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THE EFFECT OF SPATIAL DISTRIBUTION ON THE EFFECTIVE BEHAVIOR OF COMPOSITE MATERIALS AND CRACKED MEDIA

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ABSTRACT

Estimates of the Hashin–Shtrikman type are developed for the overall moduli of composites consisting of a matrix containing one or more populations of inclusions, when the spatial correlations of inclusion locations take particular "ellipsoidal" forms. Inclusion shapes can be selected independently of the shapes adopted for the spatial correlations. The formulae that result are completely explicit and easy to use. They are likely to be useful, in particular, for composites that have undergone a prior macroscopically uniform large deformation. To the extent that the statistics that are assumed may not be realized exactly, the formulae provide approximations. Since, however, they are derived as variational approximations for composites with some explicit statistics that *are* realizable, they are free from some of the drawbacks of competitor approximations such as that of Mori and Tanaka (1973 *Acta Metall.* **21,** 571–574), which can generate tensors of effective moduli which fail to satisfy a necessary symmetry requirement. The new formulae are also the only ones known that take explicit account, at least approximately, of inclusion shape and spatial distribution independently.

1. INTRODUCTION

To motivate this work, consider a steel containing hard particles whose distribution was isotropic at the stage of solidification. Suppose, then, that the steel is hot-rolled, to generate a large extension in the rolling direction, coupled with a large reduction in thickness and (perhaps) zero transverse strain. What are the overall stiffnesses of the resulting material? The material is anisotropic, both through particle deformation and alignment, and through the spatial distribution of the particles. That the latter cannot reasonably be ignored can be recognized by considering the idealized case of rigid, spherical particles: then, all of the anisotropy comes from their spatial distribution and realistic estimates of effective stiffness *have* to take account of this.

It is the purpose of this article to develop a simple prescription, based on the Hashin–Shtrikman variational structure in the form developed by Willis (1977, 1978), which provides estimates for the overall response of such systems. In its original form given in 1977, it takes account of inclusion shape and distribution jointly through an integral of the two-point correlation functions of the medium. In 1978, an expression

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was given which contained separately the influence of inclusion shape and distribution. The resulting integrals, however, may be hard to evaluate—and cannot be evaluated at all unless the requisite data are provided. In the early work, the data that were considered could be realized, conceptually at least, by taking a geometrically isotropic composite (for instance, a matrix containing an isotropic distribution of spherical inclusions) and subjecting this to an affine transformation defined by a matrix Z^{-1}. The spheres then become ellipsoids and their pair distribution function, which originally depended just upon their separation, now depends on coordinates $x = (x_1, x_2, x_3)$ through the combination $\rho = |Zx|$. In this case, the integrals are easily evaluated and the resulting explicit formulae only require specification of volume fractions of each type of material. Such shape information has recently been adopted by Ponte Castañeda and Zaidman (1994) for modeling the effective nonlinear ductile behavior of a matrix containing voids, which are allowed to change shape as a consequence of a finite deformation history. The contribution of the present work is to note that the integrals that appear in the Hashin–Shtrikman variational formula are easily evaluated when, for example, a composite contains a population of ellipsoidal inclusions, with a pair distribution function that has "ellipsoidal symmetry" in the sense described above but with a *different* ellipsoid from the one that defines the inclusion shapes. This includes, for example, the case of spherical inclusions distributed with ellipsoidal symmetry, which might perhaps be regarded as an approximation to the distribution of the rigid spheres in the example given above. (Then, Z^{-1} could be identified with the global transformation induced by the rolling. This would give the correct "far-field" statistics but would be approximate in having neglected interactions between close neighbors.)

Although the assumed statistics may not be realized exactly in any particular composite, they do at least have two virtues: the resulting formulae are *completely explicit* and easy to use; and the estimates, since they are derived from statistical distributions that are (mathematically and physically) realizable, always make physical sense.

An alternative approximate scheme which is already popular for estimating effective stiffness is that of Mori and Tanaka (1973) [see also Taya and Mura (1981), Tandon and Weng (1986) and Benveniste (1987)]. Its popularity is probably based upon the simplicity of its application. It has been observed (Weng, 1992) that it reduces precisely to the Hashin–Shtrikman-based formula of Willis (1977) in the case of a matrix containing a population of aligned ellipsoidal inclusions, and this, apparently, is considered a virtue. There are, however, cases where the Mori–Tanaka prescription is simply not appropriate. In the example given above, of spherical inclusions distributed anisotropically, it would predict isotropic overall behavior (assuming isotropy of the matrix and inclusions separately). Also, for composites containing inclusions of several shapes or orientations, it can yield estimates for the tensor of overall moduli that violate the symmetry requirement $C_{ijkl} = C_{klij}$ to which the composite *must* conform, given that its constituents have this symmetry. The formulae given here [see, for instance, (3.25) with (3.26)] are *just as simple to use* as the Mori–Tanaka prescription; they reduce to the expression given by Willis (1977)—and so also to Mori–Tanaka—when the pair distribution function has the same ellipsoidal symmetry as the inclusions, and *always* display the necessary symmetry for the effective moduli. Furthermore, they provide, in many cases, strict bounds for the effective moduli of any composite that realizes the statistical assumptions exactly.

Several examples are given for illustration. These show the separate effects of inclusion shape and spatial distribution. The new estimates are compared with various other approximations, including those derived from the self-consistent scheme, the differential self-consistent scheme and the Mori–Tanaka scheme. The cases of a matrix weakened by cracks or strengthened by rigid platelets are treated, as examples without too many free parameters. The new formula provides strict bounds in such cases (for composites with the assumed statistics). Some of these bounds are violated by the estimates obtained from the differential and Mori–Tanaka schemes. This may demonstrate no more than that these estimates actually apply to other statistical distributions but *at least* they demonstrate the need to keep explicit track of the statistics. It is not, in fact, known to what (if any) statistics these approximations correspond, for the flat inclusions considered here.

Much progress has recently been made in estimating the nonlinear effective response of composites using, as a building block, results obtained from the linear theory. The new results presented here also have this potential application. In particular, the treatment of Ponte Castañeda and Zaidman (1994) for a porous ductile material with evolving microstructures could now be extended to estimate the yield behavior of a matrix containing several populations of inclusions, whose shapes and spatial distributions evolve independently. Such applications will be reported separately.

2. EFFECTIVE PROPERTIES AND HASHIN–SHTRIKMAN ESTIMATES

In this work, a *composite* is a heterogeneous material with two distinct length scales: a macroscopic one which characterizes the overall dimensions of the specimen and the scale of variation of the applied loading conditions, and a microscopic one which characterizes the size of the typical heterogeneity (e.g., inclusions, fibers, cracks, etc.). By *effective* properties, it is meant the relation between the averages of the local stress and strain fields within the composite. Rigorous definitions of effective properties in terms of the energy of the composite are given below, together with estimates of the Hashin–Shtrikman type for these effective properties.

2.1. *Effective properties*

The local constitutive properties of the linear elastic composite, assumed to occupy a region in space Ω, are described by its modulus tensor

$$\mathbf{L}(x) = \sum_{r=1}^{N} \chi^{(r)}(x)\mathbf{L}^{(r)}, \tag{2.1}$$

and associated strain-energy density function

$$W(x,\varepsilon) = \tfrac{1}{2}\varepsilon \cdot \mathbf{L}(x)\varepsilon, \tag{2.2}$$

where ε is the infinitesimal strain tensor, $\mathbf{L}^{(r)}$ denotes the modulus tensor of phase r

and $\chi^{(r)}$ denotes the characteristic function of phase r (equal to 1 if x is in phase r and 0 otherwise). Note that the volume averages (over Ω) of the functions $\chi^{(r)}$ are the volume fractions of the phases, denoted $c^{(r)}$. The stress–strain relation is then given by

$$\sigma = \frac{\partial W}{\partial \varepsilon}(x, \varepsilon) = \mathbf{L}(x)\varepsilon. \tag{2.3}$$

Following Hill (1963), the effective behavior of the composite is determined by its effective energy function

$$\tilde{W}(\bar{\varepsilon}) = \min_{\varepsilon \in K} \frac{1}{|\Omega|} \int_{\Omega} W(x, \varepsilon)\, \mathrm{d}x, \tag{2.4}$$

where K is a set of kinematically admissible strains consistent with an average strain $\bar{\varepsilon}$ in the composite, given by

$$K = \{\varepsilon \,|\, \text{there exists } u, \text{ such that } \varepsilon = \tfrac{1}{2}[\nabla u + (\nabla u)^T] \text{ in } \Omega, \text{ and } u = \bar{\varepsilon}x \text{ on } \partial\Omega\}. \tag{2.5}$$

Because of linearity, $\tilde{W}(\bar{\varepsilon}) = \frac{1}{2}\bar{\varepsilon}\cdot\tilde{\mathbf{L}}\bar{\varepsilon}$, where $\tilde{\mathbf{L}}$ is the effective modulus tensor of the composite. Hill (1963) showed that

$$\bar{\sigma} = \frac{\partial \tilde{W}}{\partial \bar{\varepsilon}} = \tilde{\mathbf{L}}\bar{\varepsilon}. \tag{2.6}$$

2.2. *Hashin–Shtrikman estimates*

Unless the microstructure is very simple, as for a periodic composite, for example, it is extremely difficult in general to compute the effective potential $\tilde{W}$ exactly. An alternative approach is to consider classes of random microstructures with prescribed statistics and to attempt to obtain estimates for $\tilde{W}$ or $\tilde{\mathbf{L}}$ depending on these statistics. The simplest example is provided by the so-called Voigt and Reuss estimates, which express the effective modulus tensor $\tilde{\mathbf{L}}$, respectively, in terms of the arithmetic and harmonic averages of the phase moduli $\mathbf{L}^{(r)}$, weighted by the phase volume fractions $c^{(r)}$. Hill (1952) showed that the Voigt and Reuss estimates are in fact rigorous upper and lower bounds, respectively, for $\tilde{\mathbf{L}}$ within the class of composite materials with prescribed volume fractions $c^{(r)}$.

Hashin and Shtrikman (1962, 1963) proposed estimates that were based on an alternative variational representation for $\tilde{W}$ which allowed the incorporation of the hypothesis of statistical isotropy. The alternative variational representation made use of polarization fields relative to a homogeneous reference material with modulus tensor $\mathbf{L}^{(0)}$. These authors demonstrated that their estimates yield in fact upper and lower bounds for $\tilde{\mathbf{L}}$, which are sharper than the Voigt–Reuss bounds, if $\mathbf{L}^{(0)}$ is chosen to be equal to the "maximum" and "minimum" of the $\mathbf{L}^{(r)}$, respectively. Extensions of the Hashin–Shtrikman (HS) variational principles in various contexts were provided by several authors over the years (e.g., Walpole, 1966; Willis, 1977; Milton and Kohn, 1988). For completeness, a brief derivation of the HS variational principles, which incidentally also works for nonlinear composites, is given below following Talbot and Willis (1985) and Ponte Castañeda and Willis (1988).

Defining $W^{(0)}$ to be the strain-energy function associated with a reference material with uniform modulus tensor $\mathbf{L}^{(0)}$, and assuming that $\mathbf{L}^{(0)} \leqslant \min_r \{\mathbf{L}^{(r)}\}$, in the sense that $\varepsilon \cdot (\mathbf{L}^{(0)} - \mathbf{L}^{(r)})\varepsilon \leqslant 0$ for all $\varepsilon \neq 0$, $r = 1, \ldots, N$, the definition of the Legendre transform gives

$$(W - W^{(0)})^*(x, \tau) = \max_{\varepsilon} \{\tau \cdot \varepsilon - [W(x, \varepsilon) - W^{(0)}(\varepsilon)]\}, \tag{2.7}$$

from which it follows that

$$W(x, \varepsilon) \geqslant W^{(0)}(\varepsilon) + \tau \cdot \varepsilon - (W - W^{(0)})^*(x, \tau), \tag{2.8}$$

for any choice of τ. Therefore, by the definition of $\tilde{W}$, one has that

$$\tilde{W}(\bar{\varepsilon}) \geqslant \min_{\varepsilon \in K} \int_{\Omega} [W^{(0)}(\varepsilon) + \tau \cdot \varepsilon] \, \mathrm{d}x - \int_{\Omega} (W - W^{(0)})^*(x, \tau) \, \mathrm{d}x, \tag{2.9}$$

where the volume of Ω has been taken to be unity for simplicity of expression.

For any chosen "polarization field" τ, the strain field ε that attains the minimum in (2.9) has the representation

$$\varepsilon = \bar{\varepsilon} - \Gamma^{(0)}\tau \tag{2.10}$$

where $\Gamma^{(0)}$ is a linear integral operator

$$\Gamma^{(0)}\tau = \int_{\Omega} \Gamma^{(0)}(x, x')[\tau(x') - \bar{\tau}] \, \mathrm{d}x', \tag{2.11}$$

whose kernel is related to the Green's function $G^{(0)}$ for the domain Ω, with modulus tensor $\mathbf{L}^{(0)}$, via

$$\Gamma^{(0)}_{ijkl} = \left. \frac{\partial^2 G^{(0)}_{ik}}{\partial x_j \, \partial x'_l} \right|_{(ij),(kl)}. \tag{2.12}$$

In (2.11), $\bar{\tau}$ represents the mean value of τ over Ω.

It is useful to select the polarization field to be piecewise constant as follows

$$\tau(x) = \sum_{r=1}^{N} \chi^{(r)}(x)\tau^{(r)}. \tag{2.13}$$

Then, $\bar{\tau} = \sum_{r=1}^{N} c^{(r)}\tau^{(r)}$ and the finite-body kernel in (2.11) can be replaced by the corresponding kernel constructed from the infinite body Green's function under the *no long-range order* hypothesis for the distribution of the phases (Willis, 1981). The kernel now becomes a function of $(x - x')$ only: this replacement is adopted henceforth.

Thus, making use of (2.11), (2.13), and of the fact that the average of $\Gamma^{(0)}\tau$ is zero [cf. (2.10)], it can be shown that (2.9) gives

$$\tilde{W}(\bar{\varepsilon}) \geqslant \frac{1}{2}\bar{\varepsilon}\cdot\mathbf{L}^{(0)}\bar{\varepsilon}+\bar{\tau}\cdot\bar{\varepsilon}-\frac{1}{2}\sum_{r=1}^{N} c^{(r)}\tau^{(r)}\cdot[(\mathbf{L}^{(r)}-\mathbf{L}^{(0)})^{-1}\tau^{(r)}]$$
$$-\frac{1}{2}\sum_{r=1}^{N}\sum_{s=1}^{N}\Delta\tau^{(r)}\cdot[\mathbf{A}^{(rs)}\Delta\tau^{(s)}], \quad (2.14)$$

where $\Delta\tau^{(r)} = \tau^{(r)}-\tau^{(1)}$, and where the parameters

$$\mathbf{A}^{(rs)} = \int_{\Omega}\mathrm{d}x\int_{\Omega}\mathrm{d}x'\chi^{(r)}(x)[\chi^{(s)}(x')-c^{(s)}]\Gamma^{(0)}(x-x') \quad (2.15)$$

$(r, s = 2,\ldots,N)$ depend only on the microstructure and on $\mathbf{L}^{(0)}$, and can be shown to be symmetric in the superscripts r, s (see Section 3). The above estimate may be optimized over the polarizations $\tau^{(r)}$ $(r = 1,\ldots,N)$ to give

$$\tilde{W}(\bar{\varepsilon}) \geqslant \tfrac{1}{2}\bar{\varepsilon}\cdot\mathbf{L}^{(0)}\bar{\varepsilon}+\tfrac{1}{2}\bar{\varepsilon}\cdot\bar{\tau}, \quad (2.16)$$

where the optimal polarizations satisfy the relations

$$(\mathbf{L}^{(1)}-\mathbf{L}^{(0)})^{-1}\tau^{(1)}-\frac{1}{c^{(1)}}\sum_{r=2}^{N}\sum_{s=2}^{N}\mathbf{A}^{(rs)}(\tau^{(s)}-\tau^{(1)}) = \bar{\varepsilon}$$
$$(\mathbf{L}^{(r)}-\mathbf{L}^{(0)})^{-1}\tau^{(r)}+\frac{1}{c^{(r)}}\sum_{s=2}^{N}\mathbf{A}^{(rs)}(\tau^{(s)}-\tau^{(1)}) = \bar{\varepsilon}, \quad (r = 2,\ldots,N). \quad (2.17)$$

The lower bound given by (2.16) assumes that $\mathbf{L}^{(0)} \leqslant \min_r\{\mathbf{L}^{(r)}\}$. If, instead, it is assumed that $\mathbf{L}^{(0)} \geqslant \max_r\{\mathbf{L}^{(r)}\}$, change of the max to a min in (2.7) leads to an upper bound for W in (2.8). Then, (2.16), with (2.17), still applies, except that the sign of the inequality must be changed to give an upper bound for $\tilde{W}$. However, if neither hypothesis on $\mathbf{L}^{(0)}$ is satisfied, the right-hand side of (2.16) still provides a (stationary) variational estimate for $\tilde{W}$, which is neither an upper nor a lower bound on $\tilde{W}$. Note that the aforementioned symmetry of the geometric parameters $\mathbf{A}^{(rs)}$ ensures the corresponding symmetry of $\tilde{\mathbf{L}}$.

The estimate for $\tilde{W}$ provided by (2.16), together with (2.17), is explicit, except for the geometric parameters $\mathbf{A}^{(rs)}$, which need to be determined for specific classes of microstructures. Such an estimate is given in the following section for the class of particulate microstructures with prescribed two-point correlation functions for the statistics of the distribution of centers of arbitrarily shaped inclusions.

3. PARTICULATE MICROSTRUCTURES

This section deals with the special case of N-phase particulate microstructures consisting of a random distribution of inclusions of $N-1$ different types in a matrix of a homogenous phase. The matrix will be denoted by superscript 1, so that its modulus tensor and concentration are $\mathbf{L}^{(1)}$ and $c^{(1)}$, respectively. The inclusions may differ in size, shape and orientation. It will be assumed that there are $n^{(r)}$ inclusions of type $r(r = 2,\ldots,N-1)$, with modulus tensor $\mathbf{L}^{(r)}$, occupying identical regions of space

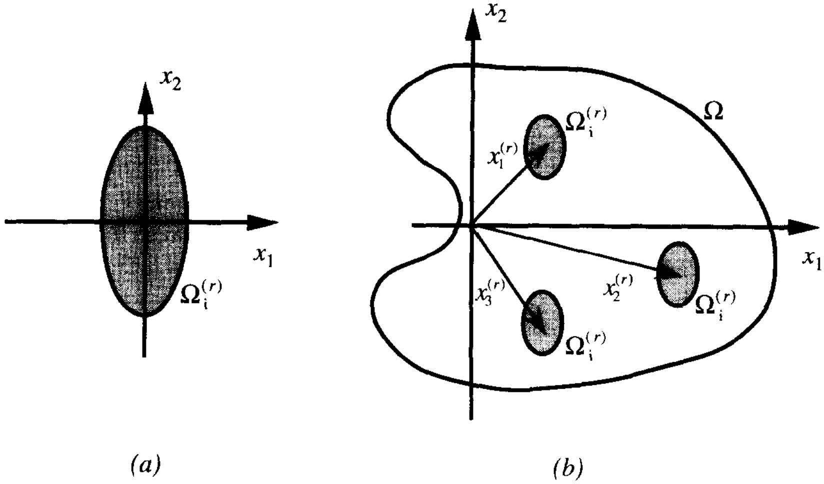

Fig. 1. Particulate microstructures. (a) Reference inclusion of type r occupying reference domain $\Omega_i^{(r)}$. (b) The inclusions of type r are distributed randomly in Ω with centers at points $x_\alpha^{(r)}$ ($\alpha = 1,\dots,n^{(r)}$).

in Ω with reference domain $\Omega_i^{(r)}$, centered at random points $x_\alpha^{(r)}$ ($\alpha = 1,\dots,n^{(r)}$) (see Fig. 1). Denoting by $\chi_i^{(r)}(x)$ the characteristic function of the reference inclusion $\Omega_i^{(r)}$ [i.e. $\chi_i^{(r)} = 1$ if $x \in \Omega_i^{(r)}$, and 0 otherwise], it follows that the characteristic function of phase r (the region of Ω occupied by all inclusions of type r) is given by

$$\chi^{(r)}(x) = \sum_{\alpha=1}^{n^{(r)}} \chi_i^{(r)}(x - x_\alpha^{(r)}), \tag{3.1}$$

which for later reference is rewritten in the form

$$\chi^{(r)}(x) = \int_\Omega \mathrm{d}z[\chi_i^{(r)}(x-z)\varphi^{(r)}(z)], \tag{3.2}$$

where

$$\varphi^{(r)}(z) = \sum_{\alpha=1}^{n^{(r)}} \delta(z - x_\alpha^{(r)}) \tag{3.3}$$

is the random density field generated by the set of random points $x_\alpha^{(r)}$ ($\alpha = 1,\dots,n^{(r)}$) (Stratonovich, 1963). Note that the volume fraction of phase $r(r = 2,\dots,N-1)$ is given by

$$c^{(r)} = n^{(r)} \frac{|\Omega_i^{(r)}|}{|\Omega|}. \tag{3.4}$$

The random density field $\varphi^{(r)}$ is a useful quantity because it may be related to the probability density fields through ensemble averaging, denoted by the brackets $\langle \cdot \rangle$. Thus,

$$\langle \varphi^{(r)}(z)\rangle = p^{(r)}(z),$$
$$\langle \varphi^{(r)}(z)\varphi^{(s)}(z')\rangle = \delta_{rs}p^{(r)}(z)\delta(z-z')+p^{(rs)}(z,z'), \tag{3.5}$$

where $p^{(r)}(z)$ denotes the probability density for finding an inclusion of type r centered at z, and $p^{(rs)}(z,z')$ correspondingly denotes the joint probability density for finding an inclusion of type r centered at z an inclusion of type s centered at z'. In this work, it will be assumed that the composite is statistically homogeneous, so that $p^{(r)}(z) = p^{(r)}$, a constant equal to the number of inclusions per unit volume (i.e. $p^{(r)} = n^{(r)}/|\Omega|$), and $p^{(rs)}(z,z') = p^{(rs)}(z-z')$.

3.1. *Microstructural parameters* $\mathbf{A}^{(rs)}$

With the above preliminaries, it makes sense to compute the ensemble average of the microstructural parameters $\mathbf{A}^{(rs)}$ $(r,\ s = 2,\dots,N)$. Thus, making use of (3.2), definition (2.15) implies

$$\langle \mathbf{A}^{(rs)}\rangle = \int_\Omega \mathrm{d}x\int_\Omega \mathrm{d}x'\int_\Omega \mathrm{d}z\int_\Omega \mathrm{d}z'\boldsymbol{\Gamma}^{(0)}(x-x')\chi_{\rm i}^{(r)}(x-z)\chi_{\rm i}^{(s)}(x'-z')\langle\varphi^{(r)}(z)\varphi^{(s)}(z')\rangle$$
$$-\int_\Omega \mathrm{d}x\int_\Omega \mathrm{d}x'\int_\Omega \mathrm{d}z\boldsymbol{\Gamma}^{(0)}(x-x')\chi_{\rm i}^{(r)}(x-z)\langle\varphi^{(r)}(z)\rangle c^{(s)}. \tag{3.6}$$

Next, making use of relations (3.5), together with

$$c^{(s)} = |\Omega_{\rm i}^{(s)}|p^{(s)} = \int_\Omega \mathrm{d}z'\chi_{\rm i}^{(s)}(x'-z')p^{(s)}, \tag{3.7}$$

it follows that

$$\langle \mathbf{A}^{(rs)}\rangle = \int_\Omega \mathrm{d}x\int_\Omega \mathrm{d}x'\int_\Omega \mathrm{d}z\int_\Omega \mathrm{d}z'\boldsymbol{\Gamma}^{(0)}(x-x')\chi_{\rm i}^{(r)}(x-z)\chi_{\rm i}^{(s)}(x'-z')[p^{(rs)}(z-z')-p^{(r)}p^{(s)}]$$
$$+\delta_{rs}p^{(s)}\int_\Omega \mathrm{d}x\int_\Omega \mathrm{d}x'\int_\Omega \mathrm{d}z\boldsymbol{\Gamma}^{(0)}(x-x')\chi_{\rm i}^{(r)}(x-z)\chi_{\rm i}^{(s)}(x'-z), \tag{3.8}$$

which may be simplified further through the changes of variables $y = x-z$, $y' = x'-z'$, $z'' = z'-z$ and $y = x-z$, $y' = x'-z$, respectively, to give

$$\langle \mathbf{A}^{(rs)}\rangle = \int \mathrm{d}y\int \mathrm{d}y'\int \mathrm{d}z''\boldsymbol{\Gamma}^{(0)}(y-y'-z'')\chi_{\rm i}^{(r)}(y)\chi_{\rm i}^{(s)}(y')[p^{(rs)}(z'')-p^{(r)}p^{(s)}]$$
$$+\delta_{rs}p^{(s)}\int \mathrm{d}y\int \mathrm{d}y'\boldsymbol{\Gamma}^{(0)}(y-y')\chi_{\rm i}^{(r)}(y)\chi_{\rm i}^{(s)}(y'), \tag{3.9}$$

where the fact that $|\Omega| = 1$ has been used for performing the integration over x, and the remaining integrations have been performed formally over all of space, because their kernels ensure restrictions to finite domains for y and y', and decay on the order of a small "correlation length" for z''. In turn, this may be written as

$$\langle \mathbf{A}^{(rs)} \rangle = p^{(r)} \int_{\Omega_i^{(r)}} \mathrm{d}y \int_{\Omega_i^{(s)}} \mathrm{d}y' \int \mathrm{d}z'' \Gamma^{(0)}(y-y'-z'')[p^{(s|r)}(z'')-p^{(s)}]$$

$$+\delta_{rs} p^{(s)} \int_{\Omega_i^{(r)}} \mathrm{d}y \int_{\Omega_i^{(s)}} \mathrm{d}y' \Gamma^{(0)}(y-y'), \quad (3.10)$$

where $p^{(s|r)}(z', z)$ is the conditional probability density for finding an inclusion of type s centered at z' given that there is an inclusion of type r centered at point z; thus $p^{(s|r)}(z', z) = p^{(rs)}(z, z')/p^{(r)}(z)$.

It is important at this point to note that expression (3.9) for $\langle \mathbf{A}^{(rs)} \rangle$ makes plain the symmetry in the superscripts r and s. This follows from the fact that $p^{(rs)}(z, z') = p^{(sr)}(z', z)$, which implies that $p^{(rs)}(z'') = p^{(sr)}(-z'')$ for a statistically homogeneous composite, together with the fact that $\Gamma^{(0)}(-x) = \Gamma^{(0)}(x)$.

To make further progress, use is made of the hypothesis of ellipsoidal symmetry for the distribution of the inclusions, introduced by Willis (1977, 1978). This is a generalization of statistical isotropy and postulates that the conditional probability density function $p^{(s|r)}$ depends on z'' only through the combination $|Z_d^{(rs)} z''|$, for some matrix $Z_d^{(rs)}$, which defines an ellipsoid

$$\Omega_d^{(rs)} = \{x : |Z_d^{(rs)} x|^2 < 1\}, \quad (3.11)$$

serving to characterize the distribution of the inclusions. Statistical isotropy can then be seen to correspond to the special case where the matrix $Z_d^{(rs)}$ is replaced by the identity matrix, so that $p^{(s|r)}$ depends on z'' only through $|z''|$. Note that consistency with the symmetry requirement that $p^{(rs)}(z'') = p^{(sr)}(-z'')$ further requires that the matrices $Z_d^{(rs)}$ be symmetric in the superscripts r and s, so that $\Omega_d^{(rs)} = \Omega_d^{(sr)}$. Finally, it is reasonable to assume that the inclusions do not overlap. Under the assumed symmetry for the two-point probability density function, this implies that $p^{(s|r)}(z'') = 0$ for $z'' \in \Omega_d^{(rs)}$, where $\Omega_d^{(rs)}$ must be chosen just large enough to exclude particle overlap.

Under the above hypotheses for the statistical distribution of inclusions, it is useful to break up the integral with respect to z'' in the first term in (3.10) into two parts, the first part being an integral over $\Omega_d^{(rs)}$ and the second an integral over its complement $\Omega_d^{(rs)}$. It is shown in Appendix A that, with the above assumptions for the statistical distribution of the phases, and sufficiently fast decay in the no long-range order hypothesis, the integral over $\Omega_d^{(rs)}$ is in fact identically zero. Therefore, the expression (3.10) for $\langle \mathbf{A}^{(rs)} \rangle$ reduces to

$$\langle \mathbf{A}^{(rs)} \rangle = -p^{(r)} p^{(s)} \int_{\Omega_i^{(r)}} \mathrm{d}y \int_{\Omega_i^{(s)}} \mathrm{d}y' \int_{\Omega_d^{(rs)}} \mathrm{d}z'' \Gamma^{(0)}(y-y'-z'')$$

$$+\delta_{rs} p^{(s)} \int_{\Omega_i^{(r)}} \mathrm{d}y \int_{\Omega_i^{(s)}} \mathrm{d}y' \Gamma^{(0)}(y-y'). \quad (3.12)$$

Next it is observed that, since $y \in \Omega_i^{(r)}$ and $y' \in \Omega_i^{(s)}$ ensure that $y-y' \in \Omega_d^{(rs)}$, it follows from the work of Eshelby (1957) that

$$\int_{\Omega_d^{(rs)}} \mathrm{d}z'' \Gamma^{(0)}(y-y'-z'') = \mathbf{P}_d^{(rs)}, \quad (3.13)$$

which can be seen to be a constant tensor corresponding to the uniform strain produced inside the ellipsoidal inclusion $\Omega_{\rm d}^{(rs)}$, when it is embedded in an infinite matrix with modulus tensor $\mathbf{L}^{(0)}$ and subjected to uniform boundary conditions. This so-called **P** tensor may be computed from the equivalent expression (Willis, 1977)

$$\mathbf{P}_{\rm d}^{(rs)} = \frac{1}{4\pi|Z_{\rm d}^{(rs)}|}\int_{|\xi|=1} \mathbf{H}^{(0)}(\xi)|(Z_{\rm d}^{(rs)})^{-1}\xi|^{-3}\,{\rm d}S, \tag{3.14}$$

where $\mathbf{H}^{(0)}$ is a tensor with components $\mathbf{H}_{ijkl}^{(0)}(\xi) = B_{ki}^{(0)}(\xi)\xi_j\xi_l|_{(ij),(kl)}$, with $B^{(0)}$ the inverse of the acoustic tensor $A^{(0)}$, with components $A_{ik}^{(0)}(\xi) = \mathbf{L}_{ijkl}^{(0)}\xi_j\xi_l$.

Similarly, if the inclusions are assumed to be ellipsoidal, so that

$$\Omega_{\rm i}^{(r)} = \{x : |Z_{\rm i}^{(r)}x|^2 < 1\}(r = 2,\ldots,N), \tag{3.15}$$

it follows that

$$\int_{\Omega_{\rm i}^{(s)}} {\rm d}y'\Gamma^{(0)}(y-y') = \mathbf{P}_{\rm i}^{(s)}, \tag{3.16}$$

where $\mathbf{P}_{\rm i}^{(s)}$ is the **P** tensor associated with the ellipsoidal inclusion $\Omega_{\rm i}^{(s)}$; it has an expression analogous to (3.14) with $Z_{\rm d}^{(rs)}$ replaced by $Z_{\rm i}^{(s)}$.

In conclusion, for sets of ellipsoidal inclusions $\Omega_{\rm i}^{(r)} (r = 2,\ldots,N)$ distributed with the symmetry of the ellipsoids $\Omega_{\rm d}^{(rs)} (r, s = 2,\ldots,N)$, the expression for $\langle \mathbf{A}^{(rs)} \rangle$ simplifies to

$$\langle \mathbf{A}^{(rs)} \rangle = c^{(r)}(\delta_{rs}\mathbf{P}_{\rm i}^{(r)} - c^{(s)}\mathbf{P}_{\rm d}^{(rs)}), \tag{3.17}$$

where use has been made of the fact that $c^{(r)} = p^{(r)}|\Omega_{\rm i}^{(r)}|$. Note that $\langle \mathbf{A}^{(rs)} \rangle$ is plainly symmetric.

Finally, it is noted that if the inclusions are not ellipsoidal, expression (3.17) still holds, but the tensor $\mathbf{P}_{\rm i}^{(r)}$ must now be replaced by

$$\mathbf{P}_{\rm i}^{(r)} = \frac{1}{|\Omega_{\rm i}^{(r)}|}\int_{\Omega_{\rm i}^{(r)}} {\rm d}y \int_{\Omega_{\rm i}^{(r)}} {\rm d}y'\Gamma^{(0)}(y-y'), \tag{3.18}$$

which clearly specializes to (3.16) if $\Omega_{\rm i}^{(r)}$ is ellipsoidal in shape [with an expression analogous to (3.14)], but which requires direct computation more generally.

3.2. *Hashin–Shtrikman estimates*

For the class of particulate microstructures considered in this work, with a clearly defined matrix with modulus tensor $\mathbf{L}^{(1)}$, a good choice for the reference material in the Hashin–Shtrikman estimates of the previous section is the matrix material itself, phase 1. This means that $\mathbf{L}^{(0)}$ should be taken equal to $\mathbf{L}^{(1)}$ in relations (2.17), so that the polarization in the matrix $\tau^{(1)}$ vanishes identically. With this simplification, taking the ensemble averages of relations (2.17), and making use of relations (3.17) leads to

$$[(\mathbf{L}^{(r)} - \mathbf{L}^{(1)})^{-1} + \mathbf{P}_{\rm i}^{(r)}]\tau^{(r)} - \sum_{s=2}^{N} c^{(s)}\mathbf{P}_{\rm d}^{(rs)}\tau^{(s)} = \bar{\varepsilon}, \quad (r = 2,\ldots,N), \tag{3.19}$$

which may be solved for $\tau^{(r)}$ $(r = 2, \ldots, N)$ to obtain an estimate for the effective modulus tensor of the composite $\tilde{\mathbf{L}}$, from the ensemble average of (2.16). Such an estimate will be a lower bound for $\tilde{\mathbf{L}}$ if the matrix happens to be the most compliant phase in the composite and an upper bound if the matrix is the stiffest phase. On the other hand, if matrix phase is neither the most compliant nor the stiffest, the estimate will neither be an upper bound nor a lower bound, but it will still be a perfectly good estimate for $\tilde{\mathbf{L}}$. Comparison with experiments and with the results of computations for particulate microstructures [see, for example, Torquato (1991)] shows that the estimate based on use of the matrix as comparison material provides a reasonably good approximation to the tensor of overall moduli, at least for small to moderate proportions of inclusions. Further support is provided by the calculation of Willis (1984) who showed that an "improved quasicrystalline approximation", in which the matrix polarization (relative to any comparison material) is expressed exactly in terms of the inclusion polarizations, leads to this estimate, precisely. In addition, optimal microstructures attaining the classical Hashin–Shtrikman bounds are known [see, for example, Milton and Kohn (1988)] to be particulate in character, with the reference material occupying the matrix phase.

Relatively simple expressions are generated for the HS estimate for $\tilde{\mathbf{L}}$ when the distribution of the inclusions is taken to be the same for all inclusion pairs, so that $\mathbf{P}_{\rm d}^{(rs)} = \mathbf{P}_{\rm d}$ for $r, s = 2, \ldots, N$. It is then found that

$$\tilde{\mathbf{L}}^{(\mathrm{HS})} = \mathbf{L}^{(1)} + \left[\mathbf{I} - \sum_{r=2}^{N} c^{(r)}\mathbf{T}^{(r)}\mathbf{P}_{\rm d}\right]^{-1}\left[\sum_{r=2}^{N} c^{(r)}\mathbf{T}^{(r)}\right], \tag{3.20}$$

where

$$\mathbf{T}^{(r)} = [(\mathbf{L}^{(r)} - \mathbf{L}^{(1)})^{-1} + \mathbf{P}_{\rm i}^{(r)}]^{-1}, \tag{3.21}$$

which, for later reference, can be shown to admit the alternative representation

$$\tilde{\mathbf{L}}^{(\mathrm{HS})} = \left\{c^{(1)}\mathbf{I} + \sum_{r=2}^{N} c^{(r)}\mathbf{A}^{(r)}[\mathbf{I} + (\mathbf{L}^{(r)} - \mathbf{L}^{(1)})(\mathbf{P}_{\rm i}^{(r)} - \mathbf{P}_{\rm d})]\right\}^{-1}$$
$$\times \left\{c^{(1)}\mathbf{L}^{(1)} + \sum_{r=2}^{N} c^{(r)}\mathbf{A}^{(r)}[\mathbf{L}^{(r)} + (\mathbf{L}^{(r)} - \mathbf{L}^{(1)})(\mathbf{P}_{\rm i}^{(r)} - \mathbf{P}_{\rm d})\mathbf{L}^{(1)}]\right\}, \tag{3.22}$$

where

$$\mathbf{A}^{(r)} = [\mathbf{I} + (\mathbf{L}^{(r)} - \mathbf{L}^{(1)})\mathbf{P}_{\rm i}^{(r)}]^{-1}. \tag{3.23}$$

When all the inclusions have the same shape, so that $\mathbf{P}_{\rm i}^{(r)} = \mathbf{P}_{\rm i}$, and the distribution of inclusions has the same symmetry as the inclusions themselves, so that $\mathbf{P}_{\rm d} = \mathbf{P}_{\rm i} = \mathbf{P}$, the above expressions reduce to those given by Willis (1977, 1978) for composites with aligned ellipsoidal inclusions and ellipsoidal symmetry (with the same aspect ratios). More generally, the above expressions account for differences between the symmetry of the distribution of the inclusions and the shape of the inclusions. This feature will be discussed in more detail in the next section in the context of some specific examples. It is useful to note, however, that the effect of the symmetry of the distribution of the inclusions on the effective modulus tensor is of higher order in the volume fraction of the inclusions than the effect of the shape of the inclusions. To see this, note that relation (3.20) implies that

$$\tilde{\mathbf{L}}^{(\mathrm{HS})} = \mathbf{L}^{(1)} + \sum_{r=2}^{N} c^{(r)}\mathbf{T}^{(r)} + \left(\sum_{r=2}^{N} c^{(r)}\mathbf{T}^{(r)}\right)\mathbf{P}_{\mathrm{d}}\left(\sum_{r=2}^{N} c^{(r)}\mathbf{T}^{(r)}\right) + O[(c^{(r)})^3], \qquad (3.24)$$

which agrees with Eshelby's dilute estimates to first order in the volume fractions. Thus, the shape of the inclusions is seen to affect $\tilde{\mathbf{L}}$, through the tensor $\mathbf{P}_{\mathrm{i}}$, to first order in the volume fraction of the inclusions, whereas the corresponding symmetry of the distribution is seen to affect $\tilde{\mathbf{L}}$, through $\mathbf{P}_{\mathrm{d}}$, to second order in the volume fraction of inclusions.

In the above expressions for the HS estimate, the inclusions may be aligned, or they may have different orientations. If there is only one type of aligned inclusions, with identical moduli tensor $\mathbf{L}^{(2)}$ and shape, so that there is only one tensor $\mathbf{P}_{\mathrm{i}}^{(2)} = \mathbf{P}_{\mathrm{i}}$, the above estimate simplifies to the familiar form

$$\tilde{\mathbf{L}}^{(\mathrm{HS})} = \mathbf{L}^{(1)} + c^{(2)}[(\mathbf{L}^{(2)} - \mathbf{L}^{(1)})^{-1} + c^{(1)}\mathbf{P}]^{-1}, \qquad (3.25)$$

where $c^{(2)}$ is the total volume fraction of inclusions (with possibly different sizes) and the relevant $\mathbf{P}$ tensor is now given by

$$\mathbf{P} = \frac{1}{c^{(1)}}(\mathbf{P}_{\mathrm{i}} - c^{(2)}\mathbf{P}_{\mathrm{d}}), \qquad (3.26)$$

which, of course, reduces to the standard $\mathbf{P}$ when $\mathbf{P}_{\mathrm{d}} = \mathbf{P}_{\mathrm{i}} = \mathbf{P}$. If there is more than one type of aligned inclusions, of either different shape, or different modulus, more complicated expressions are generated; however, form (3.20) ensures the symmetry of the HS estimate for $\tilde{\mathbf{L}}$.

Finally, if the inclusions are identical in shape and randomly oriented, with identical pairwise distributions, the general estimate (3.20) may be rewritten

$$\tilde{\mathbf{L}}^{(\mathrm{HS})} = \mathbf{L}^{(1)} + (1 - c^{(1)})[\mathbf{I} - (1 - c^{(1)})\{\mathbf{T}\}\mathbf{P}_{\mathrm{d}}]^{-1}\{\mathbf{T}\}, \qquad (3.27)$$

where the brackets $\{\cdot\}$ denote an average over all orientations.

3.3. *Comparison to other estimates*

It is appropriate at this point to establish the connection between the above estimates of the Hashin–Shtrikman type and the Mori–Tanaka (MT) estimates. These were originally proposed by Mori and Tanaka (1973) and applied and developed further by a number of authors (Taya and Mura, 1981; Tandon and Weng, 1986; Benveniste, 1987). It is well known that the Mori–Tanaka method gives estimates, for composites with spherical inclusions, that are in agreement with the Hashin–Shtrikman bounds for statistically isotropic composites with isotropic phases. The MT estimate agrees with the HS upper bound if the stiffest material is placed in the matrix phase and with the lower bound if the most compliant material is placed in the matrix. The same is true for the MT estimates for two-phase composites with aligned inclusions, when compared with the HS estimates of Willis (1977) for two-phase composites with ellipsoidal symmetry. The reason, of course, is that in these two cases, the MT estimates coincide with the HS estimates of Willis (1978) for particulate composites with aligned ellipsoidal inclusions which are distributed with the same symmetry as the shape of the inclusion. [In the context of the more general estimate (3.25), the estimates of Willis (1978) correspond to the choice $\mathbf{P}_d = \mathbf{P}_i = \mathbf{P}$.]

Weng (1990) proposed an extension to the MT estimates for multi-phase composites with aligned inclusions of different shape. These may be obtained, formally, from the form (3.22) for the general HS estimates with identical pairwise distributions of the phases, by setting the terms proportional to $\mathbf{P}_i^{(r)} - \mathbf{P}_d$ equal to zero. Conceptually, this difference corresponds to the assumption implicit in the MT method of taking the distribution of the inclusions around a given inclusion to have the same symmetry as the shape of the given inclusion. This assumption violates the physical requirement that $p^{(rs)}(z, z') = p^{(sr)}(z', z)$ for any two inclusions of different shapes, and leads to the well-known asymmetry of the MT estimates for the effective modulus tensor in this case. By contrast, the above HS estimates are free of this limitation, and exhibit the physically correct symmetries for the effective modulus tensor.

Similarly, Weng (1984) and Benveniste (1987) have proposed extensions of the MT estimates for composites with randomly oriented inclusions and cracks. For the same reason discussed above in connection with composites with aligned inclusions of different shapes, regarding physically incorrect assumptions about the distributions of pairs of different types of inclusions, the MT estimates can be shown to be incorrect for non-dilute concentrations of non-spherical inclusions. Only for the case where the inclusions are spherical and anisotropic, the MT estimates agree with the HS estimates of Walpole (1969), since in this case (and only this case) the assumption of isotropic distributions for all the (spherical) inclusions is physically consistent. In the next section, some explicit comparisons, for the special cases of randomly oriented cracks and disks, are given which show that the MT estimates violate rigorous bounds of the HS type for composites with isotropic distributions of randomly oriented cracks and disks.

Some sample comparisons are also made with the self-consistent and differential self-consistent estimates in the next sections for some specific cases. It is noted in passing that generalized estimates of the self-consistent type are possible, following Willis (1977), by choosing the comparison material to be equal to the effective material [i.e. by choosing the average polarization in (2.16) identically equal to zero]. Such self-consistent estimates, generalized to account for particle distributions which exhibit different symmetry from the shape of the inclusions, may be appropriate for composite materials without a clearly defined matrix phase and where each phase appears in the form of particles embedded in a sea of other particles. This is the case, for example, for duplex steels.

4. APPLICATION TO TWO-PHASE COMPOSITES AND CRACKED MEDIA

4.1. *Aligned inclusions and cracks*

In this section, the general HS estimate (3.25) for two-phase composites, consisting of aligned self-similar ellipsoidal inclusions with given aspect ratios and centers distributed with ellipsoidal symmetry (with aspect ratios generally different from those of the inclusion) will be specialized to the cases where the inclusions are either rigid or void. This is done to keep the number of plots to a manageable level and because the cases of rigid and vacuous inclusions, corresponding to arbitrarily large contrast between the properties of the matrix and inclusion phases, are representative of more general cases with finite contrast. For the case of rigid inclusions, the HS estimate specializes to

$$\tilde{\mathbf{L}}^{(\mathrm{HS})} = \mathbf{L}^{(1)} + \frac{c^{(2)}}{c^{(1)}}\mathbf{P}^{-1}, \tag{4.1}$$

whereas for the case of vacuous inclusions the HS estimate reduces to

$$\tilde{\mathbf{L}}^{(\mathrm{HS})} = \mathbf{L}^{(1)} + c^{(2)}[c^{(1)}\mathbf{P} - \mathbf{M}^{(1)}]^{-1}, \tag{4.2}$$

where $\mathbf{M}^{(1)} = (\mathbf{L}^{(1)})^{-1}$ is the compliance tensor of the matrix phase. Expression (3.26) makes the above expressions explicit in terms of the $\mathbf{P}$ tensors for the inclusion $\mathbf{P}_{\mathrm{i}}$ and distribution $\mathbf{P}_{\mathrm{d}}$. It should be emphasized that expressions (4.1) and (4.2) are actually lower and upper bounds, respectively, for the effective modulus tensor of composite materials with the class of microstructures specified by the tensors $\mathbf{P}_{\mathrm{i}}$ and $\mathbf{P}_{\mathrm{d}}$.

For simplicity, results will be presented for the case where the properties of the matrix are assumed to be isotropic, so that, using Hill's (1965) notation, $\mathbf{L}^{(1)} = (3K^{(1)}, 2G^{(1)})$, where $K^{(1)}$ and $G^{(1)}$ denote the bulk and shear modulus of the matrix material, respectively. In addition, the inclusions will be assumed to be spheroidal in shape, with aspect ratio w such that $w \leqslant 1$ and $\geqslant 1$ correspond to oblate and prolate shapes, respectively. Under these assumptions, the $\mathbf{P}$ tensor of the inclusion may be computed explicitly in terms of $K^{(1)}$, $G^{(1)}$ and w; it is given in Appendix B. The tensor $\mathbf{P}_{\mathrm{d}}$ may be similarly computed, if the distribution of inclusions is also assumed to be spheroidal with aspect ratio w_{d}; it is identical to $\mathbf{P}_{\mathrm{i}}$, except that the inclusion aspect ratio w is replaced by w_{d}.

With the above assumptions for the properties of the matrix and inclusion, and for the shape and distribution of the inclusions, the effective modulus tensor is found to exhibit transversely isotropic symmetry, so that, using the standard notation and corresponding simplified tensor algebra for transversely isotropic tensors (Walpole, 1969), it may be expressed in the form

$$\tilde{\mathbf{L}}^{(\mathrm{HS})} = (2k, l, l', n, 2m, 2p), \tag{4.3}$$

where the superscripts and tildes have been dropped from the transversely isotropic moduli, for simplicity. In particular, k is the effective plane strain bulk modulus, m is the effective transverse shear modulus and p is the effective longitudinal shear modulus. Also note that $\tilde{\mathbf{L}}^{(\mathrm{HS})}$ is symmetric, so that $l' = l$. The corresponding effective compliance tensor may be written in the form

$$\tilde{\mathbf{M}}^{(\mathrm{HS})} = \left(\frac{n}{2kE_{11}}, -\frac{\nu_{12}}{E_{11}}, -\frac{\nu_{12}}{E_{11}}, \frac{1}{E_{11}}, \frac{1}{2m}, \frac{1}{2p}\right), \tag{4.4}$$

where $E_{11} = n - l^2/k$ and $\nu_{12} = l/2k$ denote the longitudinal (axial) Young's modulus and Poisson's ratio, respectively. (Note that it has been assumed that the axis of

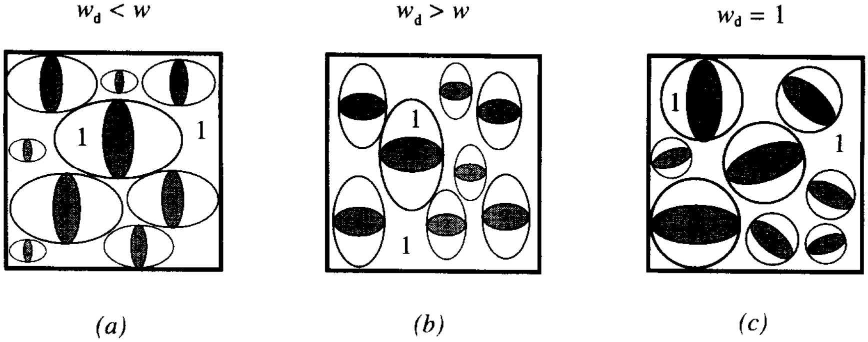

Fig. 2. Three different types of distributions containing spheroidal particles with aspect ratio w. (a) Aligned particles distributed with spheroidal symmetry with aspect ratio $w_d < w$. (b) Aligned particles distributed with spheroidal symmetry with aspect ratio $w_d > w$. (c) Randomly oriented particles with aspect ratio w distributed with spherical symmetry ($w_d = 1$).

symmetry of the composite coincides with the x_1-direction.) It is also useful to introduce an axial shear modulus $a = E_{11}/2(1+\nu_{12})$; for incompressible, transversely isotropic composites, a serves to characterize the effective response under axisymmetric loading.

Parts (a) and (b) of Fig. 2 show the two ways in which the inclusions may be distributed for the above-described class of two-phase composites with aligned inclusions. When the shape and distribution of the inclusions is such that $w_d < w$, as shown in Fig. 2(a) it is useful to introduce the variable $f^{(2)}$, denoting the proportion of spheroids Ω_d, such that

$$c^{(2)} = f^{(2)}\left(\frac{w_d}{w}\right)^2. \tag{4.5}$$

Then, for consistency with the hypothesis of impenetrability of the inclusions, or more precisely of the "security" spheroids Ω_d surrounding the inclusions, the maximum possible value of $f^{(2)}$ is 100%. It follows that for given w_d and $c^{(2)}$, the maximum value of w is given by $w|_{max} = w_d/\sqrt{c^{(2)}}$. Similarly, for given w and $c^{(2)}$, the minimum value of w_d is $w_d|_{min} = w\sqrt{c^{(2)}}$. On the other hand, when the shape and distribution of the inclusions is such that $w_d > w$, as in Fig. 2(b), the proportion of composite spheroids $f^{(2)}$ is defined by

$$c^{(2)} = f^{(2)}\left(\frac{w}{w_d}\right). \tag{4.6}$$

In this case, the minimum possible value of w for given w_d and $c^{(2)}$ is $w|_{min} = w_d c^{(2)}$, and correspondingly the maximum possible value of w_d for given w and $c^{(2)}$ is $w_d|_{max} = w/c^{(2)}$.

Results are shown in Fig. 3 for the effective transverse shear modulus m, longitudinal shear modulus p and axisymmetric (axial) modulus a of an incompressible composite with 25% volume fraction of rigid *spherical* inclusions ($w = 1$) distributed according to spheroidal symmetry with aspect ratio w_d. The results are normalized by the shear

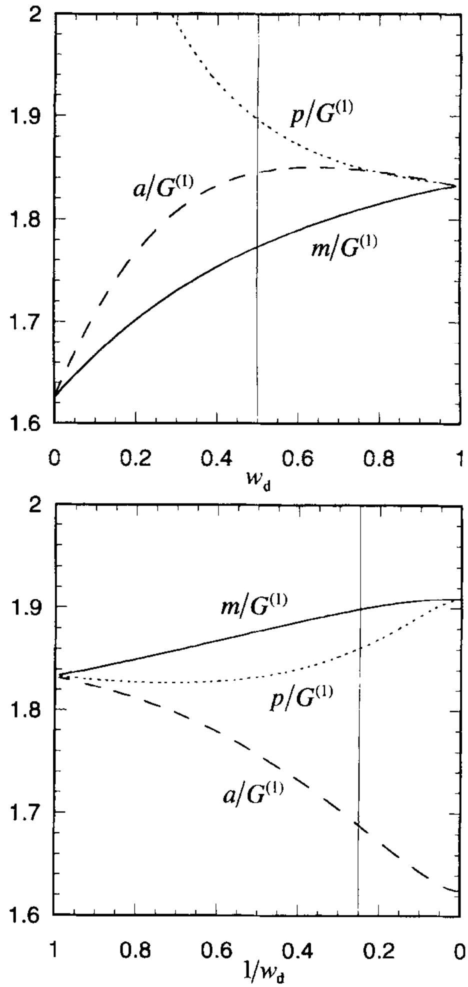

Fig. 3. Estimates for the effective transverse shear modulus m, longitudinal shear modulus p and axisymmetric modulus a of an incompressible composite with 25% (by volume) of aligned rigid spherical inclusions ($w = 1$) distributed with spheroidal symmetry with aspect ratio w_d. (a) Oblate distribution ($w_d < 1$). (b) Prolate distribution ($w_d > 1$). The estimates are rigorous lower bounds for this class of microstructures for $0.5 < w_d < 4$.

modulus of the incompressible matrix material $G^{(1)}$. Following the discussion in the previous paragraph, the results of Fig. 3(a) are only rigorously valid for $0.5 < w_d < 1$, and those of Fig. 3(b) only for $1 < w_d < 4$. It is found that oblate distributions ($w_d < 1$) of rigid spherical inclusions lead to a significant enhancement (more than 3% at $w_d = 0.5$) in the longitudinal shear modulus (p), a relatively small increase in the axial modulus (a), but to a decrease in the transverse shear modulus (m). The fact that a decrease in m should be obtained is perhaps physically intuitive given that decreasing w_d implies that the particles are further apart in the plane perpendicular to the axis of symmetry. However, the behaviors of p and a are not so easily explained. On the other hand, prolate distributions ($w_d > 1$) of rigid spherical inclusions lead to a sizable reduction (more than 8% at $w_d = 4$) in the axisymmetric modulus, to a modest enhancement in the transverse shear modulus, and to a relatively small change in the longitudinal shear modulus. As expected from the observations made in connection with asymptotic relation (3.24), the effect of the distribution of the particles is of order $(c^{(2)})^2$ (in this case 6.25%) and therefore relatively small. It is also observed that this type of distribution for spherical particles becomes progressively unrealistic with increasing values of the volume fraction of inclusions and aspect ratio of the distribution. It was selected in an attempt to isolate the effect of the distribution on effective properties.

Results are given in Fig. 4 for the opposite case where the rigid inclusions are distributed with *spherical symmetry* ($w_d = 1$), but the aspect ratio of the inclusions is allowed to vary. As in the context of the previous figure, the volume fraction of inclusions is assumed to be fixed at 25%, and is normalized relative to the shear modulus of the incompressible matrix material $G^{(1)}$. Figure 4(a) gives results for m, p and a for values of $w < 1$ and Fig. 4(b) for $w > 1$. (In addition, results are also shown for the effective shear modulus G of composites with randomly oriented inclusions of aspect ratio w, distributed with spherical symmetry, but these results will be discussed in Section 4.2.) The discussion associated with relations (4.5) and (4.6) implies that the results of Fig. 4 are only rigorously valid for values of w in the range 0.25–2. It is found that oblate ($w < 1$) rigid inclusions can significantly enhance the transverse shear modulus (m) and reduce the longitudinal shear modulus (p); however, the axisymmetric modulus (a) is not affected nearly as much. On the other hand, prolate ($w > 1$) rigid inclusions significantly enhance the axisymmetric modulus, moderately reduce the transverse shear modulus and hardly affect the longitudinal shear modulus. All of these results are fairly intuitive; for example, the fact that the axisymmetric modulus should increase (by over 30% at $w = 2$) with increasing aspect ratio follows from the fact that the long rigid fibers tend to make the composite stiffer in the axial direction. It is worth commenting here that the effect of particle aspect ratio is roughly the opposite of that of the aspect ratio of the distribution of the particles, discussed in the context of the previous figure. Also the fact that the effect of the aspect ratio of the inclusions on the effective properties of the composite is much larger in relative terms than the corresponding effect of the aspect ratio of the distribution of spherical inclusions is not surprising in view of the fact that the effect of particle shape is of order 1 in the volume fraction of inclusions, whereas the effect of particle distributions is of order 2.

Figure 5 gives the corresponding results for the case where the aspect ratio of the rigid particles is identical to that of the particle distribution (i.e. $w_d = w$). As in the previous two figures, the volume fraction of inclusions is assumed to be fixed at 25%, and is normalized relative to the shear modulus of the incompressible matrix material $G^{(1)}$. It is found that the results for this case are qualitatively similar to those of the previous figure corresponding to fixed spherical distribution and variable shape for the particles. However, the effect of the additional changes (relative to Fig. 4) in particle distribution are such that they lead to quantitatively smaller effects than the previous results for the spherical distributions of particles. This may be understood in terms of the fact that the effect of changes in the aspect ratios of the particle distributions are opposite to corresponding changes in the aspect ratios of the particle

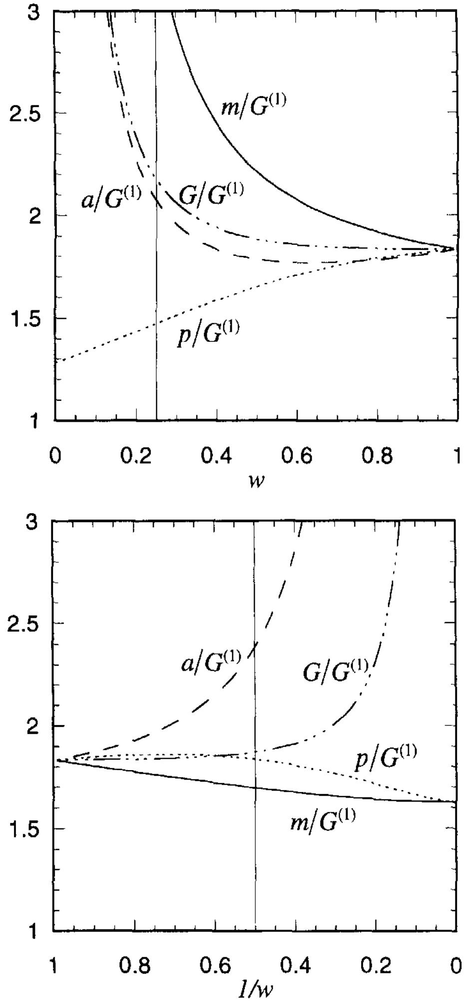

Fig. 4. Estimates for the effective transverse shear modulus m, longitudinal shear modulus p and axisymmetric modulus a of an incompressible composite with 25% (by volume) of aligned rigid spheroidal inclusions distributed with spherical symmetry ($w_d = 1$). (a) Oblate inclusions ($w < 1$). (b) Prolate inclusions ($w > 1$). In addition, estimates are given for the shear modulus G of an incompressible composite with 25% randomly oriented rigid spheroidal inclusions (with aspect ratio w) distributed with spherical symmetry ($w_d = 1$). The estimates are rigorous lower bounds for the two types of microstructures for $0.25 < w_d < 2$.

shape, although significantly smaller in relative terms. One advantageous feature of this type of distribution, originally proposed by Willis (1977, 1978), is that the limiting cases of zero and infinite aspect ratios correspond exactly to the special cases of laminated and fiber-reinforced composites. It will be recalled from the discussion associated with the two prior figures that, unlike for the present case, the limiting values of the aspect ratios were not acceptable for those cases.

Although the above-described three types of distributions may be unrealistic for

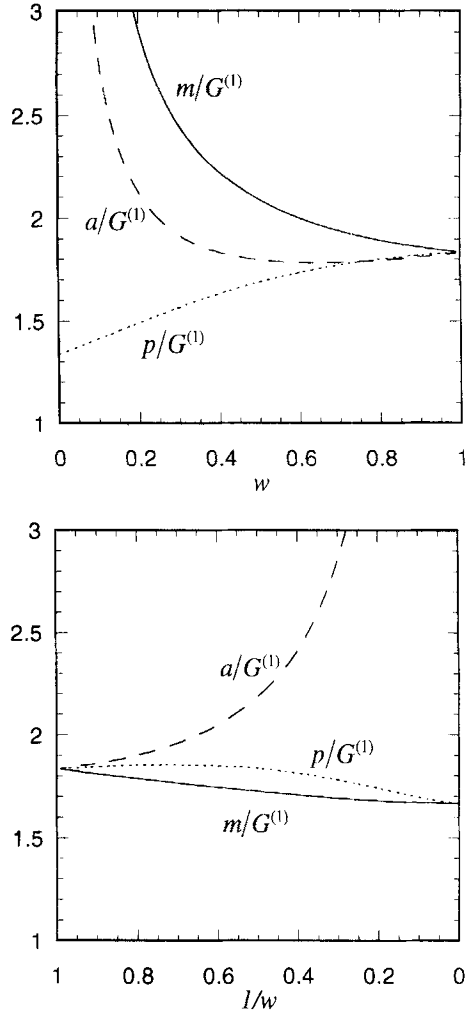

Fig. 5. Estimates for the effective transverse shear modulus m, longitudinal shear modulus p and axisymmetric modulus a of an incompressible composite with 25% (by volume) of aligned rigid spheroidal inclusions distributed with spheroidal symmetry with $w_d = w$. (a) Oblate inclusions ($w < 1$). (b) Prolate inclusions ($w > 1$). The estimates are rigorous lower bounds for this class of microstructures for all values of w_d.

most composites, they are perhaps the simplest in some sense and were chosen mainly in an attempt to dissociate the effects of particle distribution from those of particle shape. In addition, these distributions may be appropriate for some special cases. For example, the spheroidal distribution of rigid spherical particles may be the result of a forming process, where a material with an initially statistically isotropic distribution of rigid spheres is subjected to finite axisymmetric deformations. The general ellipsoidal distribution of ellipsoidal particles with the same aspect ratio as the distribution may be appropriate for certain high-concentration particle-reinforced composite materials. In general, however, it is expected that even if the particles are identical, during most forming processes the particles will lose their alignment. In this event, the estimates (4.1) and (4.2) cease to be relevant, and the more general expression (3.20) is required where each different orientation of the particle is treated as a different "phase". One simple special case of this larger class is the situation where all particle orientations are possible and are randomly distributed. This case will be considered in Section 4.2. Before pursuing this, results are given next for the important cases of materials reinforced by aligned rigid disks or weakened by aligned cracks.

It has already been observed in connection with relation (4.6) that the minimum possible value of the aspect ratio of the spheroidal inclusions, for given distribution aspect ratio w_d and inclusion volume fraction $c^{(2)}$, is $w|_{min} = w_d c^{(2)}$. This suggests considering composites with a fixed value of the proportion of distributional spheroids $f^{(2)}$, and then taking the limit as w tends to zero [which implies that $c^{(2)}$ also tends to zero]. Physically, this limit corresponds to the case of a spheroidal distribution (with aspect ratio w_d) of circular disks or cracks with density parameter $\alpha = f^{(2)}/w_d$. In this paper, the choice has been made to present the results in terms of α instead of the more commonly used (cf. Budiansky and O'Connell, 1976) density parameter $\varepsilon = p^{(2)}r^3 = 3\alpha/4\pi$, where r denotes the mean radius of the circular disks or cracks. The reason for this is that the maximum allowable value of $f^{(2)}$ is 100%, corresponding to the onset of particle overlap, which was explicitly excluded by the hypotheses made on the distribution of inclusions. Therefore, for spherical distributions ($w_d = 1$), the maximum allowable value of α is 100%. (Note that $\alpha = 1$ corresponds to $\varepsilon \cong 0.239$ for spherical distributions of particles.)

Results for rigid disks and cracks were obtained directly from expressions (4.1) and (4.2), respectively, by making use of (4.6) with α fixed, and letting $w \to 0$. The matrix material was taken to be compressible and isotropic with shear and bulk modulus $G^{(1)}$ and $K^{(1)}$. Explicit results for the cases of spherical distributions ($w_d = 1$) and "flat" distributions ($w_d = w \to 0$) are given below. For the cases of spherical distributions of disks, the results for the in-plane bulk modulus k and transverse shear modulus m are

$$\frac{k}{k^{(1)}} = 1 + \frac{120\alpha(1-\nu^{(1)})(1-2\nu^{(1)})}{15\pi(3-4\nu^{(1)}) - 16\alpha(3-5\nu^{(1)})},$$

$$\frac{m}{m^{(1)}} = 1 + \frac{240\alpha(1-\nu^{(1)})}{15\pi(7-8\nu^{(1)}) - 32\alpha(4-5\nu^{(1)})}, \tag{4.7}$$

where $\nu^{(1)}$ is the Poisson ratio of the matrix, $k^{(1)} = K^{(1)} + G^{(1)}/3$ and $m^{(1)} = G^{(1)}$. Note that these results are rigorous lower bounds for k and m, respectively, provided that $\alpha \leqslant 1$. On the other hand, for $\alpha > 1$, they are estimates which probably become progressively less accurate as α becomes larger. The corresponding results for a flat distribution of disks, as first given by Willis (1980), are

$$\frac{k}{k^{(1)}} = 1 + \frac{8\alpha(1-\nu^{(1)})(1-2\nu^{(1)})}{\pi(3-4\nu^{(1)})},$$

$$\frac{m}{m^{(1)}} = 1 + \frac{16\alpha(1-\nu^{(1)})}{\pi(7-8\nu^{(1)})}, \tag{4.8}$$

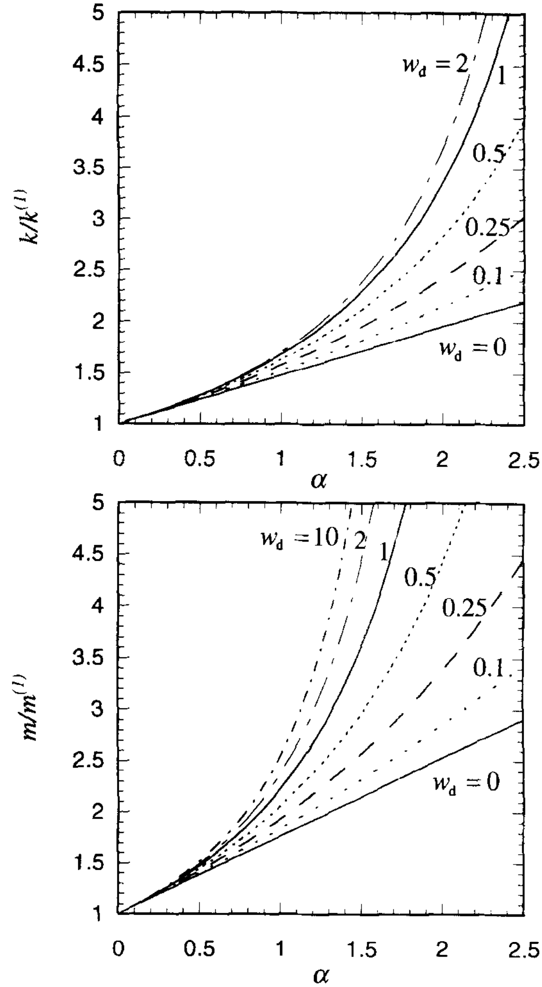

Fig. 6. Estimates for (a) the effective in-plane bulk modulus k and (b) the transverse shear modulus m of an elastic solid with Poisson ratio $\nu^{(1)} = 0.25$ reinforced by spheroidal distributions (with aspect ratio w_d) of aligned rigid circular disks as functions of the disk-density parameter α. The estimates are rigorous lower bounds for the corresponding classes of microstructures for $\alpha < 1/w_d$.

which may be seen to correspond to the results for spherical distributions with the terms depending on α in the denominator set equal to zero. The results for the other moduli in (4.3) are the same as those for the matrix material. This is seen to be a consequence of the fact that the rigid disks only produce stiffness enhancements for the in-plane hydrostatic and transverse shear modes, which makes good physical sense.

The results mentioned in the previous paragraph for composites reinforced by aligned rigid disks are compared in Fig. 6, as a function of the disk-density parameter α, for a compressible matrix with $\nu^{(1)} = 1/4$. It is observed that the results for the spherical distribution of rigid disks predict larger increases in the relevant stiffnesses (k and m) than the corresponding results for flat distributions, which are formally in agreement with the results for dilute concentrations of rigid disks and are hence linear on α. For example, at $\alpha = 100\%$, the predictions for transverse shear modulus for the spherical distribution is over 25% higher than for the flat distribution. The reason for the difference in the predictions may be understood in terms of the fact that the flat distribution assumes that the disks are spaced far away from each other in the plane perpendicular to the axis of symmetry, so that they tend to line up in the axial direction. This means that, although the density of disks may be high (because the concentration along the axial direction may be high), the interaction amongst the disks will be weak. (This explains why the result for this distribution coincides with the dilute result, even though the concentrations of disk can be high.) On the other hand, the spherical distribution implies that as the disk-density parameter α increases, there is a stronger interaction between the disks leading to the enhanced improvement in the relevant stiffnesses over the dilute prediction. Results are also given in Fig. 6 for intermediate values of the distribution aspect ratio w_d, corresponding to oblate distributions, as well as for some values of $w_d > 1$, corresponding to prolate distributions of disks. They show a progressive enhancement in the stiffening of the composite with increasing w_d, which is consistent with the above remarks. In particular, stiffer predictions are generated for the prolate distributions than for the spherical distributions as a consequence of the fact that the disks become more closely spaced in the plane perpendicular to their axes and hence interact more strongly. However, in interpreting these results it should be kept in mind that they are only rigorously valid for values of α less than $1/w_d$.

For the case of solids with spherical distributions of cracks, the results for the axial Young's modulus E_{11} and longitudinal shear modulus p are

$$\frac{E_{11}}{E_{11}^{(1)}} = 1 - \frac{60\alpha[1-(\nu^{(1)})^2]}{15\pi + 4\alpha[7-15(\nu^{(1)})^2]},$$
$$\frac{p}{p^{(1)}} = 1 - \frac{60\alpha(1-\nu^{(1)})}{15\pi(2-\nu^{(1)}) + 8\alpha(4-5\nu^{(1)})}, \tag{4.9}$$

where $E_{11}^{(1)} = 2(1+\nu^{(1)})G^{(1)}$ and $p^{(1)} = G^{(1)}$. Note that these results are rigorous upper bounds for E_{11} and p provided that $\alpha \leqslant 1$. The corresponding results for a flat distribution of cracks, as first given by Willis (1980), may be written in the form

$$\frac{E_{11}^{(1)}}{E_{11}} = 1 + \frac{4\alpha}{\pi}[1-(\nu^{(1)})^2],$$
$$\frac{p^{(1)}}{p} = 1 + \frac{4\alpha(1-\nu^{(1)})}{\pi(2-\nu^{(1)})} \tag{4.10}$$

and they are seen to be linear in α. The results for the other moduli in (4.4), for both types of distributions, are the same as those for the matrix material. This is seen to be a consequence of the fact that the cracks only affect the axial tensile and longitudinal shear modes.

These results are compared in Fig. 7 for a compressible matrix with $\nu^{(1)} = 1/4$. It is observed that the results for the spherical distribution of cracks predict larger drops

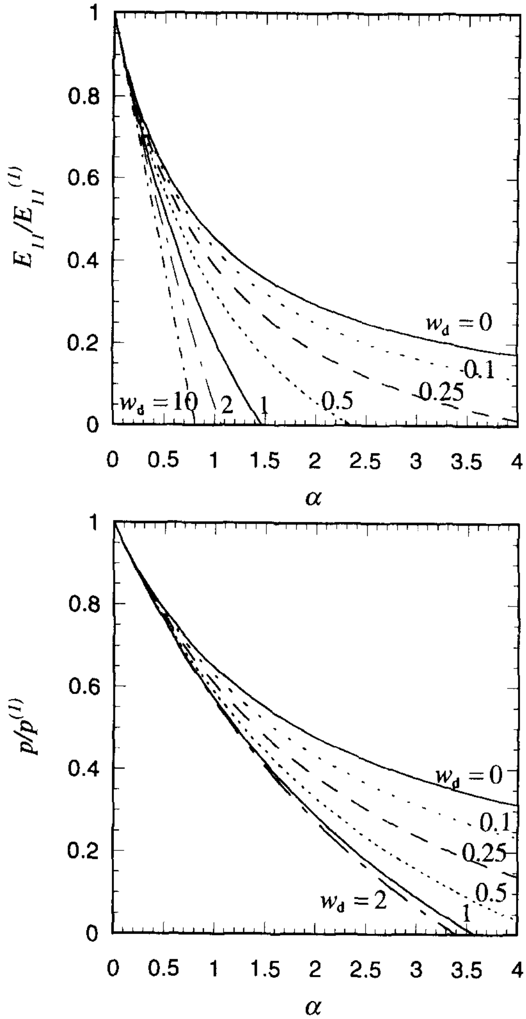

Fig. 7. Estimates for (a) the effective axial Young's modulus E_{11} and (b) the longitudinal shear modulus p of an elastic solid with Poisson ratio $\nu^{(1)} = 0.25$ weakened by spheroidal distributions (with aspect ratio w_d) of aligned circular cracks as functions of the crack-density parameter α. The estimates are rigorous upper bounds for the corresponding classes of microstructures for $\alpha < 1/w_d$.

in the relevant stiffnesses (E_{11} and p) than the corresponding results for flat distributions. For example, the predictions for axial Young's modulus for the spherical distribution is about 50% lower than for the flat distribution at $\alpha = 100\%$. As for the case of rigid disks, the reasons for this difference may be traced to the fact that the cracks interact more strongly for the spherical distribution than for the flat distribution, for which the interaction is rather weak leading to the dilute prediction. In addition, it is observed that the cracks have a more pronounced effect on the Young's modulus than for the corresponding longitudinal shear modulus. Results are also given in Fig. 7 for other values of the distribution aspect ratio w_d. They show a progressive loss of stiffness for the cracked material with increasing w_d, at fixed crack density α. In particular, the predictions for the axial Young's modulus are seen to approach zero, at progressively smaller values of α, with increasing values of $w_d > 0$. In interpreting these results it should again be kept in mind that they are only rigorously valid for values of α less than $1/w_d$, so that for example the predictions for $w_d = 10$ cease to be rigorous bounds long before ($\alpha = 0.1$) E_{11} reaches a zero value of the stiffness (near $\alpha = 0.8$). On the other hand, the prediction for E_{11} with $w_d = 0.25$, at $\alpha = 4$, where the result is still a bound, shows that the material has essentially lost all its load-carrying capacity under this loading mode.

It is relevant to mention here that formulae (4.10) for the flat distribution of cracks are identical to the formulae that would be obtained from the Mori–Tanaka procedure. This is because, as pointed out earlier, the Mori–Tanaka procedure implicitly assumes the same distribution for the inclusions as their shape, which is in agreement with the general estimate (3.25) for the case where the distribution and inclusion have the same aspect ratio. In addition, it is observed that the results for the spherical distribution of cracks are in fairly good agreement with the self-consistent estimates of Hoenig (1979), with the self-consistent estimates lying below the HS upper bounds in the range of α for which the HS bounds are rigorous. However, interestingly, the HS bounds for spherical distributions go to zero at a finite value of α [e.g. $E_{11}^{(1)} \to 0$ at $\alpha = 15\pi/32$], whereas the self-consistent estimates of Hoenig tend to zero asymptotically as $\alpha \to \infty$.

Finally, results could easily be obtained for other cases of practical interest. For example, the corresponding HS estimates for materials reinforced by rigid needles, or weakened by vacuous needles could be computed from the general expressions (4.1) and (4.2), respectively. It is found that such shapes do not produce any reinforcement or loss of stiffness relative to the matrix in the limit as the aspect ratio of the inclusions approaches an infinite value. However, large finite values of the aspect ratio do produce an effect as the results of Figs 3 and 4 have shown for the case of rigid particles. Another example would be results for composites with ellipsoidal distributions of rigid disks and cracks with general elliptical planforms. This would require the computation of the appropriate **P** tensors for the inclusion and distribution. In addition, the case of wet and sliding cracks could also be considered in this more general statistical context.

4.2. *Randomly oriented inclusions and cracks*

Paralleling the developments in the previous section, in this section the general HS estimate (3.27) for two-phase composites with randomly oriented ellipsoidal inclusions with given aspect ratios distributed according to ellipsoidal symmetry (with aspect ratios generally different from those of the inclusion) are specialized to the cases where the inclusions are either rigid or void. For the case of rigid inclusions, the HS estimate specializes to

$$\tilde{\mathbf{L}}^{(HS)} = \mathbf{L}^{(1)} + (1-c^{(1)})[\mathbf{I} - (1-c^{(1)})\{\mathbf{P}_i^{-1}\}\mathbf{P}_d]^{-1}\{\mathbf{P}_i^{-1}\}, \tag{4.11}$$

whereas for the case of vacuous inclusions the HS estimate reduces to

$$\tilde{\mathbf{L}}^{(HS)} = \mathbf{L}^{(1)} + (1-c^{(1)})[\mathbf{I} - (1-c^{(1)})\{[\mathbf{P}_i - \mathbf{M}^{(1)}]^{-1}\}\mathbf{P}_d]^{-1}\{[\mathbf{P}_i - \mathbf{M}^{(1)}]^{-1}\}, \tag{4.12}$$

where $\mathbf{P}_i$ denotes the $\mathbf{P}$ tensor of the identically shaped inclusions with variable orientation, and $\mathbf{P}_d$ is the corresponding distribution tensor. Expressions (4.11) and (4.12) are lower and upper bounds, respectively, for the effective modulus tensor of composite materials with the class of microstructures specified by the tensors $\mathbf{P}_i$ and $\mathbf{P}_d$.

As in the previous section, results will be presented for the cases where the properties of the matrix are assumed to be isotropic, with bulk and shear modulus $K^{(1)}$ and $G^{(1)}$. The inclusions are assumed to be spheroidal with aspect ratio w, so that $\mathbf{P}_i$ is as given in Appendix B in terms of $K^{(1)}$, $G^{(1)}$ and w. The distribution of inclusions will be assumed to be isotropic ($w_d = 1$), so that $\mathbf{P}_d = (1, (3/5G^{(1)})(K^{(1)}+2G^{(1)}))/(3K^{(1)}+4G^{(1)})$. Figure 2(c) gives a picture of the microstructure; recall that the security spheres surrounding the inclusions are not allowed to overlap. As is well known, the orientational average of a transversely isotropic tensor, such as (4.3), is given by $\{\mathbf{L}\} = (3K, 2G)$, where

$$K = \tfrac{1}{9}\mathbf{L}_{iijj} = \tfrac{1}{9}[4k+n+2(l+l')],$$

$$G = \tfrac{1}{10}(\mathbf{L}_{ijij}-\tfrac{1}{3}\mathbf{L}_{iijj})\mathbf{L}_{iijj} = \tfrac{1}{15}[k+n+6(m+p)-l-l']. \tag{4.13}$$

Then, the expressions (4.11) and (4.12) may be shown to lead to isotropic estimates for the effective modulus tensor of the composites of the form

$$\tilde{\mathbf{L}}^{(\mathrm{HS})} = (3K, 2G), \tag{4.14}$$

where K and G are used to denote the effective bulk and shear modulus of the composites.

Results were shown in Fig. 4 for the effective shear modulus G of an incompressible composite with 25% volume fraction of rigid spheroidal inclusions (variable w) distributed with spherical symmetry ($w_d = 1$). The results are normalized by the shear modulus of the incompressible matrix material $G^{(1)}$. As was the case for the results for the aligned particles, the results for the randomly oriented particles are only rigorously valid for $0.25 < w < 2$. It is found that, in this range of values of w, oblate ($w < 1$) rigid inclusions produce a modest increase (e.g. roughly 17% at $w = 0.25$) in the effective shear modulus of the composite, whereas prolate ($w > 1$) rigid inclusions have a very slight strengthening effect on the effective modulus. Note that self-consistent estimates for composites with randomly oriented inclusions have been proposed by Wu (1966) and Walpole (1969). Comparisons with these types of estimates will be given later in the context of cracked media.

Figure 8(a) and (b) provides plots of the effect of particle aspect ratio on the effective shear modulus of a composite reinforced by spherical distributions ($w_d = 1$) of prolate rigid inclusions and weakened by oblate voids, respectively, as functions of the density parameter α [equal to $c^{(2)}w^2$ for prolate inclusions and to $c^{(2)}/w$ for oblate inclusions]. Relative to this measure of the density of inclusions, the stiffening capability of the rigid inclusions can be seen to decrease with increasing aspect ratio and, correspondingly, the weakening effect of the voids may be seen to be reduced with decreasing aspect ratio. However, the strengthening effect of the prolate rigid inclusions may be seen to disappear in the limit as $w \to \infty$ (corresponding to needles), whereas the weakening effect of the oblate voids remains finite (and significant) in the

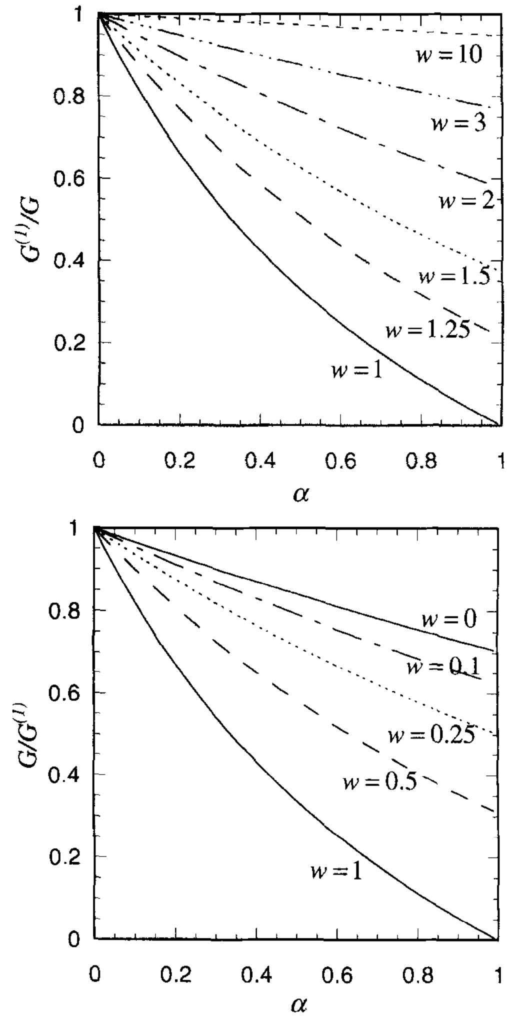

Fig. 8. (a) Estimates for the effective shear modulus G of an elastic solid with $\nu^{(1)} = 0.25$ reinforced by spherical distributions ($w_d = 1$) of randomly oriented spheroidal rigid inclusions with aspect ratio w. (b) Estimates for the effective shear modulus G of an elastic solid with $\nu^{(1)} = 0.25$ weakened by spherical distributions ($w_d = 1$) of randomly oriented spheroidal voids with aspect ratio w. The estimates are rigorous lower (upper in the figure) and upper bounds, respectively, for values of $\alpha < 1$.

limit as $w \to 0$ (corresponding to cracks). Similar observations could be made for the limits of needle-shaped voids and rigid disks.

Next the cases of randomly oriented rigid disks and cracks with spherical distributions are considered in more detail. For the case of rigid disks, the results, which are actually lower bounds, for the bulk and shear modulus may be computed from (4.11) and are given by

$$\frac{K}{K^{(1)}} = 1 + \frac{48\alpha(1-\nu^{(1)})(1-2\nu^{(1)})}{(1+\nu^{(1)})[9\pi(3-4\nu^{(1)})-16\alpha(1-2\nu^{(1)})]},$$

$$\frac{G}{G^{(1)}} = 1 + \frac{120\alpha(1-\nu^{(1)})(43-56\nu^{(1)})}{225\pi(7-8\nu^{(1)})(3-4\nu^{(1)})-16\alpha(4-5\nu^{(1)})(43-56\nu^{(1)})}, \quad (4.15)$$

where $\nu^{(1)}$ is the Poisson ratio of the matrix. For later reference, it is noted that the corresponding Mori–Tanaka estimates (Weng, 1990) are formally obtained from the above expressions by setting the terms depending on α in the denominators equal to zero.

The estimates (4.15) for composites reinforced by randomly oriented rigid disks in a compressible matrix with $\nu^{(1)} = 1/4$ are compared in Fig. 9 with the Mori–Tanaka (MT) estimates also mentioned above. It is observed that the new estimates predict a higher strengthening effect than the MT estimates. For example, at $\alpha = 1$, the new estimates for G are 5% higher than the MT estimates. Moreover, the new estimates are actually rigorous lower bounds for the class of microstructures with spherical distributions of randomly oriented rigid disks and therefore the MT estimates which violate this bound cannot be valid estimates for this class of microstructures, except for dilute concentrations of disks, in which case they agree with the dilute predictions anyway. These findings make more precise the comments made in Section 3.3 regarding the MT estimates for composites with randomly oriented inclusions, and illustrate the deficiencies associated with the MT estimates.

For solids with spherical distributions of randomly oriented cracks, the results, which are rigorous upper bounds for this class of materials, for the effective bulk and shear modulus may be computed from (4.12) and are given by

$$\frac{K}{K^{(1)}} = 1 - \frac{12\alpha[1-(\nu^{(1)})^2]}{9\pi(1-2\nu^{(1)})+4\alpha(1+\nu^{(1)})^2},$$

$$\frac{G}{G^{(1)}} = 1 - \frac{120\alpha(1-\nu^{(1)})(5-\nu^{(1)})}{225\pi(2-\nu^{(1)})+16\alpha(4-5\nu^{(1)})(5-\nu^{(1)})}. \quad (4.16)$$

These new estimates for spherical distributions of randomly oriented cracks are compared, for a compressible matrix with $\nu^{(1)} = 1/4$, in Fig. 10 with the Mori–Tanaka (MT) estimates of Benveniste (1987), as well as with the self-consistent (SC) estimates of Budiansky and O'Connell (1976) and the differential self-consistent (DSC) estimates of Zimmerman (1985) [see also Hashin (1988)]. It is observed that the new estimates predict a more compliant response in shear than the MT estimate, about the same response as the DSC estimate, but a stiffer response than the SC estimate. On the other hand, the predictions for the bulk modulus are more compliant than both the MT and DSC estimates, but still stiffer than the SC estimate. For example, at $\alpha = 1$, the new estimates for G are about 5% more compliant than the MT estimates and about 12% stiffer than the SC estimates. On the other hand, also at $\alpha = 1$, the new estimates for K are about 10 and 20% more compliant than the corresponding DSC and MT estimates, respectively. However, given that the new estimates are actually rigorous upper bounds for the class of microstructures with spherical distributions of randomly oriented cracks, it follows that the MT and DSC estimates,

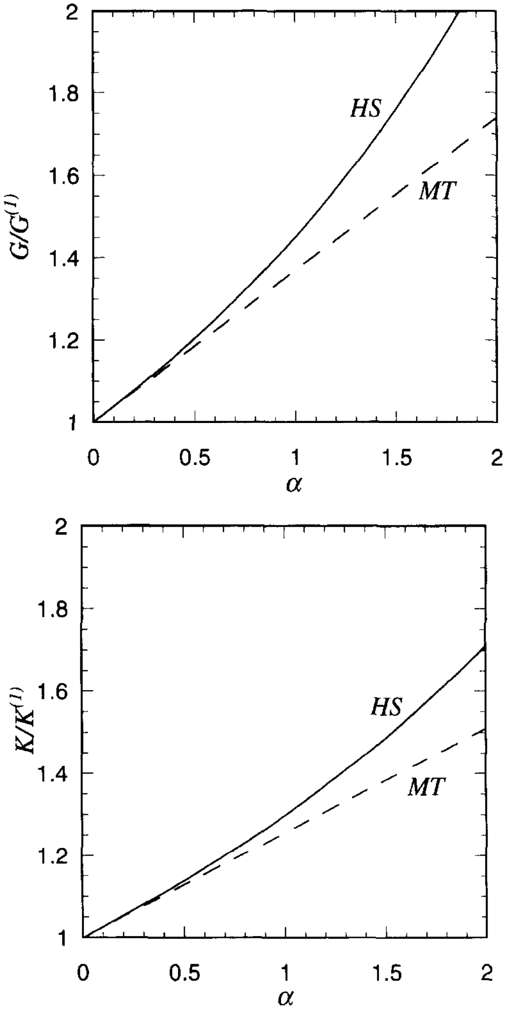

Fig. 9. Estimates for (a) the effective shear modulus G and (b) the bulk modulus K of an elastic solid with Poisson ratio $\nu^{(1)} = 0.25$ reinforced by spherical distributions ($w_d = 1$) of randomly oriented rigid circular disks as functions of the density parameter α. The new Hashin–Shtrikman (HS) estimates are rigorous lower bounds for $\alpha < 1$. The Mori–Tanaka (MT) estimates are also shown for comparison.

which violate these bounds, cannot be valid estimates for this class of microstructures, except in the dilute limit when all the results agree with the estimates of Walsh (1965).

5. CONCLUDING REMARKS

In this work, use has been made of the framework of Willis (1977, 1978) to obtain estimates of the Hashin–Shtrikman type for composites with particulate microstructures consisting of arbitrarily shaped particles which are distributed in a matrix

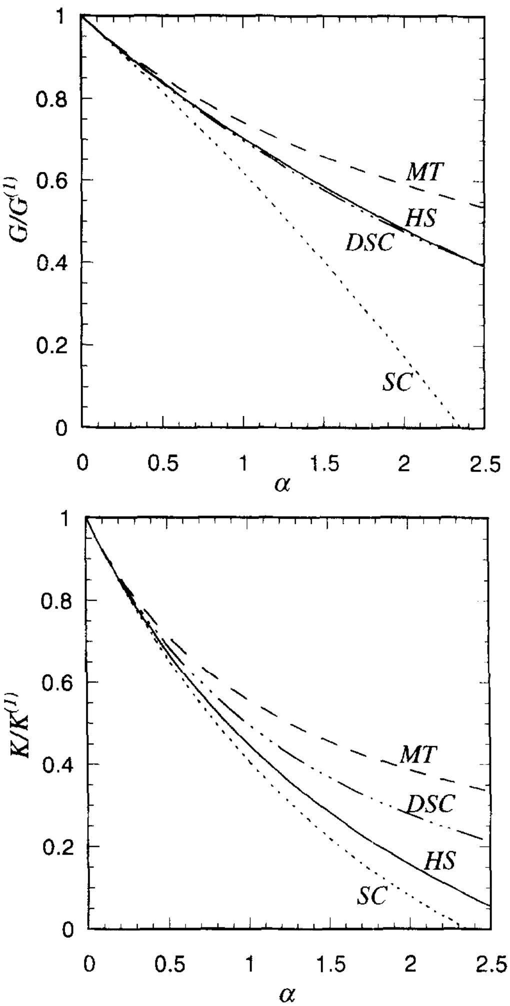

Fig. 10. Estimates for (a) the effective shear modulus G and (b) the bulk modulus K of an elastic solid with $\nu^{(1)} = 0.25$ weakened by spherical distributions ($w_d = 1$) of randomly oriented cracks as functions of the density parameter α. The new Hashin–Shtrikman (HS) estimates are rigorous upper bounds for $\alpha < 1$. The corresponding self-consistent (SC), differential self-consistent (DSC) and Mori–Tanaka (MT) estimates are also shown for comparison.

of a different phase with "ellipsoidal symmetry". When the particles are assumed to be ellipsoidal in shape, completely explicit estimates for the effective modulus tensor $\tilde{\mathbf{L}}$ [see expression (3.20) with (3.21)] are obtained in terms of two microstructural (**P**) tensors serving to characterize the shape and distribution of the particles, respectively. In particular, when the shape and distribution of the particles are chosen to have the same aspect ratio, the estimates of Willis (1977, 1978) are recovered. More generally, the new expressions make allowance for differences in the aspect ratios for the shape and distribution of the inclusions. One particular application, which had not been considered previously in the context of estimates of the Hashin–Shtrikman type, is that for composites with prescribed pairwise distributions of randomly oriented ellipsoidal inclusions. This application also leads to simple, explicit expressions for $\tilde{\mathbf{L}}$ [see, for example, the expression (3.27) for the case where all the pairwise distributions of inclusions are identical and characterized by a single **P** tensor]. Most significantly, these new expressions are found to be in disagreement with corresponding estimates of the Mori–Tanaka and differential self-consistent types, which do not specifically account for the distribution of the randomly oriented inclusions in the composite.

Finally, as already mentioned, it is important to emphasize that the analyses developed in this work may be extended to composites with nonlinear constitutive behavior in the manner originally proposed by Willis (1983) and developed by Talbot and Willis (1985). Alternatively, the variational procedure of Ponte Castañeda (1991) may be used to generate corresponding nonlinear versions of relations (3.20) and (3.27) directly from such estimates. In addition, the new estimates allow the development of corresponding estimates for composites "with evolving microstructures" (Ponte Castañeda and Zaidman, 1994), where a new internal variable is introduced to characterize independent changes in the relative positions of the particles. These possibilities are currently being pursued.

ACKNOWLEDGEMENTS

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APPENDIX A: DEMONSTRATION THAT THE INTEGRAL OVER ${}^{c}\Omega_{d}^{(rs)}$ VANISHES IDENTICALLY

The integral in question is

$$I = \int_{{}^{c}\Omega_{d}^{(rs)}} dz''\Gamma^{(0)}(y-y'-z'')[p^{(s|r)}(z'')-p^{(s)}], \tag{A.1}$$

where the $\Omega_d^{(rs)}$ are the domains defined by relation (3.11), ${}^c\Omega_d^{(rs)}$ are their complements and $\Gamma^{(0)}$ is given by (2.12). For simplicity, it will be assumed initially that the $\Omega_d^{(rs)}$ are spherical, so that

$$ {}^c\Omega_d^{(rs)} = \{z'' : |z''| > 1\}, \tag{A.2} $$

and the conditional probability functions $p^{(s|r)}$ are such that $p^{(s|r)}(z'') = p^{(s|r)}(|z''|)$.

Next, recall the identity (Willis, 1981)

$$ \Gamma^{(0)}(x) = -\frac{1}{8\pi^2}\int_{|\xi|=1} \mathrm{d}S\mathbf{H}^{(0)}(\xi)\delta''(\xi\cdot x), \tag{A.3} $$

where $\mathbf{H}^{(0)}(\xi)$ was defined in the context of relation (3.14), and which follows from the plane-wave decomposition for the three-dimensional delta function. It follows from this identity that

$$ I = -\frac{1}{8\pi^2}\int_{|z''|>1} \mathrm{d}z'' \int_{|\xi|=1} \mathrm{d}S\mathbf{H}^{(0)}(\xi)\delta''[\xi\cdot(y-y'-z'')][p^{(s|r)}(z'')-p^{(s)}], \tag{A.4} $$

which may be rewritten in the form

$$ I = -\frac{1}{8\pi^2}\int_{|\xi|=1} \mathrm{d}S\mathbf{H}^{(0)}(\xi)\frac{\partial^2}{\partial q^2}\left\{\int_{|z''|>1} \mathrm{d}z''\delta[q-\xi\cdot z''][p^{(s|r)}(z'')-p^{(s)}]\right\}\Bigg|_{q=\xi\cdot(y-y')}. \tag{A.5} $$

Finally, since $y\in\Omega_i^{(r)}, y'\in\Omega_i^{(s)}$ imply that $y-y'\in\Omega_d^{(rs)}$, so that $|q| = |\xi\cdot(y-y')| < 1$, the integral inside the curly brackets may be expressed in the form

$$ \int_{\sqrt{1-q^2}}^{\infty} 2\pi r\,\mathrm{d}r\{p^{(s|r)}[(q^2+r^2)^{1/2}]-p^{(s)}\}, \tag{A.6} $$

which, with the change of variables $u=(q^2+r^2)^{1/2}$, gives

$$ \int_1^{\infty} 2\pi u\,\mathrm{d}u[p^{(s|r)}(u)-p^{(s)}]. \tag{A.7} $$

This is a bounded constant, provided that sufficiently fast decay is assumed for $p^{(s|r)}(u)-p^{(s)}$ as $u\to\infty$ in the context of the no long-range order hypothesis. It follows that the second derivative with respect to q in (A.5) vanishes, and therefore that

$$ I = 0. \tag{A.8} $$

The same result may be proved for the more general case of ellipsoidal domains $\Omega_d^{(rs)}$, as given by (3.11), by making use of the changes of variables $z' = Z_d^{(rs)}z''$ and $\xi = (Z_d^{(rs)})^T\zeta$, so that $\zeta\cdot z' = \xi\cdot z''$, and repeating the steps that led from (A.4) to (A.8). The details are omitted for conciseness.

APPENDIX B: P TENSOR FOR SPHEROIDAL INCLUSIONS

The **P** tensor for spheroidal inclusions with aspect ratio w embedded in an isotropic matrix with shear and bulk modulus $G^{(1)}$ and $K^{(1)}$ exhibits transversely isotropic symmetry, so that it may be written in the form

$$ \mathbf{P} = (2k_p, l_p, l_p, n_p, 2m_p, 2p_p), \tag{B.1} $$

where

$$ k_p = \frac{[7h(w)-2w^2-4w^2h(w)]G^{(1)}+3[h(w)-2w^2+2w^2h(w)]K^{(1)}}{8(1-w^2)G^{(1)}(4G^{(1)}+3K^{(1)})}, $$

$$l_p = \frac{(G^{(1)}+3K^{(1)})[2w^2-h(w)-2w^2h(w)]}{4(1-w^2)G^{(1)}(4G^{(1)}+3K^{(1)})},$$

$$n_p = \frac{[6-5h(w)-8w^2+8w^2h(w)]G^{(1)}+3[h(w)-2w^2+2w^2h(w)]K^{(1)}}{2(1-w^2)G^{(1)}(4G^{(1)}+3K^{(1)})},$$

$$m_p = \frac{[15h(w)-2w^2-12w^2h(w)]G^{(1)}+3[3h(w)-2w^2]K^{(1)}}{16(1-w^2)G^{(1)}(4G^{(1)}+3K^{(1)})},$$

$$p_p = \frac{2[4-3h(w)-2w^2]G^{(1)}+3[2-3h(w)+2w^2-3w^2h(w)]K^{(1)}}{8(1-w^2)G^{(1)}(4G^{(1)}+3K^{(1)})}. \quad \text{(B.2)}$$

In the above relations,

$$h(w) = \frac{w[\arccos(w)-w\sqrt{1-w^2}]}{(1-w^2)^{3/2}}, \quad \text{(B.3)}$$

for oblate spheroids ($w < 1$), and

$$h(w) = \frac{w[w\sqrt{w^2-1}-\operatorname{arccosh}(w)]}{(w^2-1)^{3/2}}, \quad \text{(B.4)}$$

for prolate spheroids ($w > 1$). The limit $w \to 1$ gives $h(1) = 2/3$.

It is noted that asymptotic expressions for the above **P** tensor in the limits as $w \to 0$ and ∞, have been given by Willis (1980). These are useful in the computation of the effective modulus tensors for the cases of rigid disks and cracks.