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A MICROMECHANICS-BASED NONLOCAL CONSTITUTIVE EQUATION AND ESTIMATES OF REPRESENTATIVE VOLUME ELEMENT SIZE FOR ELASTIC COMPOSITES

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ABSTRACT

A variational formulation is employed to derive a micromechanics-based, explicit nonlocal constitutive equation relating the ensemble averages of stress and strain for a class of random linear elastic composite materials. For two-phase composites with any isotropic and statistically uniform distribution of phases (which themselves may have arbitrary shape and anisotropy), we show that the leading-order correction to a macroscopically homogeneous constitutive equation involves a term proportional to the second gradient of the ensemble average of strain. This nonlocal constitutive equation is derived in explicit closed form for isotropic material in the one case in which there exists a well-founded physical and mathematical basis for describing the material's statistics: a matrix reinforced (or weakened) by a random dispersion of nonoverlapping identical spheres. By assessing, when the applied loading is spatially-varying, the magnitude of the nonlocal term in this constitutive equation compared to the portion of the equation that relates ensemble average stresses and strains through a constant "overall" modulus tensor, we derive quantitative estimates for the minimum representative volume element (RVE) size, defined here as that over which the usual macroscopically homogeneous "effective modulus" constitutive models for composites can be expected to apply. Remarkably, for a maximum error of 5% of the constant "overall" modulus term, we show that the minimum RVE size is at most twice the reinforcement diameter for any reinforcement concentration level, for several sets of matrix and reinforcement moduli characterizing large classes of important structural materials. Such estimates seem essential for determining the minimum structural component size that can be treated by macroscopically homogeneous composite material constitutive representations, and also for the development of a fundamentally-based macroscopic fracture mechanics theory for composites. Finally, we relate our nonlocal constitutive equation explicitly to the ensemble average strain energy, and show how it is consistent with the stationary energy principle.

1. INTRODUCTION

A powerful approach to the mathematical modeling of the stress and deformation response of composite materials when subjected to applied loading is one that seeks to derive information on the macroscopic, or overall, constitutive behavior. In this approach, details of the (generally) complex, strongly heterogeneous microstructure

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are not considered directly, which would inevitably involve enormous numerical calculations, would be specimen-specific, and from which generalized information would be difficult to elicit. (Some progress has been made numerically by assuming periodic microstructures, but such are difficult to realize in practice.) Instead, one analyzes material elements having a length scale (the macroscale) that is sufficiently large compared to the microstructural length scale (the microscale) that the material can be accurately treated as being homogeneous with spatially constant "average" or "overall" properties. The most firmly based of such approaches are those employing variational principles which supply strict upper and lower bounds on the overall constitutive moduli. Perhaps the most widely and profitably used such principles are those of Hashin and Shtrikman (1962a, b), as summarized and generalized by Willis (1977, 1981, 1982, 1983). The latter three articles are also reviews that provide detailed background and reference related work by other authors.

A question of fundamental importance to the application of such constitutive models that describe the composite response via homogeneous "overall" properties is the minimum size of a material representative volume element (RVE). There appear to be two significantly different ways to define "representative volume element". One definition arises from the perspective that in order to characterize macroscopic composite constitutive response, one must recognize the statistical nature of the microstructure of actual composites. This perspective leads to the conclusion that the smallest RVE for which a macroscopic "effective" constitutive theory could apply is one that is sufficiently large to be statistically representative of the composite—that is, to include effectively a sampling of all possible microstructural configurations that occur in the composite. This is the perspective generally adopted, and it leads to statements of the type that the RVE must include a very large number of the composite's microheterogeneities (such as grains, inclusions, voids, cracks, fibers, etc.). Such statements are invariably qualitative; thus articles on the topic of overall composite response generally state that the RVE must be chosen "sufficiently large" compared to the microstructural size for the approach to be valid.

There is another, perhaps more pragmatic, definition of "representative volume element": the smallest material volume element of the composite for which the usual spatially constant "overall modulus" macroscopic constitutive representation is a sufficiently accurate model to represent mean constitutive response. *We adopt this definition for RVE in the present paper.* [It is important to emphasize that attention here is focused on the mean response (defined precisely in Section 2), upon which possibly large fluctuations associated with local microstructural detail are superimposed.] In remarkable contrast to the large RVE sizes (with respect to microconstituent size) implied by the statistical perspective described in the first definition above, it will emerge that quantitative estimates of RVE size for the definition we adopt here are very much smaller than these, for the entire range of reinforcement concentration level. An important related question is: When loading applied to the composite generates, in material modeled by an "overall modulus" constitutive relation, stress and strain fields that vary too rapidly for consistency with the minimum RVE size introduced above, what form should the macroscopic constitutive equation take?

In the present work, we develop an explicit nonlocal macroscopic constitutive model based on the micromechanics of the composite material from a variational characterization of Hashin–Shtrikman type. We show that for two-phase composites with any isotropic and statistically uniform *distribution* of phases (which themselves may have arbitrary shape and anisotropy), the leading-order correction to a macroscopically homogeneous constitutive equation consists of an additional term proportional to the second gradient of the ensemble average of strain. We obtain an explicit closed-form expression of this nonlocal constitutive equation when the phases are isotropic, in terms of an integral of the two-point distribution function of the phases. This is explicitly evaluated in the only case for which a physically and mathematically sensible model for the two-point distribution function is available: a matrix reinforced (or weakened) by a random dispersion of nonoverlapping spheres. In this case, Markov and Willis (1995) have shown how to express the two-point distribution function in terms of a single integral involving the radial distribution function for the sphere centers. We evaluate this integral in closed form when the radial distribution function is that derived by Wertheim (1963) from the statistical mechanics model of Percus and Yevick (1958). The resulting nonlocal constitutive equation is then employed to deduce explicit quantitative estimates for the minimum RVE size needed for the macroscopic constitutive response to be represented accurately by the usual homogeneous overall modulus.

The only related work of which we are aware [apart from a very compressed general discussion by Willis (1983)] is that of Diener *et al.* (1984), who derived bounds on the *Fourier transform* of the nonlocal overall operator that relates the ensemble averages of stress and strain, in the special case of a two-phase isotropic composite comprised of nearly spherical grains of equal size, with two-point probabilities chosen on the basis of a simple "cell-structure" model. Also, Willis (1985) analyzed the nonlocal influence of density variations in an otherwise homogeneous medium, assuming a simple exponential form for the two-point correlation function.

The plan of the paper is as follows. In Section 2 we summarize Willis' (1977) concise derivation of the Hashin–Shtrikman variational principle for deterministic composite microstructures, and then his (Willis, 1977, 1982, 1983) generalization of this to treat random microstructures, all adapted to the infinite medium to be analyzed here. This permits formulation, in general terms, of the problem of determining a nonlocal constitutive equation, which involves the solution of a set of integral equations.

Section 3 lays out our derivation of the nonlocal constitutive equation for two-phase composites, proceeding in stages from the general situation (arbitrary phase distribution and shape, arbitrary component material anisotropy) to the one specific class for which we believe there is sufficient physical and mathematical basis for deriving explicit closed-form results: an isotropic matrix reinforced (or weakened) by a random distribution of nonoverlapping isotropic spheres of a different material.

We employ this explicit nonlocal constitutive equation in Section 4 to produce *quantitative* estimates of the minimum RVE size for which the usual constant-effective-modulus constitutive equation accurately represents composite response, by comparing the magnitude of the nonlocal to local terms in our new constitutive equation when spatially varying loading is applied. This is done for a very wide range of reinforcement concentrations, and for the bounding extremes of the "reinforcing" spheres being voids and rigid particles while the matrix material is permitted to adopt several sets of elastic moduli values that encompass large classes of structural materials. We also analyze the specific systems of an aluminum matrix reinforced by alumina particles and vice-versa.

Finally, in Section 5 we employ our nonlocal constitutive equation to derive an explicit expression for the ensemble average strain energy, for random two-phase composites with isotropic distributions of phases. We also show how the nonlocal constitutive equation is consistent with the stationary energy principle.

2. FORMULATION

We will analyze linear elastic composite materials with firmly-bonded phases. The approach will be to employ variational principles that describe the overall response of *random* composites. These are generalizations, developed by Willis (1977, 1981, 1982, 1983), to the Hashin–Shtrikman (1962a, b) variational principles.

We shall first summarize Willis' (1977) derivation of the Hashin–Shtrikman principle for any specific composite material, and then show how this is adapted to analyze random composites. Because we are concerned with relating macroscopic to local *constitutive* response under spatially-varying applied loading, and to facilitate explicit solutions, we consider an infinite body with the only applied loading being a body force vector field $\mathbf{f}(\mathbf{x})$ that decays sufficiently rapidly when $|\mathbf{x}|$ is large, where $\mathbf{x}$ is the position vector and $|\mathbf{x}|$ its magnitude. The equations governing the equilibrium stress and deformation fields in such a linear elastic composite material are then

$$\nabla \cdot \boldsymbol{\sigma} + \mathbf{f} = \mathbf{0} \quad [\partial \sigma_{ij}/\partial x_i + f_j = 0] \tag{1a}$$

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

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

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 which is a function of position $\mathbf{x}$ due to local heterogeneity, sym denotes the symmetric part and, throughout the paper, lower-case Latin subscripts denote Cartesian components and obey the Einstein summation convention (except where noted).

An effective solution approach to system (1) is to reformulate by introduction of 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

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

where $\boldsymbol{\tau}$ is the "stress polarization" tensor defined as

$$\boldsymbol{\tau} \equiv (\mathbf{L} - \mathbf{L}_0)\mathbf{e}. \tag{3}$$

Substitution of (2) into (1a) gives

$$\nabla \cdot (\mathbf{L}_0 \mathbf{e}) + (\nabla \cdot \boldsymbol{\tau} + \mathbf{f}) = \mathbf{0}, \tag{4}$$

the solution of which for $\mathbf{e}$ is (adapting Willis, 1977)

$$\mathbf{e}(\mathbf{x}) = \mathbf{e}_0(\mathbf{x}) - \int_\Omega \mathbf{\Gamma}_0(\mathbf{x}-\mathbf{x}')\boldsymbol{\tau}(\mathbf{x}')\,\mathrm{d}\mathbf{x}', \tag{5}$$

where

$$[\mathbf{\Gamma}_0(\mathbf{x}-\mathbf{x}')]_{ijkl} = \left.\frac{\partial^2[\mathbf{G}_0(\mathbf{x}-\mathbf{x}')]_{jk}}{\partial x_i\,\partial x_l'}\right|_{(ij),(kl)}, \tag{6}$$

the notation indicates symmetrization on (ij) and (kl), Ω denotes the (infinite) domain of the body, and the singularity of $\mathbf{\Gamma}_0$ is interpreted in the sense of generalized functions. Here, $\mathbf{G}_0(\mathbf{x})$ is the infinite-homogeneous-body Green's function, whose components are defined by the differential equation

$$\frac{\partial^2[\mathbf{G}_0(\mathbf{x})]_{jm}}{\partial x_i\,\partial x_l}c_{ijkl} + \delta_{km}\,\delta(\mathbf{x}) = 0, \tag{7}$$

where c_{ijkl} are the components of $\mathbf{L}_0$, δ_{km} is the Kronecker delta and $\delta(\mathbf{x})$ is the three-dimensional Dirac delta function. Eliminating $\mathbf{e}$ in (5) via (3) gives

$$(\mathbf{L}(\mathbf{x})-\mathbf{L}_0)^{-1}\boldsymbol{\tau}(\mathbf{x}) + \int_\Omega \mathbf{\Gamma}_0(\mathbf{x}-\mathbf{x}')\boldsymbol{\tau}(\mathbf{x}')\,\mathrm{d}\mathbf{x}' = \mathbf{e}_0(\mathbf{x}). \tag{8}$$

Willis (1977) observed that the self-adjointness of (8) leads directly to the Hashin–Shtrikman variational principle

$$\delta\mathscr{H}(\boldsymbol{\tau}) = 0, \tag{9}$$

where $\mathscr{H}(\boldsymbol{\tau}^*)$ is the functional

$$\mathscr{H}(\boldsymbol{\tau}^*) \equiv \int_\Omega \left[\boldsymbol{\tau}^*(\mathbf{x})(\mathbf{L}(\mathbf{x})-\mathbf{L}_0)^{-1}\boldsymbol{\tau}^*(\mathbf{x}) + \boldsymbol{\tau}^*(\mathbf{x})\int_\Omega \mathbf{\Gamma}_0(\mathbf{x}-\mathbf{x}')\boldsymbol{\tau}^*(\mathbf{x}')\,\mathrm{d}\mathbf{x}' - 2\boldsymbol{\tau}^*(\mathbf{x})\mathbf{e}_0(\mathbf{x})\right]\mathrm{d}\mathbf{x}, \tag{10}$$

and $\boldsymbol{\tau}^*$ is any choice for $\boldsymbol{\tau}$. It follows via (8) that the stationary value of $\mathscr{H}$, attained when $\boldsymbol{\tau}^* = \boldsymbol{\tau}$, is

$$\mathscr{H}(\boldsymbol{\tau}) = -\int_\Omega \boldsymbol{\tau}(\mathbf{x})\mathbf{e}_0(\mathbf{x})\,\mathrm{d}\mathbf{x}. \tag{11}$$

The above formulation provides a method for finding solutions to the stress and deformation fields *locally* in an inhomogeneous composite (but such problems are, in general, prohibitively difficult). To adapt the formulation to provide estimates for macroscopic, mean or ensemble average fields, we follow Willis (1977, 1983) and treat random composites. We identify by α individual members of a sample space $\mathscr{S}$, 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 $\mathbf{x}$ [that is, the *ensemble average* of $\chi_r(\mathbf{x};\alpha)$] is

$$P_r(\mathbf{x}) = \langle \chi_r(\mathbf{x}) \rangle \equiv \int_{\mathscr{S}} \chi_r(\mathbf{x};\alpha) p(\alpha)\, \mathrm{d}\alpha, \tag{12}$$

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

$$P_{rs}(\mathbf{x}, \mathbf{x}') = \langle \chi_r(\mathbf{x}) \chi_s(\mathbf{x}') \rangle \equiv \int_{\mathscr{S}} \chi_r(\mathbf{x};\alpha) \chi_s(\mathbf{x}';\alpha) p(\alpha)\, \mathrm{d}\alpha. \tag{13}$$

If each phase r is homogeneous with moduli $\mathbf{L}_r$, where $r = 1, 2, \ldots, n$, then $\mathbf{L}(\mathbf{x})$ of (1c) in sample α, and its ensemble average, are

$$\mathbf{L}(\mathbf{x};\alpha) = \sum_{r=1}^{n} \mathbf{L}_r \chi_r(\mathbf{x};\alpha) \Rightarrow \langle \mathbf{L}(\mathbf{x}) \rangle = \sum_{r=1}^{n} \mathbf{L}_r P_r(\mathbf{x}). \tag{14}$$

Now, the variational approach summarized above is applied by choosing the most general trial fields for $\boldsymbol{\tau}^*$ allowed by restriction to one- and two-point correlations in (10) (see Willis, 1982), since higher-order correlations are extremely difficult to determine in practice

$$\boldsymbol{\tau}^*(\mathbf{x};\alpha) = \sum_{r=1}^{n} \boldsymbol{\tau}_r(\mathbf{x}) \chi_r(\mathbf{x};\alpha). \tag{15}$$

We further assume the material to be statistically uniform, meaning that $P_r(\mathbf{x})$ and $P_{rs}(\mathbf{x}, \mathbf{x}')$ are insensitive to translations; thus $P_r(\mathbf{x})$ reduces to a constant, P_r, and $P_{rs}(\mathbf{x}, \mathbf{x}') = P_{rs}(\mathbf{x} - \mathbf{x}')$. For such materials, one typically makes 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. In this case, P_r becomes the volume average of $\chi_r(\mathbf{x})$ and thus is simply the volume concentration c_r of phase r. Then, substitution of $(14)_1$ and (15) into (10) and ensemble averaging gives

$$\begin{aligned} \langle \mathscr{H}(\boldsymbol{\tau}^*) \rangle = {} & \sum_{r=1}^{n} c_r \int_{\Omega} \boldsymbol{\tau}_r(\mathbf{x}) [(\mathbf{L}_r - \mathbf{L}_0)^{-1} \boldsymbol{\tau}_r(\mathbf{x}) - 2\mathbf{e}_0(\mathbf{x})]\, \mathrm{d}\mathbf{x} \\ & + \sum_{r=1}^{n} \sum_{s=1}^{n} \int_{\Omega} \boldsymbol{\tau}_r(\mathbf{x}) \left[\int_{\Omega} \boldsymbol{\Gamma}_0(\mathbf{x} - \mathbf{x}') \boldsymbol{\tau}_s(\mathbf{x}') P_{rs}(\mathbf{x} - \mathbf{x}')\, \mathrm{d}\mathbf{x}' \right] \mathrm{d}\mathbf{x}. \end{aligned} \tag{16}$$

This is stationary when

$$(\mathbf{L}_r - \mathbf{L}_0)^{-1} \boldsymbol{\tau}_r(\mathbf{x}) c_r + \sum_{s=1}^{n} \int_{\Omega} \boldsymbol{\Gamma}_0(\mathbf{x} - \mathbf{x}') \boldsymbol{\tau}_s(\mathbf{x}') P_{rs}(\mathbf{x} - \mathbf{x}')\, \mathrm{d}\mathbf{x}' = \mathbf{e}_0(\mathbf{x}) c_r, \quad r = 1, 2, \ldots, n. \tag{17}$$

Substituting (15) (for $\boldsymbol{\tau}$) into (5) and ensemble averaging yields the approximation

$$\langle \mathbf{e} \rangle(\mathbf{x}) = \mathbf{e}_0(\mathbf{x}) - \sum_{s=1}^{n} c_s \int_\Omega \mathbf{\Gamma}_0(\mathbf{x}-\mathbf{x}')\boldsymbol{\tau}_s(\mathbf{x}')\,\mathrm{d}\mathbf{x}', \tag{18}$$

substitution of which into (17) gives finally

$$(\mathbf{L}_r - \mathbf{L}_0)^{-1}\boldsymbol{\tau}_r(\mathbf{x})c_r + \sum_{s=1}^{n} \int_\Omega \mathbf{\Gamma}_0(\mathbf{x}-\mathbf{x}')[P_{rs}(\mathbf{x}-\mathbf{x}') - c_r c_s]\boldsymbol{\tau}_s(\mathbf{x}')\,\mathrm{d}\mathbf{x}' = \langle \mathbf{e} \rangle(\mathbf{x})c_r, \quad r = 1,2,\ldots,n. \tag{19}$$

Observe that (19) is a set of n integral equations for $\boldsymbol{\tau}_r(\mathbf{x})$ in terms of $\langle \mathbf{e} \rangle(\mathbf{x})$.

Our goal is to find a constitutive equation that relates $\langle \boldsymbol{\sigma} \rangle(\mathbf{x})$ to $\langle \mathbf{e} \rangle(\mathbf{x})$, valid when these do indeed vary with $\mathbf{x}$. Taking the ensemble average of (2) gives

$$\langle \boldsymbol{\sigma} \rangle(\mathbf{x}) = \mathbf{L}_0 \langle \mathbf{e} \rangle(\mathbf{x}) + \langle \boldsymbol{\tau} \rangle(\mathbf{x}). \tag{20}$$

Approximating $\boldsymbol{\tau}(\mathbf{x};\alpha)$ by (15), ensemble averaging of that equation yields

$$\langle \boldsymbol{\tau} \rangle(\mathbf{x}) = \sum_{r=1}^{n} c_r \boldsymbol{\tau}_r(\mathbf{x}). \tag{21}$$

Thus our desired constitutive equation is (20) with (21), so we must solve the integral equations (19) for $\boldsymbol{\tau}_r(\mathbf{x})$.

3. DERIVATION OF A NONLOCAL CONSTITUTIVE EQUATION FOR TWO-PHASE COMPOSITES

3.1. *General structure*

To facilitate explicit results, we will now restrict the analysis to two-phase composites, for which (see Willis, 1982)

$$P_{rs}(\mathbf{x}-\mathbf{x}') - c_r c_s = c_r(\delta_{rs} - c_s)h(\mathbf{x}-\mathbf{x}'), \quad \text{(no sum)} \tag{22}$$

where $h(\mathbf{x}-\mathbf{x}')$ is the two-point correlation function. Then (19) becomes

$$(\mathbf{L}_r - \mathbf{L}_0)^{-1}\boldsymbol{\tau}_r(\mathbf{x})c_r + \sum_{s=1}^{2} c_r(\delta_{rs} - c_s) \int_\Omega [\mathbf{\Gamma}_0(\mathbf{x}-\mathbf{x}')h(\mathbf{x}-\mathbf{x}')]\boldsymbol{\tau}_s(\mathbf{x}')\,\mathrm{d}\mathbf{x}' = c_r \langle \mathbf{e} \rangle(\mathbf{x}), \quad r = 1,2. \tag{23}$$

We will employ Fourier transforms to solve (23). The three-dimensional Fourier transform of a function $f(\mathbf{x})$ [which decays sufficiently rapidly for $|\mathbf{x}| \to \infty$ for convergence of (24)] and its inverse are defined as

$$\tilde{f}(\boldsymbol{\xi}) \equiv \int_\Omega f(\mathbf{x})\,\mathrm{e}^{\mathrm{i}\boldsymbol{\xi}\cdot\mathbf{x}}\,\mathrm{d}\mathbf{x}, \quad f(\mathbf{x}) = \frac{1}{8\pi^3}\int_\Omega \tilde{f}(\boldsymbol{\xi})\mathrm{e}^{-\mathrm{i}\boldsymbol{\xi}\cdot\mathbf{x}}\,\mathrm{d}\boldsymbol{\xi}, \tag{24}$$

where $\mathrm{i} = \sqrt{-1}$, $\boldsymbol{\xi}$ is a vector, and $\boldsymbol{\xi}\cdot\mathbf{x}$ denotes scalar product. Thus, taking the 3-D Fourier transform of (23) gives

$$(\mathbf{L}_r-\mathbf{L}_0)^{-1}\tilde{\boldsymbol{\tau}}_r(\boldsymbol{\xi})c_r+\sum_{s=1}^{2}c_r(\delta_{rs}-c_s)(\tilde{\boldsymbol{\Gamma}}_0*\tilde{h})(\boldsymbol{\xi})\tilde{\boldsymbol{\tau}}_s(\boldsymbol{\xi}) = c_r\langle\tilde{\mathbf{e}}\rangle(\boldsymbol{\xi}),\quad r=1,2,\tag{25}$$

having noted that the integral term in (23) is a convolution, and that the Fourier transform of the bracketed term is itself the convolution

$$(\tilde{\boldsymbol{\Gamma}}_0*\tilde{h})(\boldsymbol{\xi})\equiv\frac{1}{8\pi^3}\int_{\Omega}\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi}-\boldsymbol{\xi}')\tilde{h}(\boldsymbol{\xi}')\,\mathrm{d}\boldsymbol{\xi}'\equiv\boldsymbol{\Gamma},\tag{26}$$

which will, as indicated, henceforth be written for compactness simply as $\boldsymbol{\Gamma}$. Rewriting (25) as

$$\sum_{s=1}^{2}\tilde{\mathbf{T}}_{rs}^{-1}(\boldsymbol{\xi})\tilde{\boldsymbol{\tau}}_s(\boldsymbol{\xi}) = c_r\langle\tilde{\mathbf{e}}\rangle(\boldsymbol{\xi}),\quad r=1,2\tag{27}$$

where

$$\tilde{\mathbf{T}}_{rs}^{-1}(\boldsymbol{\xi})\equiv(\mathbf{L}_r-\mathbf{L}_0)^{-1}c_r\,\delta_{rs}+c_r(\delta_{rs}-c_s)\boldsymbol{\Gamma},\quad\text{(no sum)}\tag{28}$$

(27) can be solved for $\tilde{\boldsymbol{\tau}}_r(\boldsymbol{\xi})$ to give

$$\tilde{\boldsymbol{\tau}}_r(\boldsymbol{\xi}) = \sum_{s=1}^{2}\tilde{\mathbf{T}}_{rs}(\boldsymbol{\xi})c_s\langle\tilde{\mathbf{e}}\rangle(\boldsymbol{\xi}),\quad r=1,2.\tag{29}$$

Thus we must invert (28). Doing so, noting carefully that it is a matrix of 4th-rank tensors whose products do not commute, we find

$$\tilde{\mathbf{T}}_{rs}(\boldsymbol{\xi}) = \delta\mathbf{L}_r(\boldsymbol{\Gamma}^{-1}+c_1\,\delta\mathbf{L}_2+c_2\,\delta\mathbf{L}_1)^{-1}\left[\frac{\delta_{rs}}{c_s}\boldsymbol{\Gamma}^{-1}+\delta\mathbf{L}_1+\delta\mathbf{L}_2-\delta\mathbf{L}_r\right],\quad\text{(no sum)}\tag{30}$$

where here and in the sequel,

$$\delta\mathbf{L}_r\equiv\mathbf{L}_r-\mathbf{L}_0.\tag{31}$$

Now the Fourier transform of (21) is, for two-phase composites,

$$\langle\tilde{\boldsymbol{\tau}}\rangle(\boldsymbol{\xi}) = \sum_{r=1}^{2}c_r\tilde{\boldsymbol{\tau}}_r(\boldsymbol{\xi}).\tag{32}$$

Substituting (29) into this gives

$$\langle\tilde{\boldsymbol{\tau}}\rangle(\boldsymbol{\xi}) = \sum_{r=1}^{2}\sum_{s=1}^{2}c_r\tilde{\mathbf{T}}_{rs}(\boldsymbol{\xi})c_s\langle\tilde{\mathbf{e}}\rangle(\boldsymbol{\xi})\equiv\langle\tilde{\mathbf{T}}\rangle(\boldsymbol{\xi})\langle\tilde{\mathbf{e}}\rangle(\boldsymbol{\xi}).\tag{33}$$

Employing (30) to carry out the sum appearing in (33) results in

$$\begin{aligned}\langle\tilde{\mathbf{T}}\rangle(\boldsymbol{\xi}) &= c_1\,\delta\mathbf{L}_1(\boldsymbol{\Gamma}^{-1}+c_1\,\delta\mathbf{L}_2+c_2\,\delta\mathbf{L}_1)^{-1}(\boldsymbol{\Gamma}^{-1}+\delta\mathbf{L}_2)\\&\quad+c_2\,\delta\mathbf{L}_2(\boldsymbol{\Gamma}^{-1}+c_1\,\delta\mathbf{L}_2+c_2\,\delta\mathbf{L}_1)^{-1}(\boldsymbol{\Gamma}^{-1}+\delta\mathbf{L}_1).\end{aligned}\tag{34}$$

Finally, to obtain $\langle\boldsymbol{\tau}\rangle(\mathbf{x})$, as (20) shows is needed for the macroscopic constitutive equation, take the inverse Fourier transform of (33)

$$\langle\boldsymbol{\tau}\rangle(\mathbf{x}) = \int_\Omega \langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')\langle\mathbf{e}\rangle(\mathbf{x}')\,\mathrm{d}\mathbf{x}'. \tag{35}$$

The actual evaluation of the right-hand side of (35) is extremely difficult for general $\langle\mathbf{e}\rangle(\mathbf{x}')$. Since we desire an explicit representation for $\langle\boldsymbol{\tau}\rangle(\mathbf{x})$, we will approximate $\langle\mathbf{e}\rangle(\mathbf{x}')$ by the first three terms of its Taylor expansion

$$\begin{aligned}\langle\mathbf{e}\rangle(\mathbf{x}') &\approx \langle\mathbf{e}\rangle(\mathbf{x}) + (\mathbf{x}'-\mathbf{x})\boldsymbol{\nabla}\langle\mathbf{e}\rangle(\mathbf{x}) + \tfrac{1}{2}(\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})\boldsymbol{\nabla}\boldsymbol{\nabla}\langle\mathbf{e}\rangle(\mathbf{x}) \\ &= \langle\mathbf{e}\rangle(\mathbf{x}) + (x_i'-x_i)\frac{\partial}{\partial x_i}\langle\mathbf{e}\rangle(\mathbf{x}) + \tfrac{1}{2}(x_j'-x_j)(x_k'-x_k)\frac{\partial^2}{\partial x_j\,\partial x_k}\langle\mathbf{e}\rangle(\mathbf{x}).\end{aligned} \tag{36}$$

Then (35) becomes

$$\begin{aligned}\langle\boldsymbol{\tau}\rangle(\mathbf{x}) &= \left[\int_\Omega \langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')\,\mathrm{d}\mathbf{x}'\right]\langle\mathbf{e}\rangle(\mathbf{x}) + \left[\int_\Omega \langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')(\mathbf{x}'-\mathbf{x})\,\mathrm{d}\mathbf{x}'\right]\boldsymbol{\nabla}\langle\mathbf{e}\rangle(\mathbf{x}) \\ &\quad + \left[\int_\Omega \langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')(\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})\,\mathrm{d}\mathbf{x}'\right]\tfrac{1}{2}\boldsymbol{\nabla}\boldsymbol{\nabla}\langle\mathbf{e}\rangle(\mathbf{x}).\end{aligned} \tag{37}$$

The bracketed integrals in (37) fortunately do not require that the inverse Fourier transform of $\langle\tilde{\mathbf{T}}\rangle(\boldsymbol{\xi})$ be determined explicitly; instead, notice from $(24)_1$ that

$$\int_\Omega \langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')\,\mathrm{d}\mathbf{x}' = \int_\Omega \langle\mathbf{T}\rangle(\mathbf{x})\,\mathrm{d}\mathbf{x} = \langle\tilde{\mathbf{T}}\rangle(\boldsymbol{\xi}=\mathbf{0}), \tag{38}$$

$$\int_\Omega \langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')(\mathbf{x}'-\mathbf{x})\,\mathrm{d}\mathbf{x}' = -\int_\Omega \langle\mathbf{T}\rangle(\mathbf{x})\mathbf{x}\,\mathrm{d}\mathbf{x} = i(\boldsymbol{\nabla}_\xi\langle\tilde{\mathbf{T}}\rangle)(\boldsymbol{\xi}=\mathbf{0}), \tag{39}$$

$$\int_\Omega \langle\mathbf{T}\rangle(\mathbf{x}-\mathbf{x}')(\mathbf{x}'-\mathbf{x})(\mathbf{x}'-\mathbf{x})\,\mathrm{d}\mathbf{x}' = \int_\Omega \langle\mathbf{T}\rangle(\mathbf{x})\mathbf{x}\mathbf{x}\,\mathrm{d}\mathbf{x} = -(\boldsymbol{\nabla}_\xi\boldsymbol{\nabla}_\xi\langle\tilde{\mathbf{T}}\rangle)(\boldsymbol{\xi}=\mathbf{0}), \tag{40}$$

where $\boldsymbol{\nabla}_\xi = \partial/\partial\boldsymbol{\xi}$.

3.2. *Comparison with previous results for constant* $\langle\mathbf{e}\rangle$

Before obtaining explicit new results for the general case when $\langle\mathbf{e}\rangle(\mathbf{x})$ does indeed vary with $\mathbf{x}$, let us make contact with previously published results for the case when $\langle\mathbf{e}\rangle$ does *not* vary with position. In this case, (35) reduces precisely to (37) with only the first right-hand-side term, and combining this with (20) and (38) gives

$$\langle\boldsymbol{\sigma}\rangle = \mathbf{L}_0\langle\mathbf{e}\rangle + \langle\boldsymbol{\tau}\rangle = [\mathbf{L}_0 + \langle\tilde{\mathbf{T}}\rangle(\mathbf{0})]\langle\mathbf{e}\rangle \equiv \hat{\mathbf{L}}\langle\mathbf{e}\rangle, \tag{41}$$

where this determines $\hat{\mathbf{L}}$, the constant "overall" or "effective" modulus tensor. To evaluate $\langle\tilde{\mathbf{T}}\rangle(\mathbf{0})$, examination of (34) shows that we will need to evaluate $\boldsymbol{\Gamma}$ at $\boldsymbol{\xi}=\mathbf{0}$, which from (26) is

$$\boldsymbol{\Gamma}(\mathbf{0}) = (\tilde{\boldsymbol{\Gamma}}_0 * \tilde{h})(\mathbf{0}) = \frac{1}{8\pi^3}\int_\Omega \tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi}')\tilde{h}(\boldsymbol{\xi}')\,\mathrm{d}\boldsymbol{\xi}' = \left[\frac{1}{8\pi^3}\int_\Omega \tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\tilde{h}(\boldsymbol{\xi})\mathrm{e}^{-i\boldsymbol{\xi}\cdot\mathbf{x}}\,\mathrm{d}\boldsymbol{\xi}\right]_{\mathbf{x}=0}$$

$$= \left[\int_\Omega \boldsymbol{\Gamma}_0(\mathbf{x}-\mathbf{x}')h(\mathbf{x}')\,\mathrm{d}\mathbf{x}'\right]_{\mathbf{x}=0} = \int_\Omega \boldsymbol{\Gamma}_0(\mathbf{x}')h(\mathbf{x}')\,\mathrm{d}\mathbf{x}' \equiv \mathbf{P}'. \tag{42}$$

Here we have used the Fourier transform of a convolution, and the facts that $\boldsymbol{\Gamma}_0(-\mathbf{x}) = \boldsymbol{\Gamma}_0(\mathbf{x})$ and $\tilde{\boldsymbol{\Gamma}}_0(-\boldsymbol{\xi}) = \tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})$. Thus, from (41) with (34) and (42), we find the effective modulus tensor for any two-phase composite with constant $\langle \mathbf{e} \rangle$ but arbitrary phase geometry, distribution and anisotropy to be

$$\boxed{\begin{aligned}\hat{\mathbf{L}} = \mathbf{L}_0 + c_1\,\delta\mathbf{L}_1(\mathbf{P}'^{-1} + c_1\,\delta\mathbf{L}_2 + c_2\,\delta\mathbf{L}_1)^{-1}(\mathbf{P}'^{-1} + \delta\mathbf{L}_2)\\ + c_2\,\delta\mathbf{L}_2(\mathbf{P}'^{-1} + c_1\,\delta\mathbf{L}_2 + c_2\,\delta\mathbf{L}_1)^{-1}(\mathbf{P}'^{-1} + \delta\mathbf{L}_1).\end{aligned}} \tag{43}$$

Willis (1982) employed a Radon transform approach to derive a Hashin–Shtrikman estimate for $\hat{\mathbf{L}}$ when $\langle \mathbf{e} \rangle$ is constant, for the same very general class of two-phase composites as that just described. He found [(6.21) and (6.22)], with $\mathbf{P}'$ defined as in (42),

$$\begin{aligned}
\hat{\mathbf{L}} &= \left\{\sum_{r=1}^{2} c_r[\mathbf{I} + \delta\mathbf{L}_r\mathbf{P}']^{-1}\right\}^{-1} \sum_{s=1}^{2} c_s[\mathbf{I} + \delta\mathbf{L}_s\mathbf{P}']^{-1}\mathbf{L}_s \\
&= \mathbf{L}_0 + \left\{\sum_{r=1}^{2} c_r[\mathbf{I} + \delta\mathbf{L}_r\mathbf{P}']^{-1}\right\}^{-1} \sum_{s=1}^{2} c_s[\mathbf{I} + \delta\mathbf{L}_s\mathbf{P}']^{-1}\,\delta\mathbf{L}_s \\
&= \mathbf{L}_0 + (\mathbf{P}'^{-1} + \delta\mathbf{L}_2)(\mathbf{P}'^{-1} + c_1\,\delta\mathbf{L}_2 + c_2\,\delta\mathbf{L}_1)^{-1}c_1\,\delta\mathbf{L}_1 \\
&\quad + (\mathbf{P}'^{-1} + \delta\mathbf{L}_1)(\mathbf{P}'^{-1} + c_1\,\delta\mathbf{L}_2 + c_2\,\delta\mathbf{L}_1)^{-1}c_2\,\delta\mathbf{L}_2 \\
&= \mathbf{L}_0 + [(\mathbf{P}'^{-1} + c_1\,\delta\mathbf{L}_2 + c_2\,\delta\mathbf{L}_1) + c_2(\delta\mathbf{L}_2 - \delta\mathbf{L}_1)](\mathbf{P}'^{-1} + c_1\,\delta\mathbf{L}_2 + c_2\,\delta\mathbf{L}_1)^{-1}c_1\,\delta\mathbf{L}_1 \\
&\quad + [(\mathbf{P}'^{-1} + c_1\,\delta\mathbf{L}_2 + c_2\,\delta\mathbf{L}_1) + c_1(\delta\mathbf{L}_1 - \delta\mathbf{L}_2)](\mathbf{P}'^{-1} + c_1\,\delta\mathbf{L}_2 + c_2\,\delta\mathbf{L}_1)^{-1}c_2\,\delta\mathbf{L}_2,
\end{aligned} \tag{44}$$

which, multiplying out the products and collecting all terms premultiplied by $\delta\mathbf{L}_1$ and all premultiplied by $\delta\mathbf{L}_2$, can be shown to reduce precisely to (43). Here, $\mathbf{I}$ is the fourth-rank identity tensor. Thus, we have confirmed that our results from the present Fourier transform approach agree with those from Willis' (1982) alternate Radon transform approach when $\langle \mathbf{e} \rangle$ is constant.

3.3. *Results when phase distribution is isotropic*

Now, to facilitate more explicit results for the general case that $\langle \mathbf{e} \rangle(\mathbf{x})$ *does* vary with $\mathbf{x}$, we henceforth restrict attention to composites that consist of isotropic distributions of phases, although the phases themselves may still have arbitrary anisotropy and shape. This means that the two-point correlation function $h(\mathbf{x})$ introduced in (22) satisfies

$$h(\mathbf{x}) = h(|\mathbf{x}|) \Rightarrow \tilde{h}(\boldsymbol{\xi}) = \tilde{h}(|\boldsymbol{\xi}|). \tag{45}$$

In order to evaluate (37), and hence (38)–(40), these and (34) show that we need explicit expressions for $\boldsymbol{\Gamma}$ and its first and second derivatives with respect to $\boldsymbol{\xi}$, evaluated at $\boldsymbol{\xi} = \mathbf{0}$. [Our earlier result (42) for $\boldsymbol{\Gamma}(\mathbf{0})$ will simplify now in view of (45).] The derivations of these are detailed in the Appendix, where we find

$$\boldsymbol{\Gamma}(\mathbf{0}) = \frac{1}{4\pi}\int_{|\boldsymbol{\xi}|=1} \tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\,\mathrm{d}S \equiv \mathbf{P}, \tag{46}$$

$$\frac{\partial \boldsymbol{\Gamma}}{\partial \xi_m}(\mathbf{0}) \equiv \mathbf{0}, \tag{47}$$

$$\frac{\partial^2 \boldsymbol{\Gamma}}{\partial \xi_m\,\partial \xi_n}(\mathbf{0}) = \frac{1}{4\pi}\int_{|\boldsymbol{\xi}|=1} \tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})[3\xi_m\xi_n - \delta_{mn}]\,\mathrm{d}S\left[\int_0^\infty h(r)r\,\mathrm{d}r\right]. \tag{48}$$

To evaluate (39), we compute from (34), using the notation $\partial(\ )/\partial\xi_m \equiv (\ )_{,m}$

$$\begin{aligned}\langle\tilde{\mathbf{T}}\rangle_{,m}(\boldsymbol{\xi}) &= c_1\,\delta\mathbf{L}_1\mathbf{K}\boldsymbol{\Gamma}^{-1}\boldsymbol{\Gamma}_{,m}\boldsymbol{\Gamma}^{-1}\mathbf{K}(\boldsymbol{\Gamma}^{-1}+\delta\mathbf{L}_2)\\ &\quad + c_2\,\delta\mathbf{L}_2\mathbf{K}\boldsymbol{\Gamma}^{-1}\boldsymbol{\Gamma}_{,m}\boldsymbol{\Gamma}^{-1}\mathbf{K}(\boldsymbol{\Gamma}^{-1}+\delta\mathbf{L}_1) - (c_1\,\delta\mathbf{L}_1 + c_2\,\delta\mathbf{L}_2)\mathbf{K}\boldsymbol{\Gamma}^{-1}\boldsymbol{\Gamma}_{,m}\boldsymbol{\Gamma}^{-1},\end{aligned} \tag{49}$$

where we have used the fact that for any tensor $\mathbf{A}$, $(\mathbf{A}^{-1})_{,i} = -\mathbf{A}^{-1}\mathbf{A}_{,i}\mathbf{A}^{-1}$, and have defined

$$\mathbf{K} \equiv (\boldsymbol{\Gamma}^{-1} + c_1\,\delta\mathbf{L}_2 + c_2\,\delta\mathbf{L}_1)^{-1}. \tag{50}$$

Now applying (47) to (49) gives

$$\boxed{\langle\tilde{\mathbf{T}}\rangle_{,m}(\mathbf{0}) \equiv \mathbf{0}}\ , \tag{51}$$

meaning that the second right-hand-side term in (37) is identically zero.

To determine the last term in (37), we must evaluate (40), which requires taking another derivative of (49). Doing so and applying (47) results in

$$\boxed{\begin{aligned}\langle\tilde{\mathbf{T}}\rangle_{,mn}(\mathbf{0}) &= c_1\,\delta\mathbf{L}_1\mathbf{K}\boldsymbol{\Gamma}^{-1}\boldsymbol{\Gamma}_{,mn}\boldsymbol{\Gamma}^{-1}\mathbf{K}(\boldsymbol{\Gamma}^{-1}+\delta\mathbf{L}_2)\\ &\quad + c_2\,\delta\mathbf{L}_2\mathbf{K}\boldsymbol{\Gamma}^{-1}\boldsymbol{\Gamma}_{,mn}\boldsymbol{\Gamma}^{-1}\mathbf{K}(\boldsymbol{\Gamma}^{-1}+\delta\mathbf{L}_1) - (c_1\,\delta\mathbf{L}_1 + c_2\,\delta\mathbf{L}_2)\mathbf{K}\boldsymbol{\Gamma}^{-1}\boldsymbol{\Gamma}_{,mn}\boldsymbol{\Gamma}^{-1}\end{aligned}}\ , \tag{52}$$

where the terms $\boldsymbol{\Gamma}$ and $\boldsymbol{\Gamma}_{,mn}$ are evaluated at $\boldsymbol{\xi} = \mathbf{0}$, as given by (46) and (48), respectively.

To summarize, we have found a nonlocal constitutive representation for two-phase composites having an isotropic distribution of arbitrarily-shaped, arbitrarily anisotropic phases, which has an error of $O(|\mathbf{x}-\mathbf{x}'|^4)$ compared to the exact result involving (35). This is, from (20) and (37)–(41),

$$\boxed{\langle\boldsymbol{\sigma}\rangle(\mathbf{x}) = \hat{\mathbf{L}}\langle\mathbf{e}\rangle(\mathbf{x}) - \tfrac{1}{2}\langle\tilde{\mathbf{T}}\rangle_{,mn}(\mathbf{0})\frac{\partial^2\langle\mathbf{e}\rangle(\mathbf{x})}{\partial x_m\,\partial x_n}}\ , \tag{53}$$

where $\hat{\mathbf{L}}$ is given by (43) with $\mathbf{P}$ of (46) replacing $\mathbf{P}'$, and $\langle\tilde{\mathbf{T}}\rangle_{,mn}(\mathbf{0})$ is given by (52) with (46) and (48).

These results simplify substantially if the comparison moduli $\mathbf{L}_0$ are chosen to be the same as the moduli of one of the phases, say

$$\mathbf{L}_0 = \mathbf{L}_2 \Rightarrow \delta\mathbf{L}_2 = \mathbf{0}. \tag{54}$$

For a matrix with moduli $\mathbf{L}_2$ containing inclusions with moduli $\mathbf{L}_1$, this choice is not only convenient but it has some theoretical support: Willis (1984) provided a very general analysis based on a "self-consistent quasi-crystalline approximation," in which an integral equation for the ensemble average of the polarization outside an inclusion, conditional on the presence of an inclusion at a specified location, was solved exactly. The result was that the self-consistent estimate for overall response was independent of the choice of comparison medium, and therefore the same as the estimate delivered by choosing the comparison medium to have the properties of the matrix. The choice (54) is therefore adopted for the remainder of this work. Then, (43) and (52) simplify to

$$\boxed{\hat{\mathbf{L}} = \mathbf{L}_0 + c_1[(\delta\mathbf{L}_1)^{-1} + c_2\mathbf{P}]^{-1}} \tag{55}$$

$$\boxed{\langle\tilde{\mathbf{T}}\rangle_{,mn}(\mathbf{0}) = -c_1c_2[(\delta\mathbf{L}_1)^{-1} + c_2\mathbf{P}]^{-1}\mathbf{\Gamma}_{,mn}[(\delta\mathbf{L}_1)^{-1} + c_2\mathbf{P}]^{-1}}\,. \tag{56}$$

3.4. *Simplification when both phases are isotropic*

In the interest of obtaining completely explicit expressions, we now examine the case in which both phases of the composite are isotropic, although still of arbitrary shape. The components of the comparison material modulus tensor $\mathbf{L}_0$ (and hence now $\mathbf{L}_2$) are

$$c_{ijkl} = \lambda\,\delta_{ij}\,\delta_{kl} + \mu(\delta_{ik}\,\delta_{jl} + \delta_{il}\,\delta_{jk}), \tag{57}$$

where, λ, μ are the Lamé moduli; they are related to the bulk modulus κ as

$$\lambda = \kappa - \tfrac{2}{3}\mu. \tag{58}$$

To determine $\tilde{\mathbf{\Gamma}}_0(\boldsymbol{\xi})$ explicitly in this case, note first from (A.5) of the Appendix and (57) that

$$[\mathbf{L}_0(\boldsymbol{\xi})]_{jk} = (\lambda+\mu)\xi_j\xi_k + \mu|\boldsymbol{\xi}|^2\,\delta_{jk} = |\boldsymbol{\xi}|^2[(\lambda+2\mu)\hat{\xi}_j\hat{\xi}_k + \mu(\delta_{jk} - \hat{\xi}_j\hat{\xi}_k)], \tag{59}$$

where $\hat{\xi}_i = \xi_i/|\boldsymbol{\xi}|$. Since the last of (59) is in spectral form, its inverse is

$$[\mathbf{L}_0^{-1}(\boldsymbol{\xi})]_{jk} = \frac{1}{|\boldsymbol{\xi}|^2}\left[\frac{1}{\lambda+2\mu}\hat{\xi}_j\hat{\xi}_k + \frac{1}{\mu}(\delta_{jk} - \hat{\xi}_j\hat{\xi}_k)\right]. \tag{60}$$

Finally, using (58) and (60) in (A.6) of the Appendix gives

$$[\tilde{\mathbf{\Gamma}}_0(\boldsymbol{\xi})]_{ijkl} = \frac{1}{\mu|\boldsymbol{\xi}|^4}\left\{\frac{|\boldsymbol{\xi}|^2}{4}(\xi_i\,\delta_{jk}\xi_l + \xi_j\,\delta_{ik}\xi_l + \xi_i\,\delta_{jl}\xi_k + \xi_j\,\delta_{il}\xi_k) - \frac{3\kappa+\mu}{3\kappa+4\mu}\xi_i\xi_j\xi_k\xi_l\right\}. \tag{61}$$

Upcoming calculations are most conveniently performed using Hill's (1965) *symbolic* notation for fourth-order isotropic tensors, viz.

$$\mathbf{L}_0 = (3\kappa, 2\mu), \tag{62}$$

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

$$\mathbf{L}_0\mathbf{L}_1 = [(3\kappa)(3\kappa_1),(2\mu)(2\mu_1)]. \tag{63}$$

Under our present assumptions, $\mathbf{P}$ of (46) is an isotropic fourth-order tensor, with from (57) and (58) the representation (since, e.g. $P_{ijkl} = P_{jikl}$)

$$P_{ijkl} = (\kappa_P - \tfrac{2}{3}\mu_P)\,\delta_{ij}\,\delta_{kl} + \mu_P(\delta_{ik}\,\delta_{jl} + \delta_{il}\,\delta_{jk}) = \frac{1}{4\pi}\int_{|\xi|=1} [\tilde{\mathbf{\Gamma}}_0(\xi)]_{ijkl}\,\mathrm{d}S. \tag{64}$$

Following Willis (1982), for each of the invariants P_{iikk} and P_{ijij}, the integrand in (64), given by (61), is independent of ξ, so one computes from (64)

$$P_{iikk} = 9\kappa_P = \frac{3}{3\kappa + 4\mu}, \quad P_{ijij} = 3\kappa_P + 10\mu_P = \frac{3\kappa + 7\mu}{\mu(3\kappa + 4\mu)}, \tag{65}$$

so that in Hill's notation

$$\mathbf{P} = \left(\frac{1}{3\kappa + 4\mu}, \frac{3(\kappa + 2\mu)}{5\mu(3\kappa + 4\mu)}\right). \tag{66}$$

We also need $\Gamma_{ijkl,mn}(\mathbf{0})$ in explicit form. Under present assumptions, this is an isotropic sixth-order tensor. The most general isotropic sixth-order tensor has the representation (see, e.g. Jaunzemis, 1967)

$$\begin{aligned} I_{ijklmn} = {} & C_1\,\delta_{ij}\,\delta_{kl}\,\delta_{mn} + C_2\,\delta_{ij}\,\delta_{km}\,\delta_{ln} + C_3\,\delta_{ij}\,\delta_{kn}\,\delta_{lm} + C_4\,\delta_{ik}\,\delta_{jl}\,\delta_{mn} \\ & + C_5\,\delta_{ik}\,\delta_{jm}\,\delta_{ln} + C_6\,\delta_{ik}\,\delta_{jn}\,\delta_{lm} + C_7\,\delta_{il}\,\delta_{jk}\,\delta_{mn} + C_8\,\delta_{il}\,\delta_{jm}\,\delta_{kn} \\ & + C_9\,\delta_{il}\,\delta_{jn}\,\delta_{km} + C_{10}\,\delta_{im}\,\delta_{jk}\,\delta_{ln} + C_{11}\,\delta_{im}\,\delta_{jl}\,\delta_{kn} + C_{12}\,\delta_{im}\,\delta_{jn}\,\delta_{kl} \\ & + C_{13}\,\delta_{in}\,\delta_{jk}\,\delta_{lm} + C_{14}\,\delta_{in}\,\delta_{jl}\,\delta_{km} + C_{15}\,\delta_{in}\,\delta_{jm}\,\delta_{kl}, \end{aligned} \tag{67}$$

where C_1–C_{15} are independent constants. Observe from (48) with (61) that $\Gamma_{ijkl,mn}(\mathbf{0})$ has the symmetries [dropping $(\mathbf{0})$]

$$\Gamma_{ijkl,mn} = \Gamma_{jikl,mn} = \Gamma_{ijlk,mn} = \Gamma_{ijkl,nm} = \Gamma_{klij,mn}\,; \tag{68}$$

applying these symmetries to (67) shows that the number of independent constants reduces to four, and that $\Gamma_{ijkl,mn}(\mathbf{0})$ admits the representation

$$\begin{aligned} \frac{1}{H}\Gamma_{ijkl,mn}(\mathbf{0}) = {} & A_1\,\delta_{ij}\,\delta_{kl}\,\delta_{mn} + A_2(\delta_{ik}\,\delta_{jl}\,\delta_{mn} + \delta_{il}\,\delta_{jk}\,\delta_{mn}) \\ & + A_3(\delta_{ij}\,\delta_{km}\,\delta_{ln} + \delta_{ij}\,\delta_{kn}\,\delta_{lm} + \delta_{im}\,\delta_{jn}\,\delta_{kl} + \delta_{in}\,\delta_{jm}\,\delta_{kl}) \\ & + A_4(\delta_{ik}\,\delta_{jm}\,\delta_{ln} + \delta_{ik}\,\delta_{jn}\,\delta_{lm} + \delta_{il}\,\delta_{jm}\,\delta_{kn} + \delta_{il}\,\delta_{jn}\,\delta_{km} \\ & + \delta_{im}\,\delta_{jk}\,\delta_{ln} + \delta_{im}\,\delta_{jl}\,\delta_{kn} + \delta_{in}\,\delta_{jk}\,\delta_{lm} + \delta_{in}\,\delta_{jl}\,\delta_{km}), \end{aligned} \tag{69}$$

having defined

$$H \equiv \int_0^\infty h(r) r \, \mathrm{d}r. \tag{70}$$

To evaluate A_1–A_4, we compute the following four invariants of $\Gamma_{ijkl,mn}(\mathbf{0})$, from (69) and from (48) with (61), observing that for each of these the integrand of the spherical integral in (48) is independent of ξ on $|\xi| = 1$,

$$\begin{aligned}
\frac{1}{H}\Gamma_{iikk,mm}(\mathbf{0}) &= 27A_1+18A_2+36A_3+24A_4 = 0 \\
\frac{1}{H}\Gamma_{ijij,mm}(\mathbf{0}) &= 9A_1+36A_2+12A_3+48A_4 = 0 \\
\frac{1}{H}\Gamma_{iikl,kl}(\mathbf{0}) &= 9A_1+6A_2+42A_3+48A_4 = \frac{6}{3\kappa+4\mu} \\
\frac{1}{H}\Gamma_{ijki,jk}(\mathbf{0}) &= 3A_1+12A_2+24A_3+66A_4 = \frac{3\kappa+16\mu}{2\mu(3\kappa+4\mu)}.
\end{aligned} \tag{71}$$

Solving the algebraic system (71) gives

$$\begin{aligned}
A_1 &= \frac{4}{105}\frac{3\kappa+\mu}{\mu(3\kappa+4\mu)}, \quad A_2 = -\frac{1}{35}\frac{3\kappa+8\mu}{\mu(3\kappa+4\mu)}, \\
A_3 &= -\frac{1}{35}\frac{3\kappa+\mu}{\mu(3\kappa+4\mu)}, \quad A_4 = \frac{3}{140}\frac{3\kappa+8\mu}{\mu(3\kappa+4\mu)}.
\end{aligned} \tag{72}$$

To render the nonlocal constitutive relation as explicit as possible, we employ Hill's symbolic notation to compute the following combination that appears in (55) and (56)

$$\begin{aligned}
\mathbf{B} \equiv [(\delta\mathbf{L}_1)^{-1}+c_2\mathbf{P}]^{-1} &= \left[\Big(3(\kappa_1-\kappa), 2(\mu_1-\mu)\Big)^{-1} + c_2\left(\frac{1}{3\kappa+4\mu}, \frac{3(\kappa+2\mu)}{5\mu(3\kappa+4\mu)}\right)\right]^{-1} \\
&= \left(\frac{3(\kappa_1-\kappa)(3\kappa+4\mu)}{3\kappa+4\mu+3c_2(\kappa_1-\kappa)}, \frac{10\mu(\mu_1-\mu)(3\kappa+4\mu)}{5\mu(3\kappa+4\mu)+6c_2(\mu_1-\mu)(\kappa+2\mu)}\right) \equiv (3\kappa_B, 2\mu_B),
\end{aligned} \tag{73}$$

meaning that

$$B_{ijkl} = (\kappa_B - \tfrac{2}{3}\mu_B)\delta_{ij}\delta_{kl} + \mu_B(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}). \tag{74}$$

Using (73), one computes for (55) the explicit result

$$\boxed{\begin{aligned}
\hat{\mathbf{L}} \equiv \mathbf{L}_0 + \langle\tilde{\mathbf{T}}\rangle(\mathbf{0}) \equiv (3\kappa_L, 2\mu_L) = \Bigg(&3\frac{\kappa(3\kappa_1+4\mu)+4c_1\mu(\kappa_1-\kappa)}{3\kappa_1+4\mu-3c_1(\kappa_1-\kappa)}, \\
&2\mu\frac{\mu(9\kappa+8\mu)(1-c_1)+3\kappa\mu_1(2+3c_1)+4\mu\mu_1(3+2c_1)}{5\mu(3\kappa+4\mu)+6(1-c_1)(\mu_1-\mu)(\kappa+2\mu)}\Bigg).
\end{aligned}} \tag{75}$$

(We have verified that this is identical to (7.1) of Willis (1982) when the choice $\mathbf{L}_0 = \mathbf{L}_2$ is made.) Finally, to determine (56) explicitly, we employ (69) with (72)–(74)

$$
\begin{aligned}
-\frac{1}{c_1 c_2 H}\langle \tilde{T}\rangle_{ijkl,mn}(\mathbf{0}) &= B_{ijop}\left[\frac{1}{H}\Gamma_{opqr,mn}(\mathbf{0})\right]B_{qrkl} \\
&= D_1\,\delta_{ij}\,\delta_{kl}\,\delta_{mn} + D_2(\delta_{ik}\,\delta_{jl}\,\delta_{mn} + \delta_{il}\,\delta_{jk}\,\delta_{mn}) \\
&\quad + D_3(\delta_{ij}\,\delta_{km}\,\delta_{ln} + \delta_{ij}\,\delta_{kn}\,\delta_{lm} + \delta_{im}\,\delta_{jn}\,\delta_{kl} + \delta_{in}\,\delta_{jm}\,\delta_{kl}) \\
&\quad + D_4(\delta_{ik}\,\delta_{jm}\,\delta_{ln} + \delta_{ik}\,\delta_{jn}\,\delta_{lm} + \delta_{il}\,\delta_{jm}\,\delta_{kn} + \delta_{il}\,\delta_{jn}\,\delta_{km} \\
&\quad + \delta_{im}\,\delta_{jk}\,\delta_{ln} + \delta_{im}\,\delta_{jl}\,\delta_{kn} + \delta_{in}\,\delta_{jk}\,\delta_{lm} + \delta_{in}\,\delta_{jl}\,\delta_{km}),
\end{aligned}
\tag{76}
$$

where one computes, defining $\delta\kappa \equiv \kappa_1 - \kappa$, $\delta\mu \equiv \mu_1 - \mu$ and using (71) to simplify,

$$
\begin{aligned}
D_1 &= \tfrac{4}{3}(3A_1 + 2A_2 + 2A_3)(3\kappa_B - 2\mu_B)\mu_B + 4A_1\mu_B^2 \\
&= 8\mu\,\delta\mu(3\kappa + 4\mu)\frac{5(3\kappa+4\mu)[2\,\delta\mu(3\kappa+8\mu) - 21\,\delta\kappa\mu] - 12c_2\,\delta\kappa\,\delta\mu(3\kappa+\mu)}{21(3c_2\,\delta\kappa + 3\kappa + 4\mu)[5\mu(3\kappa+4\mu) + 6c_2\,\delta\mu(\kappa+2\mu)]^2}, \\
D_2 &= 4A_2\mu_B^2 = \frac{-20\mu(\delta\mu)^2(3\kappa+4\mu)(3\kappa+8\mu)}{7[5\mu(3\kappa+4\mu) + 6c_2\,\delta\mu(\kappa+2\mu)]^2}, \\
D_3 &= 6A_3\kappa_B\mu_B + \tfrac{8}{3}A_4\mu_B(3\kappa_B - 2\mu_B) = -\tfrac{3}{4}D_1, \\
D_4 &= 4A_4\mu_B^2 = -\tfrac{3}{4}D_2.
\end{aligned}
\tag{77}
$$

Thus, our nonlocal constitutive representation for two-phase composites having an isotropic distribution of isotropic phases is (53) with (75)–(77). Notice that all of the results are completely explicit except for the evaluation of H, given by the integral in (70). To determine this, one must have explicit information about the two-point correlation function $h(r)$ of the random composite material under consideration. Expressions for $h(r)$ based on sound physical and mathematical reasoning are scarce. As an example, in the next section we evaluate H for what may be the only case yet extant of such a well-founded physical and mathematical model of a two-point correlation function.

3.5. *Explicit results for a matrix reinforced by a random dispersion of nonoverlapping identical spheres*

We now further specialize our two-phase composite, already specialized to an isotropic distribution of isotropic phases, to the case of an isotropic matrix (Phase 2) reinforced (or weakened, depending on the moduli chosen) by a random dispersion of nonoverlapping identical spheres (Phase 1). Markov and Willis (1995) have recently shown that the two-point correlation function of a random dispersion of non-overlapping spheres can be expressed as a simple one-tuple integral containing the radial distribution function. In the present notation, their results are

$$h(r) = h^{ws}(r) + h^*(r), \tag{78}$$

where $h^{ws}(r)$ is the "well-stirred" approximation, which they showed is tenable at most when $c_1 \leqslant 1/8$, and thus $h^*(r)$ is a correction term. For spheres of radius a, and defining $\rho \equiv r/a$, they found (and we have verified)

$$h^{ws}(\rho) = \begin{cases} \dfrac{1}{1-c_1}\left[(1-c_1)-\dfrac{3\rho}{4}+\dfrac{(1+3c_1)\rho^3}{16}-\dfrac{9c_1\rho^4}{160}+\dfrac{c_1\rho^6}{2240}\right], & 0 \leqslant \rho \leqslant 2, \\ \dfrac{c_1}{1-c_1}\dfrac{(\rho-4)^4(36-34\rho-16\rho^2-\rho^3)}{2240\rho}, & 2 \leqslant \rho \leqslant 4, \\ 0, \quad 4 \leqslant \rho, \end{cases} \tag{79}$$

and

$$h^*(\rho) = \begin{cases} \dfrac{c_1}{1-c_1}\displaystyle\int_2^{\rho+2} F(\rho,\eta)\beta(\eta)\,\mathrm{d}\eta, \quad 0 \leqslant \rho \leqslant 2, \\ \dfrac{c_1}{1-c_1}\left[\displaystyle\int_2^{\rho} F(-\rho,-\eta)\beta(\eta)\,\mathrm{d}\eta+\int_{\rho}^{\rho+2} F(\rho,\eta)\beta(\eta)\,\mathrm{d}\eta\right], \quad 2 \leqslant \rho \leqslant 4, \\ \dfrac{c_1}{1-c_1}\left[\displaystyle\int_{\rho-2}^{\rho} F(-\rho,-\eta)\beta(\eta)\,\mathrm{d}\eta+\int_{\rho}^{\rho+2} F(\rho,\eta)\beta(\eta)\,\mathrm{d}\eta\right], \quad 4 \leqslant \rho, \end{cases} \tag{80}$$

where

$$F(\rho,\eta) = \frac{3\eta(2+\rho-\eta)^3[4-6(\rho-\eta)+(\rho-\eta)^2]}{160\rho}, \tag{81}$$

and

$$\beta(\eta) = g(\eta)-1, \tag{82}$$

where $g(\eta)$ is the radial distribution function (to be discussed later).

Now, for our nonlocal constitutive equation we need

$$H \equiv \int_0^{\infty} h(r)r\,\mathrm{d}r = a^2\int_0^{\infty} h(\rho)\rho\,\mathrm{d}\rho = a^2\int_0^{\infty}[h^{ws}(\rho)+h^*(\rho)]\rho\,\mathrm{d}\rho. \tag{83}$$

Using (79), the first part of the last integral in (83) is easily evaluated

$$\int_0^{\infty} h^{ws}(\rho)\rho\,\mathrm{d}\rho = \frac{1}{1-c_1}\left\{\int_0^2\left[(1-c_1)\rho-\frac{3\rho^2}{4}+\frac{(1+3c_1)\rho^4}{16}-\frac{9c_1\rho^5}{160}+\frac{c_1\rho^7}{2240}\right]\mathrm{d}\rho + \int_2^4 \frac{c_1(\rho-4)^4(36-34\rho-16\rho^2-\rho^3)}{2240}\,\mathrm{d}\rho\right\}$$

$$= \frac{2-9c_1}{5(1-c_1)}. \tag{84}$$

To evaluate the second part of the last integral in (83), substitute from (80)

$$\int_0^\infty h^*(\rho)\rho\,\mathrm{d}\rho = \frac{c_1}{1-c_1}\left\{\int_0^2\left[\int_2^{\rho+2} F(\rho,\eta)\beta(\eta)\,\mathrm{d}\eta\right]\rho\,\mathrm{d}\rho\right.$$

$$+\int_2^4\left[\int_2^\rho F(-\rho,-\eta)\beta(\eta)\,\mathrm{d}\eta+\int_\rho^{\rho+2} F(\rho,\eta)\beta(\eta)\,\mathrm{d}\eta\right]\rho\,\mathrm{d}\rho$$

$$\left.+\int_4^\infty\left[\int_{\rho-2}^\rho F(-\rho,-\eta)\beta(\eta)\,\mathrm{d}\eta+\int_\rho^{\rho+2} F(\rho,\eta)\beta(\eta)\,\mathrm{d}\eta\right]\rho\,\mathrm{d}\rho\right\}. \quad (85)$$

Since $F(\rho,\eta)$ is known explicitly for the full range of its arguments, while as discussed later this is not true (at least analytically) for $\beta(\eta)$, let us interchange the order of integration of the integrals appearing in (85). Thus,

$$\int_0^2\left[\int_2^{\rho+2} F(\rho,\eta)\beta(\eta)\,\mathrm{d}\eta\right]\rho\,\mathrm{d}\rho = \int_2^4\left[\int_{\eta-2}^2 F(\rho,\eta)\rho\,\mathrm{d}\rho\right]\beta(\eta)\,\mathrm{d}\eta, \quad (86a)$$

$$\int_2^4\left[\int_2^\rho F(-\rho,-\eta)\beta(\eta)\,\mathrm{d}\eta+\int_\rho^{\rho+2} F(\rho,\eta)\beta(\eta)\,\mathrm{d}\eta\right]\rho\,\mathrm{d}\rho$$

$$= \int_2^4\left[\int_\eta^4 F(-\rho,-\eta)\rho\,\mathrm{d}\rho\right]\beta(\eta)\,\mathrm{d}\eta+\int_2^4\left[\int_2^\eta F(\rho,\eta)\rho\,\mathrm{d}\rho\right]\beta(\eta)\,\mathrm{d}\eta$$

$$+\int_4^6\left[\int_{\eta-2}^4 F(\rho,\eta)\rho\,\mathrm{d}\rho\right]\beta(\eta)\,\mathrm{d}\eta, \quad (86b)$$

$$\int_4^\infty\left[\int_{\rho-2}^\rho F(-\rho,-\eta)\beta(\eta)\,\mathrm{d}\eta+\int_\rho^{\rho+2} F(\rho,\eta)\beta(\eta)\,\mathrm{d}\eta\right]\rho\,\mathrm{d}\rho$$

$$= \int_2^4\left[\int_4^{\eta+2} F(-\rho,-\eta)\rho\,\mathrm{d}\rho\right]\beta(\eta)\,\mathrm{d}\eta+\int_4^\infty\left[\int_\eta^{\eta+2} F(-\rho,-\eta)\rho\,\mathrm{d}\rho\right]\beta(\eta)\,\mathrm{d}\eta$$

$$+\int_4^6\left[\int_4^\eta F(\rho,\eta)\rho\,\mathrm{d}\rho\right]\beta(\eta)\,\mathrm{d}\eta+\int_6^\infty\left[\int_{\eta-2}^\eta F(\rho,\eta)\rho\,\mathrm{d}\rho\right]\beta(\eta)\,\mathrm{d}\eta. \quad (86c)$$

Substituting (86) into (85) and combining terms, (85) finally reduces to

$$\int_0^\infty h^*(\rho)\rho\,\mathrm{d}\rho = \frac{c_1}{1-c_1}\int_2^\infty\left[\int_{\eta-2}^\eta F(\rho,\eta)\rho\,\mathrm{d}\rho+\int_\eta^{\eta+2} F(-\rho,-\eta)\rho\,\mathrm{d}\rho\right]\beta(\eta)\,\mathrm{d}\eta$$

$$= \frac{c_1}{1-c_1}\int_2^\infty \eta\beta(\eta)\,\mathrm{d}\eta = \frac{c_1}{1-c_1}\int_2^\infty \eta[g(\eta)-1]\,\mathrm{d}\eta, \quad (87)$$

having evaluated the integrals in the bracket via substitution of (81), and then substituting finally from (82).

It remains to evaluate the integral involving the radial distribution function $g(\rho)$ in (87). The best-known statistical mechanics model for the radial distribution function of a dispersion of hard spheres is that of Percus and Yevick (1958), who obtained an integral equation for $g(\rho)$. Wertheim (1963) derived an exact solution for this integral equation, expressed as a closed-form Laplace transform. Interestingly, this will prove sufficient to provide an explicit result for H. Wertheim's solution is, written in terms of $\rho = r/a$ [instead of his $x = r/(2a)$],

$$\left.\begin{aligned} g(\rho) &= 0, \quad 0 \leqslant \rho \leqslant 2 \\ \int_0^\infty \rho g(\rho) \mathrm{e}^{-s\rho}\,\mathrm{d}\rho &= \frac{8sL(2s)}{12c_1[L(2s)+M(2s)\,\mathrm{e}^{2s}]} \end{aligned}\right\}, \tag{88}$$

where

$$L(t) = 12c_1\left[\left(1+\frac{c_1}{2}\right)t+(1+2c_1)\right],$$

$$M(t) = (1-c_1)^2t^3+6c_1(1-c_1)t^2+18c_1^2t-12c_1(1+2c_1). \tag{89}$$

Using the first equation of (88), the last integral in (87) becomes

$$\int_2^\infty \rho[g(\rho)-1]\,\mathrm{d}\rho = \int_0^\infty \rho[g(\rho)-1]\,\mathrm{d}\rho+2, \tag{90}$$

and noting that

$$\int_0^\infty \rho\,\mathrm{e}^{-s\rho}\,\mathrm{d}\rho = \frac{1}{s^2}, \tag{91}$$

we find from (88)–(91)

$$\begin{aligned} \int_0^\infty \rho[g(\rho)-1]\,\mathrm{d}\rho &= \lim_{s\to 0}\int_0^\infty \rho[g(\rho)-1]\,\mathrm{e}^{-s\rho}\,\mathrm{d}\rho = \lim_{s\to 0}\left\{\frac{8sL(2s)}{12c_1[L(2s)+M(2s)\,\mathrm{e}^{2s}]}-\frac{1}{s^2}\right\} \\ &= \lim_{s\to 0}\left\{\frac{8s^3L(2s)-12c_1[L(2s)+M(2s)\,\mathrm{e}^{2s}]}{12c_1s^2[L(2s)+M(2s)\,\mathrm{e}^{2s}]}\right\} = -\frac{10-2c_1+c_1^2}{5(1+2c_1)}. \end{aligned} \tag{92}$$

Summarizing the results from (83)–(92), we have found

$$H \equiv \int_0^\infty h(r)r\,\mathrm{d}r = \frac{a^2}{1-c_1}\left\{\frac{2-9c_1}{5}+c_1\left[2-\frac{10-2c_1+c_1^2}{5(1+2c_1)}\right]\right\}$$

$$\boxed{H = a^2\frac{(2-c_1)(1-c_1)}{5(1+2c_1)}}\,. \tag{93}$$

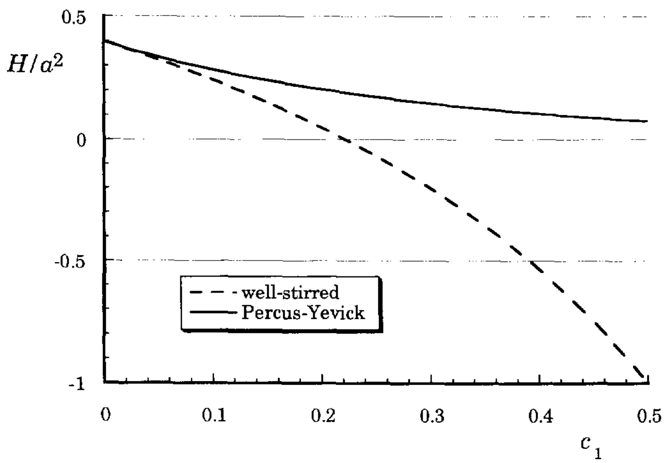

Fig. 1. The statistical factor H (normalized by a^2) as a function of reinforcement volume concentration c_1, comparing the result (84) obtained solely from the well-stirred approximation to the complete form (93) incorporating the Percus–Yevick correction.

Figure 1 compares H/a^2 from the approximate expression (84) that used only the well-stirred approximation for $h(\rho)$, with the complete form (93) that incorporates the correction due to the Percus–Yevick model.

4. QUANTITATIVE ESTIMATES OF MINIMUM RVE SIZE

One principal objective of the present work is to obtain *quantitative* estimates of the minimum size of a representative volume element of composite material for which the usual constitutive model, that relates average stress to average strain through a *constant* "overall" or "effective" modulus tensor, is sensible. That is, essentially all extant models of the constitutive response of elastic composite materials develop constitutive equations of the form

$$\langle \boldsymbol{\sigma} \rangle = \hat{\mathbf{L}} \langle \mathbf{e} \rangle, \tag{94}$$

where $\hat{\mathbf{L}}$ is a constant tensor that characterizes the macroscopic response of the composite. We derived an example $\hat{\mathbf{L}}$ in (43), valid for any two-phase composite, via the Hashin–Shtrikman variational principle and Fourier transforms, confirming an alternate derivation previously performed by Willis (1982). The typical statement made is the highly qualitative one which notes that (94) is expected to be valid for material volume elements that are much larger than the microscopic scale, or "microscale", of the composite. We think it important to derive quantitative estimates of the minimum size of a material representative volume element for which a model of the type (94) accurately describes constitutive response. Such knowledge will have several significant implications; for example, it will permit estimates of the smallest composite material structural component size than one might hope to analyze by employing a macroscopic constitutive equation of type (94). It should also allow assessment of the minimum requirements for which a macroscopic theory of fracture, of the type already successfully employed for "homogeneous" materials, might be applicable to composites. An exploration of this latter question will be reported elsewhere.

Our approach to addressing this issue will be to employ the explicit nonlocal constitutive equation we derived in the previous section. We will consider ensemble-averaged strain fields that vary with position, $\langle \mathbf{e} \rangle(\mathbf{x})$, and determine at what wavelength this variation will cause the nonlocal term in the constitutive equation to produce a nonnegligible correction to the local term. This will provide an estimate of minimum RVE size. Thus, the quantitative results we obtain will be for the type of composite for which we were able to render the nonlocal constitutive equation completely explicit: namely, two-phase composites consisting of an isotropic matrix reinforced (or weakened) by a random dispersion of isotropic spherical particles.

The nonlocal constitutive equation for this specific class of composites is (53) with (75)–(77) and (93). Thus we will determine the RVE length scale by comparing the second right-hand-side term in (53) with the first.

To obtain explicit results, we shall consider the constitutive response for two specific simple cases of straining: a normal strain that varies with position in its direction of straining, and a shear strain that varies with position in the plane of shear. These strain fields are clearly compatible with continuous, single-valued displacement fields. For the normal strain case, we assume

$$\langle e \rangle_{11}(\mathbf{x}) = \varepsilon \sin \frac{2\pi x_1}{l_N}; \quad \text{all other } \langle e \rangle_{ij}(\mathbf{x}) \equiv 0, \tag{95}$$

where $|\varepsilon| \ll 1$ is a pure number. Then (53) gives for the 11-component of ensemble average stress

$$\begin{aligned} \langle \sigma \rangle_{11}(\mathbf{x}) &= \hat{L}_{1111} \langle e \rangle_{11}(\mathbf{x}) - \tfrac{1}{2} \langle \tilde{T} \rangle_{1111,11}(\mathbf{0}) \frac{\partial^2 \langle e \rangle_{11}(\mathbf{x})}{\partial x_1^2} \\ &= \left\{ \hat{L}_{1111} + \frac{2\pi^2}{l_N^2} \langle \tilde{T} \rangle_{1111,11}(\mathbf{0}) \right\} \varepsilon \sin \frac{2\pi x_1}{l_N}. \end{aligned} \tag{96}$$

Therefore, defining $\alpha \times 100$ as the error percentage of the constant effective modulus term, that is, the percentage correction provided by the nonlocal term, we have

$$\frac{2\pi^2}{l_N^2} \langle \tilde{T} \rangle_{1111,11}(\mathbf{0}) = \alpha \hat{L}_{1111}. \tag{97}$$

This means that once a desired error limit α is specified to which constitutive response is captured purely by the effective modulus term, i.e. by a constitutive equation of the form (94), then (97) gives the smallest wavelength l_N of the ensemble average normal strain variation for which this error limit is not exceeded. The implication is that l_N provides the minimum representative volume element size for which a constitutive model having form (94) will have error α for normal strain variations. Solving (97),

$$\boxed{l_N = \pi\left|\frac{2\langle\tilde{T}\rangle_{1111,11}(\mathbf{0})}{\alpha\hat{L}_{1111}}\right|^{1/2} = 2\pi a\left|\frac{3c_1(1-c_1)^2(2-c_1)(D_1+2D_2)}{5\alpha(1+2c_1)(3\kappa_L+4\mu_L)}\right|^{1/2}}, \tag{98}$$

where D_1, D_2 are given by (77) and κ_L, μ_L by (75).

For the shear strain case we assume

$$\langle e\rangle_{12}(\mathbf{x}) = \langle e\rangle_{21}(\mathbf{x}) = \varepsilon\sin\frac{2\pi x_1}{l_S};\quad \text{all other } \langle e\rangle_{ij}(\mathbf{x}) \equiv 0. \tag{99}$$

Then (53) gives for the 12-component of ensemble average stress

$$\begin{aligned}\langle\sigma\rangle_{12}(\mathbf{x}) &= 2\hat{L}_{1212}\langle e\rangle_{12}(\mathbf{x}) - \langle\tilde{T}\rangle_{1212,11}(\mathbf{0})\frac{\partial^2\langle e\rangle_{12}(\mathbf{x})}{\partial x_1^2}\\ &= \left\{2\hat{L}_{1212} + \frac{4\pi^2}{l_S^2}\langle\tilde{T}\rangle_{1212,11}(\mathbf{0})\right\}\varepsilon\sin\frac{2\pi x_1}{l_S},\end{aligned} \tag{100}$$

which leads to

$$\boxed{l_S = \pi\left|\frac{2\langle\hat{T}\rangle_{1212,11}(\mathbf{0})}{\alpha\hat{L}_{1212}}\right|^{1/2} = \pi a\left|\frac{c_1(1-c_1)^2(2-c_1)D_2}{5\alpha(1+2c_1)\mu_L}\right|^{1/2}}. \tag{101}$$

In order to obtain explicit results for specific material types, recall that the isotropic elastic moduli are related as

$$\kappa = \frac{2(1+\nu)}{3(1-2\nu)}\mu, \tag{102}$$

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 will compute minimum RVE sizes using results (98) and (101) for matrix materials having each of these values of ν, for the extreme cases of the "reinforcing" spheres being voids ($\kappa_1 = \mu_1 = 0$) and being rigid particles ($\kappa_1 = \mu_1 = \infty$). For each of these cases, specification of ν_m, the matrix value, which relates κ_m and μ_m via (102), renders (98) and (101) independent of the other matrix elastic modulus. That is, for the cases of a matrix reinforced by voids or by rigid particles, (98) and (101) show that if the matrix Poisson's ratio is specified, the minimum RVE length can be determined explicitly, and is thus valid for all values of the other matrix elastic modulus (μ_m, say). To have intermediate results for each of these matrix types, we will also analyze the specific systems of an aluminum matrix reinforced with alumina particles, and an alumina matrix reinforced with aluminum particles. In these cases, we need to know in addition to the Poisson's ratio values that $\mu_{\text{alumina}}/\mu_{\text{aluminum}} \approx 6.65$. All these results are displayed in Tables 1 and 2, being computed for the case $\alpha = 0.05$, that is, the tables report the minimum RVE size (normalized by reinforcement *diameter*) for which an error of no more than 5% will accrue if ensemble average stress

Table 1. *Minimum RVE sizes from* (98) *for a random distribution of spherical "reinforcements", normalized by reinforcement diameter, for 5% error of* $\hat{\mathbf{L}}$ *relating normal stress to normal strain*

	Minimum RVE size, $l_N/(2a)$					
	$\nu_m = 0.2$			$\nu_m = 0.33$		
c_1	Voids	Rigid particles	Al_2O_3/Al	Voids	Rigid particles	Al/Al_2O_3
0.025	1.005	1.005	0.6699	1.018	0.8513	0.5825
0.05	1.345	1.345	0.8957	1.363	1.144	0.7818
0.1	1.710	1.710	1.134	1.729	1.462	0.9972
0.15	1.887	1.887	1.245	1.906	1.623	1.103
0.2	1.967	1.967	1.289	1.984	1.701	1.150
0.25	1.987	1.987	1.291	2.003	1.727	1.160
0.3	1.968	1.968	1.265	1.981	1.718	1.145
0.35	1.921	1.921	1.220	1.933	1.684	1.112
0.4	1.854	1.854	1.160	1.864	1.632	1.065

Table 2. *Minimum RVE sizes from* (101) *for a random distribution of spherical "reinforcements", normalized by reinforcement diameter, for 5% error of* $\hat{\mathbf{L}}$ *relating shear stress to shear strain*

	Minimum RVE size, $l_S/(2a)$					
	$\nu_m = 0.2$			$\nu_m = 0.33$		
c_1	Voids	Rigid particles	Al_2O_3/Al	Voids	Rigid particles	Al/Al_2O_3
0.025	0.5502	0.5502	0.4063	0.4889	0.5547	0.4025
0.05	0.7367	0.7367	0.5438	0.6555	0.7415	0.5381
0.1	0.9363	0.9363	0.6900	0.8353	0.9397	0.6812
0.15	1.033	1.033	0.7592	0.9241	1.034	0.7479
0.2	1.077	1.077	0.7881	0.9654	1.075	0.7747
0.25	1.088	1.088	0.7919	0.9775	1.084	0.7768
0.3	1.078	1.078	0.7787	0.9699	1.071	0.7624
0.35	1.052	1.052	0.7535	0.9485	1.044	0.7363
0.4	1.016	1.016	0.7195	0.9170	1.006	0.7017

and strain are related by the constant effective modulus tensor $\hat{\mathbf{L}}$ of (75). To obtain $(l_N/2a)$ or $(l_S/2a)$ for a different error limit, say α^*, (98) and (101) show that one must simply multiply the appropriate result in Table 1 or 2 by $(0.05/\alpha^*)^{1/2}$.

Several very interesting conclusions can be drawn from the results in Tables 1 and 2. For quite good accuracy (i.e. 5% error) of a constitutive model based solely on a constant effective modulus tensor, the minimum RVE size is remarkably small: *approximately two reinforcement diameters for any reinforcement type and for all reinforcement volume fractions shown.* Even for high accuracy—a maximum error of

1%—the minimum RVE size for the most demanding case studied is only ≈ 4.5 reinforcement diameters. The tables show that for all systems considered, the minimum RVE size increases with increasing reinforcement volume fraction until it attains a maximum at about $c_1 \approx 0.25$, after which it decreases slightly with further increases in c_1. Comparing Table 1 to Table 2 shows that for equal accuracy of a constant effective modulus constitutive model for normal and shearing response, the RVE must be significantly larger in the case of normal straining. The tables both show that for materials with $\nu_m = 0.2$, the minimum RVE size is identical for both voids and rigid particles. Finally, observe that the cases of void and rigid particle reinforcements appear to be extreme tests of a constant-effective-modulus constitutive model, in that the minimum RVE size required for the same accuracy of such a constitutive model is always substantially larger in these cases as compared to the aluminum matrix/alumina particles and alumina matrix/aluminum particles cases.

5. IMPLICATIONS FOR ENSEMBLE AVERAGE STRAIN ENERGY IN RANDOM COMPOSITES

Here we relate our nonlocal constitutive equation explicitly to the ensemble average strain energy, for random two-phase composites with isotropic distributions of phases. We also show how the nonlocal constitutive equation is consistent with the stationary energy principle.

Equations (2) and (11) yield

$$\mathscr{H}(\boldsymbol{\tau}) = \int_\Omega (\boldsymbol{\sigma}_0 \mathbf{e} - \boldsymbol{\sigma}\mathbf{e}_0)\,d\mathbf{x}. \tag{103}$$

Application of the divergence theorem, noting that both $\boldsymbol{\sigma}$ and $\boldsymbol{\sigma}_0$ satisfy (1a), gives

$$\mathscr{H}(\boldsymbol{\tau}) = -\int_\Omega \mathbf{f}(\mathbf{u}-\mathbf{u}_0)\,d\mathbf{x} = -\int_\Omega [(\nabla\cdot\boldsymbol{\sigma})\mathbf{u} - (\nabla\cdot\boldsymbol{\sigma}_0)\mathbf{u}_0]\,d\mathbf{x} = \int_\Omega (\boldsymbol{\sigma}\mathbf{e} - \boldsymbol{\sigma}_0\mathbf{e}_0)\,d\mathbf{x}. \tag{104}$$

Thus, the strain energy $\mathscr{E}$ in the composite is

$$\mathscr{E} = \frac{1}{2}\int_\Omega \boldsymbol{\sigma}\mathbf{e}\,d\mathbf{x} = \frac{1}{2}\int_\Omega \boldsymbol{\sigma}_0\mathbf{e}_0\,d\mathbf{x} + \frac{1}{2}\mathscr{H}(\boldsymbol{\tau}). \tag{105}$$

Exactly similar manipulations, noting that $\langle\boldsymbol{\sigma}\rangle$ also satisfies (1a), may be deployed to demonstrate that the ensemble average strain energy is

$$\langle\mathscr{E}\rangle = \frac{1}{2}\int_\Omega \langle\boldsymbol{\sigma}\mathbf{e}\rangle\,d\mathbf{x} = \frac{1}{2}\int_\Omega \langle\boldsymbol{\sigma}\rangle\langle\mathbf{e}\rangle\,d\mathbf{x} = \frac{1}{2}\int_\Omega \boldsymbol{\sigma}_0\mathbf{e}_0\,d\mathbf{x} + \frac{1}{2}\langle\mathscr{H}(\boldsymbol{\tau})\rangle. \tag{106}$$

Precisely the same structure applies to the variational approximation, based upon seeking a stationary value for $\langle\mathscr{H}(\boldsymbol{\tau}^*)\rangle$, amongst polarizations $\boldsymbol{\tau}^*$ of the form (15). Thus,

$$\langle \mathscr{E} \rangle \approx \frac{1}{2}\int_{\Omega} \langle \mathbf{e}\rangle(\mathbf{x})\left[\mathbf{L}_0\langle \mathbf{e}\rangle(\mathbf{x}) + \int_{\Omega} \mathbf{T}(\mathbf{x}-\mathbf{x}')\langle \mathbf{e}\rangle(\mathbf{x}')\,\mathrm{d}\mathbf{x}'\right]\mathrm{d}\mathbf{x} \tag{107}$$

in general or, employing the asymptotic form (53) for $\langle \boldsymbol{\sigma}\rangle$, which is applicable when $\langle \mathbf{e}\rangle(\mathbf{x})$ varies slowly,

$$\langle \mathscr{E} \rangle \approx \frac{1}{2}\int_{\Omega} \langle \mathbf{e}\rangle(\mathbf{x})\left[\hat{\mathbf{L}}\langle \mathbf{e}\rangle(\mathbf{x}) - \frac{1}{2}\langle \tilde{\mathbf{T}}\rangle_{,mn}(\mathbf{0})\frac{\partial^2\langle \mathbf{e}\rangle(\mathbf{x})}{\partial x_m\,\partial x_n}\right]\mathrm{d}\mathbf{x}. \tag{108}$$

Integration by parts of the second term gives

$$\langle \mathscr{E} \rangle \approx \frac{1}{2}\int_{\Omega} \left[\langle \mathbf{e}\rangle(\mathbf{x})\hat{\mathbf{L}}\langle \mathbf{e}\rangle(\mathbf{x}) + \frac{1}{2}\frac{\partial\langle \mathbf{e}\rangle(\mathbf{x})}{\partial x_m}\langle \tilde{\mathbf{T}}\rangle_{,mn}(\mathbf{0})\frac{\partial\langle \mathbf{e}\rangle(\mathbf{x})}{\partial x_n}\right]\mathrm{d}\mathbf{x}. \tag{109}$$

The energy functional for the ensemble-averaged version of the problem defined by (1) is

$$\Phi(\langle \mathbf{u}\rangle) = \frac{1}{2}\int_{\Omega}\int_{\Omega} \langle \mathbf{e}\rangle(\mathbf{x})[\mathbf{L}_0\,\delta(\mathbf{x}-\mathbf{x}') + \mathbf{T}(\mathbf{x}-\mathbf{x}')]\langle \mathbf{e}\rangle(\mathbf{x}')\,\mathrm{d}\mathbf{x}'\,\mathrm{d}\mathbf{x} - \int_{\Omega} \mathbf{f}(\mathbf{x})\langle \mathbf{u}\rangle(\mathbf{x})\,\mathrm{d}\mathbf{x}. \tag{110}$$

This is stationary with respect to variations in $\langle \mathbf{u}\rangle(\mathbf{x})$ when

$$\nabla\cdot\langle \boldsymbol{\sigma}\rangle + \mathbf{f} = \mathbf{0}, \tag{111}$$

with

$$\langle \boldsymbol{\sigma}\rangle(\mathbf{x}) = \mathbf{L}_0\langle \mathbf{e}\rangle(\mathbf{x}) + \int_{\Omega} \mathbf{T}(\mathbf{x}-\mathbf{x}')\langle \mathbf{e}\rangle(\mathbf{x}')\,\mathrm{d}\mathbf{x}'. \tag{112}$$

Similarly, in the "slowly-varying" approximation (109),

$$\Phi(\langle \mathbf{u}\rangle) = \frac{1}{2}\int_{\Omega}\left[\langle \mathbf{e}\rangle(\mathbf{x})\hat{\mathbf{L}}\langle \mathbf{e}\rangle(\mathbf{x}) + \frac{1}{2}\frac{\partial\langle \mathbf{e}\rangle(\mathbf{x})}{\partial x_m}\langle \tilde{\mathbf{T}}\rangle_{,mn}(\mathbf{0})\frac{\partial\langle \mathbf{e}\rangle(\mathbf{x})}{\partial x_n}\right]\mathrm{d}\mathbf{x} - \int_{\Omega} \mathbf{f}(\mathbf{x})\langle \mathbf{u}\rangle(\mathbf{x})\,\mathrm{d}\mathbf{x}. \tag{113}$$

This is stationary with respect to variations in $\langle \mathbf{u}\rangle(\mathbf{x})$ when (111) is satisfied, with $\langle \boldsymbol{\sigma}\rangle$ related to $\langle \mathbf{e}\rangle$ by (53). It is possible, too, in the spirit of "rational mechanics", to define stresses

$$s_{ij}(\mathbf{x}) = \frac{\partial W}{\partial\langle e\rangle_{ij}}, \quad t_{ijm}(\mathbf{x}) = \frac{\partial W}{\partial\langle e\rangle_{ij,m}}, \tag{114}$$

where

$$W = \tfrac{1}{2}[\langle \mathbf{e}\rangle\hat{\mathbf{L}}\langle \mathbf{e}\rangle + \tfrac{1}{2}\langle \mathbf{e}\rangle_{,m}\langle \tilde{\mathbf{T}}\rangle_{,mn}(\mathbf{0})\langle \mathbf{e}\rangle_{,n}]. \tag{115}$$

The Euler–Lagrange equation then takes the form

$$s_{ij,j} - t_{ijm,jm} + f_i = 0, \tag{116}$$

which is no more than a re-write of (111).

The medium thus behaves *as though* it possesses a local energy density function $W(\langle \mathbf{e} \rangle, \nabla \langle \mathbf{e} \rangle)$. This form, however, was obtained by integration by parts. Whether or not $W(\langle \mathbf{e} \rangle, \nabla \langle \mathbf{e} \rangle)$ can be identified in any precise sense with the mean local energy density $\frac{1}{2}\langle \boldsymbol{\sigma} \mathbf{e} \rangle$ is unknown. We have not accomplished any such identification for a composite with random microstructure.

It is emphasized that the nonlocal constitutive relations developed in this work have been derived under the assumption that the point under consideration is not close to the boundary of the body. In principle, some similar analysis could be performed, using the half-space Green's function instead of that for the whole space, to obtain the corresponding relation for points close to a smooth boundary. If this were done, the boundary conditions on the mean fields would be exactly those imposed on displacement or traction in the original problem. In the absence of such analysis, any attempt to solve a problem for a finite body, using the constitutive relation (53) everywhere, would require some additional boundary conditions. The best candidate conditions would be those that arise naturally in the derivation of the principle that $\Phi(\langle \mathbf{u} \rangle)$ should be stationary. One such set would be to prescribe displacement $\mathbf{u}$ and its normal derivative $\partial \mathbf{u}/\partial n$ at the boundary; another would be to prescribe $s_{ij}n_j$ and $t_{ijm,m}n_j$. Such relations would not yield accurate estimates for mean fields close to the boundary, since these could only be resolved by a local analysis, but they would yield a set of equations from which the mean field in the interior of the body could be estimated. The question of the exact choice of boundary conditions for the solution of boundary value problems is beyond the scope of this study.

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APPENDIX: DETERMINATION OF $\boldsymbol{\Gamma}(\mathbf{0})$ AND ITS DERIVATIVES FOR ISOTROPIC DISTRIBUTIONS OF PHASES

As noted in Section 3.3, in order to evaluate (37) explicitly, (38)–(40) with (34) show that we will need $\boldsymbol{\Gamma}$ and its first and second derivatives with respect to $\boldsymbol{\xi}$, evaluated at $\boldsymbol{\xi} = \mathbf{0}$. These are derived here.

The definition of $\boldsymbol{\Gamma}$, (26), shows that we will first need information about $\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})$. From (6),

$$[\boldsymbol{\Gamma}_0(\mathbf{x})]_{ijkl} = -\left.\frac{\partial^2[\mathbf{G}_0(\mathbf{x})]_{jk}}{\partial x_i\,\partial x_l}\right|_{(ij),(kl)}. \tag{A.1}$$

Taking the Fourier transform of this gives, since $\mathbf{G}_0(\mathbf{x})$ and $\nabla\mathbf{G}_0(\mathbf{x})$ both $\to\mathbf{0}$ for $|\mathbf{x}| \to \infty$,

$$[\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})]_{ijkl} = \xi_i\xi_l[\tilde{\mathbf{G}}_0(\boldsymbol{\xi})]_{jk}|_{(ij),(kl)}. \tag{A.2}$$

Next, Fourier transforming (7) gives

$$-[\tilde{\mathbf{G}}_0(\boldsymbol{\xi})]_{jm}\xi_i\xi_l c_{ijkl} + \delta_{km} = 0, \tag{A.3}$$

from which

$$[\tilde{\mathbf{G}}_0(\boldsymbol{\xi})]_{jk} = [\mathbf{L}_0^{-1}(\boldsymbol{\xi})]_{jk}, \tag{A.4}$$

having defined

$$[\mathbf{L}_0(\boldsymbol{\xi})]_{jk} \equiv \xi_i\xi_l c_{ijkl}. \tag{A.5}$$

Thus, from (A.2) and (A.4)

$$[\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})]_{ijkl} = \xi_i[\mathbf{L}_0^{-1}(\boldsymbol{\xi})]_{jk}\xi_l|_{(ij),(kl)}. \tag{A.6}$$

Observe from (A.5) and (A.6) that $\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})$ is homogeneous of degree zero in $\boldsymbol{\xi}$, and that

$$\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi}) = \tilde{\boldsymbol{\Gamma}}_0(-\boldsymbol{\xi}). \tag{A.7}$$

Our earlier result (42) for $\boldsymbol{\Gamma}(\mathbf{0})$ will now simplify in view of (45). Using the beginning of (42), applying (45) and the homogeneity of $\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})$, and then introducing spherical coordinates (ρ, α, ω) in $\boldsymbol{\xi}$-space (note that ρ and α have other meanings in the main body of the paper)

$$\boldsymbol{\Gamma}(\mathbf{0}) = (\tilde{\boldsymbol{\Gamma}}_0 * \tilde{h})(\mathbf{0}) = \frac{1}{8\pi^3}\int_\Omega \tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\tilde{h}(\boldsymbol{\xi})\,\mathrm{d}\boldsymbol{\xi} = \frac{1}{8\pi^3}\int_\Omega \tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi}/|\boldsymbol{\xi}|)\tilde{h}(|\boldsymbol{\xi}|)\,\mathrm{d}\boldsymbol{\xi}$$

$$= \frac{1}{8\pi^3}\int_0^{2\pi}\int_0^{\pi}\tilde{\boldsymbol{\Gamma}}_0(\alpha,\omega)\sin\alpha\,\mathrm{d}\alpha\,\mathrm{d}\omega\int_0^{\infty}\tilde{h}(\rho)\rho^2\,\mathrm{d}\rho. \tag{A.8}$$

Observing thus that the angular and radial parts of the integral are mutually independent as grouped above, that the angular integral may be expressed as the surface integral over a sphere of radius one in $\boldsymbol{\xi}$-space, and that the radial integral $\tilde{h}(\rho)$ may be rewritten as the total $\boldsymbol{\xi}$-space integral of $\tilde{h}$ divided by the angular contribution of 4π, (A.8) becomes

$$\boldsymbol{\Gamma}(\mathbf{0}) = \frac{1}{4\pi}\int_{|\xi|=1}\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\,\mathrm{d}S\left[\frac{1}{8\pi^3}\int_\Omega \tilde{h}(\boldsymbol{\xi})\,\mathrm{d}\boldsymbol{\xi}\right] = \frac{1}{4\pi}\int_{|\xi|=1}\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\,\mathrm{d}S\left[\frac{1}{8\pi^3}\int_\Omega \tilde{h}(\boldsymbol{\xi})\,\mathrm{e}^{-\mathrm{i}\boldsymbol{\xi}\cdot\mathbf{x}}\,\mathrm{d}\boldsymbol{\xi}\right]_{\mathbf{x}=0}$$

$$\boxed{\boldsymbol{\Gamma}(\mathbf{0}) = \frac{1}{4\pi}\int_{|\xi|=1}\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\,\mathrm{d}S \equiv \mathbf{P}}\,, \tag{A.9}$$

where we have used the fact that $h(0) = 1$ [e.g. from (22) and the fact that $P_{12}(\mathbf{0}) = 0$].

To evaluate the first derivative of $\boldsymbol{\Gamma}$, employ (26) with the commutativity property of convolutions

$$\frac{\partial\boldsymbol{\Gamma}}{\partial\xi_m} = \frac{1}{8\pi^3}\int_\Omega \frac{\partial\tilde{h}(|\boldsymbol{\xi}-\boldsymbol{\xi}'|)}{\partial\xi_m}\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi}')\,\mathrm{d}\boldsymbol{\xi}' = \frac{1}{8\pi^3}\int_\Omega \tilde{h}'(|\boldsymbol{\xi}-\boldsymbol{\xi}'|)\frac{(\xi_m-\xi'_m)}{|\boldsymbol{\xi}-\boldsymbol{\xi}'|}\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi}')\,\mathrm{d}\boldsymbol{\xi}', \tag{A.10}$$

where h' denotes the derivative of h with respect to its argument. Thus,

$$\frac{\partial\boldsymbol{\Gamma}}{\partial\xi_m}(\mathbf{0}) = -\frac{1}{8\pi^3}\int_\Omega \tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\frac{\xi_m}{|\boldsymbol{\xi}|}\tilde{h}'(|\boldsymbol{\xi}|)\,\mathrm{d}\boldsymbol{\xi} = -\frac{1}{8\pi^3}\int_0^{2\pi}\int_0^{\pi}\tilde{\boldsymbol{\Gamma}}_0(\alpha,\omega)\frac{\xi_m}{|\boldsymbol{\xi}|}\sin\alpha\,\mathrm{d}\alpha\,\mathrm{d}\omega\int_0^{\infty}\tilde{h}'(\rho)\rho^2\,\mathrm{d}\rho. \tag{A.11}$$

Again, we observe that the radial and angular parts of the integral separate as indicated, and that the angular part may be represented as the surface integral over a unit sphere. Thus,

$$\frac{\partial\boldsymbol{\Gamma}}{\partial\xi_m}(\mathbf{0}) = -\frac{1}{8\pi^3}\int_{|\xi|=1}\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\xi_m\,\mathrm{d}S\left[\int_0^{\infty}\tilde{h}'(\rho)\rho^2\,\mathrm{d}\rho\right]. \tag{A.12}$$

Now note from (A.5) and (A.6) that the surface integral in (A.12) can be written more explicitly as

$$\int_{|\xi|=1}[\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})]_{ijkl}\xi_m\,\mathrm{d}S = \int_{|\xi|=1}\xi_i\{[\boldsymbol{\xi}\mathbf{L}_0\boldsymbol{\xi}]^{-1}\}_{jk}\xi_l\xi_m\,\mathrm{d}S\bigg|_{(ij),(kl)}. \tag{A.13}$$

Because the range of integration in (A.13) is symmetric (it is a unit sphere), and since every term in the integrand will involve a product of an odd number of $\boldsymbol{\xi}$ terms, the integral in (A.13) is identically zero. Thus, (A.12) becomes

$$\boxed{\frac{\partial\boldsymbol{\Gamma}}{\partial\xi_m}(\mathbf{0}) \equiv \mathbf{0}}\,. \tag{A.14}$$

To evaluate the second derivative of $\boldsymbol{\Gamma}$, employ (A.10)

$$\frac{\partial^2\boldsymbol{\Gamma}}{\partial\xi_m\,\partial\xi_n}(\mathbf{0}) = \frac{1}{8\pi^3}\int_\Omega \tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})\left\{\delta_{mn}\frac{\tilde{h}'(|\boldsymbol{\xi}|)}{|\boldsymbol{\xi}|} + \frac{\xi_m\xi_n}{|\boldsymbol{\xi}|^2}\left[\tilde{h}''(|\boldsymbol{\xi}|) - \frac{\tilde{h}'(|\boldsymbol{\xi}|)}{|\boldsymbol{\xi}|}\right]\right\}\mathrm{d}\boldsymbol{\xi}. \tag{A.15}$$

Expressing this integral in spherical coordinates, separations into radial and angular portions can again be effected

$$\frac{\partial^2 \boldsymbol{\Gamma}}{\partial \xi_m \partial \xi_n}(\mathbf{0}) = \frac{\delta_{mn}}{8\pi^3}\int_0^{2\pi}\int_0^{\pi} \tilde{\boldsymbol{\Gamma}}_0(\alpha,\omega)\sin\alpha\,\mathrm{d}\alpha\,\mathrm{d}\omega \int_0^{\infty} \frac{\tilde{h}'(\rho)}{\rho}\rho^2\,\mathrm{d}\rho$$

$$+\frac{1}{8\pi^3}\int_0^{2\pi}\int_0^{\pi} \tilde{\boldsymbol{\Gamma}}_0(\alpha,\omega)\frac{\xi_m\xi_n}{|\boldsymbol{\xi}|^2}\sin\alpha\,\mathrm{d}\alpha\,\mathrm{d}\omega \int_0^{\infty}\left[\tilde{h}''(\rho)-\frac{\tilde{h}'(\rho)}{\rho}\right]\rho^2\,\mathrm{d}\rho. \quad \text{(A.16)}$$

To evaluate the radial integral appearing in the first right-side term, we begin with an integration by parts, noting that $\rho\tilde{h}(\rho) \to 0$ for $\rho \to \infty$

$$\int_0^{\infty} \tilde{h}'(\rho)\rho\,\mathrm{d}\rho = -\int_0^{\infty}\tilde{h}(\rho)\,\mathrm{d}\rho = -\int_0^{\infty}\frac{\tilde{h}(\rho)}{\rho^2}\rho^2\,\mathrm{d}\rho = -\frac{1}{4\pi}\int_{\Omega}\frac{\tilde{h}(|\boldsymbol{\xi}|)}{|\boldsymbol{\xi}|^2}\mathrm{d}\boldsymbol{\xi}$$

$$= -2\pi^2\left[\frac{1}{8\pi^3}\int_{\Omega}\frac{\tilde{h}(|\boldsymbol{\xi}|)}{|\boldsymbol{\xi}|^2}\mathrm{e}^{-\mathrm{i}\boldsymbol{\xi}\cdot\mathbf{x}}\,\mathrm{d}\boldsymbol{\xi}\right]_{\mathbf{x}=0}. \quad \text{(A.17)}$$

This inverse Fourier transform is carried out by recalling that the Poisson equation in fundamental form and its solution (Green's function) are

$$\nabla^2\Phi(\mathbf{x}) = \delta(\mathbf{x}) \Rightarrow \Phi(\mathbf{x}) = -\frac{1}{4\pi r}, \quad \text{(A.18)}$$

where $r = |\mathbf{x}|$. Fourier transforming the first equation in (A.18) gives

$$-\xi_i\xi_i\tilde{\Phi}(\boldsymbol{\xi}) = 1 \Rightarrow \tilde{\Phi}(\boldsymbol{\xi}) = -\frac{1}{|\boldsymbol{\xi}|^2}. \quad \text{(A.19)}$$

Comparing (A.18) and (A.19), the inverse Fourier transform of $1/|\boldsymbol{\xi}|^2$ is seen to be $(1/4\pi r)$. Using this and the Fourier convolution inverse transform, (A.17) becomes

$$\int_0^{\infty}\tilde{h}'(\rho)\rho\,\mathrm{d}\rho = -2\pi^2\left[\int_{\Omega}h(|\mathbf{x}-\mathbf{x}'|)\frac{\mathrm{d}\mathbf{x}'}{4\pi|\mathbf{x}'|}\right]_{\mathbf{x}=0}$$

$$= -\frac{\pi}{2}\int_{\Omega}\frac{h(|\mathbf{x}'|)}{|\mathbf{x}'|}\mathrm{d}\mathbf{x}' = -\frac{\pi}{2}4\pi\int_0^{\infty}\frac{h(r)}{r}r^2\,\mathrm{d}r = -2\pi^2\int_0^{\infty}h(r)r\,\mathrm{d}r. \quad \text{(A.20)}$$

From the last term in (A.16), we also need the integral of $\tilde{h}''(\rho)\rho^2$. To evaluate this, notice that the three-dimensional Laplacian of $\tilde{h}(\rho)$ is

$$\frac{\partial^2\tilde{h}}{\partial\xi_k\,\partial\xi_k} \equiv \nabla^2\tilde{h}(\rho) = \tilde{h}''(\rho)+\frac{2}{\rho}\tilde{h}'(\rho). \quad \text{(A.21)}$$

Thus,

$$\int_0^{\infty}\left[\tilde{h}''(\rho)+\frac{2}{\rho}\tilde{h}'(\rho)\right]\rho^2\,\mathrm{d}\rho = \int_0^{\infty}\frac{\partial^2\tilde{h}(|\boldsymbol{\xi}|)}{\partial\xi_k\,\partial\xi_k}|\boldsymbol{\xi}|^2\,\mathrm{d}|\boldsymbol{\xi}| = \frac{1}{4\pi}\int_{\Omega}\frac{\partial^2\tilde{h}}{\partial\xi_k\,\partial\xi_k}\mathrm{d}\boldsymbol{\xi}$$

$$= 2\pi^2\left[\frac{1}{8\pi^3}\int_{\Omega}\frac{\partial^2\tilde{h}}{\partial\xi_k\,\partial\xi_k}\mathrm{e}^{-\mathrm{i}\boldsymbol{\xi}\cdot\mathbf{x}}\,\mathrm{d}\boldsymbol{\xi}\right]_{\mathbf{x}=0} = 2\pi^2[-x_kx_kh(|\mathbf{x}|)]_{\mathbf{x}=0} = 0, \quad \text{(A.22)}$$

having used the facts that $\tilde{h}(\infty) = 0$, $(\nabla_{\xi}\tilde{h})(\infty) = \mathbf{0}$ and $h(0) = 1$.

Using (A.20) and (A.22), and reasoning similar to that used previously, (A.16) is seen to reduce to

$$\boxed{\frac{\partial^2\boldsymbol{\Gamma}}{\partial\xi_m\,\partial\xi_n}(\mathbf{0}) = \frac{1}{4\pi}\int_{|\boldsymbol{\xi}|=1}\tilde{\boldsymbol{\Gamma}}_0(\boldsymbol{\xi})[3\xi_m\xi_n-\delta_{mn}]\,\mathrm{d}S\left[\int_0^{\infty}h(r)r\,\mathrm{d}r\right]}. \quad \text{(A.23)}$$