PERGAMON

Journal of the Mechanics and Physics of Solids
48 (2000) 1359–1387

JOURNAL OF THE
MECHANICS AND
PHYSICS OF SOLIDS

www.elsevier.com/locate/jmps

Micromechanics-based variational estimates for a higher-order nonlocal constitutive equation and optimal choice of effective moduli for elastic composites

W.J. Drugan

Department of Engineering Physics, University of Wisconsin-Madison, 1500 Engineering Drive, Madison, WI 53706-1687, USA

Received 23 February 1999; received in revised form 22 September 1999

Abstract

A generalization of the Hashin–Shtrikman variational formulation to random composites, due to J.R. Willis, is employed to derive micromechanics-based variational estimates for a higher-order nonlocal constitutive equation relating the ensemble averages of stress and strain, for a class of random linear elastic composite materials. We analyze two-phase composites with any isotropic and statistically uniform distribution of phases (which themselves may have arbitrary shape and anisotropy), within a formulation accounting for one- and two-point probabilities, and derive an explicit nonlocal constitutive equation that includes terms up through the fourth gradient of average strain. The analysis is carried out first for an arbitrary comparison medium. Then, a new approach is outlined and applied which employs the nonlocal correction to determine the optimal choice of comparison medium, and hence the optimal effective modulus tensor (as well as the optimal tensor coefficients of the nonlocal terms) for the amount of statistical information employed. The new higher order analysis provides a highly accurate nonlocal constitutive equation, valid down to quite small volume size scales and to rather strong variations of average strain with position. Among several applications illustrated, it permits accurate analytical assessment of the remarkably small predictions derived by Drugan and Willis (1996. Journal of the Mechanics and Physics of Solids 44, 497–524) of the minimum representative volume element (RVE) size needed for accuracy of the standard constant-

E-mail address: drugan@engr.wisc.edu (W.J. Drugan).

PII: S0022-5096(99)00088-5

effective-modulus macroscopic constitutive equation for elastic matrix-inclusion composites that have spherical inclusions/voids. It also affords an analytical assessment of the improved (i.e., reduced) minimum RVE size scale, compared to a standard constant-effective-modulus constitutive equation, to which the leading-order nonlocal constitutive equation derived by Drugan and Willis applies. This improvement is shown to be dramatic in some example cases.

Keywords: Nonlocal constitutive equations; Voids and inclusions; Constitutive behavior; Inhomogeneous material; Variational calculus

1. Introduction

When the average strain field in a composite material sample varies sufficiently slowly with position compared to the microstructural size scale of the composite, the material's macroscopic constitutive response is well-approximated as homogenous, even though the material may be strongly heterogeneous on the microscale. The determination of estimates for, and bounds on, such 'effective' or 'overall' constitutive moduli for composite materials has comprised a large share of composite material constitutive modeling; see, e.g., the reviews of Willis (1981, 1982, 1983) and Ponte Castañeda and Suquet (1998).

When the conditions summarized informally above are not met, the composite material's response cannot adequately be characterized as homogeneous. The questions of *how* rigorously to improve the constitutive modeling in such cases, and precisely *when* such constitutive improvement is necessary, were addressed analytically from a micromechanics basis by Drugan and Willis (1996). They employed Willis' (1977) generalization of the Hashin and Shtrikman (1962a, 1962b) variational formulation to derive, for random linear elastic composites characterized by up through two-point statistical information, the explicit leading-order micromechanics-based nonlocal correction to the standard constant-effective-modulus constitutive equation relating ensemble-average stress and strain. They also employed this improved constitutive equation to provide quantitative estimates of minimum representative volume element (RVE) size, defined to mean the minimum size of a volume element of composite material, away from specimen surfaces, for which spatially-varying ensemble-averaged stress and strain are accurately related by a constant effective modulus tensor.

Here, we introduce and apply a new approach for the determination of optimal choice of the comparison moduli, and hence of the effective moduli as well as the tensor coefficients of the nonlocal terms, when retaining up through two-point statistical information. This involves derivation of the nonlocal correction in terms of arbitrary comparison moduli; the optimal comparison moduli are then hypothesized to be those that minimize the nonlocal correction term — i.e., that minimize the size of the representative volume element over which the 'effective' modulus tensor accurately applies.

We also show, within the micromechanics-based formulation Drugan and Willis developed, that a second-order nonlocal correction term for the macroscopic constitutive equation can be derived and that it admits a reasonably concise and explicit representation. This is determined completely, in closed form, for the optimal choice of comparison moduli derived, in the case of an isotropic matrix reinforced (weakened) by an isotropic distribution of nonoverlapping spherical particles (voids). The general higher-order nonlocal constitutive equation permits more accurate quantitative estimates of the minimum RVE size needed for accuracy of the standard constant-effective-modulus constitutive equation, which refine but largely confirm Drugan and Willis' results. It also facilitates the first analytical assessment of the increased accuracy (i.e., reduced RVE size as compared to the standard effective modulus constitutive equation) provided by Drugan and Willis' (1996) leading-order nonlocal constitutive equation.

As with many fundamentally-based advances in the mechanics of composite materials in particular, and in several areas of solid mechanics in general, the research described here owes much to the theoretical foundations laid down by John Willis. It is dedicated to him on his 60th Anniversary, with the hope that he will continue to provide deep and elegant mathematical frameworks for addressing important problems in solid mechanics.

2. General formulation

We consider random linear elastic composite materials with firmly-bonded phases which may have arbitrary anisotropy and be present in arbitrary concentrations. Since we seek to describe macroscopic *constitutive* response, we shall analyze an infinite body subject only to applied loading through a body force vector field $\mathbf{f}(\mathbf{x})$ that decays sufficiently rapidly for large magnitudes $|\mathbf{x}|$ of the position vector $\mathbf{x}$. For a specific composite sample (a specific realization of the random composite's microstructure), the equations governing equilibrium stress and deformation fields within the linearized theory of elasticity are:

$$\nabla \bullet \boldsymbol{\sigma}(\mathbf{x}) + \mathbf{f}(\mathbf{x}) = \mathbf{0} \quad \left[\frac{\partial \sigma_{ij}(\mathbf{x})}{\partial x_i} + f_j(\mathbf{x}) = 0\right], \tag{1a}$$

$$\mathbf{e}(\mathbf{x}) = \mathrm{sym}\big[\nabla \mathbf{u}(\mathbf{x})\big] \quad \left[e_{ij}(\mathbf{x}) = \tfrac{1}{2}\big(\partial u_i(\mathbf{x})/\partial x_j + \partial u_j(\mathbf{x})/\partial x_i\big)\right], \tag{1b}$$

$$\boldsymbol{\sigma}(\mathbf{x}) = \mathbf{L}(\mathbf{x})\mathbf{e}(\mathbf{x}) \quad \big[\sigma_{ij}(\mathbf{x}) = L_{ijkl}(\mathbf{x})e_{kl}(\mathbf{x})\big], \tag{1c}$$

where $\boldsymbol{\sigma}$, $\mathbf{e}$ are the stress and infinitesimal strain tensors, $\mathbf{u}$ the displacement vector, $\mathbf{L}$ the fourth-rank elastic modulus tensor and 'sym' denotes the symmetric part. Eqs. (1) are given both in direct and index notation to define the meanings of these notations; throughout the paper, lower-case Latin subscripts denote

Cartesian components of tensors and obey the Einstein summation convention except where noted.

To formulate the Hashin–Shtrikman variational principle corresponding to system (1) for a specific composite realization, one introduces a *homogeneous* 'comparison' body with moduli (independent of $\mathbf{x}$) $\mathbf{L}_0$ (and with solutions $\boldsymbol{\sigma}_0$, $\mathbf{e}_0$, $\mathbf{u}_0$ to the same applied $\mathbf{f}(\mathbf{x})$) so that

$$\boldsymbol{\sigma}(\mathbf{x}) = \mathbf{L}_0\mathbf{e}(\mathbf{x}) + \boldsymbol{\tau}(\mathbf{x}), \tag{2a}$$

$$\boldsymbol{\tau}(\mathbf{x}) \equiv \left[\mathbf{L}(\mathbf{x}) - \mathbf{L}_0\right]\mathbf{e}(\mathbf{x}), \tag{2b}$$

where $\boldsymbol{\tau}$ is the 'stress polarization' tensor. Substitution of Eq. (2a) in Eq. (1a) gives:

$$\nabla \bullet (\mathbf{L}_0\mathbf{e}) + (\nabla \bullet \boldsymbol{\tau} + \mathbf{f}) = \mathbf{0}. \tag{3}$$

Willis (1977) showed that the solution of Eq. (3) can be expressed as

$$\mathbf{e}(\mathbf{x}) = \mathbf{e}_0(\mathbf{x}) - \int \boldsymbol{\Gamma}_0(\mathbf{x} - \mathbf{x}')\boldsymbol{\tau}(\mathbf{x}')\,\mathrm{d}\mathbf{x}', \tag{4}$$

where

$$\left[\boldsymbol{\Gamma}_0(\mathbf{x} - \mathbf{x}')\right]_{ijkl} \equiv \left.\frac{\partial^2\left[\mathbf{G}_0(\mathbf{x} - \mathbf{x}')\right]_{jk}}{\partial x_i \partial x_l'}\right|_{(ij),\,(kl)}, \tag{5}$$

the notation indicates symmetrization on (ij) and (kl), the integral in Eq. (4) is over all space, and the singularity of $\boldsymbol{\Gamma}_0$ is interpreted in the sense of generalized functions. Here, $\mathbf{G}_0(\mathbf{x})$ is the infinite-homogeneous-body Green's function, which solves

$$\frac{\partial^2\left[\mathbf{G}_0(\mathbf{x})\right]_{jm}}{\partial x_i \partial x_l} c_{ijkl} + \delta_{km}\delta(\mathbf{x}) = 0, \tag{6}$$

where c_{ijkl} are the components of $\mathbf{L}_0$, δ_{km} is the Kronecker delta and $\delta(\mathbf{x})$ is the three-dimensional (3D) Dirac delta function. Substituting for $\mathbf{e}$ in Eq. (4) from Eq. (2b) gives

$$(\mathbf{L}(\mathbf{x}) - \mathbf{L}_0)^{-1}\boldsymbol{\tau}(\mathbf{x}) + \int \boldsymbol{\Gamma}_0(\mathbf{x} - \mathbf{x}')\boldsymbol{\tau}(\mathbf{x}')\,\mathrm{d}\mathbf{x}' = \mathbf{e}_0(\mathbf{x}). \tag{7}$$

Willis (1977) observed that self-adjointness of Eq. (7) leads directly to the Hashin–Shtrikman variational principle for $\boldsymbol{\tau}(\mathbf{x})$:

$$\delta\left\{\int\left[\boldsymbol{\tau}(\mathbf{x})(\mathbf{L}(\mathbf{x}) - \mathbf{L}_0)^{-1}\boldsymbol{\tau}(\mathbf{x}) + \boldsymbol{\tau}(\mathbf{x})\int \boldsymbol{\Gamma}_0(\mathbf{x} - \mathbf{x}')\boldsymbol{\tau}(\mathbf{x}')\,\mathrm{d}\mathbf{x}' - 2\boldsymbol{\tau}(\mathbf{x})\mathbf{e}_0(\mathbf{x})\right]\mathrm{d}\mathbf{x}\right\} = 0. \tag{8}$$

The solution to Eq. (8) for $\boldsymbol{\tau}(\mathbf{x})$ then provides the *local* stress and deformation fields in the specific composite realization, via Eqs. (2). However, this is in general prohibitively difficult; further, we seek results of more general applicability.

To obtain these, we follow Willis (1977, 1983) and recast the above formulation in terms of ensemble averages for random composites. Let α denote individual members of a sample space S, define by $p(\alpha)$ the probability density of α in S, and define a characteristic function $\chi_r(\mathbf{x};\alpha)=1$ when $\mathbf{x}$ lies in phase r, and $=0$ otherwise. Then the probability $P_r(\mathbf{x})$ of finding phase r at $\mathbf{x}$ (i.e., the *ensemble average of* $\chi_r(\mathbf{x};\alpha)$) is

$$P_r(\mathbf{x}) = \langle \chi_r(\mathbf{x})\rangle \equiv \int_S \chi_r(\mathbf{x};\alpha)p(\alpha)\,\mathrm{d}\alpha, \tag{9}$$

and the (two-point) probability $P_{rs}(\mathbf{x},\ \mathbf{x}')$ of finding simultaneously phase r at $\mathbf{x}$ and phase s at $\mathbf{x}'$ is

$$P_{rs}(\mathbf{x},\mathbf{x}') = \langle \chi_r(\mathbf{x})\chi_s(\mathbf{x}')\rangle \equiv \int_S \chi_r(\mathbf{x};\alpha)\chi_s(\mathbf{x}';\alpha)p(\alpha)\,\mathrm{d}\alpha. \tag{10}$$

We shall treat composites comprised of homogeneous phases, so that each phase r has (constant) modulus tensor $\mathbf{L}_r$, where $r=1,2,\ldots,n$, with n being the total number of phases; then $\mathbf{L}(\mathbf{x})$ of Eq. (1c) in sample α, and its ensemble average, are:

$$\mathbf{L}(\mathbf{x};\alpha) = \sum_{r=1}^{n}\mathbf{L}_r\chi_r(\mathbf{x};\alpha) \Longrightarrow \langle \mathbf{L}(\mathbf{x})\rangle = \sum_{r=1}^{n}\mathbf{L}_r\mathbf{P}_r(\mathbf{x}). \tag{11}$$

As argued by Willis (1982), in most applications it is unlikely that statistical information of higher grade than two-point probabilities will be credibly known. Thus, we follow him and choose the most general trial fields for $\boldsymbol{\tau}$ that allow for up through two-point correlations:

$$\boldsymbol{\tau}(\mathbf{x};\alpha) = \sum_{r=1}^{n}\boldsymbol{\tau}_r(\mathbf{x})\chi_r(\mathbf{x};\alpha). \tag{12}$$

(Willis (1982) showed that any more general form together with the Hashin–Shtrikman variational principle will introduce further statistical information.)

We shall further restrict the class of composites analyzed to those that are statistically uniform, and we make an ergodic assumption that local configurations occur over any one specimen with the frequency with which they occur over a single neighborhood in an ensemble of specimens. For this class of materials, $P_r(\mathbf{x})$ reduces to the volume concentration c_r of phase r, and $P_{rs}(\mathbf{x},\ \mathbf{x}') = P_{rs}(\mathbf{x}-\mathbf{x}')$. Employing these assumptions together with Eqs. (9)–(12), Willis (1977, 1983) has shown that Eq. (8) can be recast as the stochastic variational principle

$$\delta\left\{\sum_{r=1}^{n} c_r \int \boldsymbol{\tau}_r(\mathbf{x})\left[(\mathbf{L}_r-\mathbf{L}_0)^{-1}\boldsymbol{\tau}_r(\mathbf{x})-2\mathbf{e}_0(\mathbf{x})\right]\mathrm{d}\mathbf{x} + \sum_{r=1}^{n}\sum_{s=1}^{n}\int \boldsymbol{\tau}_r(\mathbf{x})\left[\int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')\boldsymbol{\tau}_s(\mathbf{x}')P_{rs}(\mathbf{x}-\mathbf{x}')\,\mathrm{d}\mathbf{x}'\right]\mathrm{d}\mathbf{x}\right\}=0. \tag{13}$$

Principle (13) is stationary when (employing the ensemble average of Eq. (4) with Eq. (12))

$$(\mathbf{L}_r-\mathbf{L}_0)^{-1}\boldsymbol{\tau}_r(\mathbf{x})c_r+\sum_{s=1}^{n}\int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')\left[P_{rs}(\mathbf{x}-\mathbf{x}')-c_rc_s\right]\boldsymbol{\tau}_s(\mathbf{x}')\,\mathrm{d}\mathbf{x}'=c_r\langle\mathbf{e}\rangle(\mathbf{x}),\quad r=1,2,\ldots,n \tag{14}$$

which is a set of n integral equations for $\boldsymbol{\tau}_r(\mathbf{x})$ in terms of $\langle\mathbf{e}\rangle(\mathbf{x})$. When these are solved, $\langle\boldsymbol{\tau}\rangle(\mathbf{x})$ can be determined from the result of ensemble-averaging Eq. (12):

$$\langle\boldsymbol{\tau}\rangle(\mathbf{x})=\sum_{r=1}^{n}c_r\boldsymbol{\tau}_r(\mathbf{x}). \tag{15}$$

Finally, the constitutive equation we desire, relating the ensemble averages of stress and strain in the general case when these depend on position, is obtained by substitution of Eq. (15) into the ensemble average of Eq. (2a):

$$\langle\boldsymbol{\sigma}\rangle(\mathbf{x})=\mathbf{L}_0\langle\mathbf{e}\rangle(\mathbf{x})+\langle\boldsymbol{\tau}\rangle(\mathbf{x}). \tag{16}$$

3. General structure of higher-order nonlocal constitutive equation for two-phase composites

To facilitate derivation of a reasonably concise and explicit form of a higher-order nonlocal constitutive equation, we now specialize to the practically important class of two-phase composites, and employ our assumptions of statistical uniformity and ergodicity. Then the two-point probability can be expressed as

$$P_{rs}(\mathbf{x}-\mathbf{x}')-c_rc_s=c_r(\delta_{rs}-c_s)h(\mathbf{x}-\mathbf{x}'), \tag{17}$$

where $h(\mathbf{x}-\mathbf{x}')$ is the two-point correlation function, defined e.g. by the 12 component of Eq. (17). Using Eq. (17) in Eq. (14) gives:

$$(\mathbf{L}_r-\mathbf{L}_0)^{-1}\boldsymbol{\tau}_r(\mathbf{x})c_r+\sum_{s=1}^{2}c_r(\delta_{rs}-c_s)\int\left[\boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')h(\mathbf{x}-\mathbf{x}')\right]\boldsymbol{\tau}_s(\mathbf{x}')\,\mathrm{d}\mathbf{x}'=c_r\langle\mathbf{e}\rangle(\mathbf{x}),\quad r=1,2. \tag{18}$$

As did Drugan and Willis (1996), we shall employ Fourier transforms to solve Eq. (18), but here we shall do so to greater accuracy. The 3D Fourier transform of a function $f(\mathbf{x})$ (which decays sufficiently rapidly for $|\mathbf{x}|\to\infty$ for convergence of Eq. (19)) and its inverse are defined as

$$\tilde{f}(\boldsymbol{\xi}) \equiv \int f(\mathbf{x})\mathrm{e}^{i\boldsymbol{\xi}\bullet\mathbf{x}}\,\mathrm{d}\mathbf{x}, \quad f(\mathbf{x}) = \frac{1}{8\pi^3}\int \tilde{f}(\boldsymbol{\xi})\mathrm{e}^{-i\boldsymbol{\xi}\bullet\mathbf{x}}\,\mathrm{d}\boldsymbol{\xi}, \tag{19}$$

where $i=\sqrt{-1}$, $\boldsymbol{\xi}$ is a vector, $\boldsymbol{\xi}\bullet\mathbf{x}$ is scalar product, and the integrals are taken over all 3D space. The 3D Fourier transform of Eq. (18) is

$$(\mathbf{L}_r-\mathbf{L}_0)^{-1}\tilde{\boldsymbol{\tau}}_r(\boldsymbol{\xi})c_r + \sum_{s=1}^{2} c_r(\delta_{rs}-c_s)\big(\tilde{\boldsymbol{\Gamma}}_0 * \tilde{h}\big)(\boldsymbol{\xi})\tilde{\boldsymbol{\tau}}_s(\boldsymbol{\xi}) = c_r\langle\tilde{\mathbf{e}}\rangle(\boldsymbol{\xi}), \quad r=1,2, \tag{20}$$

having noted that the integral term in Eq. (18) is a convolution, and that the Fourier transform of the bracketed term is itself the convolution

$$\big(\tilde{\boldsymbol{\Gamma}}_0 * \tilde{h}\big)(\boldsymbol{\xi}) \equiv \frac{1}{8\pi^3}\int \tilde{\boldsymbol{\Gamma}}_0\big(\boldsymbol{\xi}-\boldsymbol{\xi}'\big)\tilde{h}\big(\boldsymbol{\xi}'\big)\,\mathrm{d}\boldsymbol{\xi}' \equiv \boldsymbol{\Gamma}. \tag{21}$$

Drugan and Willis (1996) showed how to solve Eq. (20) for $\tilde{\boldsymbol{\tau}}_r(\boldsymbol{\xi})$, and they showed that these results can be substituted into the Fourier transform of Eq. (15) to obtain

$$\langle\tilde{\boldsymbol{\tau}}\rangle(\boldsymbol{\xi}) = \sum_{r=1}^{2} c_r\tilde{\boldsymbol{\tau}}_r(\boldsymbol{\xi}) = \langle\tilde{\mathbf{T}}\rangle(\boldsymbol{\xi})\langle\tilde{\mathbf{e}}\rangle(\boldsymbol{\xi}), \tag{22}$$

where

$$\begin{aligned}\langle\tilde{\mathbf{T}}\rangle(\boldsymbol{\xi}) &= c_1\delta\mathbf{L}_1\big(\boldsymbol{\Gamma}^{-1}+c_1\delta\mathbf{L}_2+c_2\delta\mathbf{L}_1\big)^{-1}\big(\boldsymbol{\Gamma}^{-1}+\delta\mathbf{L}_2\big)\\ &\quad + c_2\delta\mathbf{L}_2\big(\boldsymbol{\Gamma}^{-1}+c_1\delta\mathbf{L}_2+c_2\delta\mathbf{L}_1\big)^{-1}\big(\boldsymbol{\Gamma}^{-1}+\delta\mathbf{L}_1\big),\end{aligned} \tag{23}$$

having defined

$$\delta\mathbf{L}_r \equiv \mathbf{L}_r - \mathbf{L}_0. \tag{24}$$

The result we need for Eq. (16) is given by the inverse Fourier transform of Eq. (22):

$$\langle\boldsymbol{\tau}\rangle(\mathbf{x}) = \int \langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')\langle\mathbf{e}\rangle(\mathbf{x}')\,\mathrm{d}\mathbf{x}'. \tag{25}$$

The exact evaluation of the right side of Eq. (25) is very difficult, not least because it requires finding the inverse Fourier transform of Eq. (23). To circumvent this

difficulty, Drugan and Willis (1996) approximated $\langle\mathbf{e}\rangle(\mathbf{x}')$ by the first three terms in its Taylor expansion, and later showed that there are then only two nonzero terms in Eq. (25). We shall improve their analysis by now retaining *five* terms in the Taylor expansion for $\langle\mathbf{e}\rangle(\mathbf{x}')$ (which we shall show results in three nonzero terms in Eq. (25)):

$$\begin{aligned}\langle\mathbf{e}\rangle(\mathbf{x}') \approx{}& \langle\mathbf{e}\rangle(\mathbf{x}) + (\mathbf{x}'-\mathbf{x})\nabla\langle\mathbf{e}\rangle(\mathbf{x}) + \frac{1}{2}(\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})\nabla\nabla\langle\mathbf{e}\rangle(\mathbf{x})\\ &+\frac{1}{6}(\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})\nabla\nabla\nabla\langle\mathbf{e}\rangle(\mathbf{x})\\ &+\frac{1}{24}(\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})\nabla\nabla\nabla\nabla\langle\mathbf{e}\rangle(\mathbf{x}) = \langle\mathbf{e}\rangle(\mathbf{x})\\ &+\left(x_i'-x_i\right)\frac{\partial}{\partial x_i}\langle\mathbf{e}\rangle(\mathbf{x}) + \frac{1}{2}\left(x_i'-x_i\right)\left(x_j'-x_j\right)\frac{\partial^2}{\partial x_i\partial x_j}\langle\mathbf{e}\rangle(\mathbf{x}) + \cdots\end{aligned} \tag{26}$$

Using this, Eq. (25) becomes:

$$\begin{aligned}\langle\boldsymbol{\tau}\rangle(\mathbf{x}) ={}& \left[\int\langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')\,\mathrm{d}\mathbf{x}'\right]\langle\mathbf{e}\rangle(\mathbf{x}) + \left[\int\left[\langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')\right](\mathbf{x}'-\mathbf{x})\,\mathrm{d}\mathbf{x}'\right]\nabla\langle\mathbf{e}\rangle(\mathbf{x}) + \cdots\\ &+\left[\int[\langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')](\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})\,\mathrm{d}\mathbf{x}'\right]\frac{1}{24}\nabla\nabla\nabla\nabla\langle\mathbf{e}\rangle(\mathbf{x}).\end{aligned} \tag{27}$$

To evaluate the integrals in Eq. (27), recall that the Fourier transform, and its derivatives, of $\langle\mathbf{T}\rangle(\mathbf{x})$ are (where $\nabla = \partial/\partial\boldsymbol{\xi}$)

$$\langle\tilde{\mathbf{T}}\rangle(\boldsymbol{\xi}) = \int\langle\mathbf{T}\rangle(\mathbf{x})\mathrm{e}^{i\boldsymbol{\xi}\bullet\mathbf{x}}\,\mathrm{d}\mathbf{x} \Longrightarrow \nabla\langle\tilde{\mathbf{T}}\rangle(\boldsymbol{\xi}=\mathbf{0}) = \int\langle\mathbf{T}\rangle(\mathbf{x})i\mathbf{x}\,\mathrm{d}\mathbf{x},$$

$$\nabla\nabla\langle\tilde{\mathbf{T}}\rangle(\mathbf{0}) = \int\langle\mathbf{T}\rangle(\mathbf{x})i^2\mathbf{x}\mathbf{x}\,\mathrm{d}\mathbf{x}, \text{ etc.} \tag{28}$$

Therefore,

$$\int\langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')\,\mathrm{d}\mathbf{x} = \int\langle\mathbf{T}\rangle(\mathbf{x})\,\mathrm{d}\mathbf{x} = \langle\tilde{\mathbf{T}}\rangle(\boldsymbol{\xi}=\mathbf{0})$$

$$\int\left[\langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')\right](\mathbf{x}'-\mathbf{x})\,\mathrm{d}\mathbf{x} = -\int\langle\mathbf{T}\rangle(\mathbf{x})\mathbf{x}\,\mathrm{d}\mathbf{x} = i\nabla\langle\tilde{\mathbf{T}}\rangle(\mathbf{0})$$

$$\vdots$$

$$\int\left[\langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')\right](\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})\,\mathrm{d}\mathbf{x}=\int\langle\mathbf{T}\rangle(\mathbf{x})\mathbf{xxxx}\,\mathrm{d}\mathbf{x}$$
$$=\nabla\nabla\nabla\nabla\langle\tilde{\mathbf{T}}\rangle(\mathbf{0}). \tag{29}$$

Thus, Eq. (29) shows that we need derivatives of $\langle\tilde{\mathbf{T}}\rangle(\boldsymbol{\xi})$ with respect to $\boldsymbol{\xi}$. We introduce the notation $\partial(\,)/\partial\xi_m \equiv (\,)_{,m}$ and for convenience define:

$$\delta\mathbf{L}_a \equiv c_1\delta\mathbf{L}_1 + c_2\delta\mathbf{L}_2, \qquad \delta\mathbf{L}_b \equiv c_1\delta\mathbf{L}_2 + c_2\delta\mathbf{L}_1, \tag{30}$$

$$\mathbf{M} \equiv [\mathbf{I} + \boldsymbol{\Gamma}\delta\mathbf{L}_b]^{-1}, \tag{31}$$

so that

$$\left(\boldsymbol{\Gamma}^{-1} + c_1\delta\mathbf{L}_2 + c_2\delta\mathbf{L}_1\right)^{-1} = \mathbf{M}\boldsymbol{\Gamma}. \tag{32}$$

Using these, Eq. (23) may be written as

$$\langle\tilde{\mathbf{T}}\rangle(\boldsymbol{\xi}) = \delta\mathbf{L}_a\mathbf{M} + \delta\mathbf{L}_1\mathbf{M}\boldsymbol{\Gamma}\delta\mathbf{L}_2, \tag{33}$$

having used the fact that $(\mathbf{M}\boldsymbol{\Gamma})_{ijkl} = (\mathbf{M}\boldsymbol{\Gamma})_{klij}$, as can be seen from Eq. (32) by noting that $\Gamma_{ijkl} = \Gamma_{klij}$ and that the modulus tensors have this symmetry also.

Our assumption of isotropic phase *distributions* leads to the facts that

$$\boldsymbol{\Gamma}_{,m}(\boldsymbol{\xi}=\mathbf{0}) = \mathbf{0}, \qquad \boldsymbol{\Gamma}_{,mno}(\boldsymbol{\xi}=\mathbf{0}) = \mathbf{0}; \tag{34}$$

the first was proved by Drugan and Willis (1996); proof of the latter follows along exactly the same lines. Thus, using Eq. (31) to calculate the derivatives of $\mathbf{M}$ with respect to $\boldsymbol{\xi}$, and then setting $\boldsymbol{\xi}=\mathbf{0}$ and applying Eq. (34), gives the results:

$$\mathbf{M}_{,i} = \mathbf{0} \tag{35a}$$

$$\mathbf{M}_{,ij} = -\mathbf{M}\boldsymbol{\Gamma}_{,ij}\delta\mathbf{L}_b\mathbf{M} \tag{35b}$$

$$\mathbf{M}_{,ijk} = \mathbf{0} \tag{35c}$$

$$\mathbf{M}_{,ijkl} = \mathbf{M}\left(6\boldsymbol{\Gamma}_{,ij}\delta\mathbf{L}_b\mathbf{M}\boldsymbol{\Gamma}_{,kl} - \boldsymbol{\Gamma}_{,ijkl}\right)\delta\mathbf{L}_b\mathbf{M}\Big|_{(ijkl)}, \tag{35d}$$

where Eq. (35b) was used to simplify Eq. (35d), and the notation in the latter indicates that it is to be symmetrized with respect to arbitrary interchange of i, j, k, l.

Now, taking $\boldsymbol{\xi}$-derivatives of representation (33) of $\langle\tilde{\mathbf{T}}\rangle(\boldsymbol{\xi})$, and then setting $\boldsymbol{\xi} = \mathbf{0}$ and applying Eq. (34) leads to the results:

$$\langle\tilde{\mathbf{T}}\rangle(\mathbf{0}) = \delta\mathbf{L}_a\mathbf{M} + \delta\mathbf{L}_1\mathbf{M}\boldsymbol{\Gamma}\delta\mathbf{L}_2$$

$$\langle\tilde{\mathbf{T}}\rangle_{,m}(\mathbf{0}) = \mathbf{0}$$

$$\langle\tilde{\mathbf{T}}\rangle_{,mn}(\mathbf{0}) = \delta\mathbf{L}_a\mathbf{M}_{,mn} + \delta\mathbf{L}_1\big(\mathbf{M}_{,mn}\boldsymbol{\Gamma} + \mathbf{M}\boldsymbol{\Gamma}_{,mn}\big)\delta\mathbf{L}_2$$

$$\langle\tilde{\mathbf{T}}\rangle_{,mno}(\mathbf{0}) = \mathbf{0}$$

$$\langle\tilde{\mathbf{T}}\rangle_{,mnop}(\mathbf{0}) = \delta\mathbf{L}_a\mathbf{M}_{,mnop} + \delta\mathbf{L}_1\big(\mathbf{M}_{,mnop}\boldsymbol{\Gamma} + 6\mathbf{M}_{,mn}\boldsymbol{\Gamma}_{,op} + \mathbf{M}\boldsymbol{\Gamma}_{,mnop}\big)\delta\mathbf{L}_2\Big|_{(mnop)}, \tag{36}$$

where it is understood that all right-side terms in Eqs. (35) and (36) are evaluated at $\boldsymbol{\xi} = \mathbf{0}$, and the $\mathbf{M}$ derivatives in Eq. (36) are given by Eq. (35).

Now, using Eq. (27) with Eqs. (29) and (36) in Eq. (16), we have found the higher-order nonlocal constitutive equation to have the form:

$$\begin{aligned}\langle\boldsymbol{\sigma}\rangle(\mathbf{x}) = {} & [\mathbf{L}_0 + \delta\mathbf{L}_a\mathbf{M} + \delta\mathbf{L}_1\mathbf{M}\boldsymbol{\Gamma}\delta\mathbf{L}_2]\langle\mathbf{e}\rangle(\mathbf{x}) \\ & - \frac{1}{2}\big[\delta\mathbf{L}_a\mathbf{M}_{,mn} + \delta\mathbf{L}_1\big(\mathbf{M}_{,mn}\boldsymbol{\Gamma} + \mathbf{M}\boldsymbol{\Gamma}_{,mn}\big)\delta\mathbf{L}_2\big]\frac{\partial^2\langle\mathbf{e}\rangle(\mathbf{x})}{\partial x_m\partial x_n} \\ & + \frac{1}{24}\big[\delta\mathbf{L}_a\mathbf{M}_{,mnop} + \delta\mathbf{L}_1\big(\mathbf{M}_{,mnop}\boldsymbol{\Gamma} + 6\mathbf{M}_{,mn}\boldsymbol{\Gamma}_{,op} + \mathbf{M}\boldsymbol{\Gamma}_{,mnop}\big)\delta\mathbf{L}_2\big] \\ & \times \frac{\partial^4\langle\mathbf{e}\rangle(\mathbf{x})}{\partial x_m\partial x_n\partial x_o\partial x_p}.\end{aligned} \tag{37}$$

Here as in Eq. (35), it is understood that all $\mathbf{M}$ and $\boldsymbol{\Gamma}$ terms are evaluated at $\boldsymbol{\xi} = \mathbf{0}$. $\mathbf{M}$ and its derivatives are given by Eqs. (31) and (35), and Drugan and Willis (1996) showed that, since $h(0) = 1$,

$$\boldsymbol{\Gamma}(\mathbf{0}) = \big(\boldsymbol{\mathcal{T}}_0 * \tilde{h}\big)(\mathbf{0}) = \frac{1}{8\pi^3}\int \boldsymbol{\mathcal{T}}_0(\boldsymbol{\xi})\tilde{h}(\boldsymbol{\xi})\,\mathrm{d}\boldsymbol{\xi} = \frac{1}{4\pi}\int_{|\boldsymbol{\xi}|=1}\boldsymbol{\mathcal{T}}_0(\boldsymbol{\xi})\,\mathrm{d}S \equiv \mathbf{P} \tag{38}$$

$$\boldsymbol{\Gamma}_{,mn}(\mathbf{0}) = \frac{1}{4\pi}\int_{|\boldsymbol{\xi}|=1}\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\big[3\xi_m\xi_n - \delta_{mn}\big]\,\mathrm{d}S\left[\int_0^\infty h(r)r\,\mathrm{d}r\right]. \tag{39}$$

In addition to these, Eqs. (35) and (37) show we shall need the new tensor $\boldsymbol{\Gamma}_{,mnop}(\mathbf{0})$. This is derived in Appendix A, with the result:

$$\begin{aligned}\Gamma_{ijkl,\,mnop}(\mathbf{0}) = {} & \frac{1}{2}\big[3P_{ijkl}\delta_{mn}\delta_{op} - 6Q_{ijklmn}\delta_{op} \\ & + R_{ijklmnop}\big]_{(mnop)}\left[\int_0^\infty h(r)r^3\,\mathrm{d}r\right],\end{aligned} \tag{40}$$

where we have defined the tensors:

$$P_{ijkl} \equiv \frac{1}{4\pi}\int_{|\boldsymbol{\xi}|=1} \left[\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\right]_{ijkl} \mathrm{d}S \tag{41}$$

$$Q_{ijklmn} \equiv \frac{3}{4\pi}\int_{|\boldsymbol{\xi}|=1} \left[\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\right]_{ijkl}\xi_m\xi_n \mathrm{d}S \tag{42}$$

$$R_{ijklmnop} \equiv \frac{15}{4\pi}\int_{|\boldsymbol{\xi}|=1} \left[\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\right]_{ijkl}\xi_m\xi_n\xi_o\xi_p \mathrm{d}S. \tag{43}$$

Thus, when the phases are arbitrarily anisotropic, the nonlocal constitutive equation is given by Eq. (37) with Eqs. (30), (31), (35), (38)–(43).

4. Explicit form of nonlocal constitutive equation when both phases are isotropic

To obtain explicit expressions, we now examine the case in which both phases of the composite are isotropic, although still of arbitrary shape. The comparison material modulus tensor $\mathbf{L}_0$ is then sensibly taken to be isotropic, with components

$$c_{ijkl} = \left(\kappa - \frac{2}{3}\mu\right)\delta_{ij}\delta_{kl} + \mu\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right), \tag{44}$$

where κ and μ are the bulk and shear moduli, respectively. Drugan and Willis (1996), e.g., showed that for such a comparison material,

$$\begin{aligned}\left[\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\right]_{ijkl} = \frac{1}{\mu|\boldsymbol{\xi}|^4}\Bigg[&\frac{|\boldsymbol{\xi}|^2}{4}\left(\xi_i\delta_{jk}\xi_l + \xi_j\delta_{ik}\xi_l + \xi_i\delta_{jl}\xi_k + \xi_j\delta_{il}\xi_k\right)\\ &- \frac{3\kappa+\mu}{3\kappa+4\mu}\xi_i\xi_j\xi_k\xi_l\Bigg],\end{aligned} \tag{45}$$

and that use of this in Eq. (41) gives

$$\boxed{P_{ijkl} = -\frac{3\kappa+\mu}{15\mu(3\kappa+4\mu)}\delta_{ij}\delta_{kl} + \frac{3\kappa+6\mu}{10\mu(3\kappa+4\mu)}\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right).} \tag{46}$$

Next, from Eqs. (42) and (45), $\mathbf{Q}$ is given by

$$\begin{aligned}Q_{ijklmn} = \frac{3}{4\pi\mu}\int_{|\boldsymbol{\xi}|=1}\Bigg[&\frac{1}{4}\left(\xi_i\delta_{jk}\xi_l + \xi_j\delta_{ik}\xi_l + \xi_i\delta_{jl}\xi_k + \xi_j\delta_{il}\delta_k\right)\\ &- \frac{3\kappa+\mu}{3\kappa+4\mu}\xi_i\xi_j\xi_k\xi_l\Bigg]\xi_m\xi_n \mathrm{d}S.\end{aligned} \tag{47}$$

Drugan and Willis (1999) analyzed this tensor in a different context, and observed

that each term in the integrand will integrate to a constant isotropic tensor, so that the five terms in Eq. (47) integrate to the following five terms, in order:

$$Q_{ijklmn} = q_1\delta_{jk}I^{(4)}_{ilmn} + q_2\delta_{ik}I^{(4)}_{jlmn} + q_3\delta_{jl}I^{(4)}_{ikmn} + q_4\delta_{il}I^{(4)}_{jkmn} + q_5 I^{(6)}_{ijklmn}, \tag{48}$$

where q_1–q_5 are constants and we define

$$I^{(4)}_{ijkl} \equiv \delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}, \tag{49}$$

$$I^{(6)}_{ijklmn} \equiv \delta_{ij}I^{(4)}_{klmn} + \delta_{ik}I^{(4)}_{jlmn} + \delta_{il}I^{(4)}_{jkmn} + \delta_{im}I^{(4)}_{jkln} + \delta_{in}I^{(4)}_{jklm}. \tag{50}$$

One now computes each of the five terms in the invariant Q_{iikkmm} from Eq. (47), and equates each of them to the appropriate term in the same invariant computed from Eq. (48), using the facts that $I^{(4)}_{iikk}=15$, $I^{(6)}_{iikkmm}=105$; the results are:

$$q_1 = q_2 = q_3 = q_4 = \frac{1}{20\mu}, \qquad q_5 = -\frac{1}{35\mu}\frac{3\kappa+\mu}{3\kappa+4\mu}. \tag{51}$$

Substituting Eq. (51) in Eq. (48) and simplifying, one finds

$$\boxed{Q_{ijklmn} = \frac{3(3\kappa+8\mu)}{140\mu(3\kappa+4\mu)}I^{(6)}_{ijklmn} - \frac{1}{20\mu}\left[\delta_{ij}I^{(4)}_{klmn} + \delta_{kl}I^{(4)}_{ijmn} - I^{(4)}_{ijkl}\delta_{mn}\right].} \tag{52}$$

Next, from Eqs. (43) and (45), **R** is given by

$$R_{ijklmnop} = \frac{15}{4\pi\mu}\int_{|\xi|=1}\left[\frac{1}{4}\left(\xi_i\delta_{jk}\xi_l + \xi_j\delta_{ik}\xi_l + \xi_i\delta_{jl}\xi_k + \xi_j\delta_{il}\xi_k\right) - \frac{3\kappa+\mu}{3\kappa+4\mu}\xi_i\xi_j\xi_k\xi_l\right]\xi_m\xi_n\xi_o\xi_p\,\mathrm{d}S. \tag{53}$$

This is an eighth-order isotropic tensor; such have 105 terms, with 91 independent constants in general (Kearsley and Fong, 1975). However, also following Drugan and Willis (1999), observe that each term in the integrand will integrate to a constant isotropic tensor, so that the five terms in Eq. (53) integrate to the following five terms, in order:

$$R_{ijklmnop} = r_1\delta_{jk}I^{(6)}_{ilmnop} + r_2\delta_{ik}I^{(6)}_{jlmnop} + r_3\delta_{jl}I^{(6)}_{ikmnop} + r_4\delta_{il}I^{(6)}_{jkmnop} + r_5 I^{(8)}_{ijklmnop}, \tag{54}$$

where r_1–r_5 are constants and

$$I^{(8)}_{ijklmnop} \equiv \delta_{ij}I^{(6)}_{klmnop} + \delta_{ik}I^{(6)}_{jlmnop} + \delta_{il}I^{(6)}_{jkmnop} + \delta_{im}I^{(6)}_{jklnop} + \delta_{in}I^{(6)}_{jklmop} + \delta_{io}I^{(6)}_{jklmnp} + \delta_{ip}I^{(6)}_{jklmno}. \tag{55}$$

One now computes each of the five terms in the invariant $R_{iikkmmpp}$ from Eq. (53), and equates each of them to the appropriate term in the same invariant computed from Eq. (54), using the fact that $I^{(8)}_{iikkmmpp} = 945$; the results are:

$$r_1 = r_2 = r_3 = r_4 = \frac{1}{28\mu}, \qquad r_5 = -\frac{1}{63\mu}\frac{3\kappa+\mu}{3\kappa+4\mu}. \tag{56}$$

Thus,

$$\boxed{\begin{aligned} R_{ijklmnop} = {} & \frac{1}{28\mu}\left(\delta_{jk} I^{(6)}_{ilmnop} + \delta_{ik} I^{(6)}_{jlmnop} + \delta_{jl} I^{(6)}_{ikmnop} + \delta_{il} I^{(6)}_{jkmnop}\right) \\ & - \frac{1}{63\mu}\frac{3\kappa+\mu}{3\kappa+4\mu} I^{(8)}_{ijklmnop}. \end{aligned}} \tag{57}$$

Let us now put all the preceding results together and exhibit the full nonlocal constitutive equation for the case of a two-phase composite comprised of an isotropic distribution of isotropic phases (which may still have arbitrary shape). First, we rewrite the constitutive equation (37) as

$$\langle\sigma\rangle_{ij}(\mathbf{x}) = \hat{L}_{ijkl}\langle e\rangle_{kl}(\mathbf{x}) + \hat{L}^{(1)}_{ijklmn}\frac{\partial^2\langle e\rangle_{kl}(\mathbf{x})}{\partial x_m \partial x_n} + \hat{L}^{(2)}_{ijklmnop}\frac{\partial^4\langle e\rangle_{kl}(\mathbf{x})}{\partial x_m \partial x_n \partial x_o \partial x_p}. \tag{58}$$

For computation of products of isotropic fourth-rank tensors such as that of Eq. (44), it is convenient to employ the *symbolic* notation of Hill (1965), which represents Eq. (44) as

$$\mathbf{L}_0 = [3\kappa, 2\mu], \tag{59}$$

and in which the product of two isotropic tensors, say $\mathbf{L}_0$ and $\mathbf{L}_1$, is

$$\mathbf{L}_0\mathbf{L}_1 = \left[(3\kappa)(3\kappa_1), (2\mu)(2\mu_1)\right]. \tag{60}$$

In this notation, $\mathbf{P} \equiv \mathbf{\Gamma}(\mathbf{0})$ is, from Eq. (46)

$$\mathbf{P} = \left[\frac{1}{3\kappa+4\mu}, \frac{3(\kappa+2\mu)}{5\mu(3\kappa+4\mu)}\right] \equiv \left[3\kappa_p, 2\mu_p\right], \tag{61}$$

and obviously from Eq. (30),

$$\begin{aligned} \delta\mathbf{L}_a &= \left[3(c_1\kappa_1 + c_2\kappa_2 - \kappa), 2(c_1\mu_1 + c_2\mu_2 - \mu)\right], \\ \delta\mathbf{L}_b &= \left[3(c_1\kappa_2 + c_2\kappa_1 - \kappa), 2(c_1\mu_2 + c_2\mu_1 - \mu)\right]. \end{aligned} \tag{62}$$

Using these with Eq. (31), one computes

$$\mathbf{M}(\mathbf{0}) = \left[\frac{3\kappa + 4\mu}{4\mu + 3(c_1\kappa_2 + c_2\kappa_1)}, \frac{5\mu(3\kappa + 4\mu)}{\mu(9\kappa + 8\mu) + 6(\kappa + 2\mu)(c_1\mu_2 + c_2\mu_1)}\right]$$
$$\equiv \left[3\kappa_M, 2\mu_M\right]. \tag{63}$$

Then, comparing the first coefficient in Eq. (58) with that in Eq. (37), one computes the known result (see, e.g., Willis, 1982):

$$\hat{\mathbf{L}} = \left[3\frac{4\mu(c_1\kappa_1 + c_2\kappa_2) + 3\kappa_1\kappa_2}{4\mu + 3(c_1\kappa_2 + c_2\kappa_1)},\right.$$
$$\left. 2\frac{\mu(9\kappa + 8\mu)(c_1\mu_1 + c_2\mu_2) + 6(\kappa + 2\mu)\mu_1\mu_2}{\mu(9\kappa + 8\mu) + 6(\kappa + 2\mu)(c_1\mu_2 + c_2\mu_1)}\right] \equiv \left[3\kappa_L, 2\mu_L\right], \tag{64}$$

so that, with κ_L, μ_L given by Eq. (64),

$$\hat{L}_{ijkl} = \left(\kappa_L - \frac{2}{3}\mu_L\right)\delta_{ij}\delta_{kl} + \mu_L\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right). \tag{65}$$

Next, let us compute the tensor $\hat{\mathbf{L}}^{(1)}$ appearing in Eq. (58). Writing this in tensor notation but with the last two indices represented explicitly, we have by comparison with Eq. (37), employing Eq. (35b):

$$\left[\hat{\mathbf{L}}^{(1)}\right]_{mn} = \frac{1}{2}\delta\mathbf{L}_a\mathbf{M}\boldsymbol{\Gamma}_{,mn}\delta\mathbf{L}_b\mathbf{M} + \frac{1}{2}\delta\mathbf{L}_1\mathbf{M}\boldsymbol{\Gamma}_{,mn}(\delta\mathbf{L}_b\mathbf{M}\mathbf{P} - \mathbf{I})\delta\mathbf{L}_2, \tag{66}$$

where again $\mathbf{M}$ and $\boldsymbol{\Gamma}_{,mn}$ are understood to be evaluated at $\boldsymbol{\xi} = \mathbf{0}$. From Eqs. (39), (41) and (42),

$$\Gamma_{ijkl,\,mn}(\mathbf{0}) = \left(Q_{ijklmn} - P_{ijkl}\delta_{mn}\right)\int_0^\infty h(r)r\,\mathrm{d}r, \tag{67}$$

where $\mathbf{P}$ and $\mathbf{Q}$ are given by Eqs. (46) and (52). Employing all these facts, Eq. (66) can be expressed explicitly as:

$$\boxed{\begin{aligned}\hat{L}^{(1)}_{ijklmn} = \frac{D}{2}\Bigg[A_1\left(I^{(6)}_{ijklmn} - \frac{7}{3}I^{(4)}_{ijkl}\delta_{mn}\right) + A_2 I^{(4)}_{ijmn}\delta_{kl} + A_3\delta_{ij}I^{(4)}_{klmn} \\ + A_4\delta_{ij}\delta_{kl}\delta_{mn}\Bigg] \times \int_0^\infty h(r)r\,\mathrm{d}r,\end{aligned}} \tag{68}$$

where

$$A_1 = \frac{15}{7}c_1(3\kappa + 8\mu)(\mu_1 - \mu_2)^2\left[4\mu + 3(c_1\kappa_2 + c_2\kappa_1)\right]$$

$$A_2 = -6\mu\kappa_2\mu_1(3\kappa - 4\mu) + 30c_1\mu(3\kappa + 4\mu)(\kappa_1 - \kappa_2)(\mu_1 - \mu_2)$$
$$- 4c_1\left(9\kappa^2 + 33\kappa\mu + 40\mu^2\right)(\mu_1 - \mu_2)^2 - 3c_1(3\kappa + 16\mu)(c_2\kappa_1 + c_1\kappa_2)$$
$$\times (\mu_1 - \mu_2)^2 - 36(\kappa + 2\mu)\mu_1\left[\kappa_2\mu_1 - \kappa(\mu_1 - \mu_2) + \kappa_1(\mu - \mu_2)\right]$$
$$- 6\mu(9\kappa + 8\mu)\left[-\kappa(\mu_1 - \mu_2) + \mu(\kappa_1 - \kappa_2) - \kappa_1\mu_2\right]$$

$$A_3 = A_2 + 12\left[\kappa_1(\mu - \mu_2) - \kappa_2(\mu - \mu_1) - \kappa(\mu_1 - \mu_2)\right]\left[\mu(9\kappa + 8\mu) + 6(\kappa + 2\mu)\right.$$
$$\left.\times (c_1\mu_2 + c_2\mu_1)\right]$$

$$A_4 = \frac{10}{3}c_1(\mu_1 - \mu_2)\Big\{5(3\kappa + 8\mu)\left[4\mu(\mu_1 - \mu_2) - 3\kappa_1\mu_2\right]$$
$$- 6\mu(9\kappa + 8\mu)(\kappa_1 - \kappa_2) + 36\mu_1\kappa_2(\kappa + 2\mu) + 3(3\kappa + 16\mu)$$
$$\times \left[c_1\kappa_2(\mu_1 - \mu_2) + \kappa_1(c_1\mu_2 + c_2\mu_1)\right]\Big\} \tag{69}$$

$$D \equiv \frac{c_2\mu(3\kappa + 4\mu)}{\left[4\mu + 3(c_1\kappa_2 + c_2\kappa_1)\right]\left[\mu(9\kappa + 8\mu) + 6(\kappa + 2\mu)(c_1\mu_2 + c_2\mu_1)\right]^2}. \tag{70}$$

We emphasize that Eq. (64) and Eqs. (68)–(70) have been derived in terms of *arbitrary* isotropic comparison moduli κ, μ, the latter results generalizing those given by Drugan and Willis (1996) who assumed a specific choice for the comparison moduli. An explicit expression for $\hat{\mathbf{L}}^{(2)}$ of Eq. (58) can be similarly derived, but is omitted here. Later we give an expression for it for a specific choice of the comparison moduli.

5. Specific results for a matrix reinforced by a random distribution of nonoverlapping identical spheres

We showed in the previous section that the nonlocal constitutive equation permits a very explicit representation when the two-phase composite consists of an isotropic distribution of isotropic phases (which may have arbitrary shapes), *except* for two radial integrals of the two-point correlation function, which appear in Eqs. (39) and (40). Evaluation of these requires a good model for the two-point correlation function. Such does exist for the case of a random dispersion of nonoverlapping identical spheres. Markov and Willis (1998) have shown that the two-point correlation function in this case can be expressed as a simple one-tuple integral containing the radial distribution function, and Percus and Yevick (1958)

devised a very realistic statistical mechanics model for the radial distribution function in this case, which Wertheim (1963) solved as a closed-form Laplace transform.

Using these, Drugan and Willis (1996) showed that the radial integral appearing in Eq. (39) is

$$\int_0^\infty h(r)r\,\mathrm{d}r = a^2\frac{(2-c_1)(1-c_1)}{5(1+2c_1)}. \tag{71}$$

We need to evaluate the new radial integral appearing in Eq. (40); this is carried out using an approach very similar to that of Drugan and Willis (1996), with the result:

$$\int_0^\infty h(r)r^3\,\mathrm{d}r = a^4\frac{2(1-c_1)\left(30-177c_1+248c_1^2-124c_1^3+14c_1^4\right)}{175(1+2c_1)^3}. \tag{72}$$

In the above, a is the radius and c_1 the volume fraction of the spheres. Using Eqs. (71) and (72), the nonlocal constitutive equation derived in the previous section is now completely explicit. It does, however, still contain the comparison moduli κ, μ. Before this constitutive equation can be employed in practice, good choices must be made for these comparison moduli. In the next section, we employ the nonlocal constitutive equation to explore what would make optimal choices for these moduli.

6. Optimal choice of the comparison moduli

Here we conduct an initial exploration of a new approach for determining the optimal choice of the comparison moduli in the variational estimate of the leading-order effective modulus tensor, and also for the full nonlocal constitutive equation. Although it is anticipated that this approach will be applicable, and hopefully valuable, in more difficult problems where the choice of optimal comparison moduli is even more challenging, such as in nonlinear composites, here we explore its predictions in the isotropic linear composites case within the present formulation incorporating up through two-point statistics.

The basic idea is to employ the micromechanics-based nonlocal constitutive equation we have derived and enquire as to which choice(s) of comparison moduli will result in the smallest nonlocal correction to the standard constant-effective-modulus constitutive equation — i.e., will minimize the RVE size over which the 'effective modulus' tensor accurately applies. Specifically, in the context of Sections 4 and 5, and with reference to Eq. (58), for which choice(s) of comparison moduli will some appropriate measure of the magnitude of the ratio $\hat{\mathbf{L}}^{(1)}/(a^2\hat{\mathbf{L}})$ be minimized? This is a difficult question in general, but let us examine an example case. Drugan and Willis (1996), in an analysis to estimate the

minimum representative volume element size to which a constant-effective-modulus constitutive equation applies, found that one of the most demanding simple cases was that of how uniaxial ensemble-average stress relates to uniaxial ensemble-average strain in the same direction and which varies sinusoidally in that direction, when all other components of average strain are zero. In this case, the nonlocal constitutive Eq. (58) reduces to (ignoring for now the higher-order nonlocal term)

$$\langle\sigma\rangle_{11}(\mathbf{x}) = \hat{L}_{1111}\langle e\rangle_{11}(\mathbf{x}) + \hat{L}^{(1)}_{111111}\frac{\partial^2\langle e\rangle_{11}(\mathbf{x})}{\partial x_1^2}. \tag{73}$$

Here, the nonlocal contribution is minimized by minimizing the absolute value of the ratio $\hat{L}^{(1)}_{111111}/(a^2\hat{L}_{1111})$. This ratio can be computed explicitly for the present case of an isotropic matrix reinforced by a random isotropic distribution of isotropic nonoverlapping identical spheres, by employing the results of the preceding sections. We shall consider two specific cases.

In the case of a matrix weakened by voids, we calculate from the preceding results

$$\frac{\hat{L}^{(1)}_{111111}}{a^2\hat{L}_{1111}} = \frac{2c_1(2-c_1)(1-c_1)\mu_2(3\kappa+4\mu)\left[3\kappa\left(63\kappa_2\mu+57c_1\kappa_2\mu_2+20\mu\mu_2\right)+4\mu\left(42\kappa_2\mu+93c_1\kappa_2\mu_2+40\mu\mu_2\right)\right]}{35(1+2c_1)\left[\mu(9\kappa+8\mu)+6c_1(\kappa+2\mu)\mu_2\right]\left[9\kappa\left(3\kappa_2\mu+5c_1\kappa_2\mu_2+4\mu\mu_2\right)+4\mu\left(6\kappa_2\mu+15c_1\kappa_2\mu_2+8\mu\mu_2\right)\right]}. \tag{74}$$

Examination of this expression shows that for all choices of the comparison moduli in the ranges $0\le\kappa\le\kappa_2$, $0\le\mu\le\mu_2$, *the choice that the comparison moduli equal the matrix moduli, namely* $\kappa=\kappa_2$, $\mu=\mu_2$, *minimizes the absolute value of this ratio for all permissible values of* κ_2, μ_2 *and for all* $0\le c_1\le 0.63$ (the full possible range of concentrations achievable for random distributions of nonoverlapping spheres; Scott, 1960). (This implies that the Hashin-Shtrikman *upper* bound for the effective modulus tensor $\hat{\mathbf{L}}$ is optimal in this case, within the present two-point-statistics formulation.) As an illustration of this, Fig. 1 shows the value of this ratio for an incompressible matrix ($\kappa_2\to\infty$) as a function of c_1 for three different choices of the comparison moduli: when they equal the matrix moduli, the void moduli, and the self-consistent moduli. The latter can be derived by setting $\kappa_L=\kappa$ and $\mu_L=\mu$ in Eq. (64) with $\kappa_1=\mu_1=0$, $\kappa_2\to\infty$; the result is (e.g., Willis, 1982)

$$\kappa = \frac{4(1-c_1)(1-2c_1)\mu_2}{c_1(3-c_1)}, \qquad \mu = \frac{3(1-2c_1)\mu_2}{(3-c_1)}. \tag{75}$$

In the case of a matrix reinforced by rigid particles,

$$\frac{\hat{L}^{(1)}_{111111}}{a^2\hat{L}_{1111}} = \frac{2c_1(2-c_1)(1-c_1)\mu(3\kappa+4\mu)(57\kappa+124\mu)}{105(1+2c_1)(\kappa+2\mu)\left[3\kappa\left(3\kappa_2+10c_1\mu+4\mu_2\right)+2\mu\left(9\kappa_2+20c_1\mu+12\mu_2\right)\right]}. \quad (76)$$

Examination of this expression shows that for all choices of the comparison moduli in the ranges $\kappa_2 \le \kappa \le \infty$, $\mu_2 \le \mu \le \infty$, *the choice that the comparison moduli equal the matrix moduli*, namely $\kappa = \kappa_2$, $\mu = \mu_2$, *minimizes the absolute value of this ratio for all permissible values of* κ_2, μ_2 *and for all* $0 \le c_1 \le 0.63$. (This implies that the Hashin–Shtrikman *lower* bound for the effective modulus tensor $\hat{\mathbf{L}}$ is optimal in this case, within the present formulation incorporating up through two-point statistics.) If this ratio is plotted (for various allowable ratios of the matrix moduli) as a function of c_1 for three different choices of the comparison moduli: when they equal the matrix moduli, the inclusion moduli, and the self-consistent moduli, the result looks very much like Fig. 1.

We have thus shown in this section that, within the assumptions of the present analysis as detailed above, in both the cases of a matrix weakened by a distribution of voids and strengthened by a distribution of rigid inclusions, the optimal choice of the comparison modulus tensor is equal to the *matrix* modulus tensor, from the point of view of minimizing the relative magnitude of the nonlocal correction term. Thus, we shall make this choice for the remainder of this article.

Experimental measurements and computational simulations appear to support this conclusion; see, e.g., the review article of Torquato (1991). To cite one specific example, the experiments of Smith (1976) on an epoxy matrix reinforced by glass

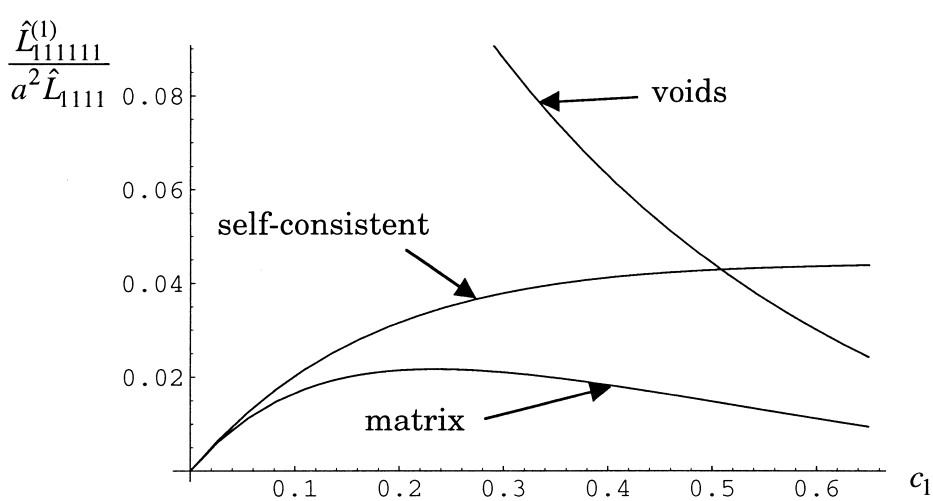

Fig. 1. Nonlocal/local coefficient ratio Eq. (74) for an incompressible matrix containing a random distribution of voids, as a function of void volume fraction, for three choices of the comparison moduli. The curve for voids continues to higher values at lower concentrations.

spheres show that the Hashin-Shtrikman *lower* bound for the effective shear modulus (obtained by choosing the comparison moduli equal to the matrix moduli in this case, as noted above) agrees quite closely with the experimentally-measured values, for the spheres concentration range $0 \le c_1 \le 0.4$.

7. Explicit form of higher-order nonlocal constitutive equation for optimal choice of comparison moduli

We shall now derive the explicit form of the full higher-order nonlocal constitutive equation for the case of an isotropic matrix reinforced/weakened by a random distribution of an isotropic second phase, *when the comparison moduli are chosen equal to the matrix moduli* (as justified in Section 6). That is, we choose

$$\mathbf{L}_0 = \mathbf{L}_2 \Longrightarrow \delta\mathbf{L}_2 = \mathbf{0}. \tag{77}$$

Then the constitutive Eq. (37) simplifies to

$$\boxed{\begin{aligned}\langle\boldsymbol{\sigma}\rangle(\mathbf{x}) &= [\mathbf{L}_2 + c_1\mathbf{B}]\langle\mathbf{e}\rangle(\mathbf{x}) + \frac{c_1c_2}{2}\mathbf{B}\boldsymbol{\Gamma}_{,mn}\mathbf{B}\frac{\partial^2\langle\mathbf{e}\rangle(\mathbf{x})}{\partial x_m\partial x_n}\\ &\quad + \frac{c_1c_2}{24}\mathbf{B}\big(6c_2\boldsymbol{\Gamma}_{,mn}\mathbf{B}\boldsymbol{\Gamma}_{,op} - \boldsymbol{\Gamma}_{,mnop}\big)\mathbf{B}\frac{\partial^4\langle\mathbf{e}\rangle(\mathbf{x})}{\partial x_m\partial x_n\partial x_o\partial x_p},\end{aligned}} \tag{78}$$

where

$$\mathbf{B} \equiv \left[(\delta\mathbf{L}_1)^{-1} + c_2\mathbf{P}\right]^{-1}. \tag{79}$$

Henceforth, the matrix moduli will be subscript-free, while the inclusion moduli will have subscript 1. The constitutive Eq. (78) has the form

$$\langle\sigma\rangle_{ij}(\mathbf{x}) = \hat{L}_{ijkl}\langle e\rangle_{kl}(\mathbf{x}) + \hat{L}^{(1)}_{ijklmn}\frac{\partial^2\langle e\rangle_{kl}(\mathbf{x})}{\partial x_m\partial x_n} + \hat{L}^{(2)}_{ijklmnop}\frac{\partial^4\langle e\rangle_{kl}(\mathbf{x})}{\partial x_m\partial x_n\partial x_o\partial x_p}. \tag{80}$$

Application of Eq. (77) to Eqs. (64) and (65) gives

$$\hat{L}_{ijkl} = \left(\kappa_L - \frac{2}{3}\mu_L\right)\delta_{ij}\delta_{kl} + \mu_L\big(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\big), \tag{81}$$

with

$$\begin{aligned}\kappa_L &= \frac{3\kappa\kappa_1 + 4\mu\big[c_1\kappa_1 + (1-c_1)\kappa\big]}{3c_1\kappa + 3\kappa_1(1-c_1) + 4\mu},\\ \mu_L &= \mu\frac{6(\kappa+2\mu)\mu_1 + (9\kappa+8\mu)\big[c_1\mu_1 + (1-c_1)\mu\big]}{\mu(9\kappa+8\mu) + 6(\kappa+2\mu)\big[c_1\mu + (1-c_1)\mu_1\big]}.\end{aligned} \tag{82}$$

Application of Eq. (77) to Eqs. (68)–(70) yields

$$\hat{L}^{(1)}_{ijklmn} = \frac{D}{2}\left[A_1\left(I^{(6)}_{ijklmn} - \frac{7}{3}I^{(4)}_{ijkl}\delta_{mn}\right) + A_2\left(\delta_{ij}I^{(4)}_{klmn} + \delta_{kl}I^{(4)}_{ijmn} - \frac{10}{3}\delta_{ij}\delta_{kl}\delta_{mn}\right)\right]\int_0^\infty h(r)r\,\mathrm{d}r, \tag{83}$$

where

$$A_1 = \frac{15}{7}c_1(3\kappa + 8\mu)(\mu_1 - \mu)^2\{4\mu + 3[c_1\kappa + (1 - c_1)\kappa_1]\}$$

$$A_2 = -c_1(\mu - \mu_1)\Big\{-9\kappa^2[(6 - c_1)\mu + (4 + c_1)\mu_1] + 8\mu\Big(20\mu(\mu - \mu_1) + 3\kappa_1[(7 - 2c_1)\mu - 2(1 - c_1)\mu_1]\Big) + 3\kappa\big(3\kappa_1[(11 - c_1)\mu - (1 - c_1)\mu_1] + 4\mu[(1 + 4c_1)\mu - (11 + 4c_1)\mu_1]\big)\Big\}$$

$$D = \frac{(1 - c_1)\mu(3\kappa + 4\mu)}{\{4\mu + 3[c_1\kappa + (1 - c_1)\kappa_1]\}\{\mu(9\kappa + 8\mu) + 6(\kappa + 2\mu)[c_1\mu + (1 - c_1)\mu_1]\}^2}. \tag{85}$$

From Eqs. (78) and (79),

$$\hat{L}^{(2)}_{ijklmnop} = \frac{c_1(1 - c_1)}{24}B_{ijqr}[6(1 - c_1)\Gamma_{qrst,\,mn}B_{stuv}\Gamma_{uvwx,\,op} - \Gamma_{qrwx,\,mnop}]B_{wxkl}, \tag{86}$$

where

$$B_{ijkl} = \left(\kappa_B - \frac{2}{3}\mu_B\right)\delta_{ij}\delta_{kl} + \mu_B(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) \tag{87}$$

$$\kappa_B = \frac{(\kappa_1 - \kappa)(3\kappa + 4\mu)}{4\mu + 3[c_1\kappa + (1 - c_1)\kappa_1]}, \qquad \mu_B = \frac{5\mu(\mu_1 - \mu)(3\kappa + 4\mu)}{5\mu(3\kappa + 4\mu) + 6(1 - c_1)(\mu_1 - \mu)(\kappa + 2\mu)} \tag{88}$$

$$\Gamma_{ijkl,\,mn}(\mathbf{0}) = (Q_{ijklmn} - P_{ijkl}\delta_{mn})\left[\int_0^\infty h(r)r\,\mathrm{d}r\right] \tag{89}$$

$$\Gamma_{ijkl,\,mnop}(\mathbf{0}) = \frac{1}{2}\big[3P_{ijkl}\delta_{mn}\delta_{op} - 6Q_{ijklmn}\delta_{op} + R_{ijklmnop}\big]_{(mnop)}\left[\int_0^\infty h(r)r^3\,\mathrm{d}r\right], \tag{90}$$

and **P**, **Q**, **R** are given by Eqs. (46), (52) and (57) in terms of the matrix moduli κ, μ. Observe from the above that the constitutive equation is completely specified except for the integrals involving $h(r)$. For the special case of the reinforcements being a random distribution of nonoverlapping identical spheres, these integrals are given by Eqs. (71) and (72).

8. More accurate estimates of minimum representative volume element size

We are now in a position to provide more accurate estimates of minimum representative volume element (RVE) size by use of the new higher-order nonlocal constitutive equation. This will permit a check on the estimates derived by Drugan and Willis (1996) for the minimum RVE size over which the standard constant-effective-modulus constitutive equation can be expected to be valid. It will also permit an analytical assessment of the improvement to RVE size facilitated by incorporating a nonlocal term into the constitutive equation. It is well to remember that all of the results to be presented are derived from our formulation that incorporates up through two-point statistics. We do not yet know the quantitative degree to which incorporation of higher-order statistical information would affect these results.

Drugan and Willis (1996) provided quantitative estimates of the minimum RVE size over which a constitutive equation of the form

$$\langle\sigma\rangle_{ij}(\mathbf{x}) = \hat{L}_{ijkl}\langle e\rangle_{kl}(\mathbf{x}) \tag{91}$$

may be expected to accurately reflect the composite's constitutive behavior. They did this by considering sinusoidally-varying ensemble-average strain and determining the smallest wavelength at which the first nonlocal term (the second right-side term in Eq. (80)) makes a non-negligible correction to the first right-side term. A tacit assumption in their analysis, of course, was that all nonlocal effects can be well-approximated by just the first nonlocal term. However, the minimum RVE sizes they found were surprisingly small, leading one to desire confirmation that retention of only one nonlocal term is indeed sufficiently accurate. Thus, here we shall re-examine the minimum RVE size for accuracy of Eq. (91), by calculating the smallest wavelength of average strain at which *both* nonlocal terms in Eq. (80) *together* make a non-negligible correction to the first right-side term.

We shall employ the same example case examined by Drugan and Willis (1996), who found it to be the most demanding among several simple cases studied: the relation of normal ensemble-average stress to normal average strain in the same

direction when this average strain varies with position in its direction of straining, and no other components of strain act. That is, the average strain field is

$$\langle e\rangle_{11}(\mathbf{x}) = \varepsilon \sin\frac{2\pi x_1}{l}, \quad \text{all other } \langle e\rangle_{ij}(\mathbf{x}) = 0, \tag{92}$$

where $|\varepsilon| \ll 1$ is a pure number, so that the 11 component of Eq. (80) is

$$\begin{aligned}\langle\sigma\rangle_{11}(\mathbf{x}) &= \hat{L}_{1111}\langle e\rangle_{11}(\mathbf{x}) + \hat{L}^{(1)}_{111111}\frac{\partial^2\langle e\rangle_{11}(\mathbf{x})}{\partial x_1^2} + \hat{L}^{(2)}_{11111111}\frac{\partial^4\langle e\rangle_{11}(\mathbf{x})}{\partial x_1^4}\\ &= \left[\hat{L}_{1111} - \frac{4\pi^2}{l^2}\hat{L}^{(1)}_{111111} + \frac{16\pi^4}{l^4}\hat{L}^{(2)}_{11111111}\right]\varepsilon\sin\frac{2\pi x_1}{l}.\end{aligned} \tag{93}$$

Thus, defining $\alpha \times 100$ as the error percentage of the constant effective modulus term, i.e., the percentage correction provided by the sum of the two nonlocal terms, we obtain the following equation for the minimum RVE size l:

$$\left| -\frac{4\pi^2}{l^2}\hat{L}^{(1)}_{111111} + \frac{16\pi^4}{l^4}\hat{L}^{(2)}_{11111111}\right| = \alpha\left|\hat{L}_{1111}\right|, \tag{94}$$

where the vertical lines denote absolute value. The correct solution to Eq. (94) is obtained by determining the first l-value that satisfies it as l decreases from a large value. The above reduces to the result of Drugan and Willis (1996) if one sets $\hat{L}^{(2)}_{11111111} = 0$.

To obtain explicit results for specific materials, recall that the isotropic elastic moduli are related as

$$\kappa = \frac{2(1+\nu)}{3(1-2\nu)}\mu, \tag{95}$$

where ν is Poisson's ratio. Large classes of important structural materials are well-characterized by two values of Poisson's ratio: glass, Al_2O_3 (alumina), WC and concrete all have $\nu \approx 0.2$, while aluminum, steels, brass, copper and titanium all have $\nu \approx 0.33$. We have employed Eq. (94) to calculate minimum RVE sizes for matrix materials having each of these values of ν, for a matrix 'reinforced' by a random distribution of identical nonoverlapping spheres, for the extreme cases of the spheres being voids ($\kappa_1 = \mu_1 = 0$) and being rigid particles ($\kappa_1 = \mu_1 = \infty$). These results are shown in boldface in Table 1, where they are compared to the results for the same cases by Drugan and Willis (1996), who retained only one nonlocal term in their constitutive equation. The numerical values given are for 5% error for use of Eq. (91); Eq. (94) shows how the results will change for a different error percentage, i.e., different α-value.

Several interesting conclusions can be drawn from the results shown. As with the Drugan and Willis estimates, the new results show that once one of the matrix elastic moduli is specified (here, ν), the RVE estimates are unaffected by the value of the other matrix modulus. Overall, the estimates derived via one nonlocal term

by Drugan and Willis (1996) are seen to be very reasonable: in most cases they are quite accurate and show correct trends with changes in modulus and concentration. In general, the Drugan and Willis results underestimate the minimum RVE sizes for voids, and overestimate them for rigid inclusions. The greatest disparities in the predictions occur in the cases of rigid particles at relatively low particle concentrations ($0.05 \leq c_1 \leq 0.2$); however, the second-order corrections are not small compared to the first-order ones in these cases. These disparities are greatly reduced for smaller α-values. Interestingly, the new results show that a puzzling feature of the Drugan and Willis results, that the RVE sizes are identical for voids and rigid particles in a matrix with $\nu = 0.2$, was an artifact

Table 1
Minimum RVE sizes for 5% error in validity of Eq. (91), normalized by 'reinforcement' sphere diameter, based on two nonlocal terms (in bold) compared to the predictions of Drugan and Willis (1996) based on one nonlocal term

	Minimum RVE size, $l/(2a)$, for 5% error			
	$\nu_m = 0.2$		$\nu_m = 0.33$	
c_1	Voids	Rigid particles	Voids	Rigid particles
0.025	1.005	1.005	1.018	0.8513
	1.177	**0.9449**	**1.232**	**0.8821**
0.05	1.345	1.345	1.363	1.144
	1.490	**0.9796**	**1.539**	**0.9218**
0.1	1.710	1.710	1.729	1.462
	1.825	**0.9323**	**1.863**	**0.8838**
0.15	1.887	1.887	1.906	1.623
	1.985	**1.422**	**2.015**	**0.8092**
0.2	1.967	1.967	1.984	1.701
	2.052	**1.728**	**2.077**	**1.402**
0.25	1.987	1.987	2.003	1.727
	2.063	**1.821**	**2.083**	**1.540**
0.3	1.968	1.968	1.981	1.718
	2.035	**1.844**	**2.052**	**1.584**
0.35	1.921	1.921	1.933	1.684
	1.982	**1.826**	**1.995**	**1.585**
0.4	1.854	1.854	1.864	1.632
	1.909	**1.780**	**1.920**	**1.556**
0.45	1.772	1.772	1.781	1.566
	1.822	**1.714**	**1.831**	**1.508**
0.5	1.679	1.679	1.686	1.489
	1.725	**1.634**	**1.732**	**1.445**
0.55	1.577	1.577	1.583	1.403
	1.619	**1.542**	**1.624**	**1.370**
0.6	1.467	1.467	1.472	1.309
	1.506	**1.441**	**1.510**	**1.286**

of their leading-order estimates: the new results show that RVE sizes are larger for voids than for rigid particles in this case. Finally, the new results confirm one overall conclusion of Drugan and Willis, namely that the minimum RVE size does not exceed about twice (2.1 times here) the diameter of the 'reinforcing' spheres for all cases and concentrations examined.

The higher-order nonlocal constitutive equation derived herein also permits another interesting calculation: an analytical assessment of the improvement to RVE size that one obtains by use of Drugan and Willis' (1996) leading-order nonlocal constitutive equation as compared to the standard constant-effective-modulus constitutive Eq. (91). The RVE sizes we have computed for validity of Eq. (91) are quite small, but they are for the relatively benign spherical voids/

Table 2
Minimum RVE size for 0.5% accuracy of the Drugan and Willis (1996) nonlocal constitutive equation (bold) versus that for the same accuracy of the constant-effective-modulus constitutive equation (91)

	Minimum RVE size, $l/(2a)$, for 0.5% error			
	$\nu_m = 0.2$		$\nu_m = 0.33$	
c_1	Voids	Rigid particles	Voids	Rigid particles
0.025	3.253	2.821	3.322	2.196
	1.520	**2.036**	**1.651**	**1.854**
0.05	4.311	4.059	4.381	3.388
	1.750	**2.280**	**1.877**	**2.077**
0.1	5.448	5.296	5.518	4.501
	1.939	**2.411**	**2.041**	**2.198**
0.15	6.001	5.890	6.066	5.050
	1.989	**2.380**	**2.066**	**2.170**
0.2	6.248	6.163	6.307	5.318
	1.975	**2.286**	**2.032**	**2.083**
0.25	6.309	6.240	6.360	5.415
	1.926	**2.162**	**1.967**	**1.970**
0.3	6.246	6.189	6.290	5.397
	1.856	**2.025**	**1.885**	**1.844**
0.35	6.095	6.048	6.132	5.298
	1.774	**1.882**	**1.794**	**1.711**
0.4	5.882	5.842	5.913	5.139
	1.683	**1.736**	**1.697**	**1.575**
0.45	5.622	5.588	5.647	4.934
	1.588	**1.589**	**1.597**	**1.438**
0.5	5.326	5.297	5.347	4.695
	1.491	**1.443**	**1.496**	**1.301**
0.55	5.001	4.977	5.018	4.426
	1.392	**1.296**	**1.395**	**1.162**
0.6	4.653	4.632	4.667	4.133
	1.293	**1.148**	**1.295**	**1.019**

inclusions treated. One anticipates that RVE sizes for more anisotropically-shaped inclusions, such as fibers, ellipsoids, cracks, etc., would lead to significantly larger minimum RVE sizes, as might other situations such as nonlinear composites. In such cases of significantly larger minimum RVE sizes for applicability of Eq. (91), it would be valuable to employ a nonlocal constitutive equation such as that of Drugan and Willis which would permit a significantly smaller RVE size for validity.

Here we perform a model calculation to give some insight into the order of improvement one might expect. As a proxy for any of the effects that would lead to larger RVE size, we shall simply insist on greater accuracy of our constitutive equations for the spherical inclusions case. Specifically, for an accuracy of 0.5%, i.e., $\alpha = 0.005$, we shall calculate the minimum RVE size for applicability of Eq. (91), using the accurate procedure just employed, and we shall compare the results to the minimum RVE size for applicability of Drugan and Willis' leading-order nonlocal constitutive equation. The latter will be computed by determining the average strain wavelength at which the last term in Eq. (80) makes a 0.5% correction to the sum of the other right-side terms. These calculations will be carried out for the same simple strain state analyzed above. Thus, Eq. (93) applies, but Eq. (94) is replaced by:

$$\left|\frac{16\pi^4}{l^4}\hat{L}^{(2)}_{11111111}\right| = \alpha\left|\hat{L}_{1111} - \frac{4\pi^2}{l^2}\hat{L}^{(1)}_{111111}\right|. \tag{96}$$

Results for voids and rigid inclusions are presented in Table 2, which shows that use of a nonlocal constitutive equation can make a dramatic reduction in the minimum RVE size: more than a factor of 4 in some cases. We re-emphasize that all the calculations performed here have incorporated information up through two-point statistics.

Acknowledgements

Support of this work by a Vilas Associate Research Award from the University of Wisconsin-Madison is gratefully acknowledged.

Appendix A. Derivation of $\mathbf{\Gamma}_{,mnop}(\mathbf{0})$ for isotropic distributions of phases

As shown in Section 3, our higher-order nonlocal constitutive equation requires determination of the fourth derivative of $\mathbf{\Gamma}$ with respect to $\boldsymbol{\xi}$, evaluated at $\boldsymbol{\xi} = \mathbf{0}$. By application of the commutative property of convolutions and the fact that the phase *distribution* is isotropic, we have from Eq. (21):

$$\frac{\partial^4 \mathbf{\Gamma}(\boldsymbol{\xi})}{\partial \xi_m \partial \xi_n \partial \xi_o \partial \xi_p} = \frac{1}{8\pi^3} \int \tilde{\mathbf{\Gamma}}_0(\boldsymbol{\xi}') \frac{\partial^4 \tilde{h}(|\boldsymbol{\xi} - \boldsymbol{\xi}'|)}{\partial \xi_m \partial \xi_n \partial \xi_o \partial \xi_p} \, \mathrm{d}\boldsymbol{\xi}'. \tag{A1}$$

Computing the fourth derivative of $\tilde{h}(|\boldsymbol{\xi} - \boldsymbol{\xi}'|)$ and then setting $\boldsymbol{\xi} = \mathbf{0}$, Eq. (A1) becomes:

$$\begin{aligned}
\mathbf{\Gamma}_{,mnop}(\mathbf{0}) = \frac{1}{8\pi^3} \int \tilde{\mathbf{\Gamma}}_0(\boldsymbol{\xi}) \Bigg\{ & 3\delta_{mn}\delta_{op} \left[\frac{\tilde{h}''(|\boldsymbol{\xi}|)}{|\boldsymbol{\xi}|^2} - \frac{\tilde{h}'(|\boldsymbol{\xi}|)}{|\boldsymbol{\xi}|^3} \right] \\
& + 6 \frac{\delta_{mn}\xi_o\xi_p}{|\boldsymbol{\xi}|^2} \left[\frac{\tilde{h}'''(|\boldsymbol{\xi}|)}{|\boldsymbol{\xi}|} - \frac{\tilde{h}''(|\boldsymbol{\xi}|)}{|\boldsymbol{\xi}|^2} + 3 \frac{\tilde{h}'(|\boldsymbol{\xi}|)}{|\boldsymbol{\xi}|^3} \right] \\
& + \frac{\xi_m\xi_n\xi_o\xi_p}{|\boldsymbol{\xi}|^4} \Bigg[\tilde{h}^{iv}(|\boldsymbol{\xi}|) - 6 \frac{\tilde{h}'''(|\boldsymbol{\xi}|)}{|\boldsymbol{\xi}|} + 15 \frac{\tilde{h}''(|\boldsymbol{\xi}|)}{|\boldsymbol{\xi}|^2} \\
& - 15 \frac{\tilde{h}'(|\boldsymbol{\xi}|)}{|\boldsymbol{\xi}|^3} \Bigg] \Bigg\} \, \mathrm{d}\boldsymbol{\xi} \Bigg|_{(mnop)}.
\end{aligned} \tag{A2}$$

Since $\tilde{\mathbf{\Gamma}}_0(\boldsymbol{\xi})$ is homogeneous of degree zero, each of the three major integrand terms can be multiplicatively separated into radial and angular portions of their spherical full-space integrals, so that Eq. (A2) may be rewritten as, with $\rho = |\boldsymbol{\xi}|$:

$$\begin{aligned}
8\pi^3 \mathbf{\Gamma}_{,mnop}(\mathbf{0}) = {} & 3\left[\delta_{mn}\delta_{op}\right]_{(mnop)} \int_{|\boldsymbol{\xi}|=\mathbf{1}} \tilde{\mathbf{\Gamma}}_0(\boldsymbol{\xi}) \, \mathrm{d}S \left\{ \int_0^\infty \left[\frac{\tilde{h}''(\rho)}{\rho^2} - \frac{\tilde{h}'(\rho)}{\rho^3} \right] \rho^2 \, \mathrm{d}\rho \right\} \\
& + 6 \int_{|\boldsymbol{\xi}|=\mathbf{1}} \tilde{\mathbf{\Gamma}}_0(\boldsymbol{\xi}) \left[\delta_{mn}\xi_o\xi_p\right]_{(mnop)} \mathrm{d}S \left\{ \int_0^\infty \left[\frac{\tilde{h}'''(\rho)}{\rho} - 3\frac{\tilde{h}''(\rho)}{\rho^2} + 3\frac{\tilde{h}'(\rho)}{\rho^3} \right] \rho^2 \, \mathrm{d}\rho \right\} \\
& + \int_{|\boldsymbol{\xi}|=\mathbf{1}} \tilde{\mathbf{\Gamma}}_0(\boldsymbol{\xi}) \xi_m\xi_n\xi_o\xi_p \, \mathrm{d}S \left\{ \int_0^\infty \left[\tilde{h}^{iv}(\rho) - 6\frac{\tilde{h}'''(\rho)}{\rho} + 15\frac{\tilde{h}''(\rho)}{\rho^2} - 15\frac{\tilde{h}'(\rho)}{\rho^3} \right] \rho^2 \, \mathrm{d}\rho \right\}.
\end{aligned} \tag{A3}$$

Let us evaluate the radial integrals appearing in Eq. (A3). Observe first that

$$\int_0^\infty \frac{\tilde{h}'(\rho)}{\rho}\,\mathrm{d}\rho = \int_0^\infty \frac{\tilde{h}'(\rho)}{\rho^3}\rho^2\,\mathrm{d}\rho = \frac{1}{4\pi}\int \frac{\tilde{h}'(|\boldsymbol{\xi}|)}{|\boldsymbol{\xi}|^3}\,\mathrm{d}\boldsymbol{\xi} = \frac{1}{4\pi}\int\left(\frac{\boldsymbol{\xi}}{|\boldsymbol{\xi}|}\bullet\nabla\tilde{h}\right)\frac{d\boldsymbol{\xi}}{|\boldsymbol{\xi}|^3}$$

$$= 2\pi^2\left[\frac{1}{8\pi^3}\int \frac{\xi_j(\partial\tilde{h}/\partial\xi_j)}{|\boldsymbol{\xi}|^4}\mathrm{e}^{-i\boldsymbol{\xi}\bullet\mathbf{x}}\,\mathrm{d}\boldsymbol{\xi}\right]_{\mathbf{x}=\mathbf{0}}. \tag{A4}$$

To determine the bracketed term, notice that

$$\frac{\partial^4}{\partial x_k\partial x_k\partial x_l\partial x_l}\left[\frac{1}{8\pi^3}\int \frac{\xi_j\left(\partial\tilde{h}/\partial\xi_j\right)}{|\boldsymbol{\xi}|^4}\mathrm{e}^{-i\boldsymbol{\xi}\bullet\mathbf{x}}\,\mathrm{d}\boldsymbol{\xi}\right] = \frac{1}{8\pi^3}\int \xi_j\frac{\partial\tilde{h}}{\partial\xi_j}\mathrm{e}^{-i\boldsymbol{\xi}\bullet\mathbf{x}}\,\mathrm{d}\boldsymbol{\xi}$$

$$= i\frac{\partial}{\partial x_j}\left[\frac{1}{8\pi^3}\int \frac{\partial\tilde{h}}{\partial\xi_j}\mathrm{e}^{-i\boldsymbol{\xi}\bullet\mathbf{x}}\,\mathrm{d}\boldsymbol{\xi}\right] = i\frac{\partial}{\partial x_j}\left[ix_jh(|\mathbf{x}|)\right] = -3h(|\mathbf{x}|) - |\mathbf{x}|h'(|\mathbf{x}|). \tag{A5}$$

Thus, the desired inverse Fourier transform — the bracketed term in Eq. (A5), which we abbreviate by Φ — is given by the solution to the inhomogeneous biharmonic equation:

$$\nabla^4\Phi = -3h(|\mathbf{x}|) - |\mathbf{x}|h'(|\mathbf{x}|). \tag{A6}$$

To solve this, we first find the Green's function for the inhomogeneous biharmonic, i.e., the solution to:

$$\nabla^4\Phi = \delta(\mathbf{x}), \tag{A7}$$

where $\delta(\mathbf{x})$ is the 3D Dirac delta. This is obtained by noting that this Green's function will be spherically symmetric, and by recalling the Green's function for the Poisson equation; Eq. (A7) thus becomes, using $r = |\mathbf{x}|$:

$$\nabla^2(\nabla^2\Phi) = \delta(\mathbf{x}) \Longrightarrow \nabla^2\Phi = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial\Phi}{\partial r}\right) = -\frac{1}{4\pi r} \Longrightarrow \Phi = -\frac{r}{8\pi}. \tag{A8}$$

Thus, using the Green's function obtained as the last of Eq. (A8), the solution to Eq. (A6) is, recalling that Φ stood for the bracketed term in Eq. (A5):

$$\frac{1}{8\pi^3}\int \frac{\xi_j(\partial\tilde{h}/\partial\xi_j)}{|\boldsymbol{\xi}|^4}\mathrm{e}^{-i\boldsymbol{\xi}\bullet\mathbf{x}}\,\mathrm{d}\boldsymbol{\xi} = \int\left[3h\left(|\mathbf{x}'|\right) + |\mathbf{x}'|h'\left(|\mathbf{x}'|\right)\right]\frac{|\mathbf{x}-\mathbf{x}'|}{8\pi}\,\mathrm{d}\mathbf{x}'. \tag{A9}$$

Using this in Eq. (A4) and performing an integration by parts on the result gives finally

$$\int_0^\infty \frac{\tilde{h}'(\rho)}{\rho}\,\mathrm{d}\rho = -\pi^2\int_0^\infty h(r)r^3\,\mathrm{d}r. \tag{A10}$$

To find the integral of $\tilde{h}''(\rho)$ that appears in Eq. (A3), we use the fact that the 3D Laplacian of $\tilde{h}(\rho)$ is

$$\frac{\partial^2 \tilde{h}}{\partial \xi_k \partial \xi_k} = \nabla^2 \tilde{h}(\rho) = \tilde{h}''(\rho) + \frac{2}{\rho}\tilde{h}'(\rho), \tag{A11}$$

so that

$$\begin{aligned}
&\int_0^\infty \left[\tilde{h}''(\rho) + \frac{2}{\rho}\tilde{h}'(\rho)\right]\frac{\rho^2}{\rho^2}\,\mathrm{d}\rho \\
&\quad = \frac{1}{4\pi}\int \frac{\partial^2 \tilde{h}(|\boldsymbol{\xi}|)}{\partial \xi_k \partial \xi_k}\frac{1}{|\boldsymbol{\xi}|^2}\,\mathrm{d}\boldsymbol{\xi} = 2\pi^2\left[\frac{1}{8\pi^3}\int \frac{\partial^2 \tilde{h}(|\boldsymbol{\xi}|)}{\partial \xi_k \partial \xi_k}\frac{1}{|\boldsymbol{\xi}|^2}\mathrm{e}^{-i\boldsymbol{\xi}\bullet\mathbf{x}}\,\mathrm{d}\boldsymbol{\xi}\right]_{\mathbf{x}=\mathbf{0}} \\
&\quad = 2\pi^2\left[\int \frac{-x_k' x_k' h(|\mathbf{x}'|)}{4\pi|\mathbf{x}-\mathbf{x}'|}\,\mathrm{d}\mathbf{x}'\right]_{\mathbf{x}=\mathbf{0}} = -2\pi^2\int_0^\infty h(r) r^3\,\mathrm{d}r.
\end{aligned} \tag{A12}$$

Here we have evaluated the inverse Fourier transform by noting that it is the convolution of the inverse transforms of the two product terms, each of which was inverted by Drugan and Willis (1996). Comparing Eq. (A12) with twice Eq. (A10) shows that

$$\int_0^\infty \tilde{h}''(\rho)\,\mathrm{d}\rho = 0. \tag{A13}$$

The remaining two radial integrals needed for Eq. (A3) can be easily evaluated by integration by parts and use of Eq. (A13); the results are:

$$\int_0^\infty \tilde{h}'''(\rho)\rho\,\mathrm{d}\rho = 0, \qquad \int_0^\infty \tilde{h}^{iv}(\rho)\rho^2\,\mathrm{d}\rho = 0. \tag{A14}$$

Thus, Eq. (A3) has simplified to the result reported in Eq. (40).

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