall incompressibility constraint given by $g\left(\overline{\lambda}_1, \overline{\lambda}_2\right) = 0$. On the other hand, using $(53)_1$, it follows from the definition of $\hat{j}^{(1)}$ that

$$\hat{j}^{(1)} = \frac{\overline{\lambda}_1\overline{\lambda}_2 - c}{1-c}, \tag{56}$$

which, together with (55), can be shown to lead to the condition

$$\bar{J} = \overline{\lambda}_1\overline{\lambda}_2 = 1, \tag{57}$$

which is nothing more than the “exact” incompressibility constraint (25), specialized to plane strain conditions. With this condition, the effective stored energy function for the *incompressible*, rigidly reinforced, neo-Hookean elastomer is generated from the left-over terms, labelled $\tilde{\Phi}$ in expression (54). The resulting explicit expression is given by (44).

It is interesting to remark that the new version of the second-order method predicts incompressible overall behavior even for rigidly reinforced neo-Hookean rubbers that have sufficiently high (but not necessarily infinite) values of μ' (i.e. for $\mu' \geq \mu^*$. This type of prediction, although perhaps not strictly correct, is probably very accurate. Physically, this is related to the fact that the overall incompressibility of the composite is expected to increase with increasing values of the volume fraction of the rigid fibers in the compressible

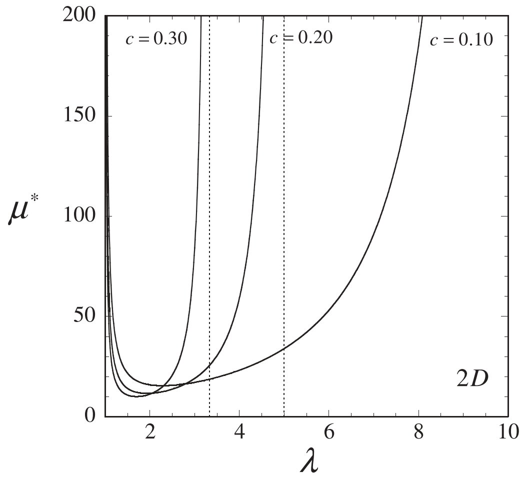


Figure 6. Plot of μ^* as a function of the stretch λ for various concentrations c of aligned rigid fibers under pure shear loading $\overline{\lambda}_1 = \lambda$ and $\overline{\lambda}_2 = 1/\lambda$.

elastomeric matrix. Figure (6) shows the limiting values μ^* at which the second-order method predicts the onset of incompressible behavior as a function of the deformation for various concentrations c of rigid fibers. Note that $\mu^* \rightarrow \infty$ as $\lambda \rightarrow 1$ and $\lambda \rightarrow 1/c$. This asymptotic behavior is consistent with the full numerical solution as well as with small-strain linearization conditions.

For completeness, the expressions for the leading order terms in the expansions of $\hat{F}_{11}^{(1)} - \overline{F}_{11}^{(1)}$ and $\hat{F}_{22}^{(1)} - \overline{F}_{22}^{(1)}$, from which the corresponding expressions for $\hat{F}_{12}^{(1)}$ and $\hat{F}_{21}^{(1)}$ can be readily determined (recall that $\hat{J}^{(1)} = 1$), are given by

$$
\begin{aligned}
\hat{F}_{11}^{(1)} - \bar{F}_{11}^{(1)} &= \frac{\sqrt{c}\left[\left(\dfrac{\overline{\lambda}_2^{(1)}}{\overline{\lambda}_1^{(1)}}\right)^2 \left(\overline{\lambda}_1 - 1\right)^2 - \left(\overline{\lambda}_2 - 1\right)^2\right]}{\sqrt{2}(1-c)\left[\left(\dfrac{\overline{\lambda}_2^{(1)}}{\overline{\lambda}_1^{(1)}}\right)^{1/2} + \left(\dfrac{\overline{\lambda}_2^{(1)}}{\overline{\lambda}_1^{(1)}}\right)^{3/2}\right]\left|\left(\overline{\lambda}_2 - 1\right) - \frac{\overline{\lambda}_2^{(1)}}{\overline{\lambda}_1^{(1)}}\left(\overline{\lambda}_1 - 1\right)\right|} \\
\hat{F}_{22}^{(1)} - \bar{F}_{22}^{(1)} &= \frac{-\sqrt{c}\left(\dfrac{\overline{\lambda}_2^{(1)}}{\overline{\lambda}_1^{(1)}}\right)\left[\left(\dfrac{\overline{\lambda}_2^{(1)}}{\overline{\lambda}_1^{(1)}}\right)^2 \left(\overline{\lambda}_1 - 1\right)^2 - \left(\overline{\lambda}_2 - 1\right)^2\right]}{\sqrt{2}(1-c)\left[\left(\dfrac{\overline{\lambda}_2^{(1)}}{\overline{\lambda}_1^{(1)}}\right)^{1/2} + \left(\dfrac{\overline{\lambda}_2^{(1)}}{\overline{\lambda}_1^{(1)}}\right)^{3/2}\right]\left|\left(\overline{\lambda}_2 - 1\right) - \frac{\overline{\lambda}_2^{(1)}}{\overline{\lambda}_1^{(1)}}\left(\overline{\lambda}_1 - 1\right)\right|}
\end{aligned}
\tag{58}
$$

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