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Two exact micromechanics-based nonlocal constitutive equations for random linear elastic composite materials

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Abstract

A Hashin–Shtrikman–Willis variational principle is employed to derive two exact micromechanics-based nonlocal constitutive equations relating ensemble averages of stress and strain for two-phase, and also many types of multi-phase, random linear elastic composite materials. By exact is meant that the constitutive equations employ the complete spatially-varying ensemble-average strain field, not gradient approximations to it as were employed in the previous, related work of Drugan and Willis (J. Mech. Phys. Solids 44 (1996) 497) and Drugan (J. Mech. Phys. Solids 48 (2000) 1359) (and in other, more phenomenological works). Thus, the nonlocal constitutive equations obtained here are valid for arbitrary ensemble-average strain fields, not restricted to slowly-varying ones as is the case for gradient-approximate nonlocal constitutive equations. One approach presented shows how to solve the integral equations arising from the variational principle directly and exactly, for a special, physically reasonable choice of the homogeneous comparison material. The resulting nonlocal constitutive equation is applicable to composites of arbitrary anisotropy, and arbitrary phase contrast and volume fraction. One exact nonlocal constitutive equation derived using this approach is valid for two-phase composites having any statistically uniform distribution of phases, accounting for up through two-point statistics and arbitrary phase shape. It is also shown that the same approach can be used to derive exact nonlocal constitutive equations for a large class of composites comprised of more than two phases, still permitting arbitrary elastic anisotropy. The second approach presented employs three-dimensional Fourier transforms, resulting in a nonlocal constitutive equation valid for arbitrary choices of the comparison modulus for isotropic composites. This approach is based on use of the general representation of an isotropic fourth-rank tensor function of a vector variable, and its inverse. The exact nonlocal constitutive equations derived from these two approaches are applied to some example

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cases, directly rationalizing some recently-obtained numerical simulation results and assessing the accuracy of previous results based on gradient-approximate nonlocal constitutive equations.

Keywords: Nonlocal constitutive equations; A. Voids and inclusions; B. Constitutive behavior; Inhomogeneous material; C. Variational calculus

1. Introduction

When a structural component comprised of an elastic composite material is large compared to the microstructural size scale of the composite, and when the geometry of the component and the applied loading on the component vary sufficiently slowly with position compared to this size scale, it is often sufficient to idealize the composite material as being homogeneous, with constant macroscopic or "effective" properties. However, when these conditions are not met, more sophisticated constitutive modeling is required to capture the actual material response.

Drugan and Willis (1996) employed Willis' (1977, 1982, 1983) generalization of a Hashin and Shtrikman (1962, 1963) variational principle to derive a micromechanics-based nonlocal constitutive equation to treat cases in which the macroscopic elastic fields vary more rapidly with position than can be adequately treated by the standard constant-effective-modulus constitutive equation. They considered an ensemble of random linear elastic composite materials having infinite extent, and derived a constitutive equation that corrects the standard one relating ensemble-average stress to ensemble-average strain by the addition of a gradient term in the ensemble-average strain. Drugan (2000) extended their results to include two strain gradient terms. In both cases, the ensemble-average strain field was assumed to vary sufficiently slowly to render sensible a Taylor expansion of this field, thus permitting approximate solution of the integral equations arising from the variational principle.

In the present work we also consider an ensemble of infinite random linear elastic composite materials characterized by the Hashin–Shtrikman–Willis variational principle. Now, however, we present two new methods to derive *exact* micromechanics-based nonlocal constitutive equations, meaning that the integral equations arising from the variational principle are solved exactly for arbitrarily-varying ensemble-average strain fields, so the resulting nonlocal constitutive equations are in terms of the full ensemble-average strain field and not a gradient approximation to it. The first method involves making a special, physically sensible choice of the constant "comparison" modulus tensor that arises in the variational principle; we show that for this choice, the integral equations arising from the variational principle can be solved directly. The resulting exact nonlocal constitutive equation is quite compact and rather simple, rendering it useful for anisotropic and complex composite materials; it applies to two-phase composites, and also to a rather broad class of multi-phase composites. The second method is restricted, at present, to composites having overall isotropic response, but it permits arbitrary choice of the (isotropic) comparison modulus tensor. The integral equations arising from the variational principle are solved exactly by means of three-dimensional

Fourier transforms, with their inversion being assisted by use of the general representation of an isotropic fourth-rank tensor function of a vector variable and its inverse.

The paper is organized as follows. Section 2 lays out the formulation for deriving constitutive response of an ensemble of random linear elastic composite materials and reviews the Hashin–Shtrikman–Willis variational principle. Section 3 shows the first new method of deriving an exact nonlocal constitutive equation, based on a specific choice of the comparison modulus tensor: first for a two-phase composite, then for a large class of multi-phase composites. Section 4 shows the second new method for deriving an exact nonlocal constitutive equation, for isotropic elastic composite materials but otherwise arbitrary choice of the comparison modulus tensor. Section 5 derives some needed results for application of the new nonlocal constitutive equations: namely, the general representation of an isotropic fourth-rank tensor function of a vector variable, the inverse of this function, and the evaluation of two such functions that arise in the nonlocal constitutive equations. Finally, Section 6 shows specific applications of the new nonlocal constitutive equations and makes comparisons of the results with recent numerical simulations of random elastic composite materials by Segurado and Llorca (2002), and with the predictions of the gradient-approximate nonlocal constitutive equation of Drugan and Willis (1996) as improved by Monetto and Drugan (2003).

It bears emphasis that although the exact nonlocal constitutive equations derived here are specifically for the case of infinite-body composite materials, facilitating use of the infinite-body Green's function in the variational principle, all of the ideas presented for deriving the exact nonlocal constitutive equation based on a special choice of the comparison modulus tensor (Section 3) go through in other cases for which the Hashin–Shtrikman–Willis variational principle applies and elastic comparison material Green's functions exist.

2. General formulation

We consider random linear elastic composite materials with firmly-bonded phases which may have arbitrary anisotropy, arbitrary contrast 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 $\mathbf{x}$. However, the key ideas to be presented here are equally applicable in non-infinite-body contexts, as will be detailed in future work.

The governing equations for quasi-statically applied body force fields in a specific composite sample α are equilibrium, geometrical compatibility and constitutive:

$$\nabla \bullet \boldsymbol{\sigma} + \mathbf{f} = \mathbf{0}, \quad \mathbf{e} = \mathrm{sym}(\nabla \mathbf{u}), \quad \boldsymbol{\sigma} = \mathbf{L}\mathbf{e}, \tag{1}$$

where $\boldsymbol{\sigma}(\mathbf{x},\alpha)$ and $\mathbf{e}(\mathbf{x},\alpha)$ are the stress and infinitesimal strain tensor fields, $\mathbf{u}(\mathbf{x},\alpha)$ is the displacement vector field, $\mathbf{L}(\mathbf{x},\alpha)$ is the fourth-rank elastic modulus tensor field, all in composite sample α, and $\mathrm{sym}(\nabla \mathbf{u})$ denotes the symmetric part of the displacement gradient. The applied body force vector field $\mathbf{f}(\mathbf{x})$ is the same in every composite sample. Here and throughout the manuscript, we employ abbreviated symbolic notation so that, for example, in index notation with summation convention, the last equation in Eq. (1) reads: $\sigma_{ij} = L_{ijkl}e_{kl}$.

Following Hashin and Shtrikman (1962, 1963), it is useful to reformulate system (1) in terms of a *homogeneous* "comparison" medium having elastic modulus tensor $\mathbf{L}_0$ (independent of $\mathbf{x}$ and α) so that

$$\boldsymbol{\sigma}(\mathbf{x},\alpha) = \mathbf{L}_0\mathbf{e}(\mathbf{x},\alpha) + \boldsymbol{\tau}(\mathbf{x},\alpha), \qquad \boldsymbol{\tau}(\mathbf{x},\alpha) \equiv [\mathbf{L}(\mathbf{x},\alpha) - \mathbf{L}_0]\mathbf{e}(\mathbf{x},\alpha), \tag{2}$$

where $\boldsymbol{\tau}(\mathbf{x},\alpha)$ is the "stress polarization" tensor field. Willis' (1977) derivation of the solution to system (1) for prescribed $\mathbf{f}(\mathbf{x})$, in terms of the Hashin–Shtrikman variational principle, shows that the strain field solution is

$$\mathbf{e}(\mathbf{x},\alpha) = \mathbf{e}_0(\mathbf{x}) - \int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')\boldsymbol{\tau}(\mathbf{x}',\alpha)\,\mathrm{d}\mathbf{x}', \tag{3}$$

where $\mathbf{e}_0(\mathbf{x})$ is the solution to the same applied $\mathbf{f}(\mathbf{x})$ in the homogeneous comparison body. Here we have defined

$$[\boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')]_{ijkl} \equiv \left.\frac{\partial^2[\mathbf{G}_0(\mathbf{x}-\mathbf{x}')]_{jk}}{\partial x_i \partial x'_l}\right|_{(ij),(kl)}, \tag{4}$$

with $\mathbf{G}_0(\mathbf{x})$ being the infinite-homogeneous-body Green's function for the comparison material and the notation indicating symmetrization on subscripts ij and kl. The tensor field $\boldsymbol{\tau}(\mathbf{x},\alpha)$ appearing in Eq. (3) satisfies the Hashin–Shtrikman variational principle:

$$\delta \int \boldsymbol{\tau}(\mathbf{x},\alpha)\left\{[\mathbf{L}(\mathbf{x},\alpha)-\mathbf{L}_0]^{-1}\boldsymbol{\tau}(\mathbf{x},\alpha) + \int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')\boldsymbol{\tau}(\mathbf{x}',\alpha)\,\mathrm{d}\mathbf{x}' - 2\mathbf{e}_0(\mathbf{x})\right\}\mathrm{d}\mathbf{x} = 0. \tag{5}$$

Willis (1982, 1983) recast the Hashin–Shtrikman formulation for a specific composite sample in terms of ensemble averages for random composites. Let α denote, as above, an individual member of a sample space $\mathscr{S}$ of composite realizations, define by $p(\alpha)$ the probability density of α in $\mathscr{S}$, 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

$$P_r(\mathbf{x}) = \langle \chi_r(\mathbf{x},\alpha)\rangle \equiv \int_{\mathscr{S}} \chi_r(\mathbf{x},\alpha)p(\alpha)\,\mathrm{d}\alpha, \tag{6}$$

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

$$P_{rs}(\mathbf{x},\mathbf{x}') = \langle \chi_r(\mathbf{x},\alpha)\chi_s(\mathbf{x}',\alpha)\rangle \equiv \int_{\mathscr{S}} \chi_r(\mathbf{x},\alpha)\chi_s(\mathbf{x}',\alpha)p(\alpha)\,\mathrm{d}\alpha. \tag{7}$$

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 $\mathbf{L}(\mathbf{x},\alpha)$ of composite sample α, and its ensemble average, are

$$\mathbf{L}(\mathbf{x},\alpha)=\sum_{r=1}^{n}\mathbf{L}_r\chi_r(\mathbf{x},\alpha) \quad\Rightarrow\quad \langle\mathbf{L}(\mathbf{x},\alpha)\rangle=\sum_{r=1}^{n}\mathbf{L}_rP_r(\mathbf{x}). \tag{8}$$

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 the stress polarization tensor field that allow for up through two-point correlations:

$$\boldsymbol{\tau}(\mathbf{x},\alpha)=\sum_{r=1}^{n}\boldsymbol{\tau}_r(\mathbf{x})\chi_r(\mathbf{x},\alpha). \tag{9}$$

[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 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, the probabilities become translation-invariant, so that $P_r(\mathbf{x})$ 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 the previous equations, Willis (1982, 1983) has shown that one obtains the following variational principle for $\boldsymbol{\tau}_r(\mathbf{x})$:

$$\delta\left\{\sum_{r=1}^{n}c_r\int\boldsymbol{\tau}_r(\mathbf{x})[(\mathbf{L}_r-\mathbf{L}_0)^{-1}\boldsymbol{\tau}_r(\mathbf{x})-2\mathbf{e}_0(\mathbf{x})]\,\mathrm{d}\mathbf{x}\right.$$
$$\left.+\sum_{r=1}^{n}\sum_{s=1}^{n}\int\boldsymbol{\tau}_r(\mathbf{x})\left[\int\boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')\boldsymbol{\tau}_s(\mathbf{x}')P_{rs}(\mathbf{x}-\mathbf{x}')\,\mathrm{d}\mathbf{x}'\right]\mathrm{d}\mathbf{x}\right\}=0. \tag{10}$$

Principle (10) is stationary when [substituting for $\mathbf{e}_0(\mathbf{x})$ from the result of putting Eq. (9) into Eq. (3) and ensemble-averaging]

$$(\mathbf{L}_r-\mathbf{L}_0)^{-1}\boldsymbol{\tau}_r(\mathbf{x})c_r+\sum_{s=1}^{n}\int\boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')[P_{rs}(\mathbf{x}-\mathbf{x}')-c_rc_s]\boldsymbol{\tau}_s(\mathbf{x}')\,\mathrm{d}\mathbf{x}'=c_r\langle\mathbf{e}\rangle(\mathbf{x}),$$
$$r=1,2,\ldots,n, \tag{11}$$

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 ensemble-averaging (9):

$$\langle\boldsymbol{\tau}\rangle(\mathbf{x})=\sum_{r=1}^{n}c_r\boldsymbol{\tau}_r(\mathbf{x}). \tag{12}$$

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. (12) into the ensemble average of the first of Eq. (2):

$$\langle\boldsymbol{\sigma}\rangle(\mathbf{x}) = \mathbf{L}_0\langle\mathbf{e}\rangle(\mathbf{x}) + \langle\boldsymbol{\tau}\rangle(\mathbf{x}). \tag{13}$$

Observe from Eqs. (11) to (13) that this is a *nonlocal* constitutive equation.

3. Exact solution for nonlocal constitutive equation for a specific choice of comparison material

Drugan and Willis (1996) and Drugan (2000) employed three-dimensional Fourier transforms to solve Eq. (11) for two-phase composites [in which case it simplifies to Eq. (15) below], and avoided the difficult Fourier transform inversion by considering slowly-varying ensemble-average strain fields that admit a Taylor expansion. Their results were therefore approximate nonlocal constitutive equations in terms of strain gradients.

Here we show that an *exact* solution of the nonlocal constitutive equation, within the Hashin–Shtrikman–Willis variational formulation as detailed above, can be found for a specific choice of the comparison modulus tensor. This exact nonlocal equation involves the full ensemble-average strain field, not gradient approximations to it, and hence is valid for arbitrarily-varying ensemble-average strain fields. We show this first for two-phase composites, and then show how to generalize the analysis to treat a broad class of composites having an arbitrary number of phases.

3.1. Two-phase composites

We first specialize to the practically important class of two-phase composites, and employ our assumptions of statistical uniformity and ergodicity. Then the two-point probabilities can be expressed as (see Willis, 1982)

$$P_{rs}(\mathbf{x}-\mathbf{x}') - c_r c_s = c_r(\delta_{rs} - c_s)h(\mathbf{x}-\mathbf{x}'), \quad \text{(no sum on indices)}, \tag{14}$$

where $h(\mathbf{x}-\mathbf{x}')$ is the two-point correlation function, defined e.g. by the 12 component of Eq. (14), and δ_{rs} is the Kronecker delta. Using Eq. (14) in Eq. (11) and dividing through by c_r gives

$$(\mathbf{L}_r - \mathbf{L}_0)^{-1}\boldsymbol{\tau}_r(\mathbf{x}) + \sum_{s=1}^{2}(\delta_{rs} - c_s)\int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')h(\mathbf{x}-\mathbf{x}')\boldsymbol{\tau}_s(\mathbf{x}')\,\mathrm{d}\mathbf{x}' = \langle\mathbf{e}\rangle(\mathbf{x}), \quad r = 1, 2. \tag{15}$$

First, write out Eq. (15) for each of $r = 1, 2$, then multiply the first by $\delta\mathbf{L}_1$ and the second by $\delta\mathbf{L}_2$, having defined $\delta\mathbf{L}_r = (\mathbf{L}_r - \mathbf{L}_0)$:

$$\boldsymbol{\tau}_1(\mathbf{x}) + c_2\delta\mathbf{L}_1\int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')h(\mathbf{x}-\mathbf{x}')[\boldsymbol{\tau}_1(\mathbf{x}') - \boldsymbol{\tau}_2(\mathbf{x}')]\,\mathrm{d}\mathbf{x}' = \delta\mathbf{L}_1\langle\mathbf{e}\rangle(\mathbf{x}), \tag{16a}$$

$$\boldsymbol{\tau}_2(\mathbf{x}) - c_1\delta\mathbf{L}_2\int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')h(\mathbf{x}-\mathbf{x}')[\boldsymbol{\tau}_1(\mathbf{x}') - \boldsymbol{\tau}_2(\mathbf{x}')]\,\mathrm{d}\mathbf{x}' = \delta\mathbf{L}_2\langle\mathbf{e}\rangle(\mathbf{x}). \tag{16b}$$

Now subtract Eq. (16b) from Eq. (16a), noting that the integrals appearing in each are identical:

$$\boldsymbol{\tau}_1(\mathbf{x}) - \boldsymbol{\tau}_2(\mathbf{x}) + (c_1\delta\mathbf{L}_2 + c_2\delta\mathbf{L}_1)\int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')h(\mathbf{x}-\mathbf{x}')[\boldsymbol{\tau}_1(\mathbf{x}') - \boldsymbol{\tau}_2(\mathbf{x}')]\,\mathrm{d}\mathbf{x}'$$
$$= (\mathbf{L}_1 - \mathbf{L}_2)\langle\mathbf{e}\rangle(\mathbf{x}). \tag{17}$$

We must solve this integral equation for the quantity $[\boldsymbol{\tau}_1(\mathbf{x}) - \boldsymbol{\tau}_2(\mathbf{x})]$; then Eq. (16) will give the solutions for the $\boldsymbol{\tau}_r(\mathbf{x})$.

Observe that an exact solution to Eq. (17) is immediately possible if we make the following choice for the comparison modulus tensor

$$\mathbf{L}_0 = c_1\mathbf{L}_2 + c_2\mathbf{L}_1, \tag{18}$$

since with this choice the term in parentheses that multiplies the integral in Eq. (17) vanishes, so that Eq. (17) reduces to

$$\boldsymbol{\tau}_1(\mathbf{x}) - \boldsymbol{\tau}_2(\mathbf{x}) = (\mathbf{L}_1 - \mathbf{L}_2)\langle\mathbf{e}\rangle(\mathbf{x}). \tag{19}$$

We then substitute Eqs. (18) and (19) into Eq. (16) to obtain the following solutions:

$$\boldsymbol{\tau}_1(\mathbf{x}) = c_1(\mathbf{L}_1 - \mathbf{L}_2)\langle\mathbf{e}\rangle(\mathbf{x}) - c_1c_2(\mathbf{L}_1 - \mathbf{L}_2)\int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')h(\mathbf{x}-\mathbf{x}')(\mathbf{L}_1 - \mathbf{L}_2)\langle\mathbf{e}\rangle(\mathbf{x}')\,\mathrm{d}\mathbf{x}', \tag{20a}$$

$$\boldsymbol{\tau}_2(\mathbf{x}) = -c_2(\mathbf{L}_1 - \mathbf{L}_2)\langle\mathbf{e}\rangle(\mathbf{x}) - c_1c_2(\mathbf{L}_1 - \mathbf{L}_2)\int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')h(\mathbf{x}-\mathbf{x}')(\mathbf{L}_1 - \mathbf{L}_2)\langle\mathbf{e}\rangle(\mathbf{x}')\,\mathrm{d}\mathbf{x}'. \tag{20b}$$

Substituting Eq. (20) into Eq. (12) gives

$$\langle\boldsymbol{\tau}\rangle(\mathbf{x}) = (\mathbf{L}_1 - \mathbf{L}_2)\Bigg[(c_1 - c_2)\langle\mathbf{e}\rangle(\mathbf{x})$$
$$- c_1c_2\int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')h(\mathbf{x}-\mathbf{x}')\,(\mathbf{L}_1 - \mathbf{L}_2)\langle\mathbf{e}\rangle(\mathbf{x}')\,\mathrm{d}\mathbf{x}'\Bigg], \tag{21}$$

so that from Eq. (13) the exact nonlocal constitutive equation is

$$\boxed{\langle\boldsymbol{\sigma}\rangle(\mathbf{x}) = [c_1\mathbf{L}_1 + c_2\mathbf{L}_2]\langle\mathbf{e}\rangle(\mathbf{x}) - c_1c_2(\mathbf{L}_1 - \mathbf{L}_2)\int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')h(\mathbf{x}-\mathbf{x}')(\mathbf{L}_1 - \mathbf{L}_2)\langle\mathbf{e}\rangle(\mathbf{x}')\,\mathrm{d}\mathbf{x}'.} \tag{22}$$

This equation is explicit and quite condensed, making it useful for complex and anisotropic composites as well as isotropic composites (for which a specific illustration will be provided later). It is valid for arbitrary anisotropies, shapes and volume fractions of the phases, and arbitrary two-point correlation functions $h(\mathbf{x}-\mathbf{x}')$, so long as statistical uniformity and ergodicity are satisfied. Note that the integral term in Eq. (22) contains contributions to both the local and nonlocal portions of the constitutive equation: for example, in cases of constant ensemble-average strain, Eq. (22) reduces to the (local) Hashin–Shtrikman estimate of the ensemble-average constitutive equation for choice (18) of the comparison modulus.

The specific choice (18) of the comparison modulus tensor needed to obtain the exact result (22) is physically reasonable: for all values of the phase volume fractions, choice (18) either equals or lies between the moduli of the phases.

Furthermore, we now show that the result (22), which we emphasize is an *exact solution* of Eq. (15) with Eqs. (12) and (13) for the specific Eq. (18) of the comparison modulus tensor, agrees exactly through second order in phase contrast $(\mathbf{L}_1 - \mathbf{L}_2)$ to the iterative solution to Eqs. (15), (12) and (13) for small phase contrast, for *arbitrary* choice of the comparison modulus tensor. To see this, observe first that the leading-order solution of Eq. (17) in $(\mathbf{L}_1 - \mathbf{L}_2)$, for arbitrary $\mathbf{L}_0$, is precisely (19). To this end, notice that the integral coefficient, $(c_1\delta\mathbf{L}_2 + c_2\delta\mathbf{L}_1)$, is $O(\mathbf{L}_1 - \mathbf{L}_2)$. [For example, for any choice of $\mathbf{L}_0$ of the form $\mathbf{L}_0 = \lambda\mathbf{L}_1 + (1-\lambda)\mathbf{L}_2$, $0 \leqslant \lambda \leqslant 1$, the integral coefficient $(c_1\delta\mathbf{L}_2 + c_2\delta\mathbf{L}_1) = (c_2 - \lambda)(\mathbf{L}_1 - \mathbf{L}_2)$.] Substitution of Eq. (19) into Eq. (16), and then the results of Eq. (16) into Eq. (12), gives, through second order in $(\mathbf{L}_1 - \mathbf{L}_2)$ for arbitrary $\mathbf{L}_0$

$$\begin{aligned}\langle\boldsymbol{\tau}\rangle(\mathbf{x}) &= (c_1\mathbf{L}_1 + c_2\mathbf{L}_2 - \mathbf{L}_0)\langle\mathbf{e}\rangle(\mathbf{x}) \\ &\quad - c_1c_2(\mathbf{L}_1 - \mathbf{L}_2)\int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')h(\mathbf{x}-\mathbf{x}')(\mathbf{L}_1 - \mathbf{L}_2)\langle\mathbf{e}\rangle(\mathbf{x}')\,\mathrm{d}\mathbf{x}',\end{aligned} \tag{23}$$

and substitution of this into Eq. (13) gives precisely Eq. (22).

3.2. A class of composites having an arbitrary number of phases

The approach just illustrated for two-phase composites does not appear to be easily generalized to arbitrary types of composites having more than two phases, since the integral equations (11) for such composites would involve multiple different two-point correlation functions. However, there is at least one large class of multiple-phase composites for which the above approach *does* go through: composites having an arbitrary number of phases for which it is sensible to group the phases into two types for the purposes of describing their correlation function. For example, consider a matrix-inclusion composite in which the matrix is treated as one phase and the inclusions are treated as the second phase, but there is an arbitrary number of different types of inclusions. The condition needing to be satisfied is that even though the inclusions are different types, this fact does not affect their statistical distribution. In such cases, here is how to calculate the two-point probabilities.

We denote one phase (e.g., the matrix) by subscript 1, and the second "phase" (which is actually comprised of multiple phases; e.g., the inclusions) by subscript I. Then, employing our assumptions of statistical uniformity and ergodicity, the two-point probabilities have the same form as for a two-phase composite, which is, rewriting (14) in terms of the notation just introduced:

$$P_{11} - c_1^2 = c_1c_Ih(\mathbf{x}-\mathbf{x}') = P_{II} - c_I^2 = -(P_{1I} - c_1c_I). \tag{24}$$

Now divide the second phase group, denoted by subscript I, into $(n-1)$ phases, denoted by subscripts 2 through n, such that among these $(n-1)$ phases *only*, the kth phase has the one-point probability p_k. The volume fraction of any phase in the overall composite is denoted by c_k. There follow the equalities:

$$c_1 + c_I = 1, \quad c_I = \sum_{k=2}^{n} c_k, \quad c_k = c_I\, p_k \ (k = 2, \ldots, n), \quad \sum_{k=2}^{n} p_k = 1. \tag{25}$$

Using Eqs. (24) and (25) we calculate:

$$P_{11} - c_1^2 = c_1 c_I h = c_1(1 - c_1)h, \tag{26a}$$

$$P_{1k} = P_{1I}\, p_k = c_1 c_I (1 - h) p_k = c_1 c_k (1 - h), \quad k = 2, \ldots, n, \tag{26b}$$

$$P_{ij} = P_{II}\, p_i p_j = (c_I^2 + c_1 c_I h) p_i p_j = c_i c_j \left(1 + \frac{c_1}{1 - c_1} h\right), \quad i, j = 2, \ldots, n. \tag{26c}$$

The results (26) can be summarized as

$$P_{rs}(\mathbf{x} - \mathbf{x}') - c_r c_s = \frac{(c_1 - \delta_{1r}) c_r (c_s - \delta_{1s})}{1 - c_1} h(\mathbf{x} - \mathbf{x}'), \quad \text{no sum on } r. \tag{27}$$

An independent check on this result can be performed by adding $c_r c_s$ to both sides and then summing on s:

$$\sum_{s=1}^{n} P_{rs}(\mathbf{x} - \mathbf{x}') = c_r \sum_{s=1}^{n} c_s + \frac{(c_1 - \delta_{1r}) c_r}{1 - c_1} h(\mathbf{x} - \mathbf{x}') \sum_{s=1}^{n} (c_s - \delta_{1s}) = c_r, \tag{28}$$

which is the correct result, having noted that the second sum in Eq. (28) equals unity, while the third equals zero.

Now, to find an exact nonlocal constitutive equation for such a material, we substitute Eq. (27) into Eq. (11), divide the result by c_r, and multiply through by $\delta\mathbf{L}_r$ to obtain, defining $\mathbf{T}(\mathbf{x} - \mathbf{x}') \equiv \boldsymbol{\Gamma}_0(\mathbf{x} - \mathbf{x}') h(\mathbf{x} - \mathbf{x}')$,

$$\boldsymbol{\tau}_r(\mathbf{x}) + \frac{(c_1 - \delta_{1r})}{1 - c_1} \delta\mathbf{L}_r \sum_{s=1}^{n} \int \mathbf{T}(\mathbf{x} - \mathbf{x}')(c_s - \delta_{1s}) \boldsymbol{\tau}_s(\mathbf{x}')\, \mathrm{d}\mathbf{x}' = \delta\mathbf{L}_r \langle \mathbf{e} \rangle(\mathbf{x}), \quad r = 1, 2, \ldots, n. \tag{29}$$

Notice that the integrand in Eq. (29) is independent of r, and hence is the same in each of the n equations; it involves the $\boldsymbol{\tau}_r(\mathbf{x})$ in the combination

$$\sum_{s=1}^{n} (c_s - \delta_{1s}) \boldsymbol{\tau}_s(\mathbf{x}) = \sum_{s=1}^{n} c_s \boldsymbol{\tau}_s(\mathbf{x}) - \boldsymbol{\tau}_1(\mathbf{x}). \tag{30}$$

Thus, generalizing the strategy employed in Section 3.1, we multiply the $r = 1$ equation of (29) by $(c_1 - 1)$, every other equation having subscript r by c_r, and then add the resulting n equations to obtain

$$\left[\sum_{r=1}^{n} c_r \boldsymbol{\tau}_r(\mathbf{x}) - \boldsymbol{\tau}_1(\mathbf{x})\right] + \frac{c_1}{1-c_1}\left[\sum_{r=1}^{n} c_r \delta\mathbf{L}_r + \frac{1-2c_1}{c_1}\,\delta\mathbf{L}_1\right] \times \int \mathbf{T}(\mathbf{x}-\mathbf{x}')\left[\sum_{s=1}^{n} c_s \boldsymbol{\tau}_s(\mathbf{x}') - \boldsymbol{\tau}_1(\mathbf{x}')\right]\mathrm{d}\mathbf{x}' = \left[\sum_{r=1}^{n} c_r \delta\mathbf{L}_r - \delta\mathbf{L}_1\right]\langle\mathbf{e}\rangle(\mathbf{x}). \quad (31)$$

The exact solution for the combination (30) of $\boldsymbol{\tau}_r(\mathbf{x})$ appearing in the integrand of Eq. (29) is obtained from Eq. (31) when the comparison modulus tensor is chosen such that the bracketed term multiplying the integral in Eq. (31) vanishes

$$\left[\sum_{r=1}^{n} c_r \delta\mathbf{L}_r + \frac{1-2c_1}{c_1}\,\delta\mathbf{L}_1\right] = 0 \;\Rightarrow\; \boxed{\mathbf{L}_0 = \frac{c_1}{1-c_1}\left[\sum_{r=1}^{n} c_r \mathbf{L}_r + \frac{1-2c_1}{c_1}\,\mathbf{L}_1\right]}. \quad (32)$$

For this choice of comparison modulus tensor, (31) gives

$$\left[\sum_{r=1}^{n} c_r \boldsymbol{\tau}_r(\mathbf{x}) - \boldsymbol{\tau}_1(\mathbf{x})\right] = \left[\sum_{r=1}^{n} c_r \mathbf{L}_r - \mathbf{L}_1\right]\langle\mathbf{e}\rangle(\mathbf{x}), \quad (33)$$

and using this in Eq. (29) gives the exact solutions for the $\boldsymbol{\tau}_r(\mathbf{x})$:

$$\boldsymbol{\tau}_r(\mathbf{x}) = (\mathbf{L}_r - \mathbf{L}_0)\left\{\langle\mathbf{e}\rangle(\mathbf{x}) - \frac{(c_1-\delta_{1r})}{1-c_1}\int \mathbf{T}(\mathbf{x}-\mathbf{x}')\left[\sum_{s=1}^{n} c_s \mathbf{L}_s - \mathbf{L}_1\right]\langle\mathbf{e}\rangle(\mathbf{x}')\,\mathrm{d}\mathbf{x}'\right\}, \quad r = 1,2,\ldots,n. \quad (34)$$

Finally, use of this in Eqs. (12) and (13) gives the exact nonlocal constitutive equation for the choice of comparison modulus tensor given in the second of Eq. (32):

$$\boxed{\langle\boldsymbol{\sigma}\rangle(\mathbf{x}) = \left[\sum_{r=1}^{n} c_r \mathbf{L}_r\right]\langle\mathbf{e}\rangle(\mathbf{x}) - \frac{c_1}{1-c_1}\left[\sum_{r=1}^{n} c_r \mathbf{L}_r - \mathbf{L}_1\right] \times \int \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')h(\mathbf{x}-\mathbf{x}')\left[\sum_{s=1}^{n} c_s \mathbf{L}_s - \mathbf{L}_1\right]\langle\mathbf{e}\rangle(\mathbf{x}')\,\mathrm{d}\mathbf{x}'.} \quad (35)$$

It is easy to verify that Eq. (35) reduces to Eq. (22) in the case of two phases.

We emphasize that the procedure employed in this section, to obtain exact nonlocal constitutive equations for two-phase and a large class of multi-phase composite materials, is essentially algebraic. Therefore, although we analyzed here infinite-body composites, the same approach will succeed for any other composite sample class for which the Hashin–Shtrikman–Willis variational principle applies and elastic comparison material Green's functions exist.

4. Exact nonlocal constitutive equation for arbitrary choice of the comparison material

Let us now consider what can be done regarding an exact nonlocal constitutive equation for *arbitrary* choice of the homogeneous comparison modulus tensor. Let us revert, for simplicity, to two-phase composites. As noted earlier, the equations determining the exact nonlocal constitutive equation within the Hashin–Shtrikman–Willis variational formulation are

$$\langle \boldsymbol{\sigma} \rangle(\mathbf{x}) = \mathbf{L}_0 \langle \mathbf{e} \rangle(\mathbf{x}) + \langle \boldsymbol{\tau} \rangle(\mathbf{x}), \qquad \langle \boldsymbol{\tau} \rangle(\mathbf{x}) = \sum_{r=1}^{2} c_r \boldsymbol{\tau}_r(\mathbf{x}), \tag{36a}$$

where the $\boldsymbol{\tau}_r(\mathbf{x})$ are obtained by solving the integral equations

$$(\mathbf{L}_r - \mathbf{L}_0)^{-1} \boldsymbol{\tau}_r(\mathbf{x}) + \sum_{s=1}^{2} (\delta_{rs} - c_s) \int [\boldsymbol{\Gamma}_0(\mathbf{x} - \mathbf{x}') h(\mathbf{x} - \mathbf{x}')] \boldsymbol{\tau}_s(\mathbf{x}')\, \mathrm{d}\mathbf{x}' = \langle \mathbf{e} \rangle(\mathbf{x}), \quad r = 1, 2. \tag{36b}$$

For a function $f(\mathbf{x})$ that decays sufficiently rapidly for convergence of Eq. (37), the three-dimensional Fourier transform and its inverse are defined as, with $i = \sqrt{-1}$

$$\tilde{f}(\boldsymbol{\xi}) = \int f(\mathbf{x}) \mathrm{e}^{\mathrm{i}\boldsymbol{\xi} \bullet \mathbf{x}}\, \mathrm{d}\mathbf{x}, \qquad f(\mathbf{x}) = \frac{1}{8\pi^3} \int \tilde{f}(\boldsymbol{\xi}) \mathrm{e}^{-\mathrm{i}\boldsymbol{\xi} \bullet \mathbf{x}}\, \mathrm{d}\boldsymbol{\xi}. \tag{37}$$

Drugan and Willis (1996) took three-dimensional Fourier transforms of Eqs. (36), and then solved for the nonlocal constitutive equation in Fourier transform space. Their result can be expressed as

$$\langle \tilde{\boldsymbol{\sigma}} \rangle(\boldsymbol{\xi}) = [\mathbf{L}_0 + \langle \tilde{\mathbf{T}} \rangle(\boldsymbol{\xi})] \langle \tilde{\mathbf{e}} \rangle(\boldsymbol{\xi}), \tag{38}$$

where they found

$$\begin{aligned} \langle \tilde{\mathbf{T}} \rangle(\boldsymbol{\xi}) = {} & c_1 \delta\mathbf{L}_1 (\boldsymbol{\Gamma}^{-1} + c_1 \delta\mathbf{L}_2 + c_2 \delta\mathbf{L}_1)^{-1} (\boldsymbol{\Gamma}^{-1} + \delta\mathbf{L}_2) \\ & + c_2 \delta\mathbf{L}_2 (\boldsymbol{\Gamma}^{-1} + c_1 \delta\mathbf{L}_2 + c_2 \delta\mathbf{L}_1)^{-1} (\boldsymbol{\Gamma}^{-1} + \delta\mathbf{L}_1), \end{aligned} \tag{39}$$

having defined

$$\boldsymbol{\Gamma} = \boldsymbol{\Gamma}(\boldsymbol{\xi}) \equiv \frac{1}{8\pi^3} \int \tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi} - \boldsymbol{\xi}') h(\boldsymbol{\xi}')\, \mathrm{d}\boldsymbol{\xi}' = \frac{1}{8\pi^3} \int \tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi}') \tilde{h}(\boldsymbol{\xi} - \boldsymbol{\xi}')\, \mathrm{d}\boldsymbol{\xi}'. \tag{40}$$

One key new step toward further progress is the recognition that Eq. (39) can be manipulated into a more condensed and revealing form, by using the index symmetries of $\boldsymbol{\Gamma}$ and the modulus tensors; the result is

$$\begin{aligned}\langle \tilde{\mathbf{T}} \rangle(\boldsymbol{\xi}) = c_1\mathbf{L}_1 + c_2\mathbf{L}_2 - \mathbf{L}_0 \\ - c_1c_2(\mathbf{L}_1 - \mathbf{L}_2)\,(\boldsymbol{\Gamma}(\boldsymbol{\xi})^{-1} + c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0)^{-1}(\mathbf{L}_1 - \mathbf{L}_2).\end{aligned} \tag{41}$$

Substituting this into Eq. (38), the full (exact) nonlocal constitutive equation in Fourier transform space is

$$\boxed{\langle \tilde{\boldsymbol{\sigma}} \rangle(\boldsymbol{\xi}) = \left[c_1\mathbf{L}_1 + c_2\mathbf{L}_2 - c_1c_2(\mathbf{L}_1 - \mathbf{L}_2)(\boldsymbol{\Gamma}(\boldsymbol{\xi})^{-1} + c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0)^{-1}(\mathbf{L}_1 - \mathbf{L}_2)\right]\langle \tilde{\mathbf{e}} \rangle(\boldsymbol{\xi}).} \tag{42}$$

This form shows clearly why the specific choice (18) for the comparison modulus tensor leads to a particularly simple exact nonlocal constitutive equation: for that choice, (42) reduces to

$$\boxed{\langle \tilde{\boldsymbol{\sigma}} \rangle(\boldsymbol{\xi}) = \left[c_1\mathbf{L}_1 + c_2\mathbf{L}_2 - c_1c_2(\mathbf{L}_1 - \mathbf{L}_2)\boldsymbol{\Gamma}(\boldsymbol{\xi})(\mathbf{L}_1 - \mathbf{L}_2)\right]\langle \tilde{\mathbf{e}} \rangle(\boldsymbol{\xi}).} \tag{43}$$

This is clearly the Fourier transform of Eq. (22).

For the general case of arbitrary comparison modulus tensor and anisotropic composites, obtaining results from Eq. (42) is complicated, while (43), or its inverse Fourier transform (22), is about as condensed and convenient as seems possible for a sensible exact nonlocal constitutive equation. We do, however, wish to obtain an exact nonlocal constitutive equation for arbitrary comparison material. This will be accomplished from Eq. (42) in the following sections for isotropic composites. The key to accomplishing this is the recognition that for isotropic composites comprised of isotropic phases, for which it is sensible to choose a comparison material that is also isotropic but otherwise arbitrary, the key quantities to be evaluated in Eq. (42), namely $\boldsymbol{\Gamma}(\boldsymbol{\xi})$ [also needed for (43)], its inverse, and $(\boldsymbol{\Gamma}(\boldsymbol{\xi})^{-1} + c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0)^{-1}$, will all be isotropic fourth-rank tensor functions of a vector variable. Thus, in the next section we will exhibit the general representation of such a tensor function, derive its inverse, and evaluate $\boldsymbol{\Gamma}(\boldsymbol{\xi})$ and $(\boldsymbol{\Gamma}(\boldsymbol{\xi})^{-1} + c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0)^{-1}$ for a sensible example choice of the two-point correlation function.

5. Representation of an isotropic fourth-rank tensor function of a vector variable and its inverse

5.1. General representation

As just noted, for isotropic composites comprised of isotropic phases, we shall need a general representation of an isotropic fourth-rank tensor function of a vector variable, for fourth-rank tensor functions that have the same index symmetries as the elastic modulus tensor. Such a function, say $\mathbf{F}(\boldsymbol{\xi})$, must satisfy

$$F_{ijkl}(\mathbf{Q}\bullet\boldsymbol{\xi}) = Q_{mi}Q_{nj}Q_{ok}Q_{pl}F_{mnop}(\boldsymbol{\xi}) \quad \text{and} \quad F_{ijkl}(\boldsymbol{\xi}) = F_{jikl}(\boldsymbol{\xi}) = F_{klij}(\boldsymbol{\xi}) \tag{44}$$

for arbitrary orthogonal second-rank tensors $\mathbf{Q}$ and arbitrary vectors $\boldsymbol{\xi}$. The most general representation of $\mathbf{F}(\boldsymbol{\xi})$ that satisfies all of Eq. (44) is

$$\begin{aligned} F_{ijkl}(\boldsymbol{\xi}) = {} & f_1(|\boldsymbol{\xi}|)\delta_{ij}\delta_{kl} + f_2(|\boldsymbol{\xi}|)\frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + f_3(|\boldsymbol{\xi}|)\frac{1}{2}(\delta_{ij}\xi_k\xi_l + \delta_{kl}\xi_i\xi_j) \\ & + f_4(|\boldsymbol{\xi}|)\frac{1}{4}(\xi_i\delta_{jk}\xi_l + \xi_j\delta_{ik}\xi_l + \xi_i\delta_{jl}\xi_k + \xi_j\delta_{il}\xi_k) + f_5(|\boldsymbol{\xi}|)\xi_i\xi_j\xi_k\xi_l, \end{aligned} \tag{45}$$

where $f_1(|\boldsymbol{\xi}|) - f_5(|\boldsymbol{\xi}|)$ are arbitrary scalar functions of the magnitude of $\boldsymbol{\xi}$. The inverse of Eq. (45) can be determined by recognizing that it also must be an isotropic fourth-rank tensor function of a vector variable satisfying all of Eq. (44), and must also satisfy

$$F_{ijmn}(\boldsymbol{\xi})F^{-1}_{mnkl}(\boldsymbol{\xi}) = F^{-1}_{ijmn}(\boldsymbol{\xi})F_{mnkl}(\boldsymbol{\xi}) = \frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}). \tag{46}$$

The result for $\mathbf{F}^{-1}(\boldsymbol{\xi})$ is identical in form to Eq. (45) except with the following five coefficient functions, expressed in terms of the original coefficient functions of $\mathbf{F}(\boldsymbol{\xi})$:

$$f_1^{-1}(|\boldsymbol{\xi}|) = \frac{|\boldsymbol{\xi}|^4 f_3^2 - 4f_1(f_2 + |\boldsymbol{\xi}|^2 f_4 + |\boldsymbol{\xi}|^4 f_5)}{2D}, \qquad f_2^{-1}(|\boldsymbol{\xi}|) = \frac{1}{f_2},$$

$$f_3^{-1}(|\boldsymbol{\xi}|) = \frac{-2f_2f_3 - |\boldsymbol{\xi}|^2 f_3^2 + 4f_1(f_4 + |\boldsymbol{\xi}|^2 f_5)}{D}, \qquad f_4^{-1}(|\boldsymbol{\xi}|) = \frac{-2f_4}{f_2(2f_2 + |\boldsymbol{\xi}|^2 f_4)},$$

$$\begin{aligned} & f_5^{-1}(|\boldsymbol{\xi}|) \\ & = \frac{2[f_2f_3(3f_3+4f_4)+2(f_1+f_2)f_4^2-4f_2(3f_1+f_2)f_5] - f_4[f_3^2-4(f_1+f_2)f_5]|\boldsymbol{\xi}|^2}{2D(2f_2 + f_4|\boldsymbol{\xi}|^2)}, \end{aligned} \tag{47}$$

where

$$D \equiv f_2\{2f_2(3f_1 + f_2) + 2[2f_1f_4 + f_2(f_3 + f_4)]|\boldsymbol{\xi}|^2 - [f_3^2 - 2(2f_1 + f_2)f_5]|\boldsymbol{\xi}|^4\}.$$

5.2. *Evaluation of* $\boldsymbol{\Gamma}(\boldsymbol{\xi})$ *and* $(\boldsymbol{\Gamma}(\boldsymbol{\xi})^{-1} + c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0)^{-1}$ *for arbitrary isotropic comparison materials and an example two-point correlation function*

Recall that $\boldsymbol{\Gamma}(\boldsymbol{\xi})$ is defined in Eq. (40). As shown e.g. by Drugan and Willis (1996), when the comparison material is isotropic with bulk modulus κ and shear modulus μ, the Fourier transform of the infinite-body $\boldsymbol{\Gamma}_0(\mathbf{x})$ of Eq. (4), which appears in the integrands in Eq. (40), is

$$[\tilde{\mathbf{\Gamma}}_0(\xi)]_{ijkl} = \frac{\xi_i\delta_{jk}\xi_l + \xi_j\delta_{ik}\xi_l + \xi_i\delta_{jl}\xi_k + \xi_j\delta_{il}\xi_k}{4\mu|\xi|^2} - \frac{3\kappa+\mu}{\mu(3\kappa+4\mu)}\frac{\xi_i\xi_j\xi_k\xi_l}{|\xi|^4}. \quad (48)$$

Note that the two scalar invariants of this are independent of ξ. Thus, using the first integral representation in Eq. (40), and using Eq. (45) to represent $\mathbf{\Gamma}(\xi)$, we calculate:

$$\begin{aligned}\mathbf{\Gamma}_{ijij}(\xi) &= 3f_1(|\xi|) + 6f_2(|\xi|) + |\xi|^2 f_3(|\xi|) + 2|\xi|^2 f_4(|\xi|) + |\xi|^4 f_5(|\xi|)\\ &= \frac{3\kappa+7\mu}{\mu(3\kappa+4\mu)},\end{aligned} \quad (49a)$$

$$\begin{aligned}\mathbf{\Gamma}_{iikk}(\xi) &= 9f_1(|\xi|) + 3f_2(|\xi|) + 3|\xi|^2 f_3(|\xi|) + |\xi|^2 f_4(|\xi|) + |\xi|^4 f_5(|\xi|)\\ &= \frac{3}{3\kappa+4\mu},\end{aligned} \quad (49b)$$

where we have used the result shown by Drugan and Willis (1996) that if $h(0)=1$ but otherwise arbitrary,

$$\frac{1}{8\pi^3}\int \tilde{h}(\xi')\,\mathrm{d}\xi' = \left[\frac{1}{8\pi^3}\int \tilde{h}(\xi')\mathrm{e}^{-\mathrm{i}\xi'\bullet\mathbf{x}}\,\mathrm{d}\xi'\right]_{\mathbf{x}=\mathbf{0}} = h(0) = 1. \quad (50)$$

Thus, conditions (49a), (49b) are valid for arbitrary two-point correlation functions having $h(0)=1$. To evaluate $\mathbf{\Gamma}(\xi)$ completely, three additional conditions are needed so that, together with Eq. (49), we have five independent conditions on the five functions appearing in Eq. (45); this requires specification of a two-point correlation function.

To facilitate completely analytical results, we will evaluate $\mathbf{\Gamma}(\xi)$ for the following choice of two-point correlation function with associated three-dimensional Fourier transform

$$h(\mathbf{x}) = \mathrm{e}^{-|\mathbf{x}|/a} \quad \Rightarrow \quad \tilde{h}(\xi) = \frac{8\pi a^3}{(1+a^2|\xi|^2)^2}. \quad (51a,b)$$

With appropriate choice of the constant a, this correlation function is a quite reasonable approximation to the two-point correlation function for a matrix containing a random distribution of nonoverlapping spherical particles/voids. We choose a so that the integral involving the two-point correlation function in the first gradient approximation to the nonlocal constitutive equation of Drugan and Willis (1996), namely [defining $r=|\mathbf{x}|$]

$$\int_0^\infty h(r)r\,\mathrm{d}r, \quad (52)$$

is identical to the value of this integral obtained by Monetto and Drugan (2003) using the Verlet and Weis (1972) correction to the Wertheim (1963) solution of the Percus and Yevick (1958) statistical mechanics model of random hard sphere distribution. This means that a is related to the sphere radius R as

$$a^2 = R^2\,\frac{5Ac_1(1+2\tilde{c}_1)+2B[(1+2\tilde{c}_1)(2+c_1)-c_1(\tilde{c}_1/c_1)^{2/3}(10-2\tilde{c}_1+\tilde{c}_1^2)]}{10B(1-c_1)(1+2\tilde{c}_1)}, \tag{53a}$$

where

$$\tilde{c}_1 = c_1 - \frac{1}{16}c_1^2, \quad A = \frac{3}{2}\,\frac{\tilde{c}_1^2(1-0.7117\tilde{c}_1-0.114\tilde{c}_1^2)}{(1-\tilde{c}_1)^4},$$
$$B = 12A\,\frac{(1-\tilde{c}_1)^2}{\tilde{c}_1(2+\tilde{c}_1)}. \tag{53b}$$

Having chosen a in this way, Fig. 1 shows a comparison of the two-point *matrix* probability $P_{22}(|\mathbf{x}-\mathbf{x}'|) = P_{22}(r)$, obtained using Eq. (14) with Eqs. (51) and (53), as a function of the distance between the two sampling points as compared to the extremely accurate results calculated from the Verlet–Weis correction to the Percus–Yevick–Wertheim solution by Torquato and Stell (1985) (the last results being in excellent agreement with the essentially exact computer simulations of Haile et al., 1985); the agreement is seen to be quite good.

The correlation function given in Eq. (51) assumes an isotropic distribution of phases, i.e., $h(\mathbf{x}) = h(|\mathbf{x}|)$, in which case the second integral form of Eq. (40) can be rewritten as

$$\boldsymbol{\Gamma}(\boldsymbol{\xi}) = \frac{1}{8\pi^3}\int_0^{\pi}\int_0^{2\pi}\tilde{\boldsymbol{\Gamma}}_0(\theta,\phi)\left[\int_0^{\infty}\tilde{h}\left(\sqrt{|\boldsymbol{\xi}|^2-2|\boldsymbol{\xi}|\rho\cos\phi+\rho^2}\right)\rho^2\,\mathrm{d}\rho\right]\sin\phi\,\mathrm{d}\theta\,\mathrm{d}\phi, \tag{54}$$

where ρ, θ, ϕ are spherical coordinates. Choosing $\boldsymbol{\xi}$ to lie purely in the 3-direction and using the representation (45) for $\boldsymbol{\Gamma}(\boldsymbol{\xi})$, we obtain from Eq. (54) with Eqs. (48) and (51b) the following three additional conditions on the functions in Eq. (45) for $\boldsymbol{\Gamma}(\boldsymbol{\xi})$:

$$\begin{aligned}\boldsymbol{\Gamma}_{1122}(\boldsymbol{\xi}) &= f_1(|\boldsymbol{\xi}|)\\ &= -\frac{3\kappa+\mu}{32\pi^2\mu(3\kappa+4\mu)}\int_0^{\pi}\int_0^{\infty}\tilde{h}\left(\sqrt{|\boldsymbol{\xi}|^2-2|\boldsymbol{\xi}|\rho\cos\phi+\rho^2}\right)\rho^2\sin^5\phi\,\mathrm{d}\rho\,\mathrm{d}\phi\\ &= -\frac{(3\kappa+\mu)}{16\mu(3\kappa+4\mu)}\,\frac{a|\boldsymbol{\xi}|(3+a^2|\boldsymbol{\xi}|^2)+(a^4|\boldsymbol{\xi}|^4-2a^2|\boldsymbol{\xi}|^2-3)\arctan(a|\boldsymbol{\xi}|)}{a^5|\boldsymbol{\xi}|^5}\end{aligned} \tag{55a}$$

$$\begin{aligned}\boldsymbol{\Gamma}_{1212}(\boldsymbol{\xi}) &= \frac{1}{2}f_2(|\boldsymbol{\xi}|)\\ &= \frac{1}{64\pi^2\mu(3\kappa+4\mu)}\int_0^{\pi}\int_0^{\infty}\tilde{h}\left(\sqrt{|\boldsymbol{\xi}|^2-2|\boldsymbol{\xi}|\rho\cos\phi+\rho^2}\right)\\ &\quad\times[3(3\kappa+5\mu)+(3\kappa+\mu)\cos 2\phi]\rho^2\sin^3\phi\,\mathrm{d}\rho\,\mathrm{d}\phi\\ &= \frac{-3(3\kappa+\mu)a|\boldsymbol{\xi}|-(15\kappa+17\mu)a^3|\boldsymbol{\xi}|^3+3(1+a^2|\boldsymbol{\xi}|^2)[3\kappa+\mu+(3\kappa+5\mu)a^2|\boldsymbol{\xi}|^2]\arctan(a|\boldsymbol{\xi}|)}{16\mu(3\kappa+4\mu)a^5|\boldsymbol{\xi}|^5}\end{aligned} \tag{55b}$$

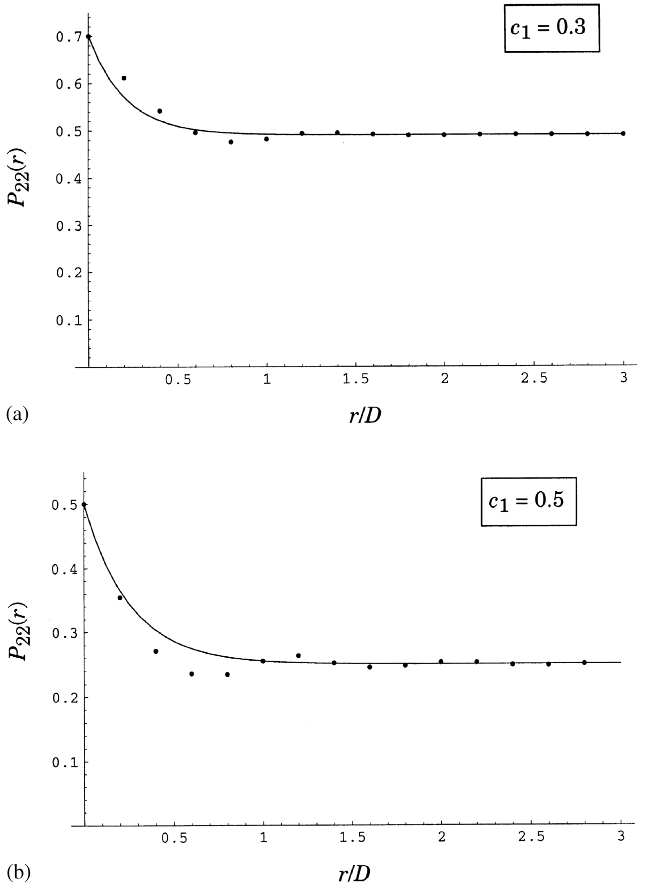


Fig. 1. Comparison of the matrix two-point probability function for (a) $c_1 = 0.3$ and (b) $c_1 = 0.5$, calculated from the exponential two-point correlation function (51) with a chosen as explained (solid line) with the effectively exact result calculated from the Verlet–Weis improvement to the Percus–Yevick model (Torquato and Stell, 1985). The separation of the two points is denoted by r, and the sphere diameter by D.

$$\begin{aligned}\Gamma_{ii11}(\xi) &= 3f_1(|\xi|) + f_2(|\xi|) + \frac{1}{2}|\xi|^2 f_3(|\xi|)\\ &= \frac{3}{8\pi^2(3\kappa+4\mu)}\int_0^{\pi}\int_0^{\infty}\tilde{h}\left(\sqrt{|\xi|^2 - 2|\xi|\rho\cos\phi + \rho^2}\right)\rho^2\sin^3\phi\,\mathrm{d}\rho\,\mathrm{d}\phi\\ &= \frac{3}{2(3\kappa+4\mu)a^3|\xi|^3}[(1+a^2|\xi|^2)\arctan(a|\xi|) - a|\xi|]. \qquad (55c)\end{aligned}$$

One check of the results (55) is to use the fact shown in the appendix of Drugan and Willis (1996) that

$$\Gamma_{ijkl}(\mathbf{0}) \equiv P_{ijkl} = -\frac{3\kappa+\mu}{15\mu(3\kappa+4\mu)}\delta_{ij}\delta_{kl} + \frac{3(\kappa+2\mu)}{10\mu(3\kappa+4\mu)}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}). \qquad (56)$$

It is easy to confirm that in the $|\boldsymbol{\xi}| \to 0$ limit, all three of the results in Eq. (55) reduce to the appropriate $\mathbf{P}$ component obtained from Eq. (56).

Finally, the five conditions (49) and (55) permit solving for the five functions $f_1(|\boldsymbol{\xi}|) - f_5(|\boldsymbol{\xi}|)$, so that the explicit representation of $\boldsymbol{\Gamma}(\boldsymbol{\xi})$ is

$$\Gamma_{ijkl}(\boldsymbol{\xi}) = f_1(|\boldsymbol{\xi}|)\delta_{ij}\delta_{kl} + f_2(|\boldsymbol{\xi}|)\,\frac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) + f_3(|\boldsymbol{\xi}|)\,\frac{1}{2}(\delta_{ij}\xi_k\xi_l + \delta_{kl}\xi_i\xi_j) + f_4(|\boldsymbol{\xi}|)\,\frac{1}{4}(\xi_i\delta_{jk}\xi_l + \xi_j\delta_{ik}\xi_l + \xi_i\delta_{jl}\xi_k + \xi_j\delta_{il}\xi_k) + f_5(|\boldsymbol{\xi}|)\xi_i\xi_j\xi_k\xi_l, \tag{57}$$

with

$$f_1(|\boldsymbol{\xi}|) = -\frac{(3\kappa+\mu)}{16\mu(3\kappa+4\mu)}\,\frac{a|\boldsymbol{\xi}|(3+a^2|\boldsymbol{\xi}|^2) + (a^4|\boldsymbol{\xi}|^4 - 2a^2|\boldsymbol{\xi}|^2 - 3)\arctan(a|\boldsymbol{\xi}|)}{a^5|\boldsymbol{\xi}|^5}, \tag{58}$$

$$f_2(|\boldsymbol{\xi}|) = \frac{-3(3\kappa+\mu)a|\boldsymbol{\xi}| - (15\kappa+17\mu)a^3|\boldsymbol{\xi}|^3 + 3(1+a^2|\boldsymbol{\xi}|^2)[3\kappa+\mu+(3\kappa+5\mu)a^2|\boldsymbol{\xi}|^2]\arctan(a|\boldsymbol{\xi}|)}{8\mu(3\kappa+4\mu)a^5|\boldsymbol{\xi}|^5},$$

$$f_3(|\boldsymbol{\xi}|) = \frac{(3\kappa+\mu)}{8\mu(3\kappa+4\mu)a^5|\boldsymbol{\xi}|^7} \times [(15+13a^2|\boldsymbol{\xi}|^2)a|\boldsymbol{\xi}| - 3(5+6a^2|\boldsymbol{\xi}|^2+a^4|\boldsymbol{\xi}|^4)\arctan(a|\boldsymbol{\xi}|)],$$

$$f_4(|\boldsymbol{\xi}|) = \frac{1}{4\mu(3\kappa+4\mu)a^5|\boldsymbol{\xi}|^7}\left\{15(3\kappa+\mu)a|\boldsymbol{\xi}| + (57\kappa+37\mu)a^3|\boldsymbol{\xi}|^3 + 4(3\kappa+4\mu)a^5|\boldsymbol{\xi}|^5 - 3\left(1+a^2|\boldsymbol{\xi}|^2\right)\left[5(3\kappa+\mu)+9(\kappa+\mu)a^2|\boldsymbol{\xi}|^2\right]\arctan(a|\boldsymbol{\xi}|)\right\},$$

$$f_5(|\boldsymbol{\xi}|) = -\frac{(3\kappa+\mu)}{16\mu(3\kappa+4\mu)a^5|\boldsymbol{\xi}|^9}\,[(105+115a^2|\boldsymbol{\xi}|^2+16a^4|\boldsymbol{\xi}|^4)a|\boldsymbol{\xi}| - 15(7+10a^2|\boldsymbol{\xi}|^2+3a^4|\boldsymbol{\xi}|^4)\arctan(a|\boldsymbol{\xi}|)].$$

Next we evaluate the function $(\boldsymbol{\Gamma}(\boldsymbol{\xi})^{-1} + c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0)^{-1}$, which is needed in the exact nonlocal constitutive equation (42). Since for isotropic composites comprised of isotropic phases this is an isotropic fourth-rank tensor function of a vector variable, its evaluation is directly accomplished by repeated use of the inverse formula (45) with Eq. (47): First, $\boldsymbol{\Gamma}(\boldsymbol{\xi})^{-1}$ has the form (45) but with coefficient functions given by Eq. (47) in terms of the coefficient functions (58) for $\boldsymbol{\Gamma}(\boldsymbol{\xi})$. To this is added the (constant) fourth-rank tensor

$$(c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0)_{ijkl} \equiv (\breve{\kappa} - 2\breve{\mu}/3)\delta_{ij}\delta_{kl} + \breve{\mu}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}), \tag{59}$$

and then the inversion formula (45) with Eq. (47) is applied to the result. The components of $(\boldsymbol{\Gamma}(\boldsymbol{\xi})^{-1} + c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0)^{-1}$ are thus given by Eq. (45), with the following coefficient functions expressed in terms of the coefficient functions (58) for $\boldsymbol{\Gamma}(\boldsymbol{\xi})$:

$$\begin{aligned}
\hat{f}_1(|\boldsymbol{\xi}|) = \frac{1}{\hat{D}}\{&4(1+2\breve{\mu}f_2)[3f_1-(3\breve{\kappa}-2\breve{\mu})f_2(3f_1+f_2)]\\
&+(3\breve{\kappa}+4\breve{\mu})[4f_1f_4|\boldsymbol{\xi}|^2+(4f_1f_5-f_3^2)|\boldsymbol{\xi}|^4]\\
&-4(3\breve{\kappa}-2\breve{\mu})f_2\{[f_3(1+2\breve{\mu}f_2)+2\breve{\mu}f_4(2f_1+f_2)]|\boldsymbol{\xi}|^2\\
&-\breve{\mu}[f_3^2-2f_5(2f_1+f_2)]|\boldsymbol{\xi}|^4\},\\
\hat{f}_2(|\boldsymbol{\xi}|) = {}&\frac{f_2}{1+2\breve{\mu}f_2},\\
\hat{f}_3(|\boldsymbol{\xi}|) = \frac{2}{\hat{D}}\{&6f_3(1+2\breve{\mu}f_2)+9\breve{\kappa}f_3^2|\boldsymbol{\xi}|^2\\
&-4[9\breve{\kappa}f_1+(3\breve{\kappa}-2\breve{\mu})f_2](f_4+f_5|\boldsymbol{\xi}|^2)\},\\
\hat{f}_4(|\boldsymbol{\xi}|) = {}&\frac{f_4}{(1+2\breve{\mu}f_2)(1+2\breve{\mu}f_2+\breve{\mu}|\boldsymbol{\xi}|^2f_4)},\\
\hat{f}_5(|\boldsymbol{\xi}|) = {}&\frac{1}{\hat{D}(1+2\breve{\mu}f_2+\breve{\mu}f_4|\boldsymbol{\xi}|^2)}\{3(1+2\breve{\mu}f_2)[4f_5(1+3\breve{\kappa}(3f_1+f_2))\\
&-12\breve{\kappa}f_3f_4-9\breve{\kappa}f_3^2]-4f_4(f_4+f_5|\boldsymbol{\xi}|^2)\\
&\times[\breve{\mu}+3\breve{\kappa}(1+3\breve{\mu}(f_1+f_2))]+9\breve{\kappa}\breve{\mu}f_3^2f_4|\boldsymbol{\xi}|^2\},
\end{aligned} \tag{60}$$

where

$$\begin{aligned}
\hat{D} \equiv 4(1+2\breve{\mu}f_2)\{&3[1+3\breve{\kappa}(3f_1+f_2)](1+2\breve{\mu}f_2)\\
&+[3\breve{\kappa}+4\breve{\mu}+18\breve{\kappa}\breve{\mu}(2f_1+f_2)](f_4|\boldsymbol{\xi}|^2+f_5|\boldsymbol{\xi}|^4)\\
&+9\breve{\kappa}f_3[(1+2\breve{\mu}f_2)|\boldsymbol{\xi}|^2-\breve{\mu}f_3|\boldsymbol{\xi}|^4]\}.
\end{aligned}$$

As a check, it is easily verified that for $\breve{\kappa}=\breve{\mu}=0$, which corresponds to the special choice (18) for the comparison modulus tensor, the results (60) reduce to those for $\boldsymbol{\Gamma}(\boldsymbol{\xi})$.

The results just given, i.e. the completely explicit representation of $(\boldsymbol{\Gamma}(\boldsymbol{\xi})^{-1}+c_1\mathbf{L}_2+c_2\mathbf{L}_1-\mathbf{L}_0)^{-1}$, render the exact nonlocal constitutive equation in Fourier transform space, (42), completely and explicitly specified for isotropic composites having the two-point correlation function (51a) and arbitrary isotropic comparison moduli. When an ensemble-average strain field is specified, (42) can be inverse Fourier transformed to obtain the exact physical-space nonlocal constitutive equation. One example of this, permitting assessment of a prior gradient-approximate nonlocal constitutive equation, is provided in Section 6.2.

6. Example evaluations of the exact nonlocal constitutive equations for isotropic composites

6.1. Evaluation of special exact nonlocal constitutive equation for bulk modulus

One interesting result that is easily obtained from the exact nonlocal constitutive equation (22) [whose Fourier transform is (43)], resulting from the special choice (18) of comparison modulus tensor, is the calculation of the nonlocal contribution to the bulk modulus of an isotropic composite having isotropic phases. In this case, we can represent the tensor $(\mathbf{L}_1 - \mathbf{L}_2)$ appearing in Eqs. (22) and (43) as

$$(\mathbf{L}_1 - \mathbf{L}_2)_{ijkl} = \bar{\lambda}\delta_{ij}\delta_{kl} + \bar{\mu}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}), \quad \bar{\lambda} = \lambda_1 - \lambda_2, \quad \bar{\mu} = \mu_1 - \mu_2. \tag{61}$$

Then we calculate for the term involving $\boldsymbol{\Gamma}(\boldsymbol{\xi})$ in Eq. (43):

$$\begin{aligned}(\mathbf{L}_1 - \mathbf{L}_2)_{ijmn}&\Gamma_{mnop}(\boldsymbol{\xi})(\mathbf{L}_1 - \mathbf{L}_2)_{opkl} \\ &= \bar{\lambda}^2\Gamma_{mmnn}(\boldsymbol{\xi})\delta_{ij}\delta_{kl} + 2\bar{\lambda}\bar{\mu}[\Gamma_{ijnn}(\boldsymbol{\xi})\delta_{kl} + \delta_{ij}\Gamma_{nnkl}(\boldsymbol{\xi})] + 4\bar{\mu}^2\Gamma_{ijkl}(\boldsymbol{\xi}).\end{aligned} \tag{62}$$

To evaluate the bulk modulus, we consider an ensemble-average strain field consisting solely of equal triaxial straining

$$\langle e\rangle_{ij}(\mathbf{x}) = \delta_{ij}\langle e\rangle(\mathbf{x}), \tag{63}$$

i.e., pure volume change, and we wish to relate the hydrostatic stress to this volume change. Then Eq. (43) becomes, using Eqs. (62) and (63)

$$\langle\tilde{\sigma}\rangle_{ii}(\boldsymbol{\xi}) = [(c_1\mathbf{L}_1 + c_2\mathbf{L}_2)_{iikk} - c_1c_2(9\bar{\lambda}^2 + 12\bar{\lambda}\bar{\mu} + 4\bar{\mu}^2)\Gamma_{iikk}(\boldsymbol{\xi})]\langle\tilde{e}\rangle(\boldsymbol{\xi}). \tag{64}$$

Now recall from Eq. (49b) that the invariant $\Gamma_{iikk}(\boldsymbol{\xi})$ appearing in Eq. (64) is independent of $\boldsymbol{\xi}$! Thus, the bracketed term in Eq. (64) is independent of $\boldsymbol{\xi}$, so that employing the definitions of $\bar{\lambda}, \bar{\mu}$ from Eq. (61), evaluating the first term in the brackets in Eq. (64), enforcing the choice (18) for the comparison moduli in Eq. (49b) and finally inverse-transforming Eq. (64), we obtain for the exact nonlocal constitutive equation:

$$\langle\sigma\rangle_{ii}(\mathbf{x}) = 9\left[c_1\kappa_1 + c_2\kappa_2 - \frac{3c_1c_2(\kappa_1 - \kappa_2)^2}{3(c_1\kappa_2 + c_2\kappa_1) + 4(c_1\mu_2 + c_2\mu_1)}\right]\langle e\rangle(\mathbf{x}). \tag{65}$$

This shows that there is *no nonlocal correction* for the bulk modulus from the exact nonlocal constitutive equation! This conclusion is valid for arbitrary isotropic random two-phase composites, since the evaluation of $\Gamma_{iikk}(\boldsymbol{\xi})$ in Eq. (49b) did not rely on use of a specific two-point correlation function. (One can also verify that the gradient correction for the bulk modulus in Drugan and Willis, 1996 vanishes.) This result provides a direct explanation for the independence of bulk modulus on volume element size in the numerical simulations of Segurado and Llorca (2002) of a matrix reinforced by a random distribution of spherical particles.

6.2. Evaluation of general exact nonlocal constitutive equation for sinusoidally-varying ensemble-average shear strain

One interesting calculation carried out by Drugan and Willis (1996) was their investigation of the "representative volume element" size needed for accuracy of the standard constant-effective-modulus constitutive equation to be valid. They did this by considering an ensemble-average strain that varied sinusoidally with position, and then determining the wavelength of the strain variation at which their nonlocal gradient term made a 5% correction (or any other desired value) to the standard local term. They obtained specific results for isotropic composites consisting of a matrix reinforced/weakened by a random distribution of identical nonoverlapping spherical particles/voids, by using the Percus–Yevick statistical mechanics model of hard sphere distribution. Monetto and Drugan (2003) improved their results by employing the more accurate Verlet–Weis modification to the Percus–Yevick model.

Here we will perform the same type of analysis by employing the general exact nonlocal constitutive equation derived in Sections 4 and 5. We will consider the nonlocal variation of the isotropic composite shear modulus, and calculate the wavelength of sinusoidally-varying shear strain at which the full nonlocal constitutive shear modulus differs by 5% from the local term. Thus, we will analyze the following ensemble-average strain field

$$\langle e\rangle_{12}(\mathbf{x}) = \varepsilon \sin\frac{2\pi x_1}{l}, \quad \text{all other } \langle e\rangle_{ij}(\mathbf{x}) \equiv 0, \tag{66}$$

where $\varepsilon \ll 1$. The three-dimensional Fourier transform of this strain field is

$$\langle \tilde{e}\rangle_{12}(\boldsymbol{\xi}) = -4\pi^3 \varepsilon \mathrm{i}\left[\delta(\beta+\xi_1) - \delta(\beta-\xi_1)\right]\delta(\xi_2)\delta(\xi_3), \quad \text{where } \beta = \frac{2\pi}{l}, \tag{67}$$

and where $\delta(\cdot)$ is the Dirac delta function. We wish to calculate the shear stress component associated with Eq. (66); that is, from Eq. (42)

$$\begin{aligned}\langle \tilde{\sigma}\rangle_{12}(\boldsymbol{\xi}) = 2\Big[&c_1\mathbf{L}_1 + c_2\mathbf{L}_2 \\ &- c_1c_2(\mathbf{L}_1-\mathbf{L}_2)(\boldsymbol{\Gamma}(\boldsymbol{\xi})^{-1} + c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0)^{-1}(\mathbf{L}_1-\mathbf{L}_2)\Big]_{1212}\langle \tilde{e}\rangle_{12}(\boldsymbol{\xi}).\end{aligned} \tag{68}$$

Application of Eq. (62) with Eq. (61) shows

$$\begin{aligned}&\left[(\mathbf{L}_1-\mathbf{L}_2)(\boldsymbol{\Gamma}(\boldsymbol{\xi})^{-1} + c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0)^{-1}(\mathbf{L}_1-\mathbf{L}_2)\right]_{1212} \\ &\quad = 4(\mu_1-\mu_2)^2\left[(\boldsymbol{\Gamma}(\boldsymbol{\xi})^{-1} + c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0)^{-1}\right]_{1212},\end{aligned} \tag{69}$$

and then use of our results at the end of Section 5.2 give

$$\begin{aligned}\left[(\boldsymbol{\Gamma}(\boldsymbol{\xi})^{-1} + c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0)^{-1}\right]_{1212} = \frac{1}{2}\hat{f}_2(|\boldsymbol{\xi}|) + \frac{1}{4}\hat{f}_4(|\boldsymbol{\xi}|)(\xi_1^2+\xi_2^2) \\ + \hat{f}_5(|\boldsymbol{\xi}|)\xi_1^2\xi_2^2.\end{aligned} \tag{70}$$

Employing these results, (68) becomes

$$\langle\tilde{\sigma}\rangle_{12}(\boldsymbol{\xi}) = 2\Big\{c_1\mu_1 + c_2\mu_2 - c_1c_2(\mu_1-\mu_2)^2[2\hat{f}_2(|\boldsymbol{\xi}|) + \hat{f}_4(|\boldsymbol{\xi}|)(\xi_1^2+\xi_2^2) + 4\hat{f}_5(|\boldsymbol{\xi}|)\xi_1^2\xi_2^2]\Big\}\langle\tilde{e}\rangle_{12}(\boldsymbol{\xi}). \tag{71}$$

Now, employing Eq. (67), it is an easy matter to perform the inverse Fourier transform of Eq. (71), given the form of Eq. (67) in terms of the Dirac delta functions. The resulting exact nonlocal constitutive equation in physical space is

$$\boxed{\begin{aligned}\langle\sigma\rangle_{12}(x_1) &= 2\Big\{c_1\mu_1 + c_2\mu_2 - c_1c_2(\mu_1-\mu_2)^2[2\hat{f}_2(\beta) + \beta^2\hat{f}_4(\beta)]\Big\}\varepsilon\sin(\beta x_1),\\ \beta &= \frac{2\pi}{l},\end{aligned}} \tag{72}$$

where from Eq. (60)

$$\hat{f}_2(\beta) = \frac{f_2(\beta)}{1+2\breve{\mu}f_2(\beta)}, \quad \hat{f}_4(\beta) = \frac{f_4(\beta)}{[1+2\breve{\mu}f_2(\beta)][1+2\breve{\mu}f_2(\beta)+\breve{\mu}\beta^2 f_4(\beta)]}, \tag{73}$$

$f_2(\beta), f_4(\beta)$ are given by Eq. (58), and from Eq. (59), $\breve{\mu} = c_1\mu_2 + c_2\mu_1 - \mu$. In all of these expressions, the isotropic moduli associated with $\mathbf{L}_1$ are (κ_1,μ_1), with $\mathbf{L}_2$ are (κ_2,μ_2) and with the comparison modulus tensor $\mathbf{L}_0$ are (κ,μ), so that the exact nonlocal constitutive equation (72) has been obtained in terms of arbitrary comparison moduli.

To compare with previous results, we now choose the comparison modulus tensor to equal the matrix modulus tensor, $\mathbf{L}_0 = \mathbf{L}_2$; this appears to be a particularly good choice, as discussed by Drugan and Willis (1996) and Drugan (2000). One confirmation of the exact nonlocal constitutive equation (72) is the fact that in the limit as the wavelength $l \to \infty$ (i.e., $\beta \to 0$), the braced term in Eq. (72) does indeed reduce to the Hashin–Shtrikman estimate for the effective (composite) shear modulus (see, e.g., Drugan, 2000, Eq. (82)):

$$\mu_{\text{eff}} = \mu_2\,\frac{6\mu_1(\kappa_2+2\mu_2) + (c_1\mu_1+c_2\mu_2)(9\kappa_2+8\mu_2)}{\mu_2(9\kappa_2+8\mu_2) + 6(\kappa_2+2\mu_2)(c_2\mu_1+c_1\mu_2)}. \tag{74}$$

Now, to determine the length l at which the nonlocal contribution in (72) makes a 5% correction to the local term (74) for the composite shear modulus, we solve for l in the following equation, having defined $\mu(\beta)$ to be the braced term in Eq. (72)—i.e., the total (local plus nonlocal) composite shear modulus, and with μ_{eff} being given by Eq. (74):

$$\frac{\mu(\beta) - \mu_{\text{eff}}}{\mu_{\text{eff}}} = 0.05. \tag{75}$$

We have obtained results for the cases of an isotropic matrix weakened by voids, and strengthened by rigid particles. In both cases, the results for l from Eq. (75) are independent of the value of the shear modulus of the matrix, but do depend on the matrix Poisson's ratio; in determining this, we have used the fact that

$$\kappa = \frac{2(1+v)}{3(1-2v)}\,\mu, \tag{76}$$

where v is Poisson's ratio. Figs. 2 and 3 show comparisons of the results calculated as just explained from the exact nonlocal constitutive equation, with those from Monetto and Drugan's (2003) improvement (by incorporating the Verlet–Weis improvement of the Percus–Yevick model) of the Drugan and Willis (1996) gradient-approximate nonlocal constitutive equation. One observes that the predictions of the gradient-approximate nonlocal constitutive equation are quite good, except when the length scale of the ensemble-average strain variation becomes on the order of the diameter of the spherical particles/voids; the comparisons show that the gradient-approximate nonlocal constitutive equation predictions are far more accurate for voids than for rigid particles.

6.3. Comparison of predictions of the two exact nonlocal constitutive equations

Finally, let us compare the predictions of the two exact nonlocal constitutive equations for two-phase composites derived in this paper, namely (22) [which results from the special choice (18) of comparison modulus tensor and whose Fourier transform is (43)], and (42) [which can be evaluated in physical space for isotropic composites and an *arbitrary* choice of the comparison modulus tensor via the results in Section 5, as shown in Section 6.2]. Comparing Eq. (43) to Eq. (42), it is clear that they differ by the term

$$c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0 \tag{77}$$

appearing in Eq. (42). We can thus assess the predictions of the special nonlocal constitutive equation (43) [(22)] by comparing the results of (42) for different choices of the comparison modulus tensor, one choice being (18) which gives (43). In this regard, we first recall from the discussion following (22) that, phrased now in terms of the Fourier transform results, (43) agrees exactly through second order in phase contrast $(\mathbf{L}_1 - \mathbf{L}_2)$ with Eq. (42) for small phase contrast, for *arbitrary* choice of the comparison modulus tensor [this can also be seen directly from Eqs. (42) and (43)]. Thus, the two exact nonlocal constitutive equations will agree well regardless of the choice of comparison modulus in Eq. (42) when the phase contrast is small.

To compare predictions when the phase contrast is not small, let us consider matrix/inclusion composites. We noted earlier that a good choice for the comparison modulus tensor in this case is the matrix modulus, $\mathbf{L}_0 = \mathbf{L}_2$; for this choice, the quantity in Eq. (77) becomes

$$c_1\mathbf{L}_2 + c_2\mathbf{L}_1 - \mathbf{L}_0 = (1-c_1)(\mathbf{L}_1 - \mathbf{L}_2). \tag{78}$$

This shows that the special exact nonlocal constitutive equation (43) will agree most closely with the general one (42) for small phase contrast (as already noted), and for the highest concentrations of inclusions/voids. Furthermore, (78) suggests that for the two extreme cases of a matrix weakened by voids or strengthened by rigid particles,

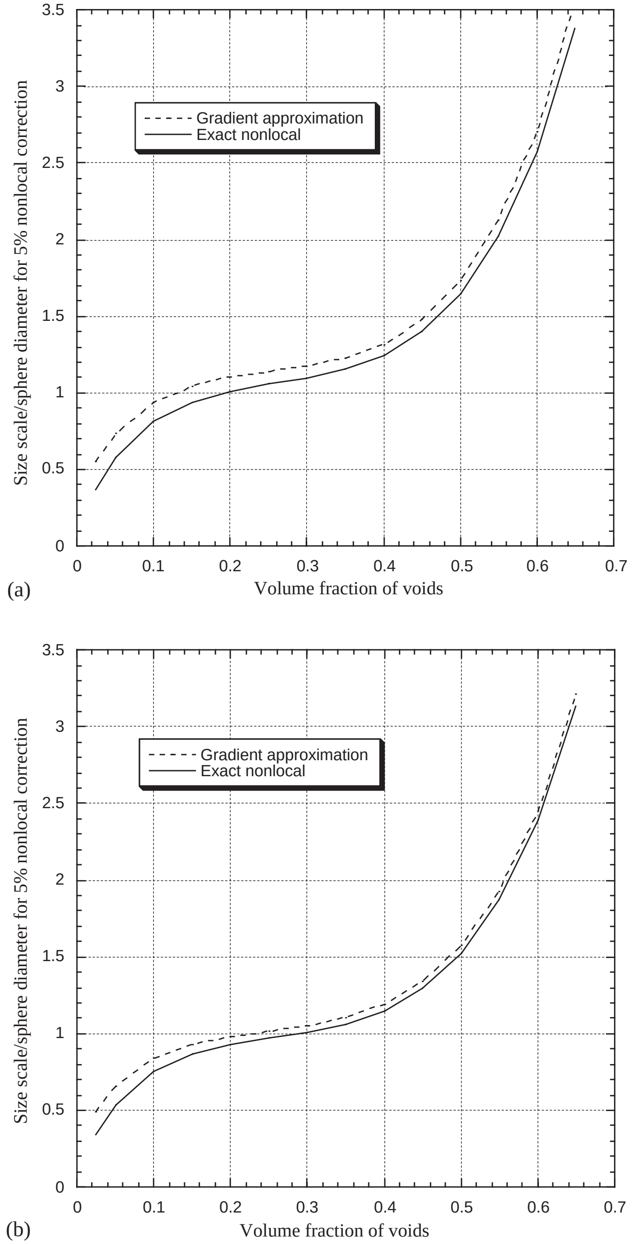
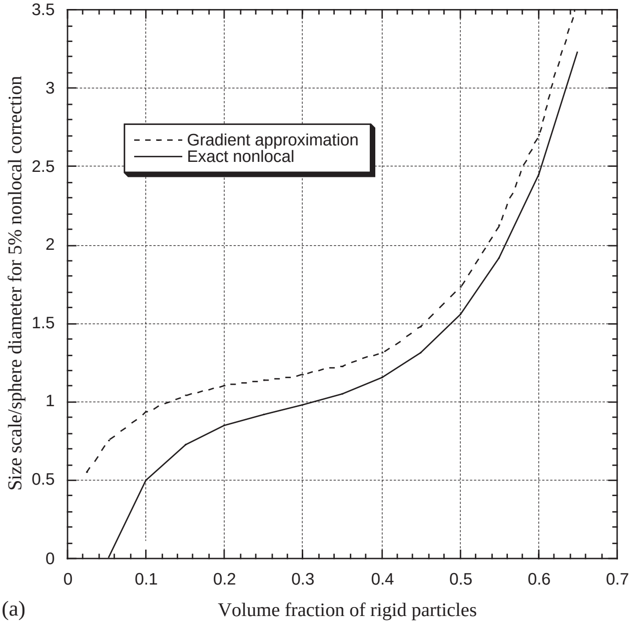
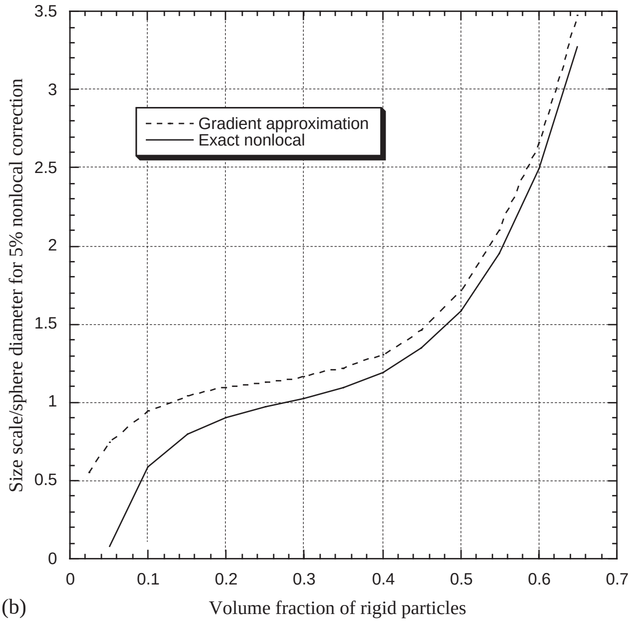


Fig. 2. Comparison of "representative volume element" size normalized by sphere diameter for the composite elastic shear modulus from the exact nonlocal constitutive equation versus the single gradient-approximate nonlocal constitutive equation of Drugan and Willis (1996) as improved by Monetto and Drugan (2003), for an isotropic matrix weakened by a random distribution of nonoverlapping identical spherical voids. (a) matrix Poisson's ratio $v = 0.2$, (b) $v = 0.33$.



(a)



(b)

Fig. 3. Comparison of "representative volume element" size normalized by sphere diameter for the composite elastic shear modulus from the exact nonlocal constitutive equation versus the single gradient-approximate nonlocal constitutive equation of Drugan and Willis (1996) as improved by Monetto and Drugan (2003), for an isotropic matrix stiffened by a random distribution of nonoverlapping identical spherical rigid particles. (a) matrix Poisson's ratio $\nu = 0.2$, (b) $\nu = 0.33$.

(43) will agree more closely with (42) for voids ($\mathbf{L}_1 = \mathbf{0}$) than for rigid particles ($\mathbf{L}_1 \to \infty$). These conclusions are confirmed by the specific comparisons shown below.

Contrasting Eq. (43) with Eq. (42) is complicated by the fact that $\boldsymbol{\Gamma}(\boldsymbol{\xi})$ is also a function of the comparison moduli, so making different choices for the comparison moduli does not only change the term (77) in Eq. (42). Furthermore, one finds that the choices $\mathbf{L}_0 = \mathbf{L}_2$ and $\mathbf{L}_0 = \mathbf{L}_1$ do *not* lead to values of the purely nonlocal term that bound the possible values of this nonlocal term, but these choices *do* lead to values of the total modulus estimate (local plus nonlocal terms) that bound all possible values of this modulus (for all choices of $\mathbf{L}_0$ lying between $\mathbf{L}_1$ and $\mathbf{L}_2$).

To show some representative quantitative results, we shall make use of the example case studied in Section 6.2 and employ the exact nonlocal constitutive equation (72), which is valid for arbitrary choice of the comparison moduli. As noted above, the braced term in Eq. (72), $\mu(\beta)$, gives the full prediction for the composite elastic shear modulus, incorporating both local and nonlocal contributions, whereas just the nonlocal term is given, for arbitrary comparison moduli, by $[\mu(\beta) - \mu(0)]$. It seems most sensible to examine how the full prediction $\mu(\beta)$ of the composite elastic shear modulus, rather than just the nonlocal term, is affected by the choice of the comparison moduli, since the local term is also significantly affected by this choice, and also since the extreme choices $\mathbf{L}_0 = \mathbf{L}_2$ and $\mathbf{L}_0 = \mathbf{L}_1$ give results that bound the full modulus prediction but not purely the nonlocal part, as noted above.

Using Eq. (72), we have calculated the full composite elastic shear modulus $\mu(\beta)$ for a sinusoidal wavelength equal to one sphere diameter, for three choices of comparison modulus: equaling the matrix modulus, the inclusion modulus, and the special choice (18) [in which case the results coincide with those of the special exact nonlocal constitutive equation (22)]. In all cases, the resulting $\mu(\beta)$ for the choice (18) lies within the results for the two other choices. Four representative plots are shown for this full composite shear modulus normalized by the matrix shear modulus; since we have already shown that the predictions will be close for small-contrast composites, we illustrate large-contrast cases. Fig. 4 shows the cases of a matrix weakened by inclusions having 1/3 and 1/10 the shear modulus of the matrix; Fig. 5 shows the cases of a matrix stiffened by inclusions having 3 and 10 times the matrix shear modulus. The matrix and inclusion Poisson's ratios all equal 0.2 in these plots; changing the Poisson's ratio has minor effects on the plots. If we regard the plots for comparison modulus = matrix modulus ($\mathbf{L}_0 = \mathbf{L}_2$) as giving the most physically realistic results (still somewhat of an open question), we observe that, in accord with the general reasoning presented above, the results from the special nonlocal constitutive equation (22) are in closest agreement with the comparison = matrix results from Eq. (72) at the highest inclusion volume fractions for both the weak and strong inclusion cases. It would seem valuable to have a simple exact nonlocal constitutive equation that is reasonably accurate at *high* inclusion concentrations; the figures show this to be the case for (22) [(43)]. Note for example that at $c_1 = 0.5$, the choice (18) is identical (obviously) to $\mathbf{L}_0 = c_1\mathbf{L}_1 + c_2\mathbf{L}_2$, not a bad choice for the comparison modulus tensor. The figures also show, in accord with prior reasoning, that the special nonlocal constitutive equation results agree significantly better in the weak inclusion cases with the comparison modulus = matrix modulus results than in the strong inclusion cases.

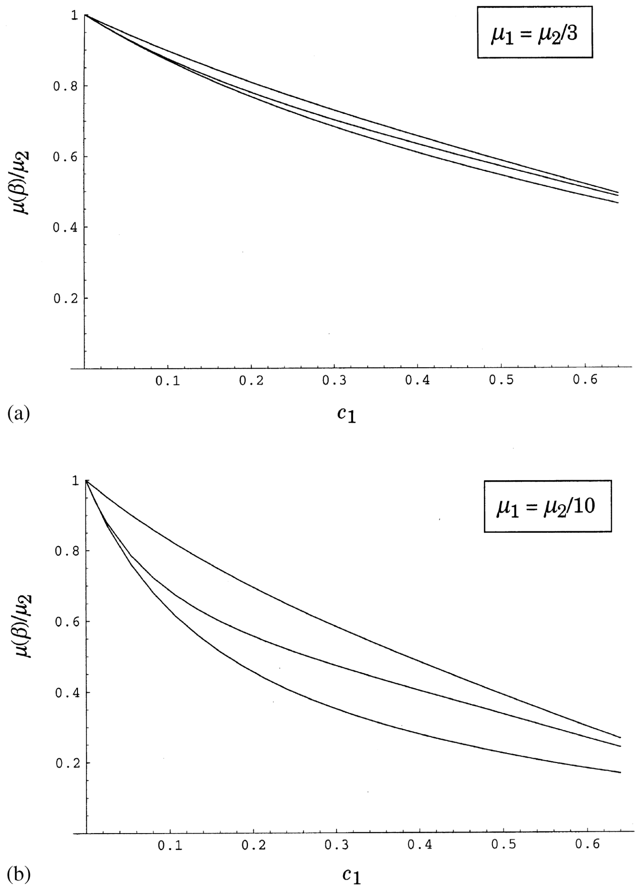


Fig. 4. Full (local plus nonlocal) composite elastic shear modulus, $\mu(\beta)$, normalized by matrix shear modulus for sinusoidal ensemble-average shear straining with wavelength equal to inclusion diameter, for the cases of inclusions having shear modulus equal to (a) 1/3 of and (b) 1/10 of the matrix shear modulus. The Poisson's ratio of all phases is 0.2. The upper curves correspond to $\mathbf{L}_0 = \mathbf{L}_2$, the lower curves to $\mathbf{L}_0 = \mathbf{L}_1$, and the intermediate curves to $\mathbf{L}_0 = c_1\mathbf{L}_2 + c_2\mathbf{L}_1$, so that the intermediate curves show the results from the special exact nonlocal constitutive equation (22) [(43)].

Finally, it seems important to emphasize that even in cases in which the special exact nonlocal constitutive equation (22) [or (35) in the case of multiple phases] is not expected to be quantitatively physically sensible due to the special comparison material choice, it remains valuable to have such an exact nonlocal constitutive equation e.g. to confirm the accuracy of numerical or approximate solution methods for nonlocal constitutive equations which permit a more realistic comparison material choice [but which can be checked by making the choice (18) and comparing with the exact result (22) or (35)].

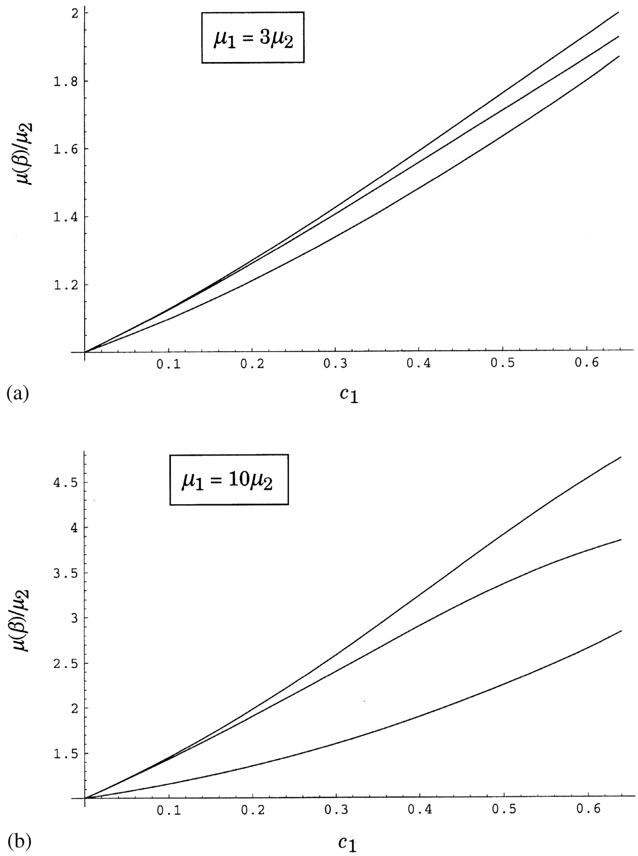


Fig. 5. Full (local plus nonlocal) composite elastic shear modulus, $\mu(\beta)$, normalized by matrix shear modulus for sinusoidal ensemble-average shear straining with wavelength equal to inclusion diameter, for the cases of inclusions having shear modulus equal to (a) 3 times and (b) 10 times the matrix shear modulus. The Poisson's ratio of all phases is 0.2. The upper curves correspond to $\mathbf{L}_0 = \mathbf{L}_1$, the lower curves to $\mathbf{L}_0 = \mathbf{L}_2$, and the intermediate curves to $\mathbf{L}_0 = c_1\mathbf{L}_2 + c_2\mathbf{L}_1$, so that the intermediate curves show the results from the special exact nonlocal constitutive equation (22) [(43)].

Acknowledgements

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