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EFFECTIVE STIFFNESS TENSOR OF COMPOSITE MEDIA—I. EXACT SERIES EXPANSIONS

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ABSTRACT

The problem of determining exact expressions for the effective stiffness tensor macroscopically anisotropic, two-phase composite media of arbitrary microstructure in arbitrary space dimension d is considered. We depart from previous treatments by introducing an integral equation for the "cavity" strain field. This leads to new, exact series expansions for the effective stiffness tensor of macroscopically anisotropic, d-dimensional, two-phase composite media in powers of the "elastic polarizabilities". The nth-order tensor coefficients of these expansions are explicitly expressed as absolutely convergent integrals over products of certain tensor fields and a determinant involving n-point correlation functions that characterize the microstructure. For the special case of macroscopically isotropic media, these series expressions may be regarded as expansions that perturb about the optimal structures that realize the Hashin–Shtrikman bounds (e.g. coated-inclusion assemblages or finite-rank laminates). Similarly, for macroscopically anisotropic media, the series expressions may be regarded as expansions that perturb about optimal structures that realize Willis' bounds. For isotropic multiphase composites, we remark on the behavior of the effective moduli as the space dimension d tends to infinity. © 1997 Elsevier Science Ltd.

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1. INTRODUCTION

We consider the problem of exactly determining the effectiveness stiffness tensor $\mathbf{C}_e$ of a macroscopically anisotropic two-phase composite medium with an arbitrary but statistically homogeneous microstructure. The fourth-order effective stiffness tensor $\mathbf{C}_e$ of such a composite is defined according to the averaged Hooke's law:

$$\langle \boldsymbol{\sigma}(\mathbf{x}) \rangle = \mathbf{C}_e : \langle \boldsymbol{\varepsilon}(\mathbf{x}) \rangle, \tag{1.1}$$

where $\boldsymbol{\sigma}(\mathbf{x})$ and $\boldsymbol{\varepsilon}(\mathbf{x})$ denote the second-order local stress and strain tensors, respectively, angular brackets denote an ensemble average, and : stands for a double contraction. The local fields in (1.1) are in principle obtained by solving the governing differential equations for elastostatics subject to appropriate boundary conditions.

For macroscopically anisotropic media in which the variations in the phase stiffness tensor are small, formal solutions to the boundary-value problem have been developed in the form of perturbation series (Dederichs and Zeller, 1973; Gubernatis and Krumhansl, 1975; Willis, 1981). Due to the nature of the integral operator, one must contend with conditionally convergent integrals. One approach to this problem is to carry out a "renormalization" procedure which amounts to identifying physically what the conditionally convergent terms ought to contribute and replacing them by convergent terms that make this contribution (McCoy, 1979).

For the special case of macroscopically isotropic media, the first few terms of this perturbation expansion have been explicitly given in terms of certain statistical correlation functions for both three-dimensional media (Beran and Molyneux, 1966; Milton and Phan-Thien, 1982) and two-dimensional media (Silnutzer, 1972; Milton, 1982). A drawback of all of these classical perturbation expansions is that they are only valid for media in which the moduli of the phases are nearly the same, albeit applicable for arbitrary volume fractions.

In this paper we develop new, exact perturbation expansions for the effective stiffness tensor of macroscopically anisotropic composite media consisting of two isotropic phases by introducing an integral equation for the so-called "cavity" strain field. The expansions are not formal but rather the nth-order tensor coefficients are given explicitly in terms of integrals over products of certain tensor fields and a determinant involving n-point statistical correlation functions that render the integrals absolutely convergent in the infinite-volume limit. Thus, no renormalization analysis is required because the procedure used to solve the integral equation systematically leads to absolutely convergent integrals. Another useful feature of the expansions is that they converge rapidly for a class of dispersions *for all volume fractions, even when the phase moduli differ significantly*.

In Section 2 we introduce an integral equation for the cavity strain field for macroscopically anisotropic two-phase media of arbitrary microstructure and space dimensionality d. The solution of this integral equation and certain averaging operations lead to exact perturbation expansions for a function of the effective stiffness tensor in powers of the "elastic polarizabilities" that for general composite media requires an infinite amount of statistical information about the microstructure. In Section 3 we specialize our results to the case of macroscopically isotropic media and, among other results, show that series expressions may be regarded as expansions that perturb about the optimal structures that realize the Hashin–Shtrikman bounds (Hashin and Shtrikman, 1963; Hashin, 1965). We also examine and discuss the truncation of the expansions after third-order terms. Finally, for isotropic multiphase composites, we remark on the behavior of the effective elastic moduli (as well as effective conductivity) as the space dimension d tends to infinity. In Section 4 we demonstrate that for macroscopically anisotropic media, the series expressions may be regarded as expansions that perturb about the optimal structures that realize Willis' (1977) bounds. In the sequel to this paper, we will more fully explore the ramifications of the present results for macroscopically isotropic composites and study approximations (based on the exact expansions) for the effective bulk and shear moduli of a class of isotropic dispersions.

2. EXACT SERIES EXPANSIONS FOR MACROSCOPICALLY ANISOTROPIC MEDIA

Motivated by the dielectric (conductivity) theory of composites (Brown, 1955; Torquato, 1985; Sen and Torquato, 1989), we formulate an integral equation for the local "cavity" strain field $\mathbf{f}(\mathbf{x})$ for a d-dimensional, ellipsoidal, macroscopically anisotropic composite specimen made up of two isotropic phases that are embedded in an infinite reference phase. The shape of the composite specimen is purposely chosen to be non-spherical since any rigorously correct expression for the effective stiffness tensor must ultimately be independent of the shape of the composite specimen in the infinite-volume limit. Indeed, the analysis described below is valid for a composite specimen of arbitrary shape. We choose to speak about an ellipsoidal shape since one can appeal to the well-known elastostatic results of Eshelby (1957) for an ellipsoidal inclusion in a matrix to understand the ensuing formalism physically.

After establishing the integral equation for the cavity strain field $\mathbf{f}(\mathbf{x})$, we then relate $\mathbf{f}(\mathbf{x})$ to the local "polarization" stress field $\mathbf{p}(\mathbf{x})$. By carefully manipulating integral equations for $\mathbf{p}(\mathbf{x})$ and $\mathbf{f}(\mathbf{x})$ and averaging, we then find series expansions for the effective stiffness tensor in powers of the elastic polarizabilities. The nth-order tensor coefficients of these expansions are explicitly expressed as integrals over products of certain tensor fields and a determinant involving certain n-point correlation functions that characterize the microstructure. The form of the determinant ensures that the integrals are independent of the shape of the ellipsoid, i.e. absolutely convergent. Some properties of the expansions are then discussed.

2.1. *Integral equation for the cavity strain field*

Consider a large but finite-sized, ellipsoidal, macroscopically anisotropic composite specimen in arbitrary space dimension d comprised of two isotropic phases. The microstructure is perfectly generally and possesses a characteristic microscopic length scale which is much smaller than the smallest semi-axes of the ellipsoid. Thus, the specimen is virtually statistically homogeneous. Ultimately, we shall take the infinite-volume limit and hence consider statistically homogeneous media.

The local stiffness tensor $\mathbf{C}(\mathbf{x})$ is given in terms of the local bulk modulus $K(\mathbf{x})$ and the local shear modulus $G(\mathbf{x})$ by the relation

$$\mathbf{C}(\mathbf{x}) = dK(\mathbf{x})\boldsymbol{\Lambda}_{\mathrm{h}} + 2G(\mathbf{x})\boldsymbol{\Lambda}_{\mathrm{s}}, \tag{2.1}$$

where in component form

$$(\Lambda_{\mathrm{h}})_{ijkl} = \frac{1}{d}\delta_{ij}\delta_{kl}, \quad i,j,k,l = 1,2,\ldots,d, \tag{2.2}$$

$$(\Lambda_{\mathrm{s}})_{ijkl} = \frac{1}{2}[\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}] - \frac{1}{d}\delta_{ij}\delta_{kl}, \quad i,j,k,l = 1,2,\ldots,d, \tag{2.3}$$

and δ_{ij} is the Kronecker delta. The tensor $\boldsymbol{\Lambda}_{\mathrm{h}}$ projects onto fields that are everywhere isotropic, i.e. hydrostatic fields, whereas the tensor $\boldsymbol{\Lambda}_{\mathrm{s}}$ projects onto fields that are everywhere trace-free, i.e. shear fields. Accordingly, we refer to the former as the *hydrostatic projection tensor* and the latter as the *shear projection tensor*. The following identities shall prove to be useful in subsequent discussions:

$$(\Lambda_h)_{ijkl}+(\Lambda_s)_{ijkl} = I_{ijkl}, \tag{2.4}$$

$$(\Lambda_h)_{ijkl}(\Lambda_s)_{ijkl} = (\Lambda_h)_{ijmn}(\Lambda_s)_{mnkl} = 0 \tag{2.5}$$

$$(\Lambda_h)_{ijkl}I_{ijkl} = (\Lambda_h)_{ijkl}(\Lambda_h)_{ijkl} = 1, \tag{2.6}$$

$$(\Lambda_s)_{ijkl}I_{ijkl} = (\Lambda_s)_{ijkl}(\Lambda_s)_{ijkl} = \frac{(d-1)(d+2)}{2}, \tag{2.7}$$

$$(\Lambda_h)_{ijmn}(\Lambda_h)_{mnkl} = (\Lambda_h)_{ijkl}, \tag{2.8}$$

$$(\Lambda_s)_{ijmn}(\Lambda_s)_{mnkl} = (\Lambda_s)_{ijkl}, \tag{2.9}$$

where

$$I_{ijkl} = \tfrac{1}{2}[\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}] \tag{2.10}$$

is the fourth-order unit tensor. We note that the local stiffness tensor can be written in terms of the phase stiffnesses

$$\mathbf{C}^{(1)} = dK_1\mathbf{\Lambda}_h+2G_1\mathbf{\Lambda}_s, \quad \mathbf{C}^{(2)} = dK_2\mathbf{\Lambda}_h+2G_2\mathbf{\Lambda}_s, \tag{2.11}$$

by the relation

$$\mathbf{C}(\mathbf{x}) = \mathbf{C}^{(1)}\chi^{(1)}(\mathbf{x})+\mathbf{C}^{(2)}\chi^{(2)}(\mathbf{x}), \tag{2.12}$$

where

$$\chi^{(p)}(\mathbf{x}) = \begin{cases} 1, & \mathbf{x} \text{ in phase } p, \\ 0, & \text{otherwise} \end{cases} \tag{2.13}$$

is the characteristic function of phase p ($p = 1, 2$).

Now let us embed this d-dimensional ellipsoidal composite specimen in an infinite *reference* phase q which is subjected to an applied strain field $\boldsymbol{\varepsilon}^0(\mathbf{x})$ at infinity (see Fig. 1). The reference phase can be chosen to be arbitrary but for our purposes we will take it either to be phase 1 or phase 2, i.e. $q = 1, 2$. The local stress $\boldsymbol{\sigma}(\mathbf{x})$ is related to the local strain $\boldsymbol{\varepsilon}(\mathbf{x})$ via Hooke's law

$$\boldsymbol{\sigma}(\mathbf{x}) = \mathbf{C}(\mathbf{x}):\boldsymbol{\varepsilon}(\mathbf{x}), \tag{2.14}$$

the strain being related to the local displacement $\mathbf{u}(\mathbf{x})$ by the differential condition

$$\boldsymbol{\varepsilon}(\mathbf{x}) = \tfrac{1}{2}[\nabla\mathbf{u}+(\nabla\mathbf{u})^{\mathrm{T}}]. \tag{2.15}$$

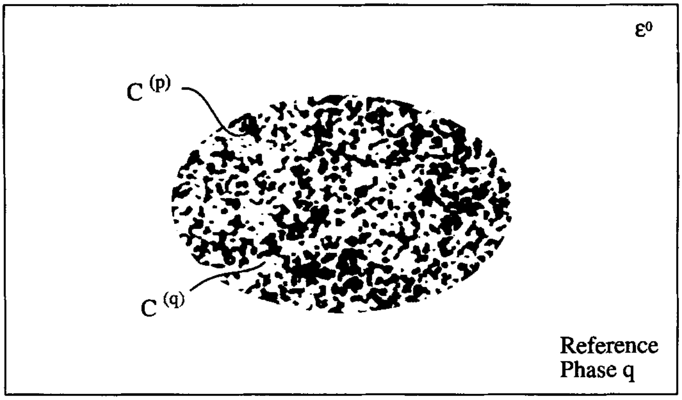
The symmetric stress field must satisfy the equilibrium requirement

$$\nabla\cdot\boldsymbol{\sigma}(\mathbf{x}) = 0. \tag{2.16}$$

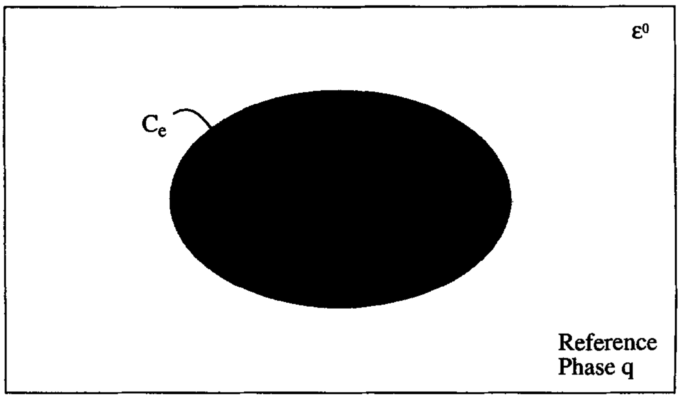
Introducing the polarization field defined by

$$\mathbf{p}(\mathbf{x}) = [\mathbf{C}(\mathbf{x})-\mathbf{C}^{(q)}]:\boldsymbol{\varepsilon}(\mathbf{x}), \tag{2.17}$$

enables us to reexpress the stress as follows:



a



b

Fig. 1. (a) Schematic of a large, d-dimensional, ellipsoidal, macroscopically anisotropic two-phase composite specimen embedded in an infinite reference phase q ($q = 1$ or 2) subjected to an applied field ε^0 at infinity. The composite consists of the polarized phase (black region) and reference phase (white region) with stiffness tensors $\mathbf{C}^{(p)}$ and $\mathbf{C}^{(q)}$, respectively, (b) after homogenization, the same ellipsoid can be viewed as having an effective stiffness tensor $\mathbf{C}_e$.

$$\boldsymbol{\sigma}(\mathbf{x}) = \mathbf{C}^{(q)}\boldsymbol{\varepsilon}(\mathbf{x}) + \mathbf{p}(\mathbf{x}). \tag{2.18}$$

The symmetric, second-order tensor $\mathbf{p}(\mathbf{x})$ is the *induced stress polarization field* relative to the medium in the absence of phase p and hence is zero in the reference phase q and non-zero in the "polarized" phase p ($p \neq q$). Throughout the paper, the indices p and q will be reserved only for the polarized and reference phases, respectively. The choice of which is the reference or polarized phase is arbitrary; all of the results are valid for any $p \neq q$, i.e. $p = 1$ and $q = 2$ or $p = 2$ and $p = 1$.

With the aid of (2.18), we can rewrite (2.16) in component form as

$$C^{(q)}_{ijkl}\frac{\partial^2 \hat{u}_k(\mathbf{x})}{\partial x_j \partial x_l} = -\frac{\partial p_{ij}(\mathbf{x})}{\partial x_j}, \tag{2.19}$$

$$\hat{u}_k(\mathbf{x}) \to 0, \quad \mathbf{x} \to \infty, \tag{2.20}$$

where $\hat{\mathbf{u}}(\mathbf{x})$ is the displacement field in excess of the displacement field at infinity $\mathbf{u}^0(\mathbf{x})$, i.e. $\hat{\mathbf{u}}(\mathbf{x}) = \mathbf{u}(\mathbf{x}) - \mathbf{u}^0(\mathbf{x})$. The infinite-space Green's function $g_{ij}^{(q)}$ is defined by requiring it to satisfy

$$C_{ijkl}^{(q)} \frac{\partial^2 g_{im}^{(q)}(\mathbf{x},\mathbf{x}')}{\partial x_j \, \partial x_l} = -\delta_{km}\delta(\mathbf{x}-\mathbf{x}'), \tag{2.21}$$

$$g_{km}^{(q)}(\mathbf{x},\mathbf{x}') \to 0, \quad \mathbf{x} \to \infty. \tag{2.22}$$

Multiplying (2.19) by the Green's function and integrating by parts yields the integral relation

$$u_i(\mathbf{x}) = u_i^0(\mathbf{x}) + \int \frac{\partial}{\partial x_l} g_{ik}^{(q)}(\mathbf{x},\mathbf{x}') p_{kl}(\mathbf{x}') \, \mathrm{d}\mathbf{x}', \tag{2.23}$$

where u_i^0 is the displacement field at infinity. Note that the presence of the polarization $\mathbf{p}$ in (2.23) implies that the integration volume extends only over the region of space occupied by the finite-sized ellipsoidal composite specimen. Integral relations of the form (2.23) have been derived previously by various investigators (Dederichs and Zeller, 1973; Gubernatis and Krumhansl, 1975; Willis, 1981) for the case of three dimensions ($d = 3$).

It is a simple matter to show that the d-dimensional Green's function that satisfies (2.21) and (2.22) is given by

$$g_{ij}^{(q)}(\mathbf{r}) = \begin{cases} \dfrac{1}{2\Omega G_q} \ln\left(\dfrac{1}{r}\right)\delta_{ij} + b_q n_i n_j, & d = 2 \\[2ex] a_q \dfrac{\delta_{ij}}{r^{d-2}} + b_q \dfrac{n_i n_j}{r^{d-2}}, & d \geqslant 3, \end{cases} \tag{2.24}$$

where

$$a_q = \frac{1}{2(d-2)\Omega G_q} \frac{dK_q + (3d-2)G_q}{dK_q + 2(d-1)G_q}, \tag{2.25}$$

$$b_q = \frac{1}{2\Omega G_q} \frac{dK_q + (d-2)G_q}{dK_q + 2(d-1)G_q}. \tag{2.26}$$

Moreover,

$$\Omega(d) = \frac{2\pi^{d/2}}{\Gamma(d/2)} \tag{2.27}$$

is the total solid angle contained in a d-dimensional sphere, $\Gamma(x)$ is the gamma function, and

$$\mathbf{r} = \mathbf{x} - \mathbf{x}', \quad \mathbf{n} = \frac{\mathbf{r}}{|\mathbf{r}|}.$$

The fact that the Green's function possesses a singularity at the point $\mathbf{x}' = \mathbf{x}$ requires one to exclude a small region containing the point $\mathbf{x}' = \mathbf{x}$ (see the Appendix). Roughly speaking the integral is convergent if this integral exists in the limit that the excluded region shrinks to zero, independent of the shape of the excluded region. According to this criterion, the integral of (2.23) is convergent.

Now to obtain the strain, one must differentiate (2.23); however, because of the singular nature of the integral one cannot simply differentiate under the integral sign. Excluding a spherical region or "cavity" from the origin in (2.23) and using the results of the Appendix, we find that the strain field is given by the integral relation

$$\varepsilon_{ij}(\mathbf{x}) = \varepsilon^0_{ij}(\mathbf{x}) + \int G^{(q)}_{ijkl}(\mathbf{r}) p_{kl}(\mathbf{x}')\, d\mathbf{x}', \tag{2.28}$$

where

$$G^{(q)}_{ijkl}(\mathbf{r}) = -A^{(q)}_{ijkl}\delta(\mathbf{r}) + H^{(q)}_{ijkl}(\mathbf{r}). \tag{2.29}$$

In relation (2.29), the constant fourth-order tensor $\mathbf{A}^{(q)}$ [that arises because of the exclusion of the spherical cavity in (2.23)] is given by

$$A^{(q)}_{ijkl} = \frac{1}{dK_q + 2(d-1)G_q}(\Lambda_{\mathrm{h}})_{ijkl} + \frac{d}{(d+2)G_q}\left[\frac{K_q + 2G_q}{dK_q + 2(d-1)G_q}\right](\Lambda_{\mathrm{s}})_{ijkl}, \tag{2.30}$$

and $\mathbf{H}^{(q)}(\mathbf{r})$ is the position-dependent fourth-order tensor given by

$$\begin{aligned} H^{(q)}_{ijkl}(\mathbf{r}) &= \frac{1}{2}[\tilde{H}^{(q)}_{ijkl}(\mathbf{r}) + \tilde{H}^{(q)}_{ijlk}(\mathbf{r})] \\ &= \frac{1}{2\Omega[dK_q + 2(d-1)G_q]}\frac{1}{r^d}\Bigg[\alpha_q\delta_{ij}\delta_{kl} - d[\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}] \\ &\quad - d\alpha_q[\delta_{ij}n_k n_l + \delta_{kl}n_i n_j] + \frac{d(d-\alpha_q)}{2}[\delta_{ik}n_j n_l + \delta_{il}n_j n_k + \delta_{jk}n_i n_l + \delta_{jl}n_i n_k] \\ &\quad + d(d+2)\alpha_q n_i n_j n_k n_l\Bigg], \end{aligned} \tag{2.31}$$

where

$$\alpha_q = dK_q/G_q + (d-2) \tag{2.32}$$

is a dimensionless parameter. In accordance with the Appendix, it is understood that integrals involving the tensor $\mathbf{H}^{(q)}$ are to be carried out with the exclusion of an infinitesimal sphere in the limit that the sphere radius shrinks to zero. The tensor $\tilde{\mathbf{H}}^{(q)}$ appearing in the first line of (2.31) is given explicitly in the Appendix. The symmetry of the polarization tensor $\mathbf{p}$ enables one to define the more symmetric tensor $\mathbf{H}^{(q)}$. Indeed, the tensor $\mathbf{H}^{(q)}$ is symmetric with respect to the first two indices and the second two indices as well as with respect to interchange of ij and kl, i.e.

$$H^{(q)}_{ijkl} = H^{(q)}_{jikl} = H^{(q)}_{ijlk} = H^{(q)}_{klij}.$$

Moreover, the integral of $\mathbf{H}^{(q)}(\mathbf{r})$ over the surface of a sphere of radius $R > 0$ is identically zero, i.e.

$$\int_{r=R} \mathbf{H}^{(q)}(\mathbf{r})\,d\Omega = 0. \tag{2.33}$$

Some contractions of the tensor $\mathbf{H}^{(q)}$ that will be of use to us in the subsequent analysis are as follows:

$$H^{(q)}_{ijkk}(\mathbf{r}) = \frac{d}{\Omega[dK_q+2(d-1)G_q]}\frac{1}{r^d}[dn_in_j-\delta_{ij}], \tag{2.34}$$

$$H^{(q)}_{iikl}(\mathbf{r}) = \frac{d}{\Omega[dK_q+2(d-1)G_q]}\frac{1}{r^d}[dn_kn_l-\delta_{kl}], \tag{2.35}$$

$$H^{(q)}_{iikk}(\mathbf{r}) = H^{(q)}_{ikik}(\mathbf{r}) = 0. \tag{2.36}$$

We shall also utilize the following scalar identities:

$$H^{(q)}_{iikl}(\mathbf{r})H^{(q)}_{klij}(\mathbf{s}) = \frac{d^3}{\Omega^2[dK_q+2(d-1)G_q]^2}\frac{1}{r^d}\frac{1}{s^d}[d(\mathbf{n}\cdot\mathbf{m})^2-1], \tag{2.37}$$

$$H^{(q)}_{ijkl}(\mathbf{r})H^{(q)}_{klij}(\mathbf{s}) = \frac{1}{4\Omega^2[dK_q+2(d-1)G_q]^2}\frac{1}{r^d}\frac{1}{s^d}\{d(d+2)\alpha_q^2[d(\mathbf{n}\cdot\mathbf{m})^4-3]$$
$$-d(5d+6))\alpha_q^2[d(\mathbf{n}\cdot\mathbf{m})^2-1]+2d^2(d-2)\alpha_q[d(\mathbf{n}\cdot\mathbf{m})^2-1]+d^3(d+2)[d(\mathbf{n}\cdot\mathbf{m})^2-1]\}, \tag{2.38}$$

where $\mathbf{m} = \mathbf{s}/|\mathbf{s}|$ is a unit vector in the direction of $\mathbf{s}$.

At this stage of the analysis we depart substantially from previous treatments by introducing an integral equation for the "cavity" strain field $\mathbf{f}$. Specifically, upon substitution of (2.29) into expression (2.28), we obtain the integral equation

$$\mathbf{f}(\mathbf{x}) = \boldsymbol{\varepsilon}^0(\mathbf{x}) + \int_\varepsilon d\mathbf{x}'\mathbf{H}^{(q)}(\mathbf{x}-\mathbf{x}') : \mathbf{p}(\mathbf{x}), \tag{2.39}$$

which is related to the usual strain by the expression

$$\mathbf{f}(\mathbf{x}) = \{\mathbf{I}+\mathbf{A}^{(q)} : [\mathbf{C}(\mathbf{x})-\mathbf{C}^{(q)}]\} : \boldsymbol{\varepsilon}(\mathbf{x}), \tag{2.40}$$

where the constant tensor $\mathbf{A}^{(q)}$ is given by (2.30). We also define

$$\int_\varepsilon d\mathbf{x}' \equiv \lim_{\varepsilon\to 0}\int_{|\mathbf{x}-\mathbf{x}'|>\varepsilon} d\mathbf{x}', \tag{2.41}$$

i.e. integration over the sample volume is carried out with the exclusion of an infinitesimally small sphere centered at $\mathbf{x}$ of radius ε, with the limit $\varepsilon \to 0$ ultimately taken. We refer to $\mathbf{f}(\mathbf{x})$ as the *cavity strain field* because, as can be seen from (2.40), it is a modified strain field, equal to the usual strain plus a contribution involving the constant tensor $\mathbf{A}^{(q)}$ which arises as a result of excluding a spherical cavity from the origin in (2.23). The cavity strain field is the elasticity analog of the Lorentz electric field used in dielectric theory (Brown, 1955; Torquato, 1985; Sen and Torquato, 1989).

Combination of the expressions (2.17) and (2.40) gives a relation between the stress polarization and cavity strain field, i.e.

$$\mathbf{p}(\mathbf{x}) = \mathscr{L}^{(q)}(\mathbf{x}):\mathbf{f}(\mathbf{x}), \tag{2.42}$$

where the fourth-order tensor $\mathscr{L}^{(q)}(\mathbf{x})$ is given by

$$\mathscr{L}^{(q)}(\mathbf{x}) = \{\mathbf{C}(\mathbf{x})-\mathbf{C}^{(q)}\}\{\mathbf{I}+\mathbf{A}^{(q)}:[\mathbf{C}(\mathbf{x})-\mathbf{C}^{(q)}]\}^{-1}. \tag{2.43}$$

Clearly, $\mathscr{L}^{(q)}(\mathbf{x})$ has the same symmetry properties as the stiffness tensor $\mathbf{C}(\mathbf{x})$. If the stiffness tensor has the isotropic form (2.1), then $\mathscr{L}^{(q)}(\mathbf{x})$ can be written as a constant tensor $\mathbf{L}^{(q)}$ multiplied by the characteristic function $\chi^{(p)}(\mathbf{x})$, i.e.

$$\mathscr{L}^{(q)}(\mathbf{x}) = \mathbf{L}^{(q)}\chi^{(p)}(\mathbf{x}), \tag{2.44}$$

where

$$\mathbf{L}^{(q)} = [dK_q+2(d-1)G_q]\left[\kappa_{pq}\mathbf{\Lambda}_{\mathrm{h}}+\frac{(d+2)G_q}{d(K_q+2G_q)}\mu_{pq}\mathbf{\Lambda}_{\mathrm{s}}\right], \tag{2.45}$$

$$\kappa_{pq} = \frac{K_p-K_q}{K_p+\dfrac{2(d-1)}{d}G_q}, \tag{2.46}$$

$$\mu_{pq} = \frac{G_p-G_q}{G_p+\dfrac{G_q[dK_q/2+(d+1)(d-2)G_q/d]}{K_q+2G_q}}. \tag{2.47}$$

Note that the coefficients κ_{pq} and μ_{pq} *are not tensors*; they are *scalar* parameters that depend on the moduli of the polarized and reference phases p and q, respectively. In analogy with dielectric theory, we refer to the scalar parameters κ_{pq} and μ_{pq} as the *bulk modulus polarizability* and the *shear modulus polarizability*, respectively.

2.2. *Exact series expansions for the effective moduli*

The effective tensor $\mathbf{L}_{\mathrm{e}}^{(q)}$ is defined via the relation linking the average polarization to the average cavity strain field, i.e.

$$\langle\mathbf{p}(\mathbf{x})\rangle = \mathbf{L}_{\mathrm{e}}^{(q)}:\langle\mathbf{f}(\mathbf{x})\rangle, \tag{2.48}$$

where

$$\mathbf{L}_{\mathrm{e}}^{(q)} = \{\mathbf{C}_{\mathrm{e}}-\mathbf{C}^{(q)}\}\{\mathbf{I}+\mathbf{A}^{(q)}:[\mathbf{C}_{\mathrm{e}}-\mathbf{C}^{(q)}]\}^{-1}. \tag{2.49}$$

The constitutive relation (2.48) is localized, i.e. it is independent of the shape of the ellipsoidal composite specimen in the infinite-volume limit. In light of (2.49), we see that the effective tensor $\mathbf{L}_{\mathrm{e}}^{(q)}$ has the same symmetry properties as the effective stiffness tensor $\mathbf{C}_{\mathrm{e}}$. Note that the constitutive relation (2.48) that defines the effective tensor $\mathbf{L}_{\mathrm{e}}^{(q)}$ is entirely equivalent to Hooke's law (1.1) that defines the effective stiffness tensor $\mathbf{C}_{\mathrm{e}}$. Keeping in mind that the tensors $\mathscr{L}^{(q)}$, $\mathbf{L}_{\mathrm{e}}^{(q)}$ and $\mathbf{H}^{(q)}$ are associated with the reference phase q, we shall temporarily drop the superscript q when referring to these tensors in the subsequent discussion.

It is desired to find an explicit expression for the effective moduli $\mathbf{L}_e$ using the solution of the integral equation (2.39) which we rewrite as

$$\mathbf{f}(1) = \boldsymbol{\varepsilon}^0(1) + \int_\varepsilon d2\mathbf{H}(1,2):\mathbf{p}(2), \tag{2.50}$$

where we have adopted the shorthand notation of representing $\mathbf{x}$ and $\mathbf{x}'$ by 1 and 2, respectively. In schematic operator form, this equation can be tersely rewritten as

$$\mathbf{f} = \boldsymbol{\varepsilon}^0 + \mathbf{Hp}, \tag{2.51}$$

where for an arbitrary operator $\boldsymbol{\Gamma}$

$$\boldsymbol{\Gamma}\mathbf{p} \equiv \int_\varepsilon d2\boldsymbol{\Gamma}(1,2):\mathbf{p}(2). \tag{2.52}$$

Multiplying the integral equation for the cavity field from the left by $\mathscr{L}(\mathbf{x})$ [defined by (2.44)] yields the equation

$$\mathbf{p} = \mathscr{L}\boldsymbol{\varepsilon}^0 + \mathscr{L}\mathbf{Hp}. \tag{2.53}$$

A solution for the polarization $\mathbf{p}$ in terms of an operator acting on the applied strain field ε^0 can be obtained by successive substitutions using (2.53) with the result

$$\begin{aligned}\mathbf{p} &= \mathscr{L}\boldsymbol{\varepsilon}^0 + \mathscr{L}\mathbf{H}\mathscr{L}\boldsymbol{\varepsilon}^0 + \mathscr{L}\mathbf{H}\mathscr{L}\mathbf{H}\mathscr{L}\boldsymbol{\varepsilon}^0 + \cdots,\\ &= \mathbf{T}\varepsilon^0,\end{aligned} \tag{2.54}$$

where the fourth-order tensor operator $\mathbf{T}$ is given by

$$\mathbf{T} = \mathscr{L}[\mathbf{I} - \mathscr{L}\mathbf{H}]^{-1}. \tag{2.55}$$

For concreteness, we write out (2.54) more explicitly as

$$\begin{aligned}\mathbf{p}(1) &= \mathscr{L}(1):\boldsymbol{\varepsilon}^0(1) + \int d2\mathscr{L}(1):\mathbf{H}(1,2):\mathscr{L}(2):\boldsymbol{\varepsilon}^0(2)\\ &\quad + \int d2\,d3\mathscr{L}(1):\mathbf{H}(1,2):\mathscr{L}(2):\mathbf{H}(2,3):\mathscr{L}(3):\boldsymbol{\varepsilon}^0(3)\\ &\quad + \cdots\\ &= \int d2\mathbf{T}(1,2):\boldsymbol{\varepsilon}^0(2).\end{aligned} \tag{2.56}$$

Ensemble averaging (2.54) yields

$$\langle\mathbf{p}\rangle = \langle\mathbf{T}\rangle\boldsymbol{\varepsilon}^0. \tag{2.57}$$

It is seen that the operator $\langle\mathbf{T}\rangle$ generally involves products of the tensor $\mathbf{H}$ which decays to zero like r^{-d} for large r. Thus, $\langle\mathbf{T}\rangle$ at best involves conditionally convergent integrals and hence must be dependent upon the shape of the ellipsoidal composite specimen. Indeed, this non-local nature of the relation between the polarization and the applied strain field is completely consistent with the well-known elastostatic results of Eshelby (1957). Eshelby showed that when an ellipsoidal inclusion in an infinite matrix of another material is subjected to a constant strain field at infinity $\boldsymbol{\varepsilon}^0$, the polarization stress field within the ellipsoid is uniform and, when expressed in terms of $\boldsymbol{\varepsilon}^0$, depends upon the shape of the ellipsoid.

Given the non-local nature of the relation (2.57), the remaining strategy is clear. In order to obtain a local relation between the average polarization $\langle \mathbf{p} \rangle$ and average cavity field $\langle \mathbf{f} \rangle$ as prescribed by (2.48), we must eliminate the applied field $\boldsymbol{\varepsilon}^0$ in favor of the appropriate average field. Thus, inverting (2.57) gives

$$\boldsymbol{\varepsilon}^0 = \langle \mathbf{T} \rangle^{-1} \langle \mathbf{p} \rangle, \tag{2.58}$$

and averaging (2.51) yields

$$\langle \mathbf{f} \rangle = \boldsymbol{\varepsilon}^0 + \mathbf{H} \langle \mathbf{p} \rangle. \tag{2.59}$$

We can now eliminate the applied field in (2.59) using (2.58) to obtain

$$\langle \mathbf{f} \rangle = \mathbf{X} \langle \mathbf{p} \rangle, \tag{2.60}$$

where

$$\mathbf{X} = \langle \mathbf{T} \rangle^{-1} + \mathbf{H}. \tag{2.61}$$

Explicitly relation (2.60) reads

$$\begin{aligned}
\langle \mathbf{f}(1) \rangle &= \int \mathrm{d}2 \mathbf{X}(1,2) \cdot \langle \mathbf{p}(2) \rangle \\
&= \langle \mathscr{L}(1) \rangle^{-1} : \langle \mathbf{p}(1) \rangle \\
&\quad - \int \mathrm{d}2 [\langle \mathscr{L}(1) \rangle^{-1} : \langle \mathscr{L}(1) : \mathbf{H}(1,2) : \mathscr{L}(2) \rangle : \langle \mathscr{L}(2) \rangle^{-1} \\
&\quad - \langle \mathscr{L}(1) \rangle^{-1} : \langle \mathscr{L}(1) \rangle : \mathbf{H}(1,2) : \langle \mathscr{L}(2) \rangle : \langle \mathscr{L}(2) \rangle^{-1}] : \langle \mathbf{p}(2) \rangle \\
&\quad - \cdots .
\end{aligned} \tag{2.62}$$

Comparing expressions (2.48) and (2.60) and returning to the notation of explicitly indicating that the reference medium is phase q, yields the desired result for the inverse of the effective tensor $\mathbf{L}_e^{(q)}$, i.e.

$$(\mathbf{L}_e^{(q)})^{-1} = \mathbf{X}. \tag{2.63}$$

It is convenient to multiply this equation by the constant fourth-order tensor $\mathbf{L}^{(q)}$ [defined by (2.44)] from the left to yield

$$\mathbf{L}^{(q)} (\mathbf{L}_e^{(q)})^{-1} = \mathbf{L}\mathbf{X}, \tag{2.64}$$

or, more explicitly,

$$\begin{aligned}
\mathbf{L}^{(q)} : (\mathbf{L}_e^{(q)})^{-1} &= \frac{\mathbf{I}}{S_1^{(p)}(1)} - \int \mathrm{d}2 \left[\frac{S_2^{(p)}(1,2) - S_1^{(p)}(1) S_1^{(p)}(2)}{S_1^{(p)}(1) S_1^{(p)}(2)} \right] \mathbf{U}^{(q)}(1,2) \\
&\quad - \int \mathrm{d}2\, \mathrm{d}3 \left[\frac{S_3^{(p)}(1,2,3)}{S_1^{(p)}(1) S_1^{(p)}(2)} - \frac{S_2^{(p)}(1,2) S_2^{(p)}(2,3)}{S_1^{(p)}(1) S_1^{(p)}(2) S_1^{(p)}(3)} \right] \mathbf{U}^{(q)}(1,2) : \mathbf{U}^{(q)}(2,3) - \cdots,
\end{aligned} \tag{2.65}$$

where

$$U^{(q)}_{ijkl}(\mathbf{r}) = L^{(q)}_{ijmn}H^{(q)}_{mnkl}(\mathbf{r})$$
$$= [dK_q+2(d-1)G_q]\left\{\left[\kappa_{pq}-\frac{(d+2)G_q}{d(K_q+2G_q)}\mu_{pq}\right]\frac{\delta_{ij}}{d}H^{(q)}_{mmkl}(\mathbf{r}) + \frac{(d+2)G_q}{d(K_q+2G_q)}\mu_{pq}H^{(q)}_{ijkl}(\mathbf{r})\right\}. \quad (2.66)$$

Here the n-point correlation function $S_n^{(p)}$ for the polarized phase p is defined according to the following ensemble average:

$$S_n^{(p)}(\mathbf{x}_1,\ldots,\mathbf{x}_n) = \langle\chi^{(p)}(\mathbf{x}_1)\ldots\chi^{(p)}(\mathbf{x}_n)\rangle. \quad (2.67)$$

The function $S_n^{(p)}(\mathbf{x}_1,\ldots,\mathbf{x}_n)$ gives the probability of simultaneously finding n points with positions $\mathbf{x}_1, \ldots, \mathbf{x}_n$ in phase p and is sometimes called the *n-point probability function*. For statistically anisotropic but homogeneous media, the $S_n^{(p)}$ depend on the relative displacements $\mathbf{x}_{ij} = \mathbf{x}_j - \mathbf{x}_i$, $1 \leqslant i < j \leqslant n$; in particular, $S_1^{(p)}$ is simply the volume fraction ϕ_p of phase p. The reason why $S_n^{(p)}$ arises in the expansion (2.65) is because the operator $\mathbf{X}$ contains averages over products of the position-dependent tensor $\mathbf{L}^{(q)}(\mathbf{x})$ which in turn depends on the characteristic function $\chi^{(p)}(\mathbf{x})$ [cf. (2.44) and (2.62)]. Note that at this stage of the analysis we have not passed to the statistically homogeneous, infinite-volume limit.

The general term of the expansion (2.65) can be easily written down as

$$\phi_p^2\mathbf{L}^{(q)} : (\mathbf{L}_e^{(q)})^{-1} = \phi_p\mathbf{I} - \sum_{n=2}^{\infty}\mathbf{B}_n^{(p)}, \quad (2.68)$$

where the tensor coefficients $\mathbf{B}_n^{(p)}$ are the following integrals over products of the $\mathbf{U}^{(q)}$ tensors and the $S_n^{(p)}$ associated with phase p as given by

$$\mathbf{B}_2^{(p)} = \int_\varepsilon \mathrm{d}2\,\mathbf{U}^{(q)}(1,2)[S_2^{(p)}(1,2)-\phi_p^2], \quad (2.69)$$

$$\mathbf{B}_n^{(p)} = (-1)^n\left(\frac{1}{\phi_p}\right)^{n-2}\int \mathrm{d}2\ldots\int \mathrm{d}n\,\mathbf{U}^{(q)}(1,2):\mathbf{U}^{(q)}(2,3):::\mathbf{U}^{(q)}(n-1,n)\Delta_n^{(p)}(1,\ldots,n), \quad n \geqslant 3, \quad (2.70)$$

and $\Delta_n^{(p)}$ is a position-dependent determinant associated with phase p given by

$$\Delta_n^{(p)} = \begin{vmatrix} S_2^{(p)}(1,2) & S_1^{(p)}(2) & \ldots & 0 & 0 \\ S_3^{(p)}(1,2,3) & S_2^{(p)}(2,3) & \ldots & 0 & 0 \\ \vdots & \vdots & \ldots & \vdots & \vdots \\ \vdots & \vdots & \ldots & \vdots & \vdots \\ \vdots & \vdots & \ldots & \vdots & \vdots \\ S_{n-1}^{(p)}(1,2,\ldots,n-1) & S_{n-2}^{(p)}(2,3,\ldots,n-1) & \ldots & S_2^{(p)}(n-2,n-1) & S_1^{(p)}(n-1) \\ S_n^{(p)}(1,2,\ldots,n) & S_{n-1}^{(p)}(2,3,\ldots,n) & \ldots & S_3^{(p)}(n-2,n-1,n) & S_2^{(p)}(n-1,n) \end{vmatrix} \quad (2.71)$$

Remarks

1. Result (2.68) is new and actually represents two different series expansion: one for $p = 1$ and $q = 2$ and the other for $p = 2$ and $q = 1$.

2. At first glance, one might surmise that the integrals of (2.69) and (2.70) are conditionally convergent because of the appearance in the integrand of the tensor $\mathbf{U}^{(q)}(\mathbf{r})$ which decays as r^{-d} for larger r. However, since the quantity within the brackets of (2.69) and the determinant $\Delta_n^{(p)}$ in (2.70) identically vanish at the boundary of the sample, because of the asymptotic properties of the $S_n^{(p)}$ (Torquato and Stell, 1982), the integrals in (2.68) are independent of the shape of the macroscopic ellipsoid (i.e. absolutely convergent), and hence any convenient shape (such as a d-dimensional sphere) may be employed in the infinite-volume limit. Moreover, when $n \geqslant 3$, the limiting process of excluding an infinitesimally small cavity about $r_{ij} = 0$ in the integrals (2.70) is no longer necessary since $\Delta_n^{(p)}$ again is identically zero for such values.

3. It is important to emphasize that the nth-order tensor coefficient $\mathbf{B}_n^{(p)}$ implicitly involves powers of the bulk modulus polarizability κ_{pq} and shear modulus polarizability μ_{pq}. To see this, one can write the product of the tensors $\mathbf{U}^{(q)}$ appearing in (2.70) in terms of the products of the tensor $\mathbf{H}^{(q)}$ via relation (2.66). Such products of $\mathbf{H}^{(q)}$ will involve the powers $\kappa_{pq}^m \mu_{pq}^{n-m}$, where m takes on integer values from 0 to n. Depending on the value of n, some of these terms will vanish identically. For example, it is easily seen that for any n, all terms involving κ_{pq}^n will vanish because of the contraction property (2.36).

4. Note that the expansion parameters κ_{pq} and μ_{pq} arise because of our choice of excluding a spherical cavity from the origin of the integral (2.23). By choosing a non-spherical cavity shape, we would have obtained a different cavity strain field and hence different expansion parameters. In a future paper, we shall fully explore the implications of excluding *non-spherical* cavities.

5. For macroscopically isotropic media, it is shown in the subsequent section that the series expressions (2.68) may be regarded as expansions that perturb about the optimal structures that realize the Hashin–Shtrikman bounds (Hashin and Shtrikman, 1963; Hashin, 1965). For macroscopically anisotropic media, the series expressions (2.68) may be regarded as expansions that perturb about the optimal structures that realize Willis' (1977) bounds. This point will be demonstrated in Section 4.

6. The n-point tensors $\mathbf{B}_n^{(p)}$ for all n generally will not possess common principal axes. This implies that for general media the principal axes of the macroscopic stiffness tensor $\mathbf{C}_e$ (which has the same symmetry properties $\mathbf{L}_e^{(q)}$) will rotate as the phase moduli ratio change, such as composites with chirality, i.e. composites with some degree of left- or right-handed asymmetry. Nonetheless, there exists a large class of media which has the symmetry required for all the $\mathbf{B}_n^{(p)}$ to possess common principal axes (e.g. a random distribution of oriented, ellipsoids or oriented cylinders in a matrix); in such instances n-point tensor multiplication is commutative.

3. EXACT SERIES EXPANSIONS FOR MACROSCOPICALLY ISOTROPIC MEDIA

In this section we specialize the previous results to macroscopically isotropic media. For such composites, it is seen from formula (2.49) that

$$\mathbf{L}_{\rm e}^{(q)} = [dK_q+2(d-1)G_q]\left[\kappa_{{\rm e}q}\mathbf{\Lambda}_{\rm h}+\frac{(d+2)G_q}{d(K_q+2G_q)}\mu_{{\rm e}q}\mathbf{\Lambda}_{\rm s}\right], \tag{3.1}$$

where the *effective polarizabilities* κ_{eq} and μ_{eq} are defined by the *scalar* relations

$$\kappa_{eq} = \frac{K_{\rm e}-K_q}{K_{\rm e}+\dfrac{2(d-1)}{d}G_q}, \quad q=1,2, \tag{3.2}$$

$$\mu_{eq} = \frac{G_{\rm e}-G_q}{G_{\rm e}+\dfrac{G_q[dK_q/2+(d+1)(d-2)G_q/d]}{K_q+2G_q}}, \quad q=1,2. \tag{3.3}$$

Therefore, series (2.68) becomes

$$\phi_p^2\left[\frac{\kappa_{pq}}{\kappa_{eq}}\mathbf{\Lambda}_{\rm h}+\frac{\mu_{pq}}{\mu_{eq}}\mathbf{\Lambda}_{\rm s}\right] = \phi_p\mathbf{I}-\sum_{n=2}^{\infty}\mathbf{B}_n^{(p)}. \tag{3.4}$$

3.1. *Bulk modulus*

In order to obtain an explicit expression for the effective bulk modulus $K_{\rm e}$, we take the quadruple dot product of the hydrostatic projection tensor $\mathbf{\Lambda}_{\rm h}$ with (3.4) and use identities (2.5) and (2.6) to yield

$$\phi_p^2\frac{\kappa_{pq}}{\kappa_{eq}} = \phi_p-\sum_{n=3}^{\infty}C_n^{(p)}, \tag{3.5}$$

where the scalar microstructural coefficients are given by

$$C_n^{(p)} = \mathbf{\Lambda}_{\rm h} \vdots \mathbf{B}_n^{(p)}, \tag{3.6}$$

as $\mathbf{B}_n^{(p)}$ is given by (2.70), and $\vdots$ denotes the quadruple dot product. Note that this series begins with the third-order term, i.e. $C_2^{(p)}$ is zero because the invariant $H_{iikk}^{(q)}$ vanishes [cf. (2.36)].

Note that when the shear moduli of the phases are equal ($G_1 = G_2$), the well-known exact result

$$\kappa_{eq} = \phi_p\kappa_{pq} \tag{3.7}$$

due to Hill (1963) immediately follows from (3.5) because

$$U_{ijkl}^{(q)}(\mathbf{r}) = [dK_q+2(d-1)G_q]\kappa_{pq}\frac{\delta_{ij}}{d}H_{mmkl}^{(q)}(\mathbf{r}) \tag{3.8}$$

and therefore each coefficient $C_n^{(p)}$ possesses the invariant $\mathbf{\Lambda}_{\rm h} \vdots \mathbf{H}^{(q)}$ which vanishes. We see that for such a composite, the effective bulk modulus $K_{\rm e}$ is independent of the microstructure in any space dimension d.

Classical perturbation expansions involve parameters of smallness that are simple differences in the phase moduli, e.g. (K_2-K_1) and (G_2-G_1) (Beran and Molyneux, 1968; Silnutzer, 1972; Milton and Phan-Thien, 1982; Milton, 1984). Such expansions have a smaller radius of convergence and hence require many terms in the series if the phase moduli are appreciably different from one another. By contrast, the expansions represented by (3.5) are *non-classical* in the sense that the expansion parameters are the polarizabilities κ_{pq} and μ_{pq} and, for certain microgeometries (described below), can converge rapidly for any values of the phase moduli.

In order to better understand the physical meaning of the expansion (3.5), it is helpful to consider the microgeometries for which the microstructural parameters $C_n^{(p)}$ vanish for any values of the phase moduli, i.e. for which class of composites is the relation

$$\kappa_{eq} = \phi_p \kappa_{pq} \tag{3.9}$$

or, equivalently,

$$\frac{K_e - K_q}{K_e + \dfrac{2(d-1)}{d} G_q} = \frac{K_p - K_q}{K_p + \dfrac{2(d-1)}{d} G_q} \phi_p \tag{3.10}$$

exact. [Note that this is the same as the Hill relation (3.7) but here we are not restricting the phase shear moduli.] For $d = 2$ and $d = 3$, expression (3.10) is recognized to coincide with the Hashin–Shtrikman bounds on the effective bulk modulus for any isotropic two-phase composite (Hashin and Shtrikman, 1963; Hashin, 1965) and hence is exact for the assemblages of coated circles ($d = 2$) and coated spheres ($d = 3$) that realize the bounds. The Hashin–Shtrikman bounds are also realized for certain finite-rank laminates in both two and three dimensions (Francfort and Murat, 1986). It is important to emphasize that for either the coated-inclusion assemblages or finite-rank laminates, one of the phases is always a disconnected, dispersed phase in a connected matrix phase (except in the trivial instance when the generally dispersed phase fills all of space). Result (3.10) is the d-dimensional generalization of the Hashin–Shtrikman bounds on K_e for any $d \geqslant 2$; for $K_2 \geqslant K_1$ and $G_2 \geqslant G_1$, it gives a lower bound for $q = 1$ and $p = 2$ and an upper bound for $q = 2$ and $p = 1$.

In light of this discussion, series (3.5) can be viewed as an expansion that perturbs around the optimal Hashin–Shtrikman structures. *Therefore, it is expected that expansion* (3.5) *will converge rapidly for any values of the phase moduli for dispersions in which the inclusions, taken to be the polarized phase, are prevented from forming large clusters*. Consequently, we contend that the first few terms of this expansion will provide an excellent approximation of the effective bulk modulus K_e of such dispersions. We demonstrate quantitatively in the sequel to this paper that this is indeed the case for a variety of ordered and disordered dispersions when (3.5) is truncated after third-order terms.

Accordingly, let us write out (3.5) through third-order terms and simplify it. We find that

$$\begin{aligned} \phi_p \frac{\kappa_{pq}}{\kappa_{eq}} &= 1 - \frac{C_3^{(p)}}{\phi_p} \\ &= 1 - \frac{(d+2) G_q \kappa_{pq} \mu_{pq}}{d(K_q + 2G_q)} \frac{M_p}{\phi_p}, \end{aligned} \tag{3.11}$$

where M_p is a three-point microstructural parameter independent of the phase moduli given by

$$M_p = \frac{d^2}{\Omega^2}\int\int \frac{\mathrm{d}\mathbf{r}}{r^d}\frac{\mathrm{d}\mathbf{s}}{s^d}[d(\mathbf{n}\cdot\mathbf{m})^2-1]\left[S_3^{(p)}(\mathbf{r},\mathbf{s})-\frac{S_2^{(p)}(\mathbf{r})S_2^{(p)}(\mathbf{s})}{\phi_p}\right] \tag{3.12}$$

and $\mathbf{n} = \mathbf{r}/|\mathbf{r}|$ and $\mathbf{m} = \mathbf{s}/|\mathbf{s}|$ are unit vectors. Here we have used the identity (2.37).

The three-point microstructural parameters M_1 and M_2 are not independent of one another; specifically, one has that

$$M_1 + M_2 = (d-1)\phi_1\phi_2. \tag{3.13}$$

This is easily shown using the fact that relation (3.11) yields exact results for the effective bulk modulus K_e through third-order in the difference in the moduli, i.e.

$$K_e = K_q + a_1^{(p)}(K_p-K_q) + a_2^{(p)}(K_p-K_q)^2 + a_3^{(p)}(K_p-K_q)^3 + b_3^{(p)}(K_p-K_q)^2(G_p-G_q), \tag{3.14}$$

where

$$a_1^{(p)} = \phi_p, \tag{3.15}$$

$$a_2^{(p)} = -\frac{d\phi_p\phi_q}{dK_q+2(d-1)G_q}, \tag{3.16}$$

$$a_3^{(p)} = \frac{d^2\phi_p\phi_q^2}{[dK_q+2(d-1)G_q]^2}, \tag{3.17}$$

$$b_3^{(p)} = \frac{2dM_p}{[dK_q+2(d-1)G_q]^2}. \tag{3.18}$$

Now since K_e remains invariant under different labels of the reference phase, relation (3.13) follows immediately.

Of course, for $d = 2$ and $d = 3$, expansions (3.14) agree with the corresponding expansions of the Silnutzer (1972) and Beran–Molyneux (1966) bounds on K_e, respectively, which are also exact through third-order in the difference in the phase moduli and involve the related three-point parameters ζ_p. Comparing (3.14) to these expansions reveals that for $d = 2$

$$\zeta_p = \frac{M_p}{\phi_1\phi_2} = \frac{4}{\pi\phi_q\phi_p}\int_0^\infty \frac{\mathrm{d}r}{r}\int_0^\infty \frac{\mathrm{d}s}{s}\int_0^\pi \mathrm{d}\theta\cos(2\theta)\left[S_3^{(p)}(r,s,t)-\frac{S_2^{(p)}(r)S_2^{(p)}(s)}{S_1^{(p)}}\right] \tag{3.19}$$

and for $d = 3$

$$\zeta_p = \frac{M_p}{2\phi_1\phi_2} = \frac{9}{2\phi_q\phi_p}\int_0^\infty \frac{\mathrm{d}r}{r}\int_0^\infty \frac{\mathrm{d}s}{s}\int_{-1}^1 \mathrm{d}(\cos\theta)P_2(\cos\theta)\left[S_3^{(p)}(r,s,t)-\frac{S_2^{(p)}(r)S_2^{(p)}(s)}{S_1^{(p)}}\right], \tag{3.20}$$

where P_2 is the Legendre polynomial of order 2 and θ is the angle opposite the side of the triangle of length t. It is well established that in two (Milton, 1982) and three

(Torquato, 1980; Milton, 1981a) dimensions, the parameters ζ_p must lie in the closed interval [0, 1]. The parameter ζ_p has been computed for a variety of model microstructures (see the review of Torquato, 1991).

For any space dimension d, we have that

$$\zeta_p = \frac{M_p}{(d-1)\phi_1\phi_2}. \tag{3.21}$$

This relation was first obtained by Torquato (1985) in the context of finding the effective conductivity, where the notation $A_3^{(p)}$ was used for M_p.

3.2. *Shear modulus*

In order to obtain an explicit expression for the effective shear modulus G_e, we take a quadruple dot product of the shear tensor $\mathbf{\Lambda}_s$ with (3.4) and use identities (2.5) and (2.7) to yield

$$\phi_p^2 \frac{\mu_{pq}}{\mu_{eq}} = \phi_p - \sum_{n=3}^{\infty} D_n^{(p)}, \tag{3.22}$$

where the scalar coefficients

$$D_n^{(p)} = \frac{2}{(d+2)(d-1)} \mathbf{\Lambda}_s \vdots \mathbf{B}_n^{(p)}, \tag{3.23}$$

and $\mathbf{B}_n^{(p)}$ is given by (2.70). Note that this series begins with the third-order term, i.e. $D_2^{(p)}$ is zero because the invariant $H_{ikik}^{(q)}$ vanishes [cf. (2.36)].

Truncating the series (3.22) after the first term yields

$$\mu_{eq} = \phi_p \mu_{pq} \tag{3.24}$$

or, equivalently,

$$\frac{G_e - G_q}{G_e + \dfrac{G_q[dK_q/2 + (d+1)(d-2)G_q/d]}{K_q + 2G_q}} = \frac{G_p - G_q}{G_p + \dfrac{G_q[dK_q/2 + (d+1)(d-2)G_q/d]}{K_q + 2G_q}} \phi_p. \tag{3.25}$$

Following the discussion on the bulk modulus, it is helpful to consider the class of microgeometries for which the formula (3.24) is exact. For $d = 2$ and $d = 3$, expression (3.24) is recognized to coincide with the Hashin–Shtrikman bounds on the effective shear modulus for any isotropic two-phase composite and hence is exact for the finite-rank, hierarchical laminate composites that realize the bounds (Francfort and Murat, 1986). Again, for such hierarchical laminates, one of the phases is always a disconnected, dispersed phase in a connected matrix phase (except in the trivial instance when the generally dispersed phase in a connected matrix phase fills all of space). Result (3.24) is the d-dimensional generalization of the Hashin–Shtrikman bounds on G_e for any $d \geqslant 2$; for the "well-ordered" case $(K_2 - K_1)(G_2 - G_1) \geqslant 0$ and $G_2 \geqslant G_1$, it gives a lower bound for $q = 1$ and $p = 2$ and an upper bound for $q = 2$ and

$p = 1$. [Bounds for the "badly-ordered" case $(K_2 - K_1)(G_2 - G_1) \leqslant 0$ were obtained by Walpole (1966) for the instance $d = 3$.]

Consequently, series (3.22) can be viewed as an expansion which perturbs around the optimal hierarchical laminates that achieve the Hashin–Shtrikman bounds. *Therefore, it is expected that expansion* (3.22) *will converge rapidly for any values of the phase moduli for dispersions in which the inclusions, taken to the polarized phase, are prevented from forming large clusters.* In the sequel to this paper, we will demonstrate quantitatively that (3.22) truncated after third-order terms provides an excellent approximation to the effective shear modulus of a variety of ordered and disordered dispersions.

Writing (3.22) through third-order terms and simplifying yields

$$\phi_p = \frac{\mu_{pq}}{\mu_{eq}} = 1 - \frac{D_3^{(p)}}{\phi_p}, \tag{3.26}$$

where

$$\begin{aligned}\frac{D_3^{(p)}}{\phi_p} &= \frac{2}{(d+2)(d-1)} \mathbf{A}_s : \mathbf{B}_n^{(p)} \\ &= \frac{2}{(d+2)(d-1)} \int\int \mathrm{d}\mathbf{r}\, \mathrm{d}\mathbf{s} \left[U_{ijkl}^{(q)}(\mathbf{r}) U_{klij}^{(q)}(\mathbf{s}) - \frac{U_{iikl}^{(q)}(\mathbf{r}) U_{klmm}^{(q)}(\mathbf{s})}{d} \right] \left[S_3^{(p)}(\mathbf{r},\mathbf{s}) \right. \\ &\quad \left. - \frac{S_2^{(p)}(\mathbf{r}) S_2^{(p)}(\mathbf{s})}{\phi_p} \right] \\ &= \frac{2G_q \kappa_{pq} \mu_{pq}}{d(d-1)(K_q + 2G_q)} \frac{M_p}{\phi_p} + \frac{(d^2-4) G_q (2K_q + 3G_q) \mu_{pq}^2}{2d(d-1)(K_q+2G_q)^2} \frac{M_p}{\phi_p} \\ &\quad + \frac{1}{2d(d-1)} \left[\frac{dK_q + (d-2)G_q}{K_q + 2G_q} \right]^2 \mu_{pq}^2 \frac{N_p}{\phi_p}. \end{aligned} \tag{3.27}$$

The quantity N_p is a microstructural parameter independent of the phase moduli given by

$$\begin{aligned} N_p = -\frac{(d+2)(5d+6)}{d^2} M_p + \frac{(d+2)^2}{\Omega^2} \int\int \frac{\mathrm{d}\mathbf{r}}{r^d} \frac{\mathrm{d}\mathbf{s}}{s^d} [d(d+2)(\mathbf{n}\cdot\mathbf{m})^4 - 3] \left[S_3^{(p)}(\mathbf{r},\mathbf{s}) \right. \\ \left. - \frac{S_2^{(p)}(\mathbf{r}) S_2^{(p)}(\mathbf{s})}{\phi_p} \right], \end{aligned} \tag{3.28}$$

where $\mathbf{n} = \mathbf{r}/|\mathbf{r}|$ and $\mathbf{m} = \mathbf{s}/|\mathbf{s}|$ are unit vectors. In obtaining (3.28), we used the identity (2.38) and definition (2.32).

As in the case of the microstructural parameters M_1 and M_2, the three-point parameters N_1 and N_2 are not independent of one another; specifically, one has that

$$N_1 + N_2 = (d-1)\phi_1 \phi_2. \tag{3.29}$$

This is shown using the fact that relation (3.28) yields exact results for the effective shear modulus G_e through third-order in the difference in the moduli, i.e.

$$G_e = G_q + c_1^{(p)}(G_p - G_q) + c_2^{(p)}(G_p - G_q)^2 + c_3^{(p)}(G_p - G_q)^3 + d_3^{(p)}(G_p - G_q)^2(K_p - K_q), \tag{3.30}$$

where

$$c_1^{(p)} = \phi_p, \tag{3.31}$$

$$c_2^{(p)} = -\frac{2d\phi_p\phi_q(K_q + 2G_q)}{(d+2)G_q[dK_q + 2(d-1)G_q]}, \tag{3.32}$$

$$c_3^{(p)} = \frac{4d^2(K_q + 2G_q)^2\phi_p\phi_q^2}{(d+2)^2G_1^2[dK_q + 2(d-1)G_q]^2} + \frac{2d(d-2)[2K_q + 3G_q]M_p}{(d+2)(d-1)G_q[dK_q + 2(d-1)G_q]^2}$$
$$+ \frac{2d}{(d-1)(d+2)^2G_q^2}\left[\frac{dK_q + (d-2)G_q}{dK_q + 2(d-1)G_q}\right]^2 N_p, \tag{3.33}$$

$$d_3^{(p)} = \frac{4dM_p}{(d+2)(d-1)[dK_q + 2(d-1)G_q]^2}. \tag{3.34}$$

Again, since G_e remains invariant under different labels of the reference phase, relation (3.30) follows immediately.

For $d = 2$ and $d = 3$, expansions (3.31) agree with the corresponding expansions of the Silnutzer (1972) and McCoy (1970) bounds on G_e, respectively, which are also exact through third-order in the difference in the phase moduli and involve the aforementioned three-point parameter ζ_p as well as another three-point parameter η_p, which also lies in the interval [0, 1] (Milton, 1981a, 1982). Comparing (3.31) to these expansions reveals that N_p and η_p are simply related to one another; specifically, for $d = 2$, we have

$$N_p = \phi_1\phi_2\eta_p \tag{3.35}$$

where

$$\eta_p = \frac{16}{\pi\phi_q\phi_p}\int_0^\infty \frac{dr}{r}\int_0^\infty \frac{ds}{s}\int_0^\pi d\theta\cos(4\theta)\left[S_3^{(p)}(r,s,t) - \frac{S_2^{(p)}(r)S_2^{(p)}(s)}{\phi_p}\right], \tag{3.36}$$

and for $d = 3$ we have

$$N_p = 2\phi_1\phi_2\eta_p, \tag{3.37}$$

where

$$\eta_p = \frac{5\zeta_2}{21} + \frac{150}{7\phi_q\phi_p}\int_0^\infty \frac{dr}{r}\int_0^\infty \frac{ds}{s}\int_{-1}^1 d(\cos\theta)P_4(\cos\theta)\left[S_3^{(p)}(r,s,t) - \frac{S_2^{(p)}(r)S_2^{(p)}(s)}{\phi_p}\right], \tag{3.38}$$

where P_4 is the Legendre polynomial of order four. The parameter η_p has been computed for a variety of model two- and three-dimensional microstructures (see the review of Torquato, 1991).

More generally, for any space dimension d, we have that

$$\eta_p = \frac{N_p}{(d-1)\phi_1\phi_2}, \tag{3.39}$$

where N_p is given by (3.28). We see that this relation has the same form as the one for ζ_p given by (3.21).

3.3. *Effective behavior in the limit of infinite dimension*

It is of interest to study the effective elastic behavior in the limit that the space dimension tends to infinity ($d \to \infty$). In this limit, we will show that the effective shear modulus G_e for any isotropic composite is given exactly by the *arithmetic average*, provided that the phase moduli are non-zero. Interestingly, the effective electric (thermal) conductivity of an isotropic composite also tends to the arithmetic average as $d \to \infty$. All of the aforementioned results turn out to apply not only to two-phase composites but to n-phase composites. By contrast, we will demonstrate that the effective bulk modulus K_e depends on the microstructure as $d \to \infty$.

The first hint of this interesting behavior as $d \to \infty$ can be gleaned by examining the exact third-order expansions (3.14) and (3.30) in this limit. We find from these relations that as $d \to \infty$,

$$\begin{aligned} K_e &= K_1 + \phi_2(K_2 - K_1) - \frac{\phi_1\phi_2}{K_1 + 2G_1}(K_2 - K_1)^2 \\ &\quad - \frac{\phi_1^2\phi_2}{(K_1 + 2G_1)^2}(K_2 - K_1)^3 + \frac{2\phi_1\phi_2\zeta_2}{(K_1 + 2G_1)^2}(K_2 - K_1)^2(G_2 - G_1), \\ G_e &= G_1 + \phi_2(G_2 - G_1) = \phi_1 G_1 + \phi_2 G_2, \end{aligned}$$

where we have taken $p = 2$ and $q = 1$. Here we used the fact that N_p is asymptotic to d as $d \to \infty$. Thus, we see that in contrast to the effective bulk modulus, the second- and third-order terms vanish in the case of the effective shear modulus. Taking the limit $d \to \infty$ in the corresponding third-order expansion of the effective conductivity σ_e (Torquato, 1985), reveals that

$$\sigma_e = \phi_1\sigma_1 + \phi_2\sigma_2,$$

where σ_1 and σ_2 are the phase conductivities.

Now the general proof for arbitrary conditions and for any number of phases requires the d-dimensional Hashin–Shtrikman–Walpole bounds for n-phase composites which are stated in Appendix B.

PROPOSITION 1. For any d-dimensional, macroscopically isotropic, n-phase composite possessing non-zero phase moduli, the effective shear modulus G_e is independent of the microstructure and is exactly given by the arithmetic average, i.e.

$$G_e = \sum_{i=1}^{n} \phi_i G_i, \tag{3.40}$$

in the limit that the space dimension becomes infinite ($d \to \infty$).

Proof. The proof is immediate given the d-dimensional bounds for n-phases on the effective shear modulus given by relation (B.2) in Appendix B. Under the stated conditions, the bounds coincide and equal expression (3.40).

PROPOSITION 2. For any d-dimensional, macroscopically isotropic, n-phase composite, the effective bulk modulus K_e generally depends on the microstructure in the limit that the space dimension becomes infinite ($d \to \infty$).

Proof. The proof follows from the fact that the d-dimensional, n-phase bounds on the effective bulk modulus given by (B.1) are realizable (see earlier discussion and Appendix B) and generally do not coincide in the limit $d \to \infty$.

PROPOSITION 3. For any d-dimensional, macroscopically isotropic, n-phase composite possessing non-zero phase conductivities, the effective conductivity σ_e is independent of the microstructure and is exactly given by the arithmetic average, i.e.

$$\sigma_e = \sum_{i=1}^{\phi} \phi_i \sigma_i, \tag{3.41}$$

in the limit that the space dimension becomes infinite ($d \to \infty$).

Proof. Under the stated conditions, the d-dimensional, n-phase bounds (B.7) on the effective conductivity coincide and equal expression (3.41).

Remarks

Interestingly, in lower dimensions ($d = 2$ or $d = 3$), it has been demonstrated that the effective conductivity of two-phase composites is more closely related to the effective bulk modulus rather than the effective shear modulus (Milton, 1984; Gibiansky and Torquato, 1995, 1996). Why do the effective conductivity and shear modulus tend to the arithmetic mean as $d \to \infty$ (implying the same constant electric or strain fields in each phase), in contrast to the behavior of the effective bulk modulus in this limit? Roughly speaking, this can be explained by considering the phase-interface continuity conditions on the electric or strain fields and the fact that the energy associated with an *isotropic composite* must be the same in any direction. For example, in the case of conduction, the components of the electric field vector are continuous in all of the d directions, except the direction normal to the interface, and thus, for an isotropic composite in the limit $d \to \infty$, the electric fields in each phase approach the same constant value. Although similar arguments apply to the shear modulus, the bulk modulus behaves differently since the quantity dK_e, rather than K_e, is an eigenvalue of the stiffness tensor and thus the energy associated with the former is unbounded for isotropic composites as $d \to \infty$. Note that these arguments apply to materials possessing non-linear stress–strain (flux–current) laws and hence both G_e and σ_e will tend to arithmetic averages as $d \to$ for *non-linear composites*.

4. REMARKS ON MACROSCOPICALLY ANISOTROPIC MEDIA

In the previous section, we showed that the series expansion for macroscopically isotropic composites may be regarded as one that perturbs around the structures that realize the isotropic Hashin–Shtrikman bounds. Based on this observation, one would expect that the expansion for macroscopically anisotropic composites can be regarded as one that perturbs around the optimal structures that realize the anisotropic generalization of the Hashin–Shtrikman bounds obtained by Willis (1977). This indeed is the case for macroscopically anisotropic composites as elaborated on below.

First let us note that the two-point tensor coefficient $\mathbf{B}_2^{(p)}$ [cf. (2.69)] does not vanish for statistically anisotropic media since the two-point probability function $S_2^{(p)}(\mathbf{r})$ depends on the distance $r = |\mathbf{r}|$ as well as the orientation of the vector $\mathbf{r}$. Recall that $\mathbf{B}_2^{(p)} = 0$ for statistically isotropic media. Now consider microgeometries, for which the n-point tensors coefficients, $\mathbf{B}_n^{(p)} = 0$ for all $n \geqslant 3$. For such composites, then (2.68) reduces exactly to

$$\phi_p^2\mathbf{L}^{(q)} : (\mathbf{L}_e^{(q)})^{-1} = \phi_p\mathbf{I} - \mathbf{B}_2^{(p)}. \tag{4.1}$$

Multiplying this relation by $(\mathbf{L}^{(q)})^{-1}$ from the left gives

$$\begin{aligned}\phi_p^2(\mathbf{L}_e^{(q)})^{-1} &= \phi_p(\mathbf{L}^{(q)})^{-1} - (\mathbf{L}^{(q)})^{-1} : \mathbf{B}_2^{(p)},\\ &= \phi_p(\mathbf{L}^{(q)})^{-1} - \int_\varepsilon \mathrm{d}2\mathbf{H}^{(q)}(1,2)[S_2^{(p)}(1,2) - \phi_p^2].\end{aligned} \tag{4.2}$$

Expression (4.2) indeed are the generalized Hashin–Shtrikman bounds for anisotropic composites derived by Willis (1977), albeit expressed in a different form than given originally by Willis. Now since the fourth-order tensor $\mathbf{\Gamma}^\infty$ of Willis is equal to $-\mathbf{G}^{(q)}$ defined by (2.29), the integral of (4.2) can be written as

$$\int_\varepsilon \mathrm{d}2\mathbf{H}^{(q)}(1,2)[S_2^{(p)}(1,2) - \phi_p^2] = \mathbf{A}^{(q)}\phi_p\phi_r - \int \mathrm{d}2\mathbf{\Gamma}^\infty(1,2)[S_2^{(p)}(1,2) - \phi_p^2], \tag{4.3}$$

where $\mathbf{A}^{(q)}$ is the constant tensor given by (2.30).

Avellaneda (1987) showed, among other results, that Willis' bounds are attainable by finite-rank laminates. Thus, the general expansion (2.68) for macroscopically anisotropic media may be regarded as one that perturbs about such laminates. Again, for such structures, one of the phases is always a disconnected, dispersed phase in a connected matrix phase (except in the trivial instance when the generally dispersed phase fills all of space).

In the special case of anisotropic composites containing oriented, similar, ellipsoidal inclusions or microstructures in which $S_2^{(p)}(\mathbf{r})$ possesses ellipsoidal symmetry, it can be shown (using the methods of the Appendix) that the integral above has the following simple form:

$$\int_\varepsilon \mathrm{d}2\mathbf{H}^{(q)}(1,2)[S_2^{(p)}(1,2) - \phi_p^2] = [\mathbf{A}^{(q)} - \mathbf{P}^{(q)}]\phi_p\phi_q, \tag{4.4}$$

where the constant fourth-order tensor $\mathbf{P}^{(q)}$ depends upon the microstructure only through the aspect ratios of the inclusions. More specifically, $\mathbf{P}^{(q)}$ can be related to the well-known Eshelby tensor $\mathbf{S}^{(q)}$ for an ellipsoidal inclusion via

$$\mathbf{P}^{(q)} = \mathbf{S}^{(q)} : (\mathbf{C}^{(q)})^{-1}, \tag{4.5}$$

where $\mathbf{C}^{(q)}$ is the stiffness tensor of the reference phase q. Willis (1977) was the first to recognize a relation of the type (4.4). (He denoted $\mathbf{P}^{(q)}$ by $\mathbf{P}_0$ and expressed the integral in terms of the aforementioned tensor $\mathbf{\Gamma}^{\infty}$.) Weng (1992) found the explicit relation (4.5) using a different procedure. Note that when the ellipsoidal inclusion becomes a sphere, the integral of (4.4) is identically zero and hence

$$\mathbf{P}^{(q)} = \mathbf{A}^{(q)}, \tag{4.6}$$

where $\mathbf{A}^{(q)}$ is given by (2.30).

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REFERENCES

Avellaneda, M. (1987) Optimal bounds and microgeometries for elastic composites. *SIAM J. Appl. Math.* **47,** 1216–1228.

Beran, M. J. and Molyneux, J. (1966) Use of classical variational principles to determine bounds for the effective bulk modulus in heterogeneous media. *Q. Appl. Math.* **24,** 107–118.

Brown, W. F. (1955) Solid mixture permittivities. *J. Chem. Phys.* **23,** 1514–1517.

Dederichs, P. H. and Zeller, R. (1973) Variational treatment of the elastic constants of disordered materials. *Z. Physik* **259,** 103–116.

Eshelby, J. D. (1957) The determination of the elastic field of an ellipsoidal inclusion and related problems. *Proc. R. Soc. Lond. A* **241,** 376–396.

Francfort, G. A. and Murat, F. (1986) Homogenization and optimal bounds in linear elasticity, *Archives Rat. Mech. Analysis* **94,** 307–334.

Gibiansky, L. V. and Torquato, S. (1995) Rigorous link between the conductivity and elastic moduli of fiber-reinforced composite materials. *Phil. Trans. R. Soc. Lond.* **343,** 243–278.

Gibiansky, L. V. and Torquato, S. (1996) Connection between the conductivity and elastic moduli of isotropic composites. *Proc. R. Soc. Lond. A* **452,** 253–283.

Gubernatis, J. E. and Krumhansl (1975) Macroscopic engineering properties of polycrystalline materials: elastic properties. *J. Appl. Phys.* **46,** 1875–1883.

Hashin, Z. (1965) On elastic behaviour of fibre reinforced materials of arbitrary transverse phase geometry. *J. Mech. Phys. Solid* **13,** 119–134.

Hashin, Z. and Shtrikman, S. (1963) A variational approach to the theory of the elastic behaviour of multiphase materials. *J. Mech. Phys. Solids* **11,** 127–140.

Hill, R. (1963) Elastic properties of reinforced solids: some theoretical results. *J. Mech. Phys. Solids* **11,** 357–373.

Kellog, O. D. (1953) *Foundations of Potential Theory*. Dover, New York.

Lurie, K. A. and Cherkaev, A. V. (1985) The problem of formation of an optimal multi-component composite. *J. Opt. Th. Appl.* **46,** 571–589.
McCoy, J. J. (1970) *Recent Advances in Engineering Sciences* 5. Gordon & Breach, New York.
McCoy, J. J. (1979) On the calculation of bulk properties of heterogeneous materials. *Q. Appl. Math.* **36,** 137–149.
Milton, G. W. (1981a) Bounds on the electromagnetic, elastic, and other properties of two-component composites. *Phys. Rev. Lett.* **46,** 542–545.
Milton, G. W. (1981b) Concerning bounds on the transport and mechanical properties of multi-component composite materials. *Appl. Phys.* **A26,** 125–130.
Milton, G. W. (1982) Bounds on the elastic and transport properties of two-component composites. *J. Mech. Phys. Solids* **30,** 177–191.
Milton, G. W. (1984) Correlation of the electromagnetic and elastic properties of composites and microgeometries corresponding with effective medium approximations. In *Physics and Chemistry of Porous Media*, ed. D. L. Johnson and P. N. Sen. AIP.
Milton, G. W. and Phan-Thien, N. (1982) New bounds on the effective moduli of two-component materials. *Proc. R. Soc. Lond. A* **380,** 305–331.
Sen, A. K. and Torquato, S. (1989) Effective electrical conductivity of two-phase disordered anisotropic composite media. *Physical Review B* **39,** 4504–4515.
Silnutzer, N. (1972) Effective constants of statistically homogeneous materials. Ph.D. dissertation, University of Pennsylvania, Philadelphia.
Torquato, S. (1980) Microscopic approach to transport in two-phase random media. Ph.D. dissertation, State University of New York, Stony Brook, New York.
Torquato, S. (1985) Electrical conductivity of two-phase disordered composite media. *J. Appl. Phys.* **58,** 3790–3797.
Torquato, S. (1991) Random heterogeneous media: microstructure and improved bounds on effective properties. *Appl. Mech. Rev.* **44,** 37–76.
Torquato, S. and Stell, G. (1982) Microstructure of two-phase random media—I. The *n*-point probability functions. *J. Chem. Phys.* **77,** 2071–2077.
Walpole, L. J. (1966) On the bounds for the overall elastic moduli of inhomogeneous systems—I. *J. Mech. Phys. Solids* **14,** 151–162.
Weng, G. J. (1992) Explicit evaluation of Willis' bounds with ellipsoidal inclusions. *Int. J. Eng. Sci.* **30,** 83–92.
Willis, J. R. (1977) Bounds and self-consistent estimates for the overall moduli of anisotropic composites. *J. Mech. Phys. Solids* **25,** 185–202.
Willis, J. R. (1981) Variational and related methods for the overall properties of composites. *Adv. Appl. Mech.* **21,** 1–78.

APPENDIX A: IMPROPER INTEGRALS INVOLVING GREEN'S FUNCTIONS

In this Appendix we discuss the treatment of *improper* integrals involving the Green's function for elasticity by generalizing the corresponding results of Kellog (1953) for potential theory. Consider improper volume integrals whose integrands, generally represented by $\mathscr{F}(\mathbf{x}')$, possess a singularity at the point $\mathbf{x}' = \mathbf{x}$. According to Kellog (1953), the integral over the volume V

$$\int_V \mathscr{F}(\mathbf{x}')\,\mathrm{d}\mathbf{x}'$$

is said to be convergent, or to exist, provided the limit

$$\lim_{\varepsilon \to 0} \int_{V-v} \mathscr{F}(\mathbf{x}')\,\mathrm{d}\mathbf{x}'$$

exists, independent of the shape of an excluded region v that contains the point $\mathbf{x}$ in its interior and has a maximum chord length no longer than ε. If the limit exists, then any convenient excluded cavity shape may be used. A d-dimensional sphere is often the most convenient shape. If v is chosen to be a sphere centered at $\mathbf{x}' = \mathbf{x}$, the limit, if it exists, is the d-dimensional analog of the Cauchy principal value of a one-dimensional improper integral.

We focus our attention on improper integrals involving the singular infinite-space Green's function $g_{ij}^{(q)}$ in d dimensions for an isotropic elastic material with bulk modulus K_q and shear modulus G_q in the absence of body forces:

$$g_{ij}^{(q)}(\mathbf{r}) = \begin{cases} \dfrac{1}{2\Omega G_q}\ln\left(\dfrac{1}{r}\right)\delta_{ij} + b_q n_i n_j, & d = 2 \\ a_q \dfrac{\delta_{ij}}{r^{d-2}} + b_q \dfrac{n_i n_j}{r^{d-2}}, & d \geqslant 3 \end{cases}, \tag{A.1}$$

where

$$a_q = \frac{1}{2(d-2)\Omega G_q}\frac{dK_q + (3d-2)G_q}{dK_q + 2(d-1)G_q}, \tag{A.2}$$

$$b_q = \frac{1}{2\Omega G_q}\frac{dK_q + (d-2)G_q}{dK_q + 2(d-1)G_q}. \tag{A.3}$$

Moreover,

$$\Omega(d) = \frac{2\pi^{d/2}}{\Gamma(d/2)} \tag{A.4}$$

is the total solid angle contained in a d-dimensional sphere, $r = |\mathbf{r}|$, and $\mathbf{n} = \mathbf{r}/r$.

Now let us consider the d-dimensional volume integrals

$$w_{ij}(\mathbf{x}) = \int_V g_{ij}^{(q)}(\mathbf{r})h(\mathbf{x}')\,\mathrm{d}\mathbf{x}', \tag{A.5}$$

$$W_{ijk}(\mathbf{x}) = \int_V \frac{\partial g_{ij}^{(q)}(\mathbf{r})}{\partial x_k}h(\mathbf{x}')\,\mathrm{d}\mathbf{x}', \tag{A.6}$$

where $h(\mathbf{x})$ is a function that is piecewise differentiable and $\mathbf{r} = \mathbf{x} - \mathbf{x}'$. Several useful theorems can now be stated.

Theorem I. Both $w_{ij}(\mathbf{x})$ and $W_{ijk}(\mathbf{x})$ exist for all $\mathbf{x}$.

Theorem II. $W_{ijk}(\mathbf{x}) = \partial w_{ij}(\mathbf{x})/\partial x_k$.

Theorem I states that the integrals (A.6) and (A.7) meet the aforementioned existence criterion. Theorem II states that $w_{ij}(\mathbf{x})$ can be differentiated under the integral sign. Generally, this operation cannot be automatically performed with improper integrals, even if both integrals exist. Kellog (1953) proved corresponding theorems for the "coulombic" terms (i.e. the first terms) in (A.1) as his interest was in potential theory. The proofs of Theorems I ad II follow in precisely the same fashion and hence will not be presented here.

The symmetrized gradient of $W_{ijk}(\mathbf{x})$ will be of central interest but to get it we cannot differentiate $W_{ijk}(\mathbf{x})$ under the integral sign because the resulting integral does not even exist, i.e. it depends on the shape of the cavity excluded at the singularity. Since $W_{ijk}(\mathbf{x})$ exists, however, we can choose any convenient shape for the excluded cavity; we will choose a d-dimensional sphere of radius ε. Thus, we are led to the following theorem.

Theorem III. The symmetrized gradient of $\mathbf{W}$ with respect to the indices i and j can be written as

$$\frac{1}{2}\left[\frac{\partial}{\partial x_j}W_{ikl}(\mathbf{x})+\frac{\partial}{\partial x_i}W_{jkl}(\mathbf{x})\right]=\lim_{\varepsilon\to 0}\int_{r>\varepsilon}\mathscr{H}^{(q)}_{ijkl}h(\mathbf{x}')\,\mathrm{d}\mathbf{x}'-A^{(q)}_{ijkl}h(\mathbf{x}), \tag{A.7}$$

where the constant tensor $\mathbf{A}^{(q)}$ is give by

$$A^{(q)}_{ijkl}=\frac{1}{dK_q+2(d-1)G_q}(\Lambda_{\mathrm{h}})_{ijkl}+\frac{d}{2(d+2)G_q}\frac{K_q+2G_q}{dK_q+2(d-1)G_q}(\Lambda_{\mathrm{s}})_{ijkl}, \tag{A.8}$$

$$\begin{aligned}\mathscr{H}^{(q)}_{ijkl}&=\frac{1}{2}\left[\frac{\partial^2 g^{(q)}_{ik}}{\partial x_j\,\partial x_l}+\frac{\partial^2 g^{(q)}_{jk}}{\partial x_i\,\partial x_l}\right]\\&=\frac{1}{r^d}\Bigg\{b_q\delta_{ij}\delta_{kl}-\frac{[a_q(d-2)-b_q]}{2}[\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}]+d\frac{[a_q(d-2)-b_q]}{2}[\delta_{ik}n_jn_l+\delta_{jk}n_in_l]\\&\quad-db_q[\delta_{ij}n_kn_l+\delta_{jl}n_in_k+\delta_{kl}n_in_j+\delta_{il}n_jn_k]+d(d+2)b_qn_in_jn_kn_l\Bigg\},\end{aligned} \tag{A.9}$$

and

$$\mathbf{n}=\frac{\mathbf{x}-\mathbf{x}'}{|\mathbf{x}-\mathbf{x}'|}$$

is the unit outward normal.

Proof. Excluding a d-dimensional spherical cavity of radius ε, we can write the third-order tensor $\mathbf{W}$ in component form as

$$W_{ijk}(\mathbf{x})=\lim_{\varepsilon\to 0}\int_{r\geqslant\varepsilon}\frac{\partial g^{(q)}_{ij}(\mathbf{r})}{\partial x_k}h(\mathbf{x}')\,\mathrm{d}\mathbf{x}',$$

where $r=|\mathbf{x}-\mathbf{x}'|$. Using the identity

$$\frac{\partial}{\partial x_i}g^{(q)}_{ij}(\mathbf{r})=-\frac{\partial}{\partial x_i'}g^{(q)}_{ij}(\mathbf{r}),$$

integrating by parts, and applying the divergence theorem yields

$$W_{ijk}(\mathbf{x})=\lim_{\varepsilon\to 0}\int_{r\geqslant\varepsilon}g^{(q)}_{ij}(\mathbf{r})\frac{\partial}{\partial x_l'}h(\mathbf{x}')\,\mathrm{d}\mathbf{x}'.$$

The surface integral over the sphere of radius $|\mathbf{x}-\mathbf{x}'|=\varepsilon$ does not appear since it vanishes as $\varepsilon\to 0$. Now can take the symmetrized gradient inside the integral sign by Theorem II to obtain

$$\begin{aligned}\frac{1}{2}\left[\frac{\partial}{\partial x_j}W_{ikl}(\mathbf{x})+\frac{\partial}{\partial x_i}W_{jkl}(\mathbf{x})\right]&=\lim_{\varepsilon\to 0}\int_{r\geqslant\varepsilon}\frac{1}{2}\left[\frac{\partial}{\partial x_j}g^{(q)}_{ik}(\mathbf{r})+\frac{\partial}{\partial x_i}g^{(q)}_{jk}(\mathbf{r})\right]\frac{\partial}{\partial x_l'}h(\mathbf{x}')\,\mathrm{d}\mathbf{x}',\\&=\lim_{\varepsilon\to 0}\int_{r\geqslant\varepsilon}\mathscr{H}^{(q)}_{ijkl}h(\mathbf{x}')\,\mathrm{d}\mathbf{x}'\\&\quad-\lim_{\varepsilon\to 0}\int_{r=\varepsilon}\frac{1}{2}\left[\frac{\partial}{\partial x_j}g^{(q)}_{ik}(\mathbf{r})+\frac{\partial}{\partial x_i}g^{(q)}_{jk}(\mathbf{r})\right]n_lr^{d-1}h(\mathbf{x}')\,\mathrm{d}\Omega,\end{aligned} \tag{A.10}$$

where we have again integrated by parts.

Using the identities

$$\delta_{ij}\int n_kn_l\,\mathrm{d}\Omega=\frac{\Omega}{d}\delta_{ij}\delta_{kl}, \tag{A.11}$$

$$\int n_i n_j n_k n_l \, \mathrm{d}\Omega = \frac{\Omega}{d(d+2)}\left[\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}\right], \tag{A.12}$$

in the angular integration above, where $\mathrm{d}\Omega$ represents an element of solid angle in d dimensions, yields Theorem III.

Defining the quantity

$$\tilde{G}^{(q)}_{ijkl} = \tilde{H}^{(q)}_{ijkl} - A^{(q)}_{ijkl}\delta(\mathbf{r}) \tag{A.13}$$

where $\delta(\mathbf{x})$ is the Dirac delta function, enables one to rewrite Theorem III in the more compact form

$$\frac{1}{2}\left[\frac{\partial}{\partial x_j}W_{ikl}(\mathbf{x}) + \frac{\partial}{\partial x_i}W_{jkl}(\mathbf{x})\right] = \lim_{\varepsilon\to 0}\int_{r\geqslant\varepsilon} \tilde{G}^{(q)}_{ijkl} h(\mathbf{x}')\,\mathrm{d}\mathbf{x}'. \tag{A.14}$$

Finally, we remark that we could have chosen to exclude a non-spherical cavity from the origin in the integral (A.6). This choice would have led to a constant tensor $\mathbf{A}^{(q)}$ different from that given by (A.8). We will study such choices in a future paper.

APPENDIX B: HASHIN–SHTRIKMAN–WALPOLE BOUNDS FOR MULTIPHASE COMPOSITES IN ARBITRARY DIMENSION d

Here we state the Hashin–Shtrikman–Walpole bounds for a d-dimensional, macroscopically isotropic composite consisting of n isotropic phases. These more general results are a simple extension of the two-phase bounds discussed in the text and therefore the details of the derivation are omitted. Let K_i and G_i be the bulk and shear moduli, respectively, of the ith phase and ϕ_i be the corresponding volume fraction. Let the largest and smallest phase bulk moduli be denoted by $K_{\max}$ and $K_{\min}$, respectively, and the largest and smallest phase shear moduli be denoted by $G_{\max}$ and $G_{\min}$, respectively. Then the effective bulk modulus K_e and effective shear modulus G_e are bounded according to the relations

$$\left[\sum_{i=1}^{n}\phi_i(K^*_{\min}+K_i)^{-1}\right]^{-1} - K^*_{\min} \leqslant K_e \leqslant \left[\sum_{i=1}^{n}\phi_i(K^*_{\max}+K_i)^{-1}\right]^{-1} - K^*_{\max}, \tag{B.1}$$

$$\left[\sum_{i=1}^{n}\phi_i(G^*_{\min}+G_i)^{-1}\right]^{-1} - G^*_{\min} \leqslant G_e \leqslant \left[\sum_{i=1}^{n}\phi_i(G^*_{\max}+G_i)^{-1}\right]^{-1} - G^*_{\max}, \tag{B.2}$$

where

$$K^*_{\min} = \frac{2(d-1)}{d}G_{\min}, \tag{B.3}$$

$$K^*_{\max} = \frac{2(d-1)}{d}G_{\max}, \tag{B.4}$$

$$G^*_{\min} = \frac{G_{\min}[dK_{\min}/2+(d+1)(d-2)G_{\min}/d]}{K_{\min}+2G_{\min}}, \tag{B.5}$$

$$G^*_{\max} = \frac{G_{\max}[dK_{\max}/2+(d+1)(d-2)G_{\max}/d]}{K_{\max}+2G_{\max}}. \tag{B.6}$$

The corresponding bounds on the effective conductivity σ_e of d-dimensional, macroscopically isotropic composite consisting of n isotropic phases is also given. Let σ_i be the conductivity of phase i, and denote by $\sigma_{\max}$ and $\sigma_{\min}$ the largest and smallest phase conductivities, respectively. We have

$$\left[\sum_{i=1}^{n} \phi_i(\sigma^*_{\min}+\sigma_i)^{-1}\right]^{-1} - \sigma^*_{\min} \leqslant \sigma_e \leqslant \left[\sum_{i=1}^{n} \phi_i(\sigma^*_{\max}+q_i)^{-1}\right]^{-1} - \sigma^*_{\max}, \tag{B.7}$$

where

$$\sigma^*_{\min} = (d-1)\sigma_{\min}, \tag{B.8}$$

$$\sigma^*_{\max} = (d-1)\sigma_{\max}. \tag{B.9}$$

Note that the multiphase bounds on the effective bulk modulus (B.1) and effective conductivity (B.7) were shown by Milton (1981b) and by Lurie and Cherkaev (1985) to be realizable (under certain conditions) by certain multi-coated circles ($d = 2$) and spheres ($d = 3$). The bounds for any dimension $d > 2$ are realizable by the corresponding d-dimensional multi-coated spheres.