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# A general hyperelastic model for incompressible fiber-reinforced elastomers

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## ABSTRACT

This work presents a new constitutive model for the effective response of fiber-reinforced elastomers at finite strains. The matrix and fiber phases are assumed to be incompressible, isotropic, hyperelastic solids. Furthermore, the fibers are taken to be perfectly aligned and distributed randomly and isotropically in the transverse plane, leading to overall transversely isotropic behavior for the composite. The model is derived by means of the “second-order” homogenization theory, which makes use of suitably designed variational principles utilizing the idea of a “linear comparison composite.” Compared to other constitutive models that have been proposed thus far for this class of materials, the present model has the distinguishing feature that it allows consideration of behaviors for the constituent phases that are more general than Neo-Hookean, while still being able to account directly for the shape, orientation, and distribution of the fibers. In addition, the proposed model has the merit that it recovers a known exact solution for the special case of incompressible Neo-Hookean phases, as well as some other known exact solutions for more general constituents under special loading conditions.

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## 1. Introduction

Fiber-reinforced, elastomer-matrix composites constitute a broadly utilized class of materials in engineering applications. In addition, fiber-reinforced-type morphologies appear naturally in a number of other “soft” matter systems of current interest. Prominent examples include nano-structured thermoplastic elastomers (see, e.g., Honeker and Thomas, 1996; Honeker et al., 2000) and soft biological tissues (see, e.g., Finlay et al., 1998; Quapp and Weiss, 1998). It is often the case that such fiber-reinforced “soft” materials are subjected to finite deformations, and it is therefore of practical interest to develop constitutive models for their mechanical behavior under such loading conditions. Beyond accounting for finite deformations, it is also desirable that these models incorporate full dependence on the constitutive behavior of the constituents (i.e., the matrix phase and the fibers), as well as on their spatial arrangement (i.e., the microstructure). In this paper, we will consider fiber-reinforced elastomers with *hyperelastic* matrix and fiber phases. In addition, we will restrict attention to microgeometries with a single family of aligned fibers which are taken to be initially circular in cross section and *randomly* and *isotropically* distributed in the undeformed configuration.

A variety of efforts have been pursued over the past few decades to model the effective behavior of fiber-reinforced hyperelastic materials. In terms of phenomenological approaches, there is the pioneering theory of Spencer (1972), in the

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context of which the fibers are treated as inextensible material line elements. Other more sophisticated phenomenological models have been constructed by augmenting existing isotropic stored-energy functions with additional terms depending on the transversely isotropic invariants associated with the fiber direction (Spencer, 1984). Examples include the models proposed by Qiu and Pence (1997), Merodio and Ogden (2005), Horgan and Saccomandi (2005), and Gasser et al. (2006). Although these models possess a number of desirable features, and in particular, they are simple and can be “calibrated” to become macroscopically unstable—via loss of strong ellipticity—for loading conditions where such instabilities are expected to occur from physical experience (Triantafyllidis and Abeyaratne, 1983), their predictive capabilities for the general response of actual fiber-reinforced elastomers remain limited. In a separate effort—essentially making use of a micromechanics approach—Guo et al. (2006) have proposed a hyperelastic model for fiber-reinforced elastomers with *incompressible Neo-Hookean* matrix phases.

On the other hand, homogenization approaches have also been used to obtain bounds and estimates for the constitutive response of these materials. In particular, there is the simple, microstructure-independent, Voigt-type bound (Ogden, 1978), and the polyconvex Reuss-type lower bound (Ponte Castañeda, 1989), as well as an estimate put forward by deBotton (2005) and deBotton et al. (2006) for fiber-reinforced elastomers with *incompressible Neo-Hookean* phases. One of the strengths of the later model (deBotton et al., 2006) is that it predicts the exact effective response of composites with the “composite cylinder assemblage” microstructure, when subjected to axisymmetric and antiplane shear loadings. Based on the “second-order” homogenization procedure (Ponte Castañeda, 1996; Ponte Castañeda and Tiberio, 2000), Lhellec et al. (2004) proposed a constitutive model, for the transverse response of incompressible hyperelastic fiber-reinforced elastomers with *periodic* microstructures, and made successful comparisons with experimental and numerical results. In addition, by making use of the more recent “second-order” homogenization theory (Ponte Castañeda, 2002; Lopez-Pamies and Ponte Castañeda, 2006a), Lopez-Pamies and Ponte Castañeda (2006b) obtained closed-form estimates for the transverse in-plane response of incompressible elastomers reinforced with *randomly* distributed, *rigid* fibers, while Brun et al. (2007) provided more general estimates for fiber-reinforced elastomers with *compressible*, isotropic (matrix and fiber) phases and *periodic* microstructures.

In this paper, we will make use of the second-order homogenization theories (Ponte Castañeda and Tiberio, 2000; Lopez-Pamies and Ponte Castañeda, 2006a) to construct a complete three-dimensional constitutive model for the overall behavior of fiber-reinforced elastomers with *incompressible*, isotropic matrix and fiber phases and *random* microstructures. More specifically, the constitutive behaviors of the matrix and fibers are assumed to be characterized by *generalized Neo-Hookean* models. This class of materials is sufficiently general to model many types of real elastomers (see, e.g., Gent, 1996; Boyce and Arruda, 2000) and, at the same time, is sufficiently simple to lead to analytical results. Furthermore, the fibers are assumed to be perfectly aligned and to be distributed randomly and isotropically in the transverse plane, leading to overall transversely isotropic behavior for the composite. The main result of this paper is given by expression (38), together with expressions (39)–(42), which provide estimates for the effective stored-energy function of the composite materials of interest.

It is relevant to mention that the two “second-order” homogenization methods (Ponte Castañeda, 1996, 2002) were established on the common basis that available estimates for the effective behavior of (suitably constructed) linear composites can be converted into corresponding estimates for the effective behavior of non-linear composites. They both have the capability to account for statistical information on the initial microstructure beyond the volume fraction, as well as for its evolution, resulting from the applied finite deformations. This point is crucial as the evolution of the microstructure may have a significant geometric softening or stiffening effect on the overall response of the material, which, in turn, may lead to the possible development of macroscopic instabilities. The first method, when it works, is simpler to use than the second. When the first method fails, the second method, using additional information about the field fluctuations, can deliver improved results at the expense of a somewhat heavier implementation. Finally, it is important to mention that in addition to the already-mentioned applications to fiber-reinforced elastomers, these homogenization methods can be employed more generally, and have already been used, for example, to construct constitutive models for the overall response of porous elastomers (Lopez-Pamies and Ponte Castañeda, 2004; Michel et al., 2007).

## 2. Problem formulation

Consider a specimen occupying a volume  $\Omega_0$  with boundary  $\partial\Omega_0$  in the reference (undeformed) configuration, and made up of a single family of aligned, cylindrical fibers with circular cross section, distributed randomly and isotropically (in the transverse plane) in a matrix phase. The orientation of the fibers in the reference configuration is taken to be characterized by the unit vector  $\mathbf{N}$ . Furthermore, it is assumed that the average diameter of the fibers is much smaller than the size of the specimen and the scale of variation of the applied load.

Both the matrix (phase 1) and the fibers (phase 2) are assumed to be made up of (different) homogeneous hyperelastic materials. Their constitutive behaviors are characterized by stored-energy functions  $W^{(1)}$  and  $W^{(2)}$ , respectively, which are assumed to be *objective*, *isotropic*, strictly rank-one convex (strongly elliptic) functions of the deformation gradient tensor  $\mathbf{F}$ . Here, we will restrict our attention to stored-energy functions  $W^{(r)}$  for the phases ( $r = 1, 2$ ) of the form

$$W^{(r)}(\mathbf{F}) = g^{(r)}(J) + h^{(r)}(J) + \frac{\kappa^{(r)}}{2}(J - 1)^2, \quad (1)$$

where  $I = \text{tr}(\mathbf{F}^T \mathbf{F})$  and  $J = \det \mathbf{F}$ . In this expression,  $g^{(r)}(I)$  and  $h^{(r)}(J)$  are twice differentiable material functions that satisfy the “linearization” conditions:  $g^{(r)}(3) = h^{(r)}(1) = 0$ ,  $g'_I{}^{(r)}(3) = \mu^{(r)}/2$ ,  $h'_J{}^{(r)}(1) = -\mu^{(r)}$ , and  $4g''_{II}{}^{(r)}(3) + h''_{JJ}{}^{(r)}(1) = \mu^{(r)}/3$ , where the parameters  $\mu^{(r)}$  and  $\kappa^{(r)}$  denote the classical shear and bulk moduli. (Here and subsequently, the subscripts  $I$  and  $J$  denote differentiation with respect to these invariants, e.g.,  $g'_I{}^{(r)}(\cdot) = (dg^{(r)}/dI)(\cdot)$ ,  $g''_{II}{}^{(r)}(\cdot) = (d^2g^{(r)}/dI^2)(\cdot)$ .) Note that upon taking the limit  $\kappa^{(r)} \rightarrow \infty$ , the hyperelastic potentials (1) reduce to the (incompressible) *generalized Neo-Hookean* stored-energy functions  $W^{(r)}(\mathbf{F}) = g^{(r)}(I)$ , together with the incompressibility constraint  $J = 1$ .

An example of a model of the type (1), which captures the limited chain extensibility of elastomers, is the *Gent (1996)* model:

$$W(\mathbf{F}) = -\frac{J_m \mu}{2} \ln \left[ 1 - \frac{I-3}{J_m} \right] - \mu \ln J + \left( \frac{\kappa}{2} - \frac{J_m + 3}{3J_m} \mu \right) (J-1)^2, \quad (2)$$

where the parameter  $J_m$  corresponds to the limit of  $I-3$  for the lock-up of the elastomer. Note that, in the limit  $J_m \rightarrow \infty$  the Gent model reduces to a compressible Neo-Hookean model.

It should be emphasized that the homogenization methods to be developed in this work could be applied for more general constitutive models for the phases, including dependence on the second invariant  $I_2$ . Indeed, generalizations of the above-mentioned constitutive models incorporating dependence on  $I_2$  have been discussed by *Horgan and Saccomandi (1999)*. However, one of the goals of this first application for *incompressible* fiber-reinforced materials is to obtain results that are as (analytically) explicit as possible, and in this respect the form (1) for the constitutive relation will greatly simplify the concomitant analyses.

The *local* stored-energy function of the fiber-reinforced elastomer may be conveniently written as

$$W(\mathbf{X}; \mathbf{F}) = (1 - \chi_0(\mathbf{X}))W^{(1)}(\mathbf{F}) + \chi_0(\mathbf{X})W^{(2)}(\mathbf{F}), \quad (3)$$

where  $\chi_0$  denotes the characteristic function of the part of  $\Omega_0$  occupied by fibers (i.e.,  $\chi_0$  is equal to one if the position vector  $\mathbf{X}$  of a material point in the reference configuration is inside a fiber, and zero otherwise) and serves to characterize the microstructure of the material in the undeformed configuration. Note that, in view of the assumed random distribution of the fibers, the dependence of  $\chi_0$  on  $\mathbf{X}$  is not known precisely, and the microstructure is only partially defined in terms of its  $n$ -point statistics. (In this work, we will make use of one- and two-point statistics, as detailed further below.) The *local* constitutive relation for the composite is then given by

$$\mathbf{S} = \frac{\partial W}{\partial \mathbf{F}}(\mathbf{X}; \mathbf{F}), \quad (4)$$

where  $\mathbf{S}$  denotes the first Piola–Kirchhoff stress tensor.

Under the hypotheses of *statistical uniformity*, and the above-mentioned *separation of length scales*, the *effective* (or macroscopic) constitutive relation for the fiber-reinforced elastomer is defined (*Hill, 1972*) as follows:

$$\bar{\mathbf{S}} = \frac{\partial \bar{W}}{\partial \bar{\mathbf{F}}}, \quad (5)$$

where  $\bar{\mathbf{S}} = \langle \mathbf{S} \rangle$ ,  $\bar{\mathbf{F}} = \langle \mathbf{F} \rangle$  are the *average stress* and *average deformation gradient*, respectively, and

$$\begin{aligned} \bar{W}(\bar{\mathbf{F}}) &= \min_{\mathbf{F} \in \mathcal{K}(\bar{\mathbf{F}})} \langle W(\mathbf{X}; \mathbf{F}) \rangle \\ &= \min_{\mathbf{F} \in \mathcal{K}(\bar{\mathbf{F}})} [(1 - c_0) \langle W^{(1)}(\mathbf{F}) \rangle^{(1)} + c_0 \langle W^{(2)}(\mathbf{F}) \rangle^{(2)}] \end{aligned} \quad (6)$$

is the *effective stored-energy function* of the composite. In the above expressions, the triangular brackets  $\langle \cdot \rangle$ ,  $\langle \cdot \rangle^{(1)}$  and  $\langle \cdot \rangle^{(2)}$  denote, respectively, volume averages over the specimen ( $\Omega_0$ ), the matrix ( $\Omega_0^{(1)}$ ) and the fibers ( $\Omega_0^{(2)}$ ), in the undeformed configuration. The scalar  $c_0 = \langle \chi_0 \rangle$  stands for the volume fraction of the fibers in the undeformed configuration. Furthermore,  $\mathcal{K}$  denotes the set of kinematically admissible deformation gradients:

$$\mathcal{K}(\bar{\mathbf{F}}) = \{ \mathbf{F} | \exists \mathbf{x} = \mathbf{x}(\mathbf{X}) \text{ with } \mathbf{F} = \text{Grad } \mathbf{x} \text{ and } J > 0 \text{ in } \Omega_0, \mathbf{x} = \bar{\mathbf{F}}\mathbf{X} \text{ on } \partial\Omega_0 \}. \quad (7)$$

In passing, it is relevant to remark that  $\bar{W}$  represents the average elastic energy stored in the composite when subjected to an affine deformation on its boundary. Note also that, by virtue of definition (6) together with the objectivity of  $W^{(1)}$  and  $W^{(2)}$ ,  $\bar{W}$  is an objective scalar function of  $\bar{\mathbf{F}}$ .

Because of the non-convexity of  $W$  on  $\mathbf{F}$ , the solution (assuming that it exists) of the Euler–Lagrange equations associated with the variational problem (6) need not be unique. However, within a sufficiently small neighborhood of  $\bar{\mathbf{F}} = \mathbf{I}$ , problem (6) is expected to lead to a well-posed linear-elastic problem with a unique solution. As the deformation progresses beyond the linearly elastic neighborhood into the finite deformation regime, the composite may reach a point at which this “principal” solution bifurcates into different energy solutions. This point corresponds to the onset of an *instability*, beyond which the applicability of the “principal” solution becomes questionable. Following the work of *Triantafyllidis* and collaborators (see, e.g., *Geymonat et al., 1993*; *Triantafyllidis et al., 2006*), it is useful to make the distinction between “microscopic” instabilities, that is, instabilities with wavelengths that are small compared to the size of the specimen, and “macroscopic” instabilities, that is, instabilities with wavelengths comparable to the size of the specimen. The computation of “microscopic” instabilities is in general an extremely difficult task, especially for random

systems. On the other hand, the computation of “macroscopic” instabilities is a much simpler endeavor, since it reduces to the detection of loss of strong ellipticity of the effective stored-energy function of the material evaluated at the above-described “principal” solution (Geymonat et al., 1993). Based on these remarks, in this work, we will not attempt to solve the variational problem (6), but instead, we will estimate the overall behavior of fiber-reinforced elastomers by means of the effective stored-energy function:

$$\widehat{W}(\bar{\mathbf{F}}) = \text{stat}_{\mathbf{F} \in \mathcal{X}(\bar{\mathbf{F}})} [(1 - c_0)(W^{(1)}(\mathbf{F}))^{(1)} + c_0(W^{(2)}(\mathbf{F}))^{(2)}], \quad (8)$$

where the stat(ionary) variational operation means evaluation at the above-described “principal” solution of the pertinent Euler–Lagrange equations. Finally, it is relevant to mention that other definitions of macroscopic behavior for hyperelastic composites have been proposed in the literature, including the notion of a “globally equivalent homogeneous” material (Chen et al., 2006).

### 2.1. Transverse isotropy

In view of the above-adopted constitutive and (micro)geometric assumptions, it follows that the effective stored-energy function (8) is a transversely isotropic scalar function of  $\bar{\mathbf{F}}$  (with symmetry axis  $\mathbf{N}$ ). Accordingly, the functional dependence of  $\widehat{W}$  on  $\bar{\mathbf{F}}$  may be conveniently expressed as

$$\widehat{W}(\bar{\mathbf{F}}) = \widehat{\phi}(\bar{\lambda}_p, \bar{\lambda}_n, \bar{\gamma}_p, \bar{\gamma}_n, \bar{\psi}_{\bar{\gamma}}), \quad (9)$$

where the scalars  $\bar{\lambda}_p$ ,  $\bar{\lambda}_n$ ,  $\bar{\gamma}_p$ ,  $\bar{\gamma}_n$ , and  $\bar{\psi}_{\bar{\gamma}}$  constitute a set of transversely isotropic invariant functions of  $\bar{\mathbf{C}} = \bar{\mathbf{F}}^T \bar{\mathbf{F}}$ , first introduced by Ericksen and Rivlin (1954), and given by

$$\begin{aligned} \bar{\lambda}_n &= (\mathbf{N} \cdot \bar{\mathbf{C}}\mathbf{N})^{1/2}, & \bar{\lambda}_p &= ((\det \bar{\mathbf{C}})^{1/2} / \bar{\lambda}_n)^{1/2}, \\ \bar{\gamma}_n &= (\mathbf{N} \cdot \bar{\mathbf{C}}\bar{\mathbf{C}}\mathbf{N} - \bar{\lambda}_n^4)^{1/2} / \bar{\lambda}_n, & \bar{\gamma}_p &= (\text{tr} \bar{\mathbf{C}} - \bar{\lambda}_n^2 - 2\bar{\lambda}_p^2 - \bar{\gamma}_n^2)^{1/2}. \end{aligned} \quad (10)$$

The invariant  $\bar{\psi}_{\bar{\gamma}}$  is a more complicated function of  $\bar{\mathbf{C}}$  and  $\mathbf{N}$  and will not be spelled out here (see, e.g., deBotton et al., 2006, for an explicit expression).

As discussed by deBotton et al. (2006), a practical implication of the representation (9) is that, in order to determine  $\widehat{W}$ , it suffices to restrict attention to deformation gradient tensors  $\bar{\mathbf{F}}$  of the form

$$\bar{F}_{ij} = \begin{bmatrix} \bar{\lambda}_p & 0 & 0 \\ \bar{\gamma}_p & \bar{\lambda}_p & 0 \\ \bar{\gamma}_n \cos \bar{\psi}_{\bar{\gamma}} & \bar{\gamma}_n \sin \bar{\psi}_{\bar{\gamma}} & \bar{\lambda}_n \end{bmatrix} \quad (11)$$

in a coordinate system  $\mathbf{e}_i$  ( $i = 1, 2, 3$ ) with  $\mathbf{e}_3 = \mathbf{N}$ . This particular type of applied deformation entails the following interpretation for the invariants in (10):  $\bar{\lambda}_n$  is the stretch along the direction of the fibers  $\mathbf{N}$ ,  $\bar{\lambda}_p$  is a measure of the dilatation in the transverse plane,  $\bar{\gamma}_n$  is the amount of antiplane shear, and  $\bar{\gamma}_p$  is the amount of shear in the transverse plane (in-plane shear). The invariant  $\bar{\psi}_{\bar{\gamma}}$  is a measure of the coupling among the other invariants and, like the orientation of  $\mathbf{e}_1$  and  $\mathbf{e}_2$  in the transverse plane, can be chosen arbitrarily whenever  $\bar{\gamma}_p = 0$  or  $\bar{\gamma}_n = 0$ . (These two cases are of special interest in this work and will be considered in detail in Sections 3.2.1 and 3.3.1.)

To conclude this section, it is appropriate to mention that the estimates to be derived next for the effective stored-energy function (8) of fiber-reinforced elastomers with matrix and fiber phases characterized by (1) will be shown in Section 4 to depend weakly on the invariant  $\bar{\psi}_{\bar{\gamma}}$ . In addition, when the matrix and fibers are taken to be incompressible—which is precisely the case of interest in this work—the overall constraint

$$\det \bar{\mathbf{F}} = \bar{\lambda}_p^2 \bar{\lambda}_n = 1 \quad (12)$$

must be satisfied. This means that, in practice, the effective stored-energy function (8) of fiber-reinforced elastomers with incompressible generalized Neo-Hookean phases can be expediently approximated as a function solely of  $\bar{\lambda}_n$ ,  $\bar{\gamma}_p$ , and  $\bar{\gamma}_n$ , namely:

$$\widehat{W}(\bar{\mathbf{F}}) \approx \widehat{\phi}(\bar{\lambda}_n^{-1/2}, \bar{\lambda}_n, \bar{\gamma}_p, \bar{\gamma}_n, 0) \doteq \widehat{\Phi}(\bar{\lambda}_n, \bar{\gamma}_p, \bar{\gamma}_n). \quad (13)$$

### 3. Second-order homogenization estimates

As already mentioned, we will make use of the second-order variational procedures of Ponte Castañeda and Tiberio (2000) and Lopez-Pamies and Ponte Castañeda (2006a) to estimate the effective stored-energy function (8) for the class of fiber-reinforced elastomers of interest in this work. The main concept behind these two methods is the construction of suitable variational principles making use of the idea of a “linear comparison composite” (LCC) with the same microstructure as the actual hyperelastic composite (see Section 3.1). Both methods have the distinguishing feature of

delivering estimates that are exact to second order in the heterogeneity contrast, provided that the corresponding estimates used for the LCC are also exact to second order in the contrast (hence their names). However, the second method makes use of the deformation field fluctuations in the linearization scheme, while the first method makes use only of the first moments, and is therefore easier to implement. For convenience, and reasons that will become more evident further below, we henceforth refer to the first procedure as the tangent second-order (TSO) method and to the second as the generalized second-order (GSO) method.

The main objective of this work is to compute an estimate for the effective stored-energy function (8) of the class of fiber-reinforced elastomers defined in the previous section in the limit of incompressible behavior for the matrix and fiber phases (i.e., in the limit as  $\kappa^{(1)} \rightarrow \infty$  and  $\kappa^{(2)} \rightarrow \infty$  in expression (1)). To this end, we could in principle make use of the more accurate GSO method. However, in the limit of incompressibility, the use of this technique for general loading conditions leads to a fairly complex set of asymptotic equations that (although easy to solve numerically) are difficult to solve explicitly. On the other hand, it will be shown below that the simpler TSO method yields quite accurate results (relative to the generally more accurate GSO method) for the special case of *combined antiplane and axisymmetric shear deformations* (i.e., for  $\bar{\gamma}_p = 0$  in (11)). For this reason, the GSO method will only be used to generate an estimate for *generalized plane-strain deformations* (i.e., for  $\bar{\gamma}_n = 0$  in (11)), when the pertinent asymptotic analysis becomes more manageable (see below). The two estimates for the complementary sets of loading conditions will then be used in Section 4 to construct—via a non-linear interpolation scheme—a complete constitutive model for general loading conditions.

### 3.1. Local and effective properties of the LCC

Consider a two-phase LCC with the same initial microstructure (i.e., the same  $\chi_0$ ) as the actual fiber-reinforced elastomer and with local stored-energy function

$$W_T(\mathbf{X}; \mathbf{F}) = (1 - \chi_0(\mathbf{X}))W_T^{(1)}(\mathbf{F}) + \chi_0(\mathbf{X})W_T^{(2)}(\mathbf{F}). \quad (14)$$

Here,  $W_T^{(r)}$  ( $r = 1, 2$ ) are quadratic potentials in  $\mathbf{F}$  given by the Taylor-like expressions

$$W_T^{(r)}(\mathbf{F}) = W^{(r)}(\mathbf{F}^{(r)}) + \mathbf{S}^{(r)}(\mathbf{F}^{(r)}) \cdot (\mathbf{F} - \mathbf{F}^{(r)}) + \frac{1}{2}(\mathbf{F} - \mathbf{F}^{(r)}) \cdot \mathbf{L}^{(r)}(\mathbf{F} - \mathbf{F}^{(r)}), \quad (15)$$

where the second-order tensors  $\mathbf{F}^{(r)}$  are constant reference deformation gradients,  $\mathbf{S}^{(r)}(\cdot) = \partial W^{(r)}(\cdot)/\partial \mathbf{F}$ , and  $\mathbf{L}^{(r)}$  are constant fourth-order tensors with major symmetry. The specific choices for  $\mathbf{F}^{(r)}$  and  $\mathbf{L}^{(r)}$  are different in the contexts of the GSO and TSO methods, and therefore the pertinent discussions are postponed for now.

Having defined the local behavior, the corresponding effective stored-energy function of the LCC,  $\widehat{W}_T$  say, can be readily determined in terms of the effective modulus tensor,  $\widehat{\mathbf{L}}$ , of a *linear-elastic* composite with the same microstructure as the actual fiber-reinforced elastomer and phase moduli  $\mathbf{L}^{(1)}$  and  $\mathbf{L}^{(2)}$ ; an explicit expression for  $\widehat{W}_T$  in terms of  $\widehat{\mathbf{L}}$  is given by Eq. (28) in Lopez-Pamies and Ponte Castañeda (2006a). In this work, we will make use of the following Willis (1977) estimate:

$$\widehat{\mathbf{L}} = \mathbf{L}^{(1)} + c_0[(1 - c_0)\mathbf{P} - (\mathbf{L}^{(1)} - \mathbf{L}^{(2)})^{-1}]^{-1}, \quad (16)$$

which is known to be exact to second order in the heterogeneity contrast and to first order in  $c_0$ , and particularly well suited for the “particulate” microstructures of interest here. The Eshelby-type tensor  $\mathbf{P}$  in (16) contains information about the shape (cylindrical and circular) and distribution (aligned and randomly and isotropically distributed in the transverse plane) of the fibers in the undeformed configuration. The components of  $\mathbf{P}$  in a coordinate system  $\mathbf{e}_i$  ( $i = 1, 2, 3$ ) with  $\mathbf{e}_3 = \mathbf{N}$  are given by

$$P_{ijkl} = \frac{1}{2\pi} \int_0^{2\pi} (L_{imkn}^{(1)} \xi_m \xi_n)^{-1} \xi_j \xi_l \, d\theta, \quad (17)$$

where  $\xi_1 = \cos \theta$ ,  $\xi_2 = \sin \theta$ , and  $\xi_3 = 0$ . It should also be noted that  $\mathbf{P}$  depends on the modulus tensor  $\mathbf{L}^{(1)}$ , but not on  $\mathbf{L}^{(2)}$ , has major symmetry, and

$$P_{i3kl} = P_{ijk3} = 0 \quad \text{for } i, j, k, l = 1, 2, 3. \quad (18)$$

### 3.2. TSO estimates

Following Ponte Castañeda and Tiberio (2000), the use of an LCC with the prescriptions  $\mathbf{F}^{(r)} = \langle \mathbf{F} \rangle^{(r)} \equiv \bar{\mathbf{F}}^{(r)}$  and  $\mathbf{L}^{(r)} = \partial^2 W^{(r)}(\bar{\mathbf{F}}^{(r)})/\partial \mathbf{F}^2$  ( $r = 1, 2$ ) leads to the following approximation for the effective stored-energy function (8):

$$\widehat{W}(\bar{\mathbf{F}}) = (1 - c_0)[W^{(r)}(\bar{\mathbf{F}}^{(1)}) + \frac{1}{2}\mathbf{S}^{(1)}(\bar{\mathbf{F}}^{(1)}) \cdot (\bar{\mathbf{F}} - \bar{\mathbf{F}}^{(1)})] + c_0[W^{(2)}(\bar{\mathbf{F}}^{(2)}) + \frac{1}{2}\mathbf{S}^{(2)}(\bar{\mathbf{F}}^{(2)}) \cdot (\bar{\mathbf{F}} - \bar{\mathbf{F}}^{(2)})]. \quad (19)$$

In this context, the variables  $\bar{\mathbf{F}}^{(1)}$  and  $\bar{\mathbf{F}}^{(2)}$ , denoting the phase averages of the deformation gradient field in the LCC, are determined by means of the system of equations:

$$\bar{\mathbf{F}} = (1 - c_0)\bar{\mathbf{F}}^{(1)} + c_0\bar{\mathbf{F}}^{(2)} \quad (20)$$

and

$$\bar{\mathbf{F}} - \bar{\mathbf{F}}^{(1)} = \mathbf{P}(\mathbf{L}^{(1)}(\bar{\mathbf{F}} - \bar{\mathbf{F}}^{(1)} + c_0(\mathbf{S}^{(1)}(\bar{\mathbf{F}}^{(1)} - \mathbf{S}^{(2)}(\bar{\mathbf{F}}^{(2)}))), \tag{21}$$

Note that estimate (19) is relatively simple to evaluate. This becomes more evident by first realizing—with the help of (18)—that  $\bar{F}_{i3}^{(1)} = \bar{F}_{i3}^{(2)} = \bar{F}_{i3}$  for  $i = 1, 2, 3$ . Then, using (20) to express the unknown components of  $\bar{\mathbf{F}}^{(2)}$  in terms of the unknown components of  $\bar{\mathbf{F}}^{(1)}$ , the problem essentially reduces to solving numerically the remaining six non-trivial scalar equations in (21) for the six remaining unknown components  $\bar{F}_{i\beta}^{(1)}$  ( $i = 1, 2, 3$ , and  $\beta = 1, 2$ ).

As opposed to the GSO estimate, the TSO estimate (19) has the disadvantage that it can become inconsistent with the overall kinematical constraint  $\det \bar{\mathbf{F}} = 1$  resulting from the incompressibility limit  $\kappa^{(1)} \rightarrow \infty$  and  $\kappa^{(2)} \rightarrow \infty$  for *general loading conditions* (see Ponte Castañeda and Tiberio, 2000; Lhellec et al., 2004; Lopez-Pamies and Ponte Castañeda, 2004 for an in-depth discussion of this deficiency). However, for some special loading conditions, namely, for combined antiplane and axisymmetric shear deformations, the TSO estimate (19) can be shown to be fully consistent with the constraint  $\det \bar{\mathbf{F}} = 1$  in the limit as the matrix and fibers are made incompressible. The explicit computation of (19) for these special conditions is spelled out in the next subsection.

### 3.2.1. Incompressible estimate for combined antiplane–axisymmetric shear

In this subsection, we outline the computation of the TSO estimate (19) for the effective stored-energy function of fiber-reinforced elastomers with *incompressible* generalized Neo-Hookean matrix and fiber phases under antiplane combined with axisymmetric shear deformations. To this end, we consider an  $\bar{\mathbf{F}}$  of the form (11) with  $\bar{\gamma}_p = \bar{\psi}_{\bar{\gamma}} = 0$  and set (without loss of generality)  $\Delta = 1/\kappa^{(1)} = 1/\kappa^{(2)}$ , where the parameter  $\Delta$  will be taken to tend to zero in order to model incompressible behavior.

Then, guided by numerical results (not shown here) for finite values of  $\kappa^{(1)}$  and  $\kappa^{(2)}$ , it is assumed that the asymptotic expansion for the six unknown components<sup>1</sup>  $\bar{F}_{i\alpha}^{(1)}$  ( $i = 1, 2, 3$ , and  $\alpha = 1, 2$ ) in the limit as  $\Delta \rightarrow 0$  is of the form

$$\bar{F}_{i\alpha}^{(1)} = \overset{\circ}{\bar{F}}_{i\alpha}^{(1)} + \check{\bar{F}}_{i\alpha}^{(1)} \Delta + O(\Delta^2), \tag{22}$$

where  $\overset{\circ}{\bar{F}}_{i\alpha}^{(1)}$  and  $\check{\bar{F}}_{i\alpha}^{(1)}$  are unknown coefficients to be determined from the asymptotic expansion of Eq. (21) in the limit as  $\Delta \rightarrow 0$ . Indeed, substituting (22) in (21) and subsequently expanding in small values of  $\Delta$  yields a system of hierarchical equations for the unknown coefficients in (22). The resulting equations of orders  $\Delta^{-1}$  and  $\Delta^0$ , which can be solved explicitly, are consistent with the *exact* overall incompressibility constraint (12), and lead to the following relations:

$$\overset{\circ}{\bar{F}}_{11}^{(1)} = \overset{\circ}{\bar{F}}_{22}^{(1)} = \bar{\lambda}_p, \quad \overset{\circ}{\bar{F}}_{12}^{(1)} = \overset{\circ}{\bar{F}}_{21}^{(1)} = \overset{\circ}{\bar{F}}_{32}^{(1)} = 0, \quad \overset{\circ}{\bar{F}}_{31}^{(1)} = \bar{\gamma}_n^{(1)}, \tag{23}$$

where the variable  $\bar{\gamma}_n^{(1)}$  is determined as the solution of the non-linear algebraic equation

$$\sqrt{g_I^{(1)}(I_N^{(1)})[g_I^{(1)}(I_N^{(1)}) + 2g_{II}^{(1)}(I_N^{(1)})(\bar{\gamma}_n^{(1)})^2]}(\bar{\gamma}_n - \bar{\gamma}_n^{(1)}) + c_0[g_I^{(2)}(I_N^{(2)})\bar{\gamma}_n^{(2)} - g_I^{(1)}(I_N^{(1)})\bar{\gamma}_n^{(1)}] = 0. \tag{24}$$

In this expression, the notations

$$\bar{\gamma}_n^{(2)} = \frac{\bar{\gamma}_n - (1 - c_0)\bar{\gamma}_n^{(1)}}{c_0}, \tag{25}$$

$I_N^{(1)} = \bar{\mathbf{F}}^{(1)} \cdot \bar{\mathbf{F}}^{(1)} = 2/\bar{\lambda}_n + \bar{\lambda}_n^2 + (\bar{\gamma}_n^{(1)})^2$ , and  $I_N^{(2)} = \bar{\mathbf{F}}^{(2)} \cdot \bar{\mathbf{F}}^{(2)} = 2/\bar{\lambda}_n + \bar{\lambda}_n^2 + (\bar{\gamma}_n^{(2)})^2$  have been introduced for convenience. Physically, the variables  $\bar{\gamma}_n^{(1)}$  and  $\bar{\gamma}_n^{(2)}$  above denote the average amount of antiplane shear in the matrix and fibers, respectively, in the incompressibility limit.

With the help of relations (22)–(25), it is a simple matter to deduce that in the limit when the matrix and fibers are taken to be incompressible, the TSO estimate (19) reduces to

$$\widehat{W}(\bar{\mathbf{F}}) = \widehat{\Phi}(\bar{\lambda}_n, 0, \bar{\gamma}_n) = (1 - c_0)[g_I^{(1)}(I_N^{(1)}) + g_I^{(1)}(I_N^{(1)})\bar{\gamma}_n^{(1)}(\bar{\gamma}_n - \bar{\gamma}_n^{(1)})] + c_0[g_I^{(2)}(I_N^{(2)}) + g_I^{(2)}(I_N^{(2)})\bar{\gamma}_n^{(2)}(\bar{\gamma}_n - \bar{\gamma}_n^{(2)})] \tag{26}$$

for combined antiplane–axisymmetric shear deformations. Thus, the computation of this estimate is seen to amount to solving numerically only one non-linear equation. Moreover, for pure axisymmetric shear ( $\bar{\gamma}_n = 0$ ), the TSO estimate (26) reduces identically to the Voigt bound, which is known to be an exact result for this mode of deformation (see Section 4.1). In addition, for Neo-Hookean phases (i.e.,  $g^{(1)}(I) = \mu^{(1)}(I - 3)/2$  and  $g^{(2)}(I) = \mu^{(2)}(I - 3)/2$ ), (26) can be shown to be *exact* for fiber-reinforced elastomers with the “composite cylinder assemblage” microstructure of Hashin (see Section 4.1 below and deBotton et al., 2006, Section 3).

<sup>1</sup> Recall that  $\bar{F}_{i3}^{(1)} = \bar{F}_{i3}$  and  $\bar{F}_{ij}^{(2)} = (\bar{F}_{ij} - (1 - c_0)\bar{F}_{ij}^{(1)})/c_0$  ( $i, j = 1, 2, 3$ ).

### 3.3. GSO estimates

Following Lopez-Pamies and Ponte Castañeda (2006a), the alternative use of an LCC with prescriptions  $\mathbf{F}^{(1)} = \bar{\mathbf{F}}$ ,  $\mathbf{F}^{(2)} = \langle \mathbf{F} \rangle^{(2)} \equiv \bar{\mathbf{F}}^{(2)}$ ,

$$L_{ijkl}^{(1)} = \bar{Q}_{rm} \bar{Q}_{jn} \bar{Q}_{sp} \bar{Q}_{lq} \bar{R}_{ir} \bar{R}_{ks} L_{mnpq}^*, \quad (27)$$

and  $\mathbf{L}^{(2)} = \partial^2 W^{(2)}(\bar{\mathbf{F}}^{(2)})/\partial \mathbf{F}^2$  leads to the following approximation for the effective stored-energy function (8):

$$\widehat{W}(\bar{\mathbf{F}}) = (1 - c_0)[W^{(1)}(\hat{\mathbf{F}}^{(1)}) - \mathbf{S}^{(1)}(\bar{\mathbf{F}}) \cdot (\hat{\mathbf{F}}^{(1)} - \bar{\mathbf{F}}^{(1)})] + c_0 W^{(2)}(\bar{\mathbf{F}}^{(2)}). \quad (28)$$

In the above expressions,  $\bar{\mathbf{R}}$  and  $\bar{\mathbf{Q}}$  are the orthogonal tensors in the decompositions  $\bar{\mathbf{F}} = \bar{\mathbf{R}}\bar{\mathbf{U}} = \bar{\mathbf{R}}\bar{\mathbf{Q}}\bar{\mathbf{D}}\bar{\mathbf{Q}}^T$ , where  $\bar{\mathbf{D}} \doteq \sum_{i=1}^3 \bar{\lambda}_i \bar{\mathbf{v}}_i \otimes \bar{\mathbf{v}}_i$  and  $\bar{\mathbf{Q}} = \bar{\mathbf{M}}_i \otimes \bar{\mathbf{v}}_i$ , with  $\bar{\mathbf{v}}_i$  denoting a Cartesian basis for the laboratory frame of reference and  $\bar{\lambda}_i, \bar{\mathbf{M}}_i$  the principal values and the corresponding directions of the macroscopic right stretch tensor  $\bar{\mathbf{U}}$ . The tensor  $\mathbf{L}^*$  is orthotropic relative to  $\bar{\mathbf{v}}_i$  and is defined in terms of seven independent moduli, denoted by  $\ell_\alpha^*$  ( $\alpha = 1, 2, \dots, 7$ ) (see relation (51) in Appendix A). Moreover,  $\bar{\mathbf{F}}^{(1)}$  and  $\bar{\mathbf{F}}^{(2)}$  denote the phase averages of the deformation gradient field in the LCC which, together with the tensor  $\hat{\mathbf{F}}^{(1)}$  and the seven unknown moduli  $\ell_\alpha^*$  in  $\mathbf{L}^*$ , are determined by the following system of coupled, non-linear, algebraic equations:

$$\bar{\mathbf{F}} = (1 - c_0)\bar{\mathbf{F}}^{(1)} + c_0\bar{\mathbf{F}}^{(2)}, \quad (29)$$

$$\bar{\mathbf{F}} - \bar{\mathbf{F}}^{(2)} = (1 - c_0)\mathbf{P}(\mathbf{L}^{(1)}(\bar{\mathbf{F}} - \bar{\mathbf{F}}^{(2)}) - \mathbf{S}^{(1)}(\bar{\mathbf{F}}) + \mathbf{S}^{(2)}(\bar{\mathbf{F}}^{(2)})), \quad (30)$$

$$\mathbf{S}^{(1)}(\hat{\mathbf{F}}^{(1)}) - \mathbf{S}^{(1)}(\bar{\mathbf{F}}) = \mathbf{L}^{(1)}(\hat{\mathbf{F}}^{(1)} - \bar{\mathbf{F}}), \quad (31)$$

$$(\hat{\mathbf{F}}^{(1)} - \bar{\mathbf{F}}) \cdot \frac{\partial \mathbf{L}^{(1)}}{\partial \ell_\alpha^*} (\hat{\mathbf{F}}^{(1)} - \bar{\mathbf{F}}) = \frac{2}{1 - c_0} \frac{\partial \widehat{W}_T}{\partial \ell_\alpha^*} = k_\alpha, \quad (32)$$

where the variables  $k_\alpha$  ( $\alpha = 1, 2, \dots, 7$ ) are given by

$$k_\alpha = -\frac{c_0}{1 - c_0} (\bar{\mathbf{F}} - \bar{\mathbf{F}}^{(2)}) \cdot \frac{\partial \mathbf{L}^{(1)}}{\partial \ell_\alpha^*} (\bar{\mathbf{F}} - \bar{\mathbf{F}}^{(2)}) - \frac{c_0}{(1 - c_0)^2} \mathbf{A} \cdot \frac{\partial \mathbf{P}}{\partial \ell_\alpha^*} \mathbf{A}, \quad (33)$$

with  $\mathbf{A} \doteq \mathbf{P}^{-1}(\bar{\mathbf{F}} - \bar{\mathbf{F}}^{(2)})$ . Note that Eq. (29)—which is nothing more than the overall average condition of the deformation gradient field—can be solved explicitly for  $\bar{\mathbf{F}}^{(1)}$  in terms of  $\bar{\mathbf{F}}^{(2)}$ . Furthermore, with the help of (18), it is not difficult to show from (30) that  $\bar{F}_{i3}^{(2)} = \bar{F}_{i3}$  ( $i = 1, 2, 3$ ). Thus, the computation of the GSO estimate (28) is seen to reduce ultimately to solving numerically a system of 22 coupled, non-linear, algebraic equations for the 22 scalar unknowns  $\bar{F}_{i\beta}^{(2)}$  ( $i = 1, 2, 3; \beta = 1, 2$ ),  $\hat{F}_{ij}^{(1)}$  ( $i, j = 1, 2, 3$ ), and  $\ell_\alpha^*$  ( $\alpha = 1, 2, \dots, 7$ ).

For general loading conditions of the form (11), the computation of the GSO estimate (28) in the incompressible limit leads to a complex set of asymptotic equations that are difficult to simplify analytically. However, for the special case of generalized plane-strain deformations (i.e.,  $\bar{\gamma}_n = 0$ ), the resulting asymptotic analysis becomes manageable (see Appendix A). The specialization of (28) to these special conditions is the substance of the next subsection.

#### 3.3.1. Incompressible estimate for generalized plane-strain deformations

To avoid loss of continuity, the pertinent (lengthy) derivations are given in Appendix A, and here we will only record the final result of the asymptotic analysis. Thus, in the limit as  $\kappa^{(1)} \rightarrow \infty$  and  $\kappa^{(2)} \rightarrow \infty$ , estimate (28) for the effective stored-energy function (8) simplifies to

$$\widehat{W}(\bar{\mathbf{F}}) = \widehat{\Phi}(\bar{\lambda}_n, \bar{\gamma}_p, 0) = (1 - c_0)g^{(1)}(I_P^{(1)}) + c_0g^{(2)}(I_P^{(2)}), \quad (34)$$

for generalized plane-strain deformations. In these expressions,

$$I_P^{(1)} = c_0 \left( \frac{\bar{\lambda}_1 - \bar{\lambda}_1^{(2)}}{(1 - c_0)\bar{\lambda}_1 \bar{\lambda}_1^{(2)}} \right)^2 [(\bar{\lambda}_1^{(2)}\bar{\lambda}_1\bar{\lambda}_n + 1)^2 + (\bar{\lambda}_1^2 + (\bar{\lambda}_1^{(2)})^2)\bar{\lambda}_n] + \left( \frac{\bar{\lambda}_1 - c_0\bar{\lambda}_1^{(2)}}{1 - c_0} \right)^2 + \left( \frac{\bar{\lambda}_2 - c_0\bar{\lambda}_2^{(2)}}{1 - c_0} \right)^2 + \bar{\lambda}_n^2 \quad (35)$$

and

$$I_P^{(2)} = (\bar{\lambda}_1^{(2)})^2 + (\bar{\lambda}_2^{(2)})^2 + \bar{\lambda}_n^2, \quad (36)$$

where  $\bar{\lambda}_1 = (\sqrt{\bar{\gamma}_p^2 + 4\bar{\lambda}_p^2} - \bar{\gamma}_p)/2$ ,  $\bar{\lambda}_2 = (\sqrt{\bar{\gamma}_p^2 + 4\bar{\lambda}_p^2} + \bar{\gamma}_p)/2$ ,  $\bar{\lambda}_2^{(2)} = 1/(\bar{\lambda}_1^{(2)}\bar{\lambda}_n)$ , with  $\bar{\lambda}_p$  satisfying the exact incompressibility constraint (12), and the variable  $\bar{\lambda}_1^{(2)}$  being the solution of the non-linear algebraic equation

$$[(1 + c_0)g_I^{(1)}(I_P^{(1)}) + (1 - c_0)g_I^{(2)}(I_P^{(2)})](\bar{\lambda}_n^{-2} - (\bar{\lambda}_1^{(2)})^4) - g_I^{(1)}(I_P^{(1)}) \left[ 2\bar{\lambda}_1^{(2)}\bar{\lambda}_1\bar{\lambda}_2^2 - (\bar{\lambda}_1^{(2)})^2 \right] - \frac{(\bar{\lambda}_1^{(2)})^4}{\bar{\lambda}_1} \bar{\lambda}_2 = 0. \quad (37)$$

Thus, it is seen that the GSO estimate (34) is quite simple in form, as its computation amounts to solving numerically just one non-linear equation. Moreover, it is worth remarking that for pure axisymmetric shear ( $\bar{\gamma}_p = 0$ ), estimate (34)—much like the TSO estimate (26)—reduces identically to the Voigt bound. In addition, for pure plane-strain deformations ( $\bar{\lambda}_p = 1$ ) and in the limit of rigid fibers, (34) recovers the estimate of Lopez-Pamies and Ponte Castañeda (2006b) for the effective behavior of elastomers reinforced by a random distribution of circular rigid particles (see relation (27) together with (33) in that reference).

**4. A constitutive model for general loading conditions**

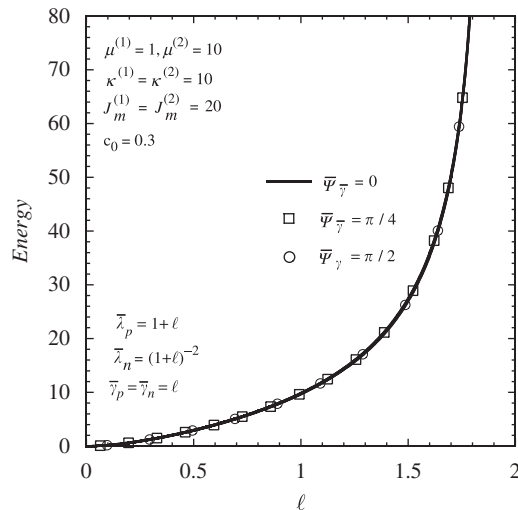
The effective stored-energy function (8) of incompressible fiber-reinforced elastomers under general loading conditions can in principle be determined (approximately) by taking the limit  $\kappa^{(1)} \rightarrow \infty$  and  $\kappa^{(2)} \rightarrow \infty$  in the second-order estimate (SOE) (28). However, as already pointed out in the preceding section, the asymptotic analysis required for the computation of this limit is quite complicated. Because of this, and keeping in mind our objective to construct an estimate for (8) that is as simple as possible, we do not attempt to carry out such an asymptotic analysis here, and instead we propose to combine in a consistent fashion the GSO result (34) for generalized plane-strain deformations with the TSO result (26) for antiplane-axisymmetric shear deformations into one estimate for general loading conditions.

Before proceeding with the details, it is helpful to make the following two remarks:

- (1) The GSO estimate (28) for the effective stored-energy function of fiber-reinforced elastomers, with generalized Neo-Hookean phases of the form (1), depends weakly on the invariant  $\bar{\psi}_{\bar{\gamma}}$ .
- (2) The TSO estimate (19) for the effective stored-energy function of incompressible fiber-reinforced elastomers is practically identical to the GSO estimate (28) for the special case of combined antiplane-axisymmetric deformations.

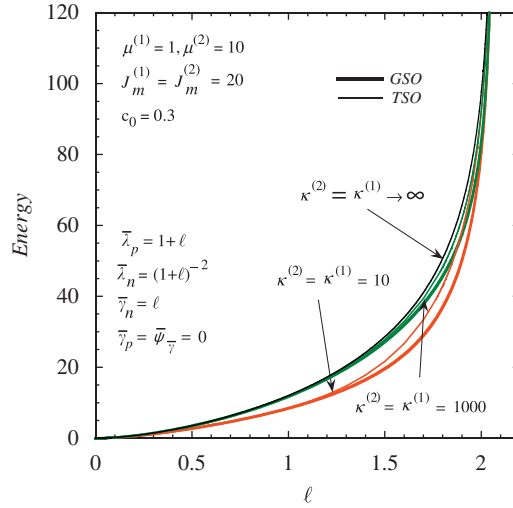
To illustrate the first remark, sample GSO results for the effective stored-energy function  $\widehat{W}(\bar{\mathbf{F}}) = \widehat{\phi}(1 + \ell, (1 + \ell)^{-2}, \ell, \ell, \bar{\psi}_{\bar{\gamma}})$  of a specific fiber-reinforced composite with compressible Gent phases (2) are plotted in Fig. 1 for  $\bar{\psi}_{\bar{\gamma}} = 0, \pi/4$  and  $\pi/2$ , as a function of the loading parameter  $\ell$ . The results clearly indicate that the GSO estimates are very much independent of the value of the invariant  $\bar{\psi}_{\bar{\gamma}}$ . To illustrate the second remark, on the other hand, sample GSO and TSO results for the effective stored-energy function  $\widehat{W}(\bar{\mathbf{F}}) = \widehat{\phi}(1 + \ell, (1 + \ell)^{-2}, 0, \ell, 0)$  of fiber-reinforced elastomers with Gent phases are plotted in Fig. 2 for various values of the bulk moduli  $\kappa^{(1)}$  and  $\kappa^{(2)}$ , as a function of the loading parameter  $\ell$ . The results in this figure clearly show that the GSO and TSO estimates are very close for large values of  $\kappa^{(1)}$  and  $\kappa^{(2)}$ .

Guided by the above two remarks, we now proceed with the combination of estimates (34) and (26) into one estimate for general loading conditions. We start out by considering a macroscopic deformation gradient of the form  $\bar{\mathbf{F}} = \bar{\mathbf{D}} + \bar{\gamma}_n \mathbf{e}_3 \otimes \mathbf{e}_1$ , where  $\bar{\mathbf{D}}$  is given by (49) in Appendix A, such that  $\det \bar{\mathbf{F}} = \det \bar{\mathbf{D}} = \bar{\lambda}_1 \bar{\lambda}_2 \bar{\lambda}_n = \bar{\lambda}_p^2 \bar{\lambda}_n = 1$ . Note that this form of  $\bar{\mathbf{F}}$  is sufficiently general for our purposes, since it depends on all three invariants  $\bar{\lambda}_n, \bar{\gamma}_p$  and  $\bar{\gamma}_n$ , but not on  $\bar{\psi}_{\bar{\gamma}}$ , and for the special cases  $\bar{\gamma}_n = 0$  and  $\bar{\gamma}_p = 0$ , it reduces identically to the loading conditions associated with estimates (34) and (26), respectively.



**Fig. 1.** The GSO estimate (28) for the effective stored-energy  $\widehat{\phi}$  of a fiber-reinforced elastomer with compressible Gent phases subjected to an  $\bar{\mathbf{F}}$  of the form (11), as a function of the loading parameter  $\ell$ , for various values of the invariant  $\bar{\psi}_{\bar{\gamma}}$ .





**Fig. 2.** The GSO (28) and TSO (19) estimates for the effective stored-energy  $\widehat{\phi}$  of fiber-reinforced elastomers with compressible Gent phases subjected to antiplane combined with axisymmetric shear loadings, as a function of the loading parameter  $\ell$ , for  $\kappa^{(1)} = \kappa^{(2)} = 10$  and  $\kappa^{(1)} = \kappa^{(2)} = 1000$ . The corresponding incompressible TSO estimate (26) is also shown.

Then, for consistency with the above-proposed form of  $\bar{\mathbf{F}}$ , we generalize the values of the quantities  $\mathbf{F}^{(1)}$ ,  $\bar{\mathbf{F}}^{(1)}$ ,  $\mathbf{F}^{(2)}$ ,  $\bar{\mathbf{F}}^{(2)}$  and  $\hat{\mathbf{F}}^{(1)}$ , involved in the computation of the GSO and TSO estimates, in the following manner:  $\mathbf{F}^{(1)} = \bar{\mathbf{D}} + \bar{\gamma}_n^{(1)} \mathbf{e}_3 \otimes \mathbf{e}_1$ ,  $\bar{\mathbf{F}}^{(1)} = \bar{\mathbf{D}}^{(1)} + \bar{\gamma}_n^{(1)} \mathbf{e}_3 \otimes \mathbf{e}_1$ ,  $\mathbf{F}^{(2)} = \bar{\mathbf{F}}^{(2)} = \bar{\mathbf{D}}^{(2)} + \bar{\gamma}_n^{(2)} \mathbf{e}_3 \otimes \mathbf{e}_1$ , where  $\bar{\mathbf{D}}^{(1)} = \text{diag}(\bar{\lambda}_1^{(1)}, \bar{\lambda}_2^{(1)}, \bar{\lambda}_n)$  and  $\bar{\mathbf{D}}^{(2)} = \text{diag}(\bar{\lambda}_1^{(2)}, \bar{\lambda}_2^{(2)}, \bar{\lambda}_n)$  are coaxial with  $\bar{\mathbf{D}}$ , and  $\hat{\mathbf{F}}^{(1)} = \hat{F}_{\alpha\beta}^{(1)} \mathbf{e}_\alpha \otimes \mathbf{e}_\beta + \bar{\lambda}_n \mathbf{e}_3 \otimes \mathbf{e}_3 + \bar{\gamma}_n^{(1)} \mathbf{e}_3 \otimes \mathbf{e}_1$  ( $\alpha, \beta = 1, 2$ ). In the same spirit, the associated invariants  $I_N^{(1)}$ ,  $I_N^{(2)}$ ,  $I_P^{(1)}$  and  $I_P^{(2)}$  are generalized as follows:  $I_N^{(1)} \doteq I^{(1)} = \mathbf{F}^{(1)} \cdot \mathbf{F}^{(1)}$ ,  $I_N^{(2)} = I_P^{(2)} \doteq I^{(2)} = \bar{\mathbf{F}}^{(2)} \cdot \bar{\mathbf{F}}^{(2)}$  and  $I_P^{(1)} \doteq \hat{I}^{(1)} = \hat{\mathbf{F}}^{(1)} \cdot \hat{\mathbf{F}}^{(1)}$ .

Next, we assume that Eq. (37) for  $\bar{\lambda}_1$  in the context of the GSO estimate (34) and Eq. (24) for  $\bar{\gamma}_n^{(1)}$  in the context of the TSO estimate (26), hold for the more general loading condition  $\bar{\mathbf{F}} = \bar{\mathbf{D}} + \bar{\gamma}_n \mathbf{e}_3 \otimes \mathbf{e}_1$ , with the only difference that the invariants  $I_N^{(1)}$ ,  $I_N^{(2)}$ ,  $I_P^{(1)}$ , and  $I_P^{(2)}$  in the relevant expressions are replaced by their more general counterparts defined above, i.e.,  $I_N^{(1)} \rightarrow I^{(1)} = \mathbf{F}^{(1)} \cdot \mathbf{F}^{(1)}$ ,  $\{I_N^{(2)}, I_P^{(2)}\} \rightarrow I^{(2)} = \bar{\mathbf{F}}^{(2)} \cdot \bar{\mathbf{F}}^{(2)}$  and  $I_P^{(1)} \rightarrow \hat{I}^{(1)} = \hat{\mathbf{F}}^{(1)} \cdot \hat{\mathbf{F}}^{(1)}$ .

Finally, making use of the above hypotheses, we obtain the following estimate for the effective stored-energy function (8) of fiber-reinforced elastomers, with incompressible generalized Neo-Hookean phases, subjected to general loading conditions:

$$\widehat{W}(\bar{\mathbf{F}}) = \widehat{\Phi}(\bar{\lambda}_n, \bar{\gamma}_p, \bar{\gamma}_n) = (1 - c_0)[g^{(1)}(\hat{I}^{(1)}) + g_I^{(1)}(I^{(1)})\bar{\gamma}_n^{(1)}(\bar{\gamma}_n - \bar{\gamma}_n^{(1)})] + c_0[g^{(2)}(I^{(2)}) + g_I^{(2)}(I^{(2)})\bar{\gamma}_n^{(2)}(\bar{\gamma}_n - \bar{\gamma}_n^{(2)})], \quad (38)$$

referred in what follows as the SOE. In this expression,

$$\begin{aligned} \hat{I}^{(1)} &= \hat{\mathbf{F}}^{(1)} \cdot \hat{\mathbf{F}}^{(1)} = \left( \frac{\bar{\lambda}_1 - c_0 \bar{\lambda}_1^{(2)}}{1 - c_0} \right)^2 + \left( \frac{\bar{\lambda}_2 - c_0 \bar{\lambda}_2^{(2)}}{1 - c_0} \right)^2 + \bar{\lambda}_n^2 + (\bar{\gamma}_n^{(1)})^2 + \\ &\quad \dots + c_0 \left( \frac{\bar{\lambda}_1 - \bar{\lambda}_1^{(2)}}{(1 - c_0) \bar{\lambda}_1 \bar{\lambda}_n} \right)^2 [(\bar{\lambda}_1^{(2)} \bar{\lambda}_1 \bar{\lambda}_n + 1)^2 + (\bar{\lambda}_1^2 + (\bar{\lambda}_1^{(2)})^2) \bar{\lambda}_n], \\ I^{(1)} &= \mathbf{F}^{(1)} \cdot \mathbf{F}^{(1)} = \bar{\lambda}_1^2 + \bar{\lambda}_2^2 + \bar{\lambda}_n^2 + (\bar{\gamma}_n^{(1)})^2, \\ I^{(2)} &= \bar{\mathbf{F}}^{(2)} \cdot \bar{\mathbf{F}}^{(2)} = (\bar{\lambda}_1^{(2)})^2 + (\bar{\lambda}_2^{(2)})^2 + \bar{\lambda}_n^2 + (\bar{\gamma}_n^{(2)})^2, \end{aligned} \quad (39)$$

where

$$\begin{aligned} \bar{\lambda}_1 &= \frac{\sqrt{\bar{\gamma}_p^2 + 4/\bar{\lambda}_n} - \bar{\gamma}_p}{2}, \quad \bar{\lambda}_2 = \frac{\sqrt{\bar{\gamma}_p^2 + 4/\bar{\lambda}_n} + \bar{\gamma}_p}{2}, \\ \bar{\lambda}_2^{(2)} &= \frac{1}{\bar{\lambda}_1^{(2)} \bar{\lambda}_n}, \quad \bar{\gamma}_n^{(2)} = \frac{\bar{\gamma}_n - (1 - c_0) \bar{\gamma}_n^{(1)}}{c_0}, \end{aligned} \quad (40)$$

and the variables  $\bar{\lambda}_1^{(2)}$  and  $\bar{\gamma}_n^{(1)}$  are the solution of the system of *coupled*, non-linear equations:

$$\sqrt{g_I^{(1)}(I^{(1)})[g_I^{(1)}(I^{(1)}) + 2g_{II}^{(1)}(I^{(1)})(\bar{\gamma}_n^{(1)})^2](\bar{\gamma}_n - \bar{\gamma}_n^{(1)}) + c_0(g_I^{(2)}(I^{(2)})\bar{\gamma}_n^{(2)} - g_I^{(1)}(I^{(1)})\bar{\gamma}_n^{(1)})} = 0 \quad (41)$$

and

$$[(1 + c_0)g_I^{(1)}(I^{(1)}) + (1 - c_0)g_I^{(2)}(I^{(2)})](\bar{\lambda}_n^{-2} - (\bar{\lambda}_1^{(2)})^4) - g_I^{(1)}(I^{(1)}) \left[ 2\bar{\lambda}_1^{(2)}\bar{\lambda}_1(\bar{\lambda}_2^2 - (\bar{\lambda}_1^{(2)})^2) - \frac{(\bar{\lambda}_1 - (\bar{\lambda}_1^{(2)})^4)\bar{\lambda}_2}{\bar{\lambda}_1} \right] = 0. \quad (42)$$

In connection with the above estimate for fiber-reinforced elastomers, it is useful to make the following remarks:

- As opposed to other constitutive models that have thus far been proposed in the literature, model (38) permits consideration of constitutive behaviors for the matrix and fibers that are more general than Neo-Hookean.
- For generalized plane-strain deformations ( $\bar{\gamma}_n = 0$ ), the stored-energy function (38) reduces identically to the GSO estimate (34). On the other hand, for antiplane-axisymmetric shear deformations ( $\bar{\gamma}_p = 0$ ), (38) reduces to the TSO estimate (26). For more general loading conditions with  $\bar{\gamma}_n \neq 0$  and  $\bar{\gamma}_p \neq 0$ , (38) can be thought of as a *non-linear interpolation* between (34) and (26).
- The computation of the constitutive model (38) amounts to solving a system of only two non-linear algebraic equations, (41) and (42), and thus it can be readily implemented into commercial numerical packages (e.g., ABAQUS) for solving structural problems of interest.
- In the limit of small deformations, (38) recovers the corresponding linear-elastic Willis estimate, which is known to be very accurate for small to moderate values of the volume fraction of fibers  $c_0$ .

#### 4.1. Comparisons with bounds and other estimates

For the fairly general class of matrix and fiber behaviors of interest in this work, the only known (non-trivial) bound for the effective stored-energy function (8) is the Voigt upper bound (Ogden, 1978):

$$\hat{\Phi}^V(\bar{\lambda}_n, \bar{\gamma}_p, \bar{\gamma}_n) = (1 - c_0)g^1(\bar{I}) + c_0g^2(\bar{I}), \quad (43)$$

where  $\bar{I} = \bar{\mathbf{F}} \cdot \bar{\mathbf{F}} = 2/\bar{\lambda}_n + \bar{\lambda}_n^{-2} + \bar{\gamma}_p^2 + \bar{\gamma}_n^2$ . It should be noted that (43) is isotropic and depends on the microstructure only through the volume fraction of the fibers  $c_0$ . In general, result (43) is expected to be too stiff, in view of the fact that it is a rigorous upper bound. Note, for instance, that in the limit of rigid fibers (43) becomes unbounded for all  $\bar{\mathbf{F}} \neq \mathbf{I}$ , that is, the composite is undeformable, which is in contradiction with physical evidence (for transverse and longitudinal shear loading). Nevertheless, there exist certain special cases for which the Voigt bound is an *exact* result, as it will be discussed below (see He et al., 2006 for a general discussion on uniform-field exact solutions in fiber-reinforced elastomers). At any rate, the proposed constitutive model (38) can be shown to satisfy the rigorous upper bound (43) for all deformations.

The Voigt bound (43) makes use of the trial field  $\mathbf{F}(\mathbf{X}) = \bar{\mathbf{F}}$  for all  $\mathbf{X}$  in  $\Omega_0$ , which in general is consistent with the equilibrium requirement at all points  $\mathbf{X}$  in  $\Omega_0^{(1)}$  (but *not* on  $\partial\Omega_0^{(1)}$ ) and all  $\mathbf{X}$  in  $\Omega_0^{(2)}$  (but *not* on  $\partial\Omega_0^{(2)}$ ). Thus, this field is an exact solution to problem (8) for all values of  $\bar{\mathbf{F}}$  whenever the traction-continuity requirement across the matrix/fibers interfaces:

$$\mathbf{S}^{(1)}(\bar{\mathbf{F}})\mathbf{e} = \mathbf{S}^{(2)}(\bar{\mathbf{F}})\mathbf{e} \quad \text{for all } \mathbf{e} \perp \mathbf{N} \text{ with } \mathbf{e} \cdot \mathbf{e} = 1 \quad (44)$$

are satisfied. For axisymmetric shear deformations, conditions (44) reduce to the constraint  $p^{(1)} - p^{(2)} = 2(g_I^{(1)}(\bar{I}) - g_I^{(2)}(\bar{I}))/\bar{\lambda}_n$ —with  $p^{(1)}$  and  $p^{(2)}$  denoting, respectively, the constant pressures in the matrix and fibers—which can be satisfied for any value of  $\bar{\lambda}_n$ . In other words, the Voigt bound (43) is exact for axisymmetric shear deformations. For loading conditions other than axisymmetric shear, (44) is equivalent to  $p^{(1)} = p^{(2)}$  and  $g_I^{(1)}(\bar{I}) = g_I^{(2)}(\bar{I})$ , which actually require equality of the stress tensors in the matrix and fibers. Although the first of these conditions can always be enforced, the second cannot be satisfied, in general. For example, in the case of composites with Neo-Hookean phases  $g_I^{(1)}(\bar{I}) = \mu^{(1)}/2 \neq \mu^{(2)}/2 = g_I^{(2)}(\bar{I})$ . However, there are materials for which the requirement  $g_I^{(1)}(\bar{I}) = g_I^{(2)}(\bar{I})$  is met for certain values of  $\bar{I}$ . An example is that of composites with Gent constituents, for which the requirement specializes to

$$\frac{\mu^{(1)}J_m^{(1)}}{J_m^{(1)} - (\bar{I} - 3)} = \frac{\mu^{(2)}J_m^{(2)}}{J_m^{(2)} - (\bar{I} - 3)}, \quad (45)$$

where  $J_m^{(1)}$  and  $J_m^{(2)}$  denote the lock-up parameters of the matrix and fibers, respectively. The above equation has a unique, physically plausible solution for  $\bar{I}$  provided that  $(J_m^{(1)}(3 + J_m^{(2)})\mu^{(1)} - J_m^{(2)}(3 + J_m^{(1)})\mu^{(2)})/(J_m^{(1)}\mu^{(1)} - J_m^{(2)}\mu^{(2)}) > 3$ . It is not difficult to show that the proposed constitutive model (38) recovers such exact solutions of the Voigt-type. Indeed, note that the values  $\bar{\lambda}_1^{(2)} = \bar{\lambda}_1$  and  $\bar{\gamma}_n^{(1)} = \bar{\gamma}_n$ , for which we have  $\hat{I}^{(1)} = I^{(1)} = I^{(2)} = \bar{I}$ , constitute a solution of the system of Eqs. (41) and (42) whenever  $g_I^{(1)}(\bar{I}) = g_I^{(2)}(\bar{I})$ , and for these special cases, the stored-energy function (38) reduces to the Voigt bound (43).

For the special case when the underlying matrix and fibers are characterized by incompressible Neo-Hookean solids, deBotton et al. (2006) derived the following estimate for the effective stored-energy function (8):

$$\widehat{\Phi}^{BHS}(\bar{\lambda}_n, \bar{\gamma}_p, \bar{\gamma}_n) = \frac{1}{2}\bar{\mu}\left(\bar{\lambda}_n + \frac{2}{\bar{\lambda}_n} - 3\right) + \frac{\tilde{\mu}}{2}(\bar{\gamma}_p^2 + \bar{\gamma}_n^2), \quad (46)$$

where  $\bar{\mu} = (1 - c_0)\mu^{(1)} + c_0\mu^{(2)}$  and

$$\tilde{\mu} = \mu^{(1)}\frac{(1 - c_0)\mu^{(1)} + (1 + c_0)\mu^{(2)}}{(1 + c_0)\mu^{(1)} + (1 - c_0)\mu^{(2)}}. \quad (47)$$

A nice feature of this estimate—beyond its simplicity—is that it is *exact* for antiplane combined with axisymmetric shear deformations ( $\bar{\gamma}_p = 0$ ) for fiber-reinforced Neo-Hookean elastomers with the “composite cylinder assemblage” microstructure of Hashin. Specializing now the constitutive model (38) to Neo-Hookean phases—namely, setting  $g^{(1)} = \mu^{(1)}(I - 3)/2$  and  $g^{(2)} = \mu^{(2)}(I - 3)/2$ —leads to

$$\widehat{\Phi}(\bar{\lambda}_n, \bar{\gamma}_p, \bar{\gamma}_n) = (1 - c_0)\frac{\mu^{(1)}}{2}(\hat{I}_p^{(1)} - 3) + c_0\frac{\mu^{(2)}}{2}(\hat{I}_p^{(2)} - 3) + \frac{\tilde{\mu}}{2}\bar{\gamma}_n^2, \quad (48)$$

where it is recalled that  $\hat{I}_p^{(1)}$  and  $\hat{I}_p^{(2)}$  are given by expressions (35) and (36), depending on the unknown  $\bar{\lambda}_1^{(2)}$  to be computed as the solution to the fourth-order polynomial equation resulting from (42) by substituting  $g_p^{(1)}(\hat{I}_p^{(1)}) = \mu^{(1)}/2$  and  $g_p^{(2)}(\hat{I}_p^{(2)}) = \mu^{(2)}/2$ . Clearly, expressions (48) and (46) are different for general loading conditions. However, for antiplane combined with axisymmetric shear deformations ( $\bar{\gamma}_p = 0$ ), it is easy to verify that (48) reduces identically to (46), which, again, is exact for “composite cylinder assemblage” microstructures. For more general loading conditions, although not identical, estimates (46) and (48) lead to fairly similar results.

## 5. Results and discussion

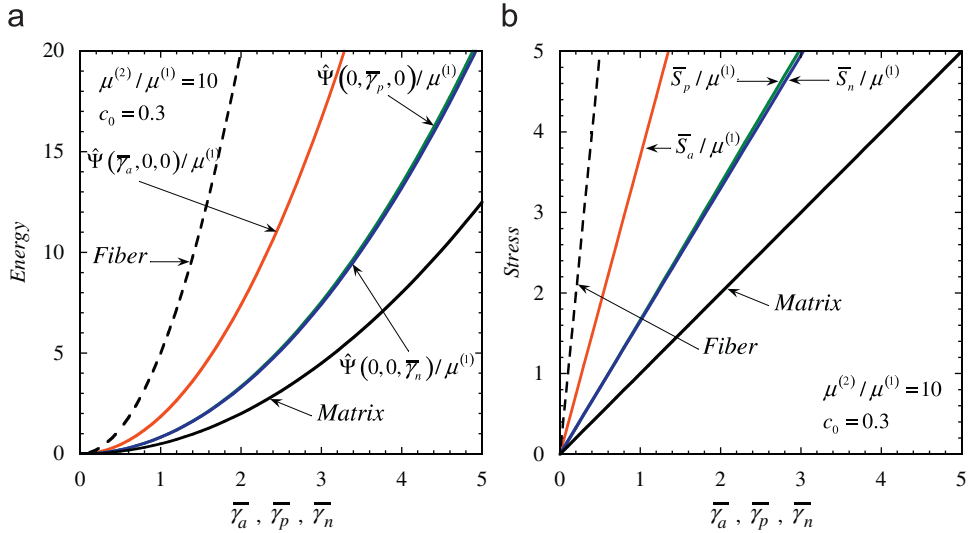
In this section, we examine in detail some important features of the predictions of the SOE (38) for the overall response of fiber-reinforced elastomers. For the purposes of the current discussion, we consider specific applications to composites with incompressible Neo-Hookean and Gent constituents. Particular emphasis is placed on the specific features of the predicted anisotropy and the coupling among the three modes of shear. In addition, the predictions of the SOE (38) are compared with the corresponding predictions of the deBotton et al. (BHS) model (46) (for Neo-Hookean phases) and the Voigt upper bound (43).

For the purpose of comparing the axisymmetric mode with the in-plane and antiplane modes, it proves useful to introduce the axisymmetric shear variable:  $\bar{\gamma}_a = \pm(\bar{\lambda}_n + 2/\bar{\lambda}_n - 3)^{1/2}$ , where the plus and minus signs correspond to  $\bar{\lambda}_n > 1$  and  $\bar{\lambda}_n < 1$ , respectively. A path in loading space is defined by  $\bar{\gamma}_a = \alpha_a\bar{\gamma}_e$ ,  $\bar{\gamma}_p = \alpha_p\bar{\gamma}_e$  and  $\bar{\gamma}_n = \alpha_n\bar{\gamma}_e$ , where  $\alpha_a$ ,  $\alpha_p$  and  $\alpha_n$  are constants, such that  $\bar{\gamma}_e \equiv (\bar{\gamma}_a^2 + \bar{\gamma}_p^2 + \bar{\gamma}_n^2)^{1/2} = (I - 3)^{1/2}$ . For our objectives here, it will be sufficient to confine our attention to loadings for which  $\bar{\gamma}_a \geq 0$ . Furthermore, we define the stored-energy function  $\widehat{\Psi}$  such that  $\widehat{\Psi}(\bar{\gamma}_a, \bar{\gamma}_p, \bar{\gamma}_n) = \widehat{\Phi}(\bar{\lambda}_n, \bar{\gamma}_p, \bar{\gamma}_n)$ , and the stress measures  $\bar{s}_a = \partial\widehat{\Psi}/\partial\bar{\gamma}_a$ ,  $\bar{s}_p = \partial\widehat{\Psi}/\partial\bar{\gamma}_p$  and  $\bar{s}_n = \partial\widehat{\Psi}/\partial\bar{\gamma}_n$ , so that the effective response of a composite for a given loading path may be conveniently represented by  $\widehat{\Psi}$  and/or  $\bar{s}_a$ ,  $\bar{s}_p$ ,  $\bar{s}_n$  as functions of  $\bar{\gamma}_e$ .

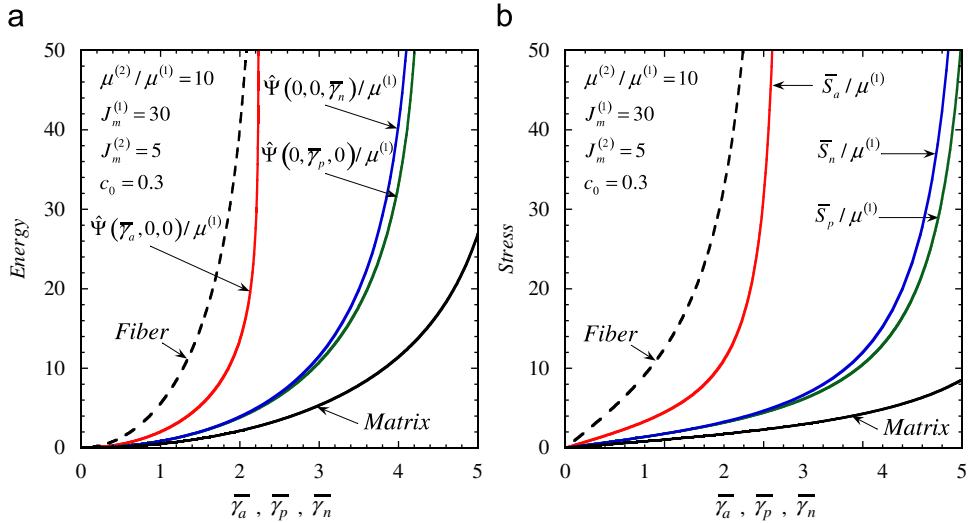
Fig. 3 shows the SOE for the response of a fiber-reinforced elastomer with incompressible Neo-Hookean phases under axisymmetric, in-plane and antiplane shear loadings. The calculations have been performed for a contrast  $\mu^{(2)}/\mu^{(1)} = 10$  and an initial volume fraction of the fibers  $c_0 = 0.3$ . Recall that the predictions of the Voigt model are identical for all three modes of shear and coincide with the predictions of the SOE and the BHS model for the axisymmetric mode, which correspond to exact solutions of the Voigt-type (see Section 4), while the predictions of the BHS model for the other two modes are the same as the SOE predictions for antiplane shear. It is observed that the response of the composite under axisymmetric shear is much stiffer than its responses under the other two modes (as predicted by the SOE and BHS models). This observation together with the fact that the predictions of the SOE and BHS models are exact for the antiplane mode and the composite cylinder assemblage microstructure illustrates how inaccurate the predictions of the Voigt model can be. In addition, according to the SOE, the in-plane mode is slightly stiffer than the antiplane mode. The difference between these predictions increases with increasing amount of shear, but it remains small even at very large deformations.

The results shown in Fig. 3 are representative of the main features of the SOE (48), and for this reason we do not include here further applications for composites with incompressible Neo-Hookean constituents. It should be noticed, however, that the difference between the SOE predictions for the responses under in-plane and antiplane shear loadings increase with increasing  $\mu^{(2)}/\mu^{(1)}$  and/or increasing  $c_0$ , although not significantly. In addition, calculations for combined loading conditions have shown that coupling effects are also insignificant. The above observations lead us to the conclusion that the predictions of the SOE and BHS models for composites with incompressible Neo-Hookean constituents are in good agreement.

Fig. 4 shows the same results as Fig. 3, but for a composite made of incompressible Gent phases with  $\mu^{(2)}/\mu^{(1)} = 10$ ,  $J_m^{(1)} = 30$ ,  $J_m^{(2)} = 5$  and  $c_0 = 0.3$ . Recall again that the SOE for the axisymmetric mode is identical to the Voigt upper bound and therefore exact. It is observed that, in accordance with the behavior of a homogeneous Gent material, there exists a range of deformations for which the effective response of the composite under consideration can be approximated reasonably well by the corresponding response of the composite with incompressible Neo-Hookean phases shown in Fig. 3.



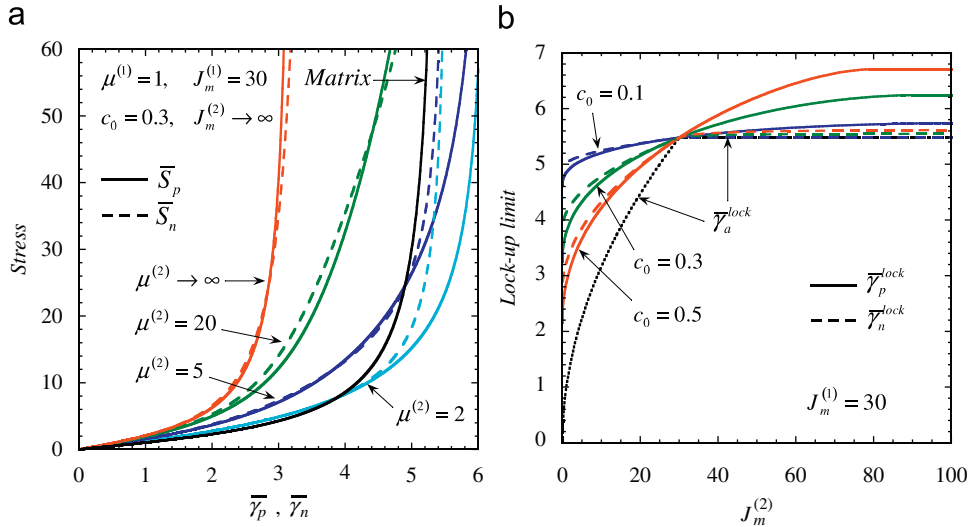
**Fig. 3.** Second-order estimates (SOEs) for the effective response of a fiber-reinforced elastomer with incompressible Neo-Hookean phases under the three different modes of shear. The results are shown as a function of the corresponding amount of shear  $\bar{\gamma}_a, \bar{\gamma}_p$  and  $\bar{\gamma}_n$ . (a) The energies  $\hat{\Psi}(\bar{\gamma}_a, 0, 0)$ ,  $\hat{\Psi}(0, \bar{\gamma}_p, 0)$  and  $\hat{\Psi}(0, 0, \bar{\gamma}_n)$ , normalized by  $\mu^{(1)}$ . (b) The corresponding (non-zero) normalized stresses  $\bar{S}_a(\bar{\gamma}_a, 0, 0)$ ,  $\bar{S}_p(0, \bar{\gamma}_p, 0)$  and  $\bar{S}_n(0, 0, \bar{\gamma}_n)$ .



**Fig. 4.** Second-order estimates (SOEs) for the effective response of a fiber-reinforced elastomer with incompressible Gent phases under the three different modes of shear. The results are shown as a function of the corresponding amount of shear  $\bar{\gamma}_a, \bar{\gamma}_p$  and  $\bar{\gamma}_n$ . (a) The energies  $\hat{\Psi}(\bar{\gamma}_a, 0, 0)$ ,  $\hat{\Psi}(0, \bar{\gamma}_p, 0)$  and  $\hat{\Psi}(0, 0, \bar{\gamma}_n)$ , normalized by  $\mu^{(1)}$ . (b) The corresponding (non-zero) normalized stresses  $\bar{S}_a(\bar{\gamma}_a, 0, 0)$ ,  $\bar{S}_p(0, \bar{\gamma}_p, 0)$  and  $\bar{S}_n(0, 0, \bar{\gamma}_n)$ .

This is because, within this range of deformations, the contribution of the parameters  $J_m^{(1)}$  and  $J_m^{(2)}$  is insignificant. It is also observed that the predictions for the axisymmetric mode are significantly stiffer than the predictions for the other two modes, and that this difference increases with increasing deformation. Finally, as opposed to the case of the composite with Neo-Hookean phases (Fig. 3), we observe that here the difference between the predicted responses under in-plane and antiplane shear is substantial at large deformations, with the predictions for the antiplane mode being stiffer.

Fig. 5(a) compares the SOE for the in-plane and antiplane shear responses of fiber-reinforced elastomers with an incompressible Gent matrix with  $\mu^{(1)} = 1$  and  $J_m^{(1)} = 30$  and incompressible Neo-Hookean fibers with  $\mu^{(2)} = 2, 5, 20, \infty$  and initial concentration  $c_0 = 0.3$ . It is observed that for all values of  $\mu^{(2)}$  there exists a range of deformations, extending well beyond the linear-elastic regime, for which the predictions for  $\bar{S}_p$  and  $\bar{S}_n$  are essentially the same. The size of this range increases with decreasing  $\mu^{(2)}$ . For larger values of the deformation, the predictions for  $\bar{S}_p$  and  $\bar{S}_n$  are different, in general. The fact that the matrix of the composites considered here is a Gent material imposes limits in the amount of the applied deformation. We observe that the stresses  $\bar{S}_p$  and  $\bar{S}_n$  increase rapidly near the corresponding lock-up limits of the composites, which turn out to be different for in-plane and antiplane shear loadings. Specifically, for the composite with



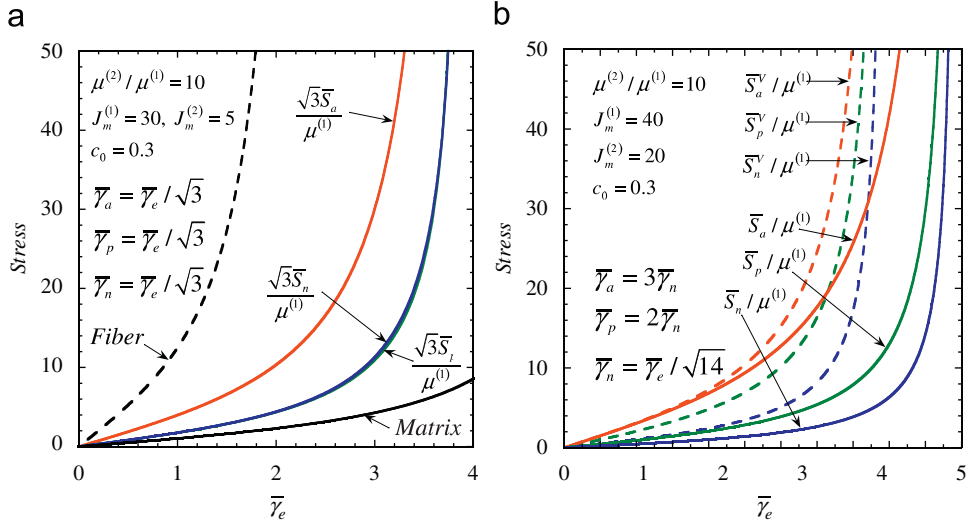
**Fig. 5.** (a) SOE predictions for the macroscopic stresses  $\bar{S}_p(0, \bar{\gamma}_p, 0)$  and  $\bar{S}_n(0, 0, \bar{\gamma}_n)$  of fiber-reinforced elastomers made out of an incompressible Gent matrix and Neo-Hookean fibers, with various values of  $\mu^{(2)}$ , as functions of the amounts of shear  $\bar{\gamma}_p$  and  $\bar{\gamma}_n$ , respectively. (b) SOE predictions for the lock-up limits  $\bar{\gamma}_a^{lock}$ ,  $\bar{\gamma}_p^{lock}$  and  $\bar{\gamma}_n^{lock}$  of axisymmetric, in-plane and antiplane shear loadings, respectively—applied to composites with incompressible Gent constituents—as functions of the lock-up parameter of the fibers,  $J_m^{(2)}$ , for various values of  $c_0$  and  $J_m^{(1)} = 30$ .

rigid fibers ( $\mu^{(2)} \rightarrow \infty$ ) it is found that  $\bar{\gamma}_p^{lock} < \bar{\gamma}_n^{lock}$ , while for the other three cases ( $\mu^{(2)} = 2, 5, 20$ ) we have  $\bar{\gamma}_n^{lock} < \bar{\gamma}_p^{lock}$  (see Fig. 5(b)). Finally, note that the intersection points of the curves representing  $\bar{S}_p, \bar{S}_n$  and the corresponding stress measure of the matrix material (subjected to simple shear) correspond to exact solutions of the Voigt-type.

Fig. 5(b) presents the SOE predictions for the limiting values of axisymmetric,  $\bar{\gamma}_a^{lock}$ , in-plane,  $\bar{\gamma}_p^{lock}$ , and antiplane,  $\bar{\gamma}_n^{lock}$ , shear loadings at which composites made out of incompressible Gent phases lock up. The results are shown as functions of  $J_m^{(2)}$  for a fixed lock-up parameter of the matrix,  $J_m^{(1)} = 30$ , and volume fractions of the fibers  $c_0 = 0.1, 0.3, 0.5$ . It is easy to show that  $\bar{\gamma}_a^{lock} = \min\{(J_m^{(1)})^{1/2}, (J_m^{(2)})^{1/2}\}$ , which, once again, is an exact result, and may be interpreted as follows: for  $J_m^{(2)} < J_m^{(1)}$  the composite locks-up because the fibers lock-up, for  $J_m^{(2)} > J_m^{(1)}$  the overall lock-up is due to the lock-up of the matrix and for  $J_m^{(2)} = J_m^{(1)}$  the composite locks-up because both phases lock-up. The lock-up limit for in-plane shear loadings is the limiting value of  $\bar{\gamma}_p$ , which, along with the corresponding limiting value of  $\bar{\lambda}_1^{(2)}$ , satisfy at least one of the conditions  $\bar{I}^{(1)} - 3 = J_m^{(1)}$  and  $\bar{I}^{(2)} - 3 = J_m^{(2)}$ . It turns out that, for values of  $J_m^{(2)}$  between zero and a “critical” value, depending on  $c_0$ , both conditions are met and they provide a system of two equations for the computation of the two unknowns, namely  $\bar{\gamma}_p^{lock}$  and the corresponding limiting value of  $\bar{\lambda}_1^{(2)}$ . The aforementioned critical value of  $J_m^{(2)}$  corresponds to the maximum  $\bar{\gamma}_p^{lock}$  obtained in this way. For  $J_m^{(2)}$  greater than this critical value, only the condition  $\bar{I}^{(1)} - 3 = J_m^{(1)}$  is satisfied and  $\bar{\gamma}_p^{lock}$  is independent of  $J_m^{(2)}$ . Note that  $\bar{\gamma}_p^{lock}$  is independent of  $\mu^{(2)}/\mu^{(1)}$  for all values of  $J_m^{(2)}$ . Similarly, the lock-up limit for antiplane shear loadings is the limiting value of  $\bar{\gamma}_n$ , which, along with the corresponding limiting value of  $\bar{\lambda}_1^{(1)}$ , satisfy at least one of the conditions  $\bar{I}^{(1)} - 3 = J_m^{(1)}$  or  $\bar{I}^{(2)} - 3 = J_m^{(2)}$ . For  $J_m^{(2)} \leq J_m^{(1)}$  both conditions are satisfied, and  $\bar{\gamma}_n^{lock} = (1 - c_0)(J_m^{(1)})^{1/2} + c_0(J_m^{(2)})^{1/2}$ . For  $J_m^{(2)} > J_m^{(1)}$ , only the first condition is met in the lock-up limit, which in this case depends weakly on  $\mu^{(2)}/\mu^{(1)}$ , and the corresponding results presented in this figure have been obtained numerically for  $\mu^{(2)}/\mu^{(1)} = 10$ . It is observed that  $\bar{\gamma}_a^{lock}$  is less than both  $\bar{\gamma}_p^{lock}$  and  $\bar{\gamma}_n^{lock}$  for all values of  $J_m^{(2)}$ . For  $J_m^{(2)} < J_m^{(1)}$ , we observe that  $\bar{\gamma}_p^{lock} < \bar{\gamma}_n^{lock}$ , while for  $J_m^{(2)} > J_m^{(1)}$  it is found that  $\bar{\gamma}_p^{lock} > \bar{\gamma}_n^{lock}$ . In addition, the difference between  $\bar{\gamma}_p^{lock}$  and  $\bar{\gamma}_n^{lock}$  increases with increasing  $|J_m^{(2)} - J_m^{(1)}|$ , which is a measure of the heterogeneity contrast at large deformations, and/or increasing  $c_0$ . Finally, for  $J_m^{(2)} = J_m^{(1)}$  and any value of  $c_0$ , it is found that  $\bar{\gamma}_a^{lock} = \bar{\gamma}_p^{lock} = \bar{\gamma}_n^{lock} = (J_m^{(1)})^{1/2}$ , which corresponds to an exact result of the Voigt-type.

Fig. 6(a) illustrates the coupling in the SOE for the response of the composite considered in Fig. 4. It is observed that, for the range of values of  $\bar{\gamma}_e$  for which the overall response of this composite is essentially identical with the response of the corresponding composite with Neo-Hookean constituents (see discussion related to Fig. 3), the stresses associated with the combined loading are actually the same as the corresponding stresses for the individual modes (i.e., the three modes are uncoupled). For larger values of  $\bar{\gamma}_e$ , the overall response of the composite under the combined loading differs significantly from its responses under the pure loadings. Specifically, we observe that, although for combined loading the axisymmetric shear stress is substantially larger than the other two stress measures, the associated difference is much smaller than the corresponding difference observed for the case of the pure modes, and the same is true for the difference between the in-plane and antiplane shear stresses. Furthermore, for the limiting value of  $\bar{\gamma}_e$  at which the composite locks-up, all stress measures become unbounded for the combined loading.

Fig. 6(b) presents the SOE and the Voigt upper bound for the effective behavior of a fiber-reinforced elastomer made out of incompressible Gent constituents when subjected to combined loading conditions. It is observed that the predictions of



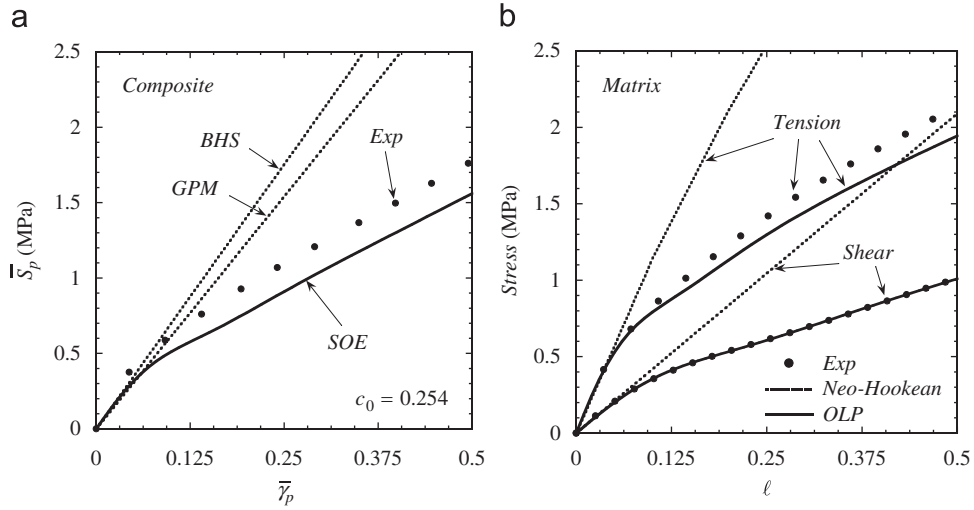
**Fig. 6.** (a) Combined loading of the composite considered in Fig. 3. The predictions of the SOE for the stresses  $\sqrt{3}\bar{S}_a(\bar{\gamma}_a, \bar{\gamma}_p, \bar{\gamma}_n)$ ,  $\sqrt{3}\bar{S}_p(\bar{\gamma}_a, \bar{\gamma}_p, \bar{\gamma}_n)$  and  $\sqrt{3}\bar{S}_n(\bar{\gamma}_a, \bar{\gamma}_p, \bar{\gamma}_n)$ , normalized by  $\mu^{(1)}$ , are plotted as functions of the loading parameter  $\bar{\gamma}_e$ . (b) Combined loading of a fiber-reinforced elastomer with incompressible Gent phases. The normalized stresses  $\bar{S}_a(\bar{\gamma}_a, \bar{\gamma}_p, \bar{\gamma}_n)$ ,  $\bar{S}_p(\bar{\gamma}_a, \bar{\gamma}_p, \bar{\gamma}_n)$  and  $\bar{S}_n(\bar{\gamma}_a, \bar{\gamma}_p, \bar{\gamma}_n)$ , as predicted by the SOE and the Voigt upper bound (distinguished by a superscript “V”), are shown as functions of the loading parameter  $\bar{\gamma}_e$ .

the two models for the effective response differ significantly, and that the differences increase with increasing deformation. The Voigt model predicts that the limiting value of  $\bar{\gamma}_e$  for which the composite locks-up is identical with the corresponding value of the material of the fibers (note that in this case  $J_m^{(2)} < J_m^{(1)}$ ), while according to the SOE model, this value is in-between the corresponding values associated with the lock-up limits of the phase materials.

Finally, Fig. 7(a) presents a comparison of the SOE with the experimental results of [Lahellec et al. \(2004\)](#) for the transverse shear response of a fiber-reinforced elastomer with a *periodic* microstructure. While this comparison is not entirely fair since the SOE is for a random microstructure, and corresponds to the softest possible estimate for this type of microstructure (since it arises from the Willis estimates (16), which is a rigorous lower bound for the effective modulus tensor), the comparison shows that the method has the capability to incorporate more general constitutive behaviors (than Neo-Hookean) for the matrix phase, leading to much better agreement with experimental results for actual rubbers. Thus, it can be seen that the SOE greatly improves on the predictions of the model of [deBotton et al. \(2006\)](#) (BHS) for reinforced Neo-Hookean solids, as well as the model of [Guo et al. \(2006\)](#) (GPM), which appears to be inconsistent with the bounding character of the Willis estimate for infinitesimal deformations. In connection with these results, it should be mentioned that the matrix phase was fitted with the OLP model ([Lopez-Pamies, 2008](#); see also [Lopez-Pamies et al., 2008](#), Eq. (33)) for a simple shear test, as depicted in Fig. 7(b). The material parameters for this (incompressible) matrix model were found to be:  $\mu_1^{(1)} = 0.6$  MPa,  $\mu_2^{(1)} = 1.664$  MPa,  $\mu_3^{(1)} = 1.915$  MPa,  $\alpha_1^{(1)} = -16.212$ ,  $\alpha_2^{(1)} = -133.471$  and  $\alpha_3^{(1)} = 0.696$ . (Note that the shear modulus of the Neo-Hookean and OLP models is  $\mu^{(1)} = \mu_1^{(1)} + \mu_2^{(1)} + \mu_3^{(1)} = 4.179$  MPa.) In Fig. 7(b), a comparison is also provided with the experimental results for the matrix material in tension, which although not quite as good as for simple shear for the parameters chosen, is still much better than the Neo-Hookean model.

## 6. Concluding remarks

In this work, we have developed a homogenization-based constitutive model for *incompressible* fiber-reinforced elastomers with *random* microstructures. In particular, the model applies to composites with a single family of aligned, cylindrical fibers distributed randomly and isotropically in the matrix phase (in the undeformed configuration). The constitutive behavior of the phases is characterized by stored-energies that are *general* functions of the first invariant of the right Cauchy–Green deformation tensor (i.e., generalized Neo-Hookean materials). The model provides a generalization of the results of [Lopez-Pamies and Ponte Castañeda \(2006b\)](#) for plane-strain (transverse in-plane) loading of fiber-reinforced elastomers (with rigid fibers), which it recovers in this limiting case. Compared to other constitutive models proposed thus far in the literature (e.g., [Guo et al., 2006](#); [deBotton et al., 2006](#)) for the materials of interest here, the present model has the unique feature that it is applicable to composites with more general matrix and fiber behaviors than Neo-Hookean while directly accounting in a rigorous manner for the microstructure. Given the well-known limitations of the Neo-Hookean model at large strains, the new model should prove useful in practical applications involving large elastic strains. Other important features of the model include the fact that it accounts for the coupling among the three possible modes of shear that are expected under general loading conditions, and that it recovers available exact solutions. More specifically, the model recovers any possible exact Voigt-type (i.e., uniform deformation field) solution, as well as the exact effective



**Fig. 7.** (a) The predictions of the SOE, BHS and GPM models for the effective in-plane shear response of an elastomer reinforced with *randomly* distributed, rigid fibers are compared with corresponding experimental (Exp) results for *periodically* distributed fibers. The SOE makes use of the OLP model for the matrix. (b) The OLP and Neo-Hookean models, used to characterize the matrix material in part (a), are compared with corresponding experimental data for *uniaxial tension* ( $\ell = \lambda - 1$ , with  $\lambda$  being the tensile stretch) and *simple shear* ( $\ell = \gamma$ , with  $\gamma$  denoting the amount of shear) tests.

response of fiber-reinforced elastomers with Neo-Hookean phases and composite cylinder assemblage microstructures subjected to axisymmetric combined with antiplane shear deformations. A further strength of the model is that—in spite of incorporating fine microstructural information—it is still relatively simple to implement, requiring only the solution of a system of two non-linear, algebraic equations. Finally, it is important to emphasize that one of the advantages of these homogenization-based models is that they can be generalized, at least in principle, to more general types of microstructures and material behaviors.

## Acknowledgments

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## Appendix A. GSO estimates for incompressible fiber-reinforced rubbers under generalized plane-strain conditions

In this appendix, we sketch the derivation for the incompressibility limit of the GSO estimate (28) under generalized plane-strain loading conditions. To this end, we first present the specialization of the general system of Eqs. (29)–(32) associated with the compressible problem to generalized plane-strain deformations, and then discuss the corresponding asymptotic analysis in the limit as  $\kappa^{(1)} = \kappa^{(2)} \rightarrow \infty$ .

The generalized plane-strain problem corresponds to an  $\bar{\mathbf{F}}$  with components (in the coordinate system  $\mathbf{e}_i$ ) given by (11) with  $\bar{\gamma}_n = 0$ . Because of the objectivity and material symmetry (invariance under rotations about  $\mathbf{N} = \mathbf{e}_3$ ) of the GSO estimate (28), there is no loss of generality in considering a diagonal  $\bar{\mathbf{F}}$  of the form

$$\bar{\mathbf{F}} = \bar{\mathbf{D}} = \bar{\lambda}_1 \mathbf{e}_1 \otimes \mathbf{e}_1 + \bar{\lambda}_2 \mathbf{e}_2 \otimes \mathbf{e}_2 + \bar{\lambda}_n \mathbf{e}_3 \otimes \mathbf{e}_3, \quad (49)$$

where

$$\bar{\lambda}_1 = \frac{\sqrt{\bar{\gamma}_p^2 + 4\bar{\lambda}_p^2} - \bar{\gamma}_p}{2} \quad \text{and} \quad \bar{\lambda}_2 = \frac{\sqrt{\bar{\gamma}_p^2 + 4\bar{\lambda}_p^2} + \bar{\gamma}_p}{2}. \quad (50)$$

In this case, we have that  $\bar{\mathbf{R}} = \bar{\mathbf{Q}} = \mathbf{I}$  and  $\mathbf{L}^{(1)} = \mathbf{L}^*$  in (27). In addition, following Lopez-Pamies and Ponte Castañeda (2006b) and Brun et al. (2007), the seven independent components of  $\mathbf{L}^*$  are chosen to be  $L_{1111}^* = \ell_1^*$ ,  $L_{2222}^* = \ell_2^*$ ,  $L_{3333}^* = \ell_3^*$ ,  $L_{1122}^* = \ell_4^*$ ,  $L_{1133}^* = \ell_5^*$ ,  $L_{2233}^* = \ell_6^*$ ,  $L_{1212}^* = \ell_7^*$ , while the other non-zero components are defined as

$$\begin{aligned} L_{2121}^* &= L_{1313}^* = L_{3131}^* = L_{2323}^* = L_{3232}^* = \ell_7^*, \\ L_{1221}^* &= \sqrt{(\ell_1^* - \ell_7^*)(\ell_2^* - \ell_7^*)} - \ell_4^*, \\ L_{1331}^* &= \sqrt{(\ell_1^* - \ell_7^*)(\ell_3^* - \ell_7^*)} - \ell_5^*, \\ L_{2332}^* &= \sqrt{(\ell_2^* - \ell_7^*)(\ell_3^* - \ell_7^*)} - \ell_6^*. \end{aligned} \quad (51)$$

Making use of the above choice for  $\mathbf{L}^{(1)}$ , it follows that the non-zero components of the microstructural tensor  $\mathbf{P}$ , as defined by (17), specialize to

$$\begin{aligned} P_{1111} &= \frac{\ell_2^* + \ell_7^*(1 + 2\sqrt{\ell_2^*/\ell_1^*})}{2\ell_7^*(\sqrt{\ell_1^*} + \sqrt{\ell_2^*})^2}, & P_{2222} &= \frac{\ell_1^* + \ell_7^*(1 + 2\sqrt{\ell_1^*/\ell_2^*})}{2\ell_7^*(\sqrt{\ell_1^*} + \sqrt{\ell_2^*})^2}, \\ P_{1212} &= \frac{\ell_2^* + \ell_7^* + 2\sqrt{\ell_1^*\ell_2^*}}{2\ell_7^*(\sqrt{\ell_1^*} + \sqrt{\ell_2^*})^2}, & P_{2121} &= \frac{\ell_1^* + \ell_7^* + 2\sqrt{\ell_1^*\ell_2^*}}{2\ell_7^*(\sqrt{\ell_1^*} + \sqrt{\ell_2^*})^2}, \\ P_{3131} &= P_{3232} = \frac{1}{2\ell_7^*}, & P_{1122} &= P_{1221} = -\frac{\sqrt{(\ell_1^* - \ell_7^*)(\ell_2^* - \ell_7^*)}}{2\ell_7^*(\sqrt{\ell_1^*} + \sqrt{\ell_2^*})^2}, \end{aligned} \quad (52)$$

$P_{2211} = P_{2112} = P_{1122}$ , where it is noted that all of these components depend only on the moduli  $\ell_1^*$ ,  $\ell_2^*$  and  $\ell_7^*$ .

In the context of the diagonal form (49), together with the above choice of  $\mathbf{L}^{(1)}$ , it follows from (29) and (30) that the tensors  $\bar{\mathbf{F}}^{(1)}$  and  $\bar{\mathbf{F}}^{(2)}$  have the same diagonal form as  $\bar{\mathbf{F}}$ , and in particular,  $\bar{\mathbf{F}}^{(1)} = \text{diag}(\bar{\lambda}_1^{(1)}, \bar{\lambda}_2^{(1)}, \bar{\lambda}_n)$  and  $\bar{\mathbf{F}}^{(2)} = \text{diag}(\bar{\lambda}_1^{(2)}, \bar{\lambda}_2^{(2)}, \bar{\lambda}_n)$ . In this connection, relation (29) may be conveniently re-written as

$$\bar{\lambda}_1^{(1)} = \frac{\bar{\lambda}_1 - c_0\bar{\lambda}_1^{(2)}}{1 - c_0} \quad \text{and} \quad \bar{\lambda}_2^{(1)} = \frac{\bar{\lambda}_2 - c_0\bar{\lambda}_2^{(2)}}{1 - c_0}. \quad (53)$$

Furthermore, the two non-trivial scalar equations in (30) reduce to

$$\begin{aligned} \bar{\lambda}_1 - \bar{\lambda}_1^{(2)} &= (1 - c_0)[P_{1111}E_{11} + P_{1122}E_{22}], \\ \bar{\lambda}_2 - \bar{\lambda}_2^{(2)} &= (1 - c_0)[P_{1122}E_{11} + P_{2222}E_{22}], \end{aligned} \quad (54)$$

with

$$\begin{aligned} E_{11} &= \ell_1^*(\bar{\lambda}_1 - \bar{\lambda}_1^{(2)}) + \ell_4^*(\bar{\lambda}_2 - \bar{\lambda}_2^{(2)}) - 2(\bar{g}_I^{(1)}\bar{\lambda}_1 - \bar{g}_I^{(2)}\bar{\lambda}_1^{(2)}) - \bar{\lambda}_2\bar{\lambda}_n[\bar{h}_J^{(1)} + \kappa^{(1)}(\bar{J} - 1)] + \bar{\lambda}_2^{(2)}\bar{\lambda}_n[\bar{h}_J^{(2)} + \kappa^{(2)}(\bar{J}^{(2)} - 1)], \\ E_{22} &= \ell_4^*(\bar{\lambda}_1 - \bar{\lambda}_1^{(2)}) + \ell_2^*(\bar{\lambda}_2 - \bar{\lambda}_2^{(2)}) - 2(\bar{g}_I^{(1)}\bar{\lambda}_2 - \bar{g}_I^{(2)}\bar{\lambda}_2^{(2)}) - \bar{\lambda}_1\bar{\lambda}_n[\bar{h}_J^{(1)} + \kappa^{(1)}(\bar{J} - 1)] + \bar{\lambda}_1^{(2)}\bar{\lambda}_n[\bar{h}_J^{(2)} + \kappa^{(2)}(\bar{J}^{(2)} - 1)]. \end{aligned} \quad (55)$$

Here,  $\bar{g}_I^{(1)} = g_I^{(1)}(\bar{I})$ ,  $\bar{h}_J^{(1)} = h_J^{(1)}(\bar{J})$ ,  $\bar{g}_I^{(2)} = g_I^{(2)}(\bar{I}^{(2)})$ ,  $\bar{h}_J^{(2)} = h_J^{(2)}(\bar{J}^{(2)})$  have been introduced for ease of notation, where  $\bar{I} = \bar{\mathbf{F}} \cdot \bar{\mathbf{F}} = 2\bar{\lambda}_p^2 + \bar{\lambda}_n^2 + \bar{\gamma}_p^2$ ,  $\bar{J} = \det \bar{\mathbf{F}} = \bar{\lambda}_p^2\bar{\lambda}_n$ , and

$$\begin{aligned} \bar{I}^{(2)} &= \bar{\mathbf{F}}^{(2)} \cdot \bar{\mathbf{F}}^{(2)} = (\bar{\lambda}_1^{(2)})^2 + (\bar{\lambda}_2^{(2)})^2 + \bar{\lambda}_n^2, \\ \bar{J}^{(2)} &= \det \bar{\mathbf{F}}^{(2)} = \bar{\lambda}_1^{(2)}\bar{\lambda}_2^{(2)}\bar{\lambda}_n. \end{aligned} \quad (56)$$

The specialization of Eqs. (31) and (32) to generalized plane-strain deformations has previously been worked out in detail by Brun et al. (2007), but for completeness we recall it here. Thus, for generalized plane-strain deformations of the form (49), the “generalized secant conditions” (31) can be shown to simplify to

$$\begin{aligned} \ell_1^*Y_{11} + \ell_4^*Y_{22} &= 2\hat{g}_I^{(1)}(Y_{11} + \bar{\lambda}_1) + [\hat{h}_J^{(1)} + \kappa^{(1)}(\hat{J}^{(1)} - 1)](Y_{22} + \bar{\lambda}_2)\bar{\lambda}_n - 2\bar{g}_I^{(1)}\bar{\lambda}_1 - [\bar{h}_J^{(1)} + \kappa^{(1)}(\bar{J} - 1)]\bar{\lambda}_2\bar{\lambda}_n, \\ \ell_4^*Y_{11} + \ell_2^*Y_{22} &= 2\hat{g}_I^{(1)}(Y_{22} + \bar{\lambda}_2) + [\hat{h}_J^{(1)} + \kappa^{(1)}(\hat{J}^{(1)} - 1)](Y_{11} + \bar{\lambda}_1)\bar{\lambda}_n - 2\bar{g}_I^{(1)}\bar{\lambda}_2 - [\bar{h}_J^{(1)} + \kappa^{(1)}(\bar{J} - 1)]\bar{\lambda}_1\bar{\lambda}_n, \\ L_{1221}^* &= -[\hat{h}_J^{(1)} + \kappa^{(1)}(\hat{J}^{(1)} - 1)]\bar{\lambda}_n, \\ \ell_7^* &= 2\hat{g}_I^{(1)} \end{aligned} \quad (57)$$

and

$$\ell_5^*Y_{11} + \ell_6^*Y_{22} = 2\hat{g}_I^{(1)}\bar{\lambda}_n - 2\bar{g}_I^{(1)}\bar{\lambda}_n - [\bar{h}_J^{(1)} + \kappa^{(1)}(\bar{J} - 1)]\bar{\lambda}_1\bar{\lambda}_2 + [\hat{h}_J^{(1)} + \kappa^{(1)}(\hat{J}^{(1)} - 1)](Y_{11} + \bar{\lambda}_1)(Y_{22} + \bar{\lambda}_2) - p_1. \quad (58)$$

Here,  $Y_{11} \doteq (\hat{F}_{11}^{(1)} - \bar{\lambda}_1)$ ,  $Y_{22} \doteq (\hat{F}_{22}^{(1)} - \bar{\lambda}_2)$ ,  $p_1 \doteq \hat{F}_{12}^{(1)}\hat{F}_{21}^{(1)}$ ,  $\hat{g}_I^{(1)} = g_I^{(1)}(\hat{I}^{(1)})$ ,  $\hat{h}_J^{(1)} = h_J^{(1)}(\hat{J}^{(1)})$ , with

$$\begin{aligned} \hat{I}^{(1)} &= \hat{\mathbf{F}}^{(1)} \cdot \hat{\mathbf{F}}^{(1)} = (Y_{11} + \bar{\lambda}_1)^2 + (Y_{22} + \bar{\lambda}_2)^2 + \bar{\lambda}_n^2 + s, \\ \hat{J}^{(1)} &= \det \hat{\mathbf{F}}^{(1)} = \bar{\lambda}_n((Y_{11} + \bar{\lambda}_1)(Y_{22} + \bar{\lambda}_2) - p_1) \end{aligned} \quad (59)$$

and  $s \doteq (\hat{F}_{12}^{(1)})^2 + (\hat{F}_{21}^{(1)})^2$ . Furthermore, Eq. (32) can be shown to reduce to

$$Y_{11} = -\frac{k_1 + f_1k_4}{\sqrt{k_1 + 4f_1(f_1k_2 + k_4/2)}}, \quad Y_{22} = -\frac{k_4/2 + 2f_1k_2}{\sqrt{k_1 + 4f_1(f_1k_2 + k_4/2)}} \quad (60)$$

and

$$p_1 = Y_{11}Y_{22} - k_4/2, \quad s = k_7 - 2f_4p_1, \quad (61)$$



where  $f_1 = \partial L_{1221}^* / \partial \ell_1^*$  and  $f_4 = \partial L_{1221}^* / \partial \ell_7^*$ . In connection with the above expressions, it is recalled that the variables  $k_\alpha$  are determined by relations (33) in the text, which after some algebraic manipulation, may be conveniently expressed as

$$\begin{aligned} k_1 &= -\frac{c_0(\bar{\lambda}_1 - \bar{\lambda}_1^{(2)})^2}{1 - c_0} - \frac{c_0}{(1 - c_0)^2} \left[ A_{11}^2 \frac{\partial P_{1111}}{\partial \ell_1^*} + 2A_{11}A_{22} \frac{\partial P_{1122}}{\partial \ell_1^*} + A_{22}^2 \frac{\partial P_{2222}}{\partial \ell_1^*} \right], \\ k_2 &= -\frac{c_0(\bar{\lambda}_2 - \bar{\lambda}_2^{(2)})^2}{1 - c_0} - \frac{c_0}{(1 - c_0)^2} \left[ A_{11}^2 \frac{\partial P_{1111}}{\partial \ell_2^*} + 2A_{11}A_{22} \frac{\partial P_{1122}}{\partial \ell_2^*} + A_{22}^2 \frac{\partial P_{2222}}{\partial \ell_2^*} \right], \\ k_4 &= -\frac{2c_0(\bar{\lambda}_1 - \bar{\lambda}_1^{(2)})(\bar{\lambda}_2 - \bar{\lambda}_2^{(2)})}{1 - c_0}, \\ k_7 &= -\frac{c_0}{(1 - c_0)^2} \left[ A_{11}^2 \frac{\partial P_{1111}}{\partial \ell_7^*} + 2A_{11}A_{22} \frac{\partial P_{1122}}{\partial \ell_7^*} + A_{22}^2 \frac{\partial P_{2222}}{\partial \ell_7^*} \right], \end{aligned} \quad (62)$$

and  $k_3 = k_5 = k_6 = 0$ , where  $A_{11}$  and  $A_{22}$  (the only non-zero components of the tensor  $\mathbf{A}$ ) are simply given by

$$\begin{aligned} A_{11} &= \frac{P_{1122}(\bar{\lambda}_2 - \bar{\lambda}_2^{(2)}) - P_{2222}(\bar{\lambda}_1 - \bar{\lambda}_1^{(2)})}{P_{1122}^2 - P_{1111}P_{2222}}, \\ A_{22} &= \frac{P_{1122}(\bar{\lambda}_1 - \bar{\lambda}_1^{(2)}) - P_{1111}(\bar{\lambda}_2 - \bar{\lambda}_2^{(2)})}{P_{1122}^2 - P_{1111}P_{2222}}. \end{aligned} \quad (63)$$

In summary, for generalized plane-strain deformations, the general system of Eqs. (29)–(32) essentially reduces to the system of six coupled, non-linear, algebraic equations formed by expressions (54) and (57), for the six unknowns  $\bar{\lambda}_1^{(2)}$ ,  $\bar{\lambda}_2^{(2)}$ ,  $\ell_1^*$ ,  $\ell_2^*$ ,  $\ell_4^*$  and  $\ell_7^*$ . (Note that Eq. (58), which establishes a connection between the moduli  $\ell_5^*$ ,  $\ell_6^*$  and the other variables of the problem, does not intervene in the computation of the GSO estimate (34) for the effective stored-energy function  $\hat{W}$  under generalized plane-strain deformations.)

Next, we carry out the asymptotic analysis of the above equations associated with the limit  $\kappa^{(1)} = \kappa^{(2)} \rightarrow \infty$ . Making use of the *ansatz* proposed by Lopez-Pamies and Ponte Castañeda (2006b) (see Appendix A in that reference), it is assumed that the asymptotic expansion for the unknown variables in the limit  $\kappa^{(1)} = \kappa^{(2)} \rightarrow \infty$  is of the form

$$\begin{aligned} \bar{\lambda}_1^{(2)} &= a_0 + a_1 \Delta^{1/3} + a_2 \Delta^{2/3} + a_3 \Delta + a_4 \Delta^{4/3} + O(\Delta^{5/3}), \\ \bar{\lambda}_2^{(2)} &= b_0 + b_1 \Delta^{1/3} + b_2 \Delta^{2/3} + b_3 \Delta + b_4 \Delta^{4/3} + O(\Delta^{5/3}), \\ \ell_1^* &= d_{-1} \Delta^{-1/3} + d_0 + d_1 \Delta^{1/3} + d_2 \Delta^{2/3} + d_3 \Delta + O(\Delta^{4/3}), \\ \ell_2^* &= e_{-1} \Delta^{-1/3} + e_0 + d_1 \Delta^{1/3} + e_2 \Delta^{2/3} + e_3 \Delta + O(\Delta^{4/3}), \\ \ell_4^* &= f_{-1} \Delta^{-1/3} + f_0 + f_1 \Delta^{1/3} + f_2 \Delta^{2/3} + f_3 \Delta + O(\Delta^{4/3}), \\ \ell_7^* &= m_0 + m_1 \Delta^{1/3} + m_2 \Delta^{2/3} + m_3 \Delta + O(\Delta^{4/3}), \end{aligned} \quad (64)$$

where  $\Delta \doteq 1/\kappa^{(1)} = 1/\kappa^{(2)}$  is a small parameter. The unknown coefficients in these expressions are to be determined from the asymptotic expansion of the system of Eqs. (54) and (57) in the limit as  $\Delta \rightarrow 0$ . To simplify the presentation of the results, it proves helpful to introduce the following notation for the expansion of the intermediate quantities involved in (54) and (57):

$$\begin{aligned} Y_{11} &= x_0 + x_1 \Delta^{1/3} + x_2 \Delta^{2/3} + x_3 \Delta + O(\Delta^{4/3}), \\ Y_{22} &= y_0 + y_1 \Delta^{1/3} + y_2 \Delta^{2/3} + y_3 \Delta + O(\Delta^{4/3}), \\ p_1 &= z_0 + z_1 \Delta^{1/3} + z_2 \Delta^{2/3} + z_3 \Delta + O(\Delta^{4/3}), \\ s &= t_0 + t_1 \Delta^{1/3} + t_2 \Delta^{2/3} + t_3 \Delta + O(\Delta^{4/3}), \\ \bar{J}^{(2)} &= j_0 + j_1 \Delta^{1/3} + j_2 \Delta^{2/3} + j_3 \Delta + j_4 \Delta^{4/3} + O(\Delta^{5/3}), \\ \hat{J}^{(1)} &= \hat{j}_0 + \hat{j}_1 \Delta^{1/3} + \hat{j}_2 \Delta^{2/3} + \hat{j}_3 \Delta + \hat{j}_4 \Delta^{4/3} + O(\Delta^{5/3}), \\ \bar{g}_f^{(2)} &= \zeta_0 + \zeta_1 \Delta^{1/3} + \zeta_2 \Delta^{2/3} + \zeta_3 \Delta + O(\Delta^{4/3}), \\ \hat{g}_f^{(1)} &= \hat{\zeta}_0 + \hat{\zeta}_1 \Delta^{1/3} + \hat{\zeta}_2 \Delta^{2/3} + \hat{\zeta}_3 \Delta + O(\Delta^{4/3}), \\ \bar{h}_j^{(2)} &= \eta_0 + \eta_1 \Delta^{1/3} + \eta_2 \Delta^{2/3} + \eta_3 \Delta + O(\Delta^{4/3}), \\ \hat{h}_j^{(1)} &= \hat{\eta}_0 + \hat{\eta}_1 \Delta^{1/3} + \hat{\eta}_2 \Delta^{2/3} + \hat{\eta}_3 \Delta + O(\Delta^{4/3}), \end{aligned} \quad (65)$$

where the coefficients in the above expressions are known functions of the coefficients in the right-hand side of (64) too cumbersome to be included here.

Substituting expressions (64) and (65) in Eqs. (54) and (57) and subsequently expanding in small values of  $\Delta$  leads to a hierarchical system of equations for the unknown coefficients introduced in (64). After some algebraic manipulation,

the equations of leading order  $O(\Delta^{-1})$  can be shown to yield the following results:

$$\bar{J} \doteq \bar{\lambda}_1 \bar{\lambda}_2 \bar{\lambda}_n \doteq \bar{\lambda}_p^2 \bar{\lambda}_n = 1, \quad \hat{j}_0 = 1, \quad \hat{j}_0 = 1. \quad (66)$$

Note that condition (66)<sub>1</sub> is precisely the exact overall incompressibility constraint, and that (66)<sub>2</sub> may alternatively be written as  $b_0 = 1/(a_0 \bar{\lambda}_n)$ . The equations of next order  $O(\Delta^{-2/3})$  imply that  $j_1 = 0$  and  $\hat{j}_1 = 0$ , which ultimately reduce to

$$b_1 = -\frac{a_1}{a_0^2 \bar{\lambda}_n} \quad \text{and} \quad e_{-1} = \bar{\lambda}_1^4 \bar{\lambda}_n^2 d_{-1}, \quad (67)$$

respectively.

At this point, in view of results (66) and (67), it is useful to recognize that the coefficients  $x_0, y_0, z_0$  and  $t_0$  take the simple form

$$\begin{aligned} x_0 &= \frac{c_0(\bar{\lambda}_1 - a_0)}{1 - c_0}, \quad y_0 = -\frac{c_0(\bar{\lambda}_1 - a_0)}{a_0(1 - c_0)\bar{\lambda}_1 \bar{\lambda}_n}, \quad z_0 = -\frac{c_0(\bar{\lambda}_1 - a_0)^2}{a_0(1 - c_0)^2 \bar{\lambda}_1 \bar{\lambda}_n}, \\ t_0 &= \frac{y_0^2}{c_0} [(a_0 \bar{\lambda}_1 \bar{\lambda}_n + 1)^2 + (\bar{\lambda}_1^2 + a_0^2) \bar{\lambda}_n], \end{aligned} \quad (68)$$

which are seen to depend *only* on the coefficient  $a_0$ , i.e., the leading order term of  $\bar{\lambda}_1^{(2)}$ . In turn, making use of (68), it is not difficult to show that the leading order term of the GSO estimate (28) does also depend *only* on the coefficient  $a_0$ . The resulting expression is given by relation (34)—together with (35) and (36)—in the text, where for clarity of notation  $a_0$  was written as  $\bar{\lambda}_1^{(2)}$ .

Next, the equations of third order  $O(\Delta^{-1/3})$  can be shown to lead to

$$j_2 = \hat{j}_2 = d_{-1} \bar{\lambda}_1 (y_0 \bar{\lambda}_1^2 \bar{\lambda}_n + x_0), \quad f_{-1} = d_{-1} \bar{\lambda}_1 \bar{\lambda}_n (y_0 \bar{\lambda}_1^2 \bar{\lambda}_n + \bar{\lambda}_1 + x_0), \quad (69)$$

while those of fourth order  $O(\Delta^0)$  render expressions for  $j_3, \hat{j}_3$  and  $f_0$  in terms of  $a_0, a_1, d_{-1}, d_0$  and  $e_0$  too cumbersome to be included here, as well as the following useful result:

$$c_0(a_0 - \bar{\lambda}_1)^2 (e_0 - d_0 \bar{\lambda}_1^4 \bar{\lambda}_n^2) = 2a_0(1 - c_0)(\bar{\lambda}_1^4 \bar{\lambda}_n^2 - 1)(2\bar{\lambda}_1(\hat{\zeta}_0 - \bar{g}_I^{(1)}) + (x_0 + \bar{\lambda}_1^2 \bar{\lambda}_n y_0) \hat{\zeta}_0). \quad (70)$$

Finally, the equations of fifth order  $O(\Delta^{1/3})$  are treated in the following manner. First, using the equations resulting from (57)<sub>1</sub> and (57)<sub>2</sub>, we are able to write down an explicit expression for  $f_1$  in terms of  $\hat{j}_4, a_0, a_1, a_2, b_2, d_{-1}, d_0, e_0, d_1$  and  $e_1$ . Then, after substituting the obtained expression for  $f_1$  in (the appropriate order equation resulting from) (54)<sub>1</sub>, we generate an explicit expression for  $j_4$  in terms of  $a_0, a_1, a_2, b_2, d_{-1}, d_0, e_0, d_1$ . Making use of these results for  $f_1$  and  $j_4$  in (54)<sub>2</sub> ultimately renders the following condition:

$$\begin{aligned} c_0(a_0 - \bar{\lambda}_1)^2 (e_0 - d_0 \bar{\lambda}_1^4 \bar{\lambda}_n^2) a_0 &= 4a_0^4 c_0 \bar{\lambda}_1 \bar{\lambda}_n (\hat{\zeta}_0 + \bar{\lambda}_1^2 \bar{\lambda}_n ((1 - c_0) \hat{\zeta}_0 + (c_0 + 1) \hat{\zeta}_0)) \\ &\quad + 2a_0^3 c_0 (1 - 5\bar{\lambda}_1^4 \bar{\lambda}_n^2) \hat{\zeta}_0 + 4a_0^2 \bar{\lambda}_1 (\bar{\lambda}_1^4 \bar{\lambda}_n^2 - 1) (\hat{\zeta}_0 - \bar{g}_I^{(1)} (1 - c_0)) \\ &\quad + 2a_0 c_0 \bar{\lambda}_1^2 (5 - \bar{\lambda}_1^4 \bar{\lambda}_n^2) \hat{\zeta}_0 - 4c_0 \bar{\lambda}_1^3 ((1 - c_0) \hat{\zeta}_0 + (1 + c_0 + \bar{\lambda}_1^2 \bar{\lambda}_n) \hat{\zeta}_0). \end{aligned} \quad (71)$$

At this stage, it is recognized that relations (70) and (71) depend on unknowns other than  $a_0$  only through the combination  $e_0 - d_0 \bar{\lambda}_1^4 \bar{\lambda}_n^2$ , and so, after eliminating  $e_0 - d_0 \bar{\lambda}_1^4 \bar{\lambda}_n^2$  from these expressions, we obtain the following equation for  $a_0$ :

$$[(1 + c_0) \hat{\zeta}_0 + (1 - c_0) \zeta_0] (\bar{\lambda}_n^2 - a_0^4) - \hat{\zeta}_0 \left[ 2a_0 \bar{\lambda}_1 (\bar{\lambda}_2^2 - a_0^2) - \frac{(\bar{\lambda}_1^4 - a_0^4) \bar{\lambda}_2}{\bar{\lambda}_1} \right] = 0. \quad (72)$$

Note that Eq. (72) is nothing more than Eq. (37) in the text with slightly different notation:  $a_0 \rightarrow \bar{\lambda}_1^{(2)}$ ,  $\hat{\zeta}_0 \rightarrow g_I^{(1)}(I_P^{(1)})$  and  $\zeta_0 \rightarrow g_I^{(2)}(I_P^{(2)})$ . As already emphasized, the solution of this equation serves to completely determine the incompressible GSO estimate (34).

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