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# IMPROVED RIGOROUS BOUNDS ON THE EFFECTIVE ELASTIC MODULI OF A COMPOSITE MATERIAL

Y. KANTOR and D. J. BERGMAN

Dept. of Physics and Astronomy, Tel Aviv University, 69978 Tel Aviv, Israel

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ABSTRACT

A NEW METHOD for deriving rigorous bounds on the effective elastic constants of a composite material is presented and used to derive a number of known as well as some new bounds. The new approach is based on a presentation of those constants as a sum of simple poles. The locations and strengths of the poles are treated as variational parameters, while different kinds of available information are translated into constraints on these parameters. Our new results include an extension of the range of validity of the Hashin–Shtrikman bounds to the case of composites made of isotropic materials but with an arbitrary microgeometry. We also use information on the effective elastic constants of one composite in order to obtain improved bounds on the effective elastic constants of another composite with the same or a similar microgeometry.

## 1. INTRODUCTION

THEORIES of the effective elastic properties of composite materials (i.e. macroscopically inhomogeneous systems made of regions or grains of different homogeneous substances) have many practical applications in technology and geophysics. Various approaches to the problem were reviewed by HASHIN (1970), WATT *et al.* (1976) (this review contains an exhaustive list of references), HALE (1976), and WILLIS (1982).

In principle, exact values for the effective elastic constants can be obtained only when the microgeometry of the composite is known precisely. In many cases, however, the precise microgeometry is unknown, e.g. when the composite has a certain randomness in its microstructure. In that case, exact theories are limited to the derivation of rigorous bounds on the elastic constants. These range from the simplest bounds of Voigt and Reuss (HILL, 1952), for which only the volume fractions of the components need to be known, through the more complicated Hashin–Shtrikman bounds and their extensions (HASHIN and SHTRIKMAN, 1963; HILL, 1963; WALPOLE, 1966a,b) for isotropic mixtures, and up to some very complicated bounds which require knowledge of the two- and three-point correlation functions (see, e.g. BERAN and MOLYNEUX, 1966; MILLER, 1969; MCCOY, 1970). Recently some of these bounds (the Hashin–Shtrikman and some of the higher order bounds) were modified and improved by MILTON and PHAN-THIEN (1982). The derivation of bounds is usually based on variational principles (e.g. HILL, 1952; HASHIN and SHTRIKMAN, 1961, 1962; BERAN and MOLYNEUX, 1966). Sometimes these principles are used in the context of a scattering-theory-like approach (e.g. DEDERICHS and ZELLER, 1973; WILLIS, 1982).

Recently we introduced a new approach to the problem of evaluation of the effective elastic constants of composite materials (KANTOR and BERGMAN, (1982a)—this will be referred to as I). We showed that any effective elastic constant of a composite can be written as a sum of simple poles, and reduced the problem to that of a systematic evaluation of the locations and weights of these poles. The usefulness of this approach was demonstrated on several well defined microgeometries (see I, and also KANTOR and BERGMAN (1982b)). In this article we will apply this pole representation to the case when only partial information is available on the microgeometry of the system. We will treat the locations and the weights of the poles as unknown parameters, and we will derive rigorous upper and lower bounds on the effective elastic constants by varying them subject to certain constraints imposed by the known information. A similar method was developed and applied by BERGMAN (1978a, b, 1982), for the derivation of bounds on the effective dielectric constants of composites.

In Section 2, we rederive the general theory of I for the elastic properties of composites in a simplified form, and thereby also introduce the main concepts to be used later. In Section 3 we use our formalism to obtain some of the simple known bounds, and we also extend the range of validity of the Hashin–Shtrikman bounds to include composites with an arbitrary (i.e. not necessarily isotropic or cubic) microgeometry and without any information about the microgeometric correlation functions. A more general and also more flexible formalism is introduced in Section 4. This is applied in Section 5 to derive improved bounds that require information of a new type about the composite. This information is in the form of known values of the effective elastic constants for a composite with the same microgeometry but different constituents. Such information can be obtained either by measurement or by another calculation. It is clearly information of a "physical" nature about the composite, as opposed to the purely geometrical information that is contained in the correlation functions which are also sometimes used to obtain improved bounds.

## 2. THE GENERAL THEORY

The position dependent local elastic stiffness tensor $C(\mathbf{r})$ of a two-component composite made of homogeneous materials with stiffness tensors $C^{(1)}$ and $C^{(2)}$ can be written with the help of a step function $\theta_1$:

$$C(\mathbf{r}) = C^{(2)} + \theta_1(\mathbf{r})(C^{(1)} - C^{(2)}) \equiv C^{(2)} + \theta_1 \delta C, \tag{2.1}$$

where

$$\theta_1(\mathbf{r}) \equiv \begin{cases} 1, & \mathbf{r} \text{ inside } C^{(1)} \text{ material}, \\ 0, & \mathbf{r} \text{ outside } C^{(1)} \text{ material}. \end{cases} \tag{2.2}$$

In (2.1), and often also in subsequent discussions, we have suppressed the tensorial indices.

The effective elastic stiffness tensor $C^{(e)}$ is usually defined by means of the volume averages of the stress tensor $\sigma$ and the strain tensor $\varepsilon$ in the inhomogeneous sample, so that

$$\langle \sigma \rangle_{av} = C^{(e)} \langle \varepsilon \rangle_{av}, \tag{2.3}$$

where $\langle\ \rangle_{av}$ denotes a volume average. Alternatively, $C^{(e)}$ may be defined by requiring that the elastic energy density that would exist in a homogeneous material with stiffness tensor $C^{(e)}$ be equal to the volume averaged energy density in the actual inhomogeneous sample when it is subjected to the same boundary conditions on the displacement vector **u**. We will use the boundary conditions (see HASHIN, 1970, pp. 44–47)

$$u_i = \varepsilon^0_{ij} x_j, \quad \text{for } \mathbf{r} = (x_1, x_2, x_3) \text{ on the boundary,} \tag{2.4}$$

where $\varepsilon^0_{ij}$ is some constant symmetric tensor (i.e. $\varepsilon^0$ has the same value over the entire boundary). Here and subsequently we use the Einstein summation convention on repeated tensorial indices. These boundary conditions would cause the strain $\varepsilon$ to be a uniform constant $\varepsilon = \varepsilon^0$ in the entire volume of a homogeneous material, while in the case of an inhomogeneous material only the volume average of the position dependent $\varepsilon(\mathbf{r})$ would be equal to $\varepsilon^0$. Thus the alternative definition of $C^{(e)}$ under the boundary conditions of (2.4) is

$$\varepsilon^0 C^{(e)} \varepsilon^0 = \langle \varepsilon C(\mathbf{r}) \varepsilon \rangle_{av} \tag{2.5}$$

for any constant symmetric tensor $\varepsilon^0$. For these boundary conditions, it can be shown that the two definitions of $C^{(e)}$, namely, (2.3) and (2.5), coincide.

We now introduce a somewhat generalized form of (2.1), allowing $C$ to depend on a continuous parameter $s$:

$$C(\mathbf{r}; s) = C^{(2)} + \frac{1}{s}\theta_1(\mathbf{r})\delta C. \tag{2.6}$$

By allowing $s$ to take arbitrary values, we are actually replacing the $C^{(1)}$ material by a different material $C^{(1)'}$, where

$$C^{(1)'} = C^{(2)} + \frac{1}{s}\delta C = \frac{1}{s}C^{(1)} + \frac{s-1}{s}C^{(2)}. \tag{2.7}$$

This replacement also makes $C^{(e)}$ a function of $s$. We note that when $s$ lies in certain ranges, the tensor $C^{(1)'}$ becomes *unphysical*, i.e. it ceases to be positive definite. From (2.5) and (2.6) we can obtain the expression for $C^{(e)}$ as (cf. I)

$$\varepsilon^0 C^{(e)} \varepsilon^0 - \varepsilon^0 C^{(2)} \varepsilon^0 = \frac{1}{sV} \int \theta_1 \varepsilon^0 \delta C \varepsilon \, \mathrm{d}V \equiv F(s). \tag{2.8}$$

In order to simplify the notation, we now introduce two definitions: For any tensor $\varepsilon$ we define a complementary tensor $\tilde{\varepsilon}$,

$$\tilde{\varepsilon} \equiv (\varepsilon \delta C)^*, \tag{2.9}$$

where the asterisk denotes complex conjugation, and we also define a scalar product between two arbitrary tensors

$$\langle \varepsilon \,|\, \varepsilon' \rangle \equiv \int \theta_1(\mathbf{r}) \varepsilon^*_{ij}(\mathbf{r}) \varepsilon'_{ij}(\mathbf{r}) \, \mathrm{d}V. \tag{2.10}$$

We can now rewrite (2.8) in the form

$$F(s) = \frac{1}{sV}\langle \tilde{\varepsilon}^0 | \varepsilon \rangle. \tag{2.11}$$

The use of non-real (i.e. complex) tensors $\varepsilon, \tilde{\varepsilon}$ is mandatory only if $s$ or $C$ are complex. Otherwise we can always restrict ourselves to real $\varepsilon, \tilde{\varepsilon}$. However, even then it is sometimes convenient to allow complex $\varepsilon, \tilde{\varepsilon}$.

The strain tensor $\varepsilon(\mathbf{r})$ in a composite material, the boundaries of which undergo the displacement (2.4), is the solution of the linear integral equation (see, e.g. WU and MCCULLOUGH (1977))

$$\varepsilon_{ij}(\mathbf{r}) = \varepsilon^0_{ij} + \frac{1}{s}\int \theta_1(\mathbf{r}') G_{ijkl}(\mathbf{r}, \mathbf{r}'; C^{(2)})' \delta C_{klmn} \varepsilon_{mn}(\mathbf{r}')\, \mathrm{d}V'. \tag{2.12}$$

Here $G$ is the tensor Green's function of the problem, which depends on $C^{(2)}$ as well as on the shape of the sample, and has the symmetries

$$G_{ijkl}(\mathbf{r}, \mathbf{r}') = G_{ijlk}(\mathbf{r}, \mathbf{r}') = G_{jikl}(\mathbf{r}, \mathbf{r}') = G_{klij}(\mathbf{r}', \mathbf{r}). \tag{2.13}$$

The integral equation (2.12) can be written in a more concise bra- and -ket notation

$$|\varepsilon\rangle = |\varepsilon^0\rangle + \frac{1}{s}\hat{G}|\varepsilon\rangle. \tag{2.14}$$

We can formally solve this equation, and substitute that solution in (2.11) to obtain

$$F(s) = \frac{1}{V}\left\langle \tilde{\varepsilon}^0 \left| \frac{1}{s - \hat{G}} \right| \varepsilon^0 \right\rangle \tag{2.15}$$

In order to make further progress, we introduce the eigenstates of the operator $\hat{G}$:

$$\hat{G}|\varepsilon^{(\alpha)}\rangle = s_\alpha |\varepsilon^{(\alpha)}\rangle. \tag{2.16}$$

Since $\hat{G}$ is a non-hermitian operator, its right eigenstates differ from its left eigenstates. However, from (2.13) and the definition of $\hat{G}$ we can easily see, that the complementary tensor $\tilde{\varepsilon}^{(\alpha)}$ of a right eigenstate $\varepsilon^{(\alpha)}$ is a left eigenstate of $\hat{G}$ with the same eigenvalue. In I we also proved that all the eigenvalues of $\hat{G}$ are real and lie in the interval $(-\infty, 1)$. Moreover we showed that

$$s_\alpha \leqslant 0, \quad \text{for } \delta C \geqslant 0, \tag{2.17a}$$

$$0 \leqslant s_\alpha < 1, \quad \text{for } \delta C \leqslant 0, \tag{2.17b}$$

where $\delta C \geqslant 0$ ($\delta C \leqslant 0$) means that $\delta C$ is a positive (negative) semi-definite tensor.†

By comparing (2.16) with (2.14) we can identify the physical significance of these eigenstates. They are elastostatic resonances of the sample, i.e. states where the sample

† The question of whether the eigenvalues $s$ are a discrete set has been discussed extensively in the context of electrostatic properties of composites (see, e.g. BERGMAN, 1983; PAPANICOLAOU, 1983a, b). A similarly general discussion of this question for elastic properties does not exist, as far as we know, though a particular case has been treated by KUPRADZE (1965) (see also I). In any case, this question is not of crucial importance for the practical derivation of bounds.

is internally deformed and strained although the boundaries are undeformed. Obviously, such resonances can occur only at values of $s = s_\alpha$ such that $C^{(1)'}$ of (2.7) is unphysical. The right and left eigenstates of $\hat{G}$ form a hopefully complete bi-orthogonal set of states (see, e.g. MORSE and FESHBACH, 1953) which can be used to expand the identity operator in the usual way

$$I = \sum_\alpha |\varepsilon^{(\alpha)}\rangle \langle \tilde{\varepsilon}^{(\alpha)}|. \tag{2.18}$$

Using this expansion, we can bring (2.15) to the form

$$F(s) = \sum_\alpha \frac{F_\alpha}{s - s_\alpha}, \tag{2.19}$$

where

$$F_\alpha \equiv \frac{1}{V} \langle \tilde{\varepsilon}^0 | \varepsilon^{(\alpha)} \rangle \langle \tilde{\varepsilon}^{(\alpha)} | \varepsilon^0 \rangle = \frac{1}{V} (\langle \tilde{\varepsilon}^0 | \varepsilon^{(\alpha)} \rangle)^2. \tag{2.20}$$

In I we showed that $F_\alpha$ is real and satisfies

$$F_\alpha \cdot s_\alpha \leqslant 0. \tag{2.21}$$

Some examples of explicit calculation of elastostatic resonances and their application to a systematic evaluation of $F_\alpha$ and $s_\alpha$ (and consequently $C^{(e)}$) for some *known* microgeometries can be found in I and in KANTOR and BERGMAN (1982b). For an *unknown* microgeometry, we will treat the poles $s_\alpha$ and their weights $F_\alpha$ in (2.19) as free parameters subject to certain constraints, two examples of which are the inequalities of (2.17) and (2.21).

## 3. SUM RULES AND SIMPLE BOUNDS

Besides the inequalities mentioned in the previous section, the parameters $F_\alpha$, $s_\alpha$ appearing in (2.19) must satisfy certain (moment) sum rules. Those may be obtained by expanding two different representations for $F(s)$, namely (2.15) and (2.19), in powers of $1/s$. Equating the expansions order by order we thus obtain the following expression for the $n$th moment of the pole spectrum

$$Q_n \equiv \sum_\alpha F_\alpha \cdot s_\alpha^n = \frac{1}{V} \langle \tilde{\varepsilon}^0 | \hat{G}^n | \varepsilon^0 \rangle. \tag{3.1}$$

In general $Q_n$ is a scalar quantity whose value depends on the choice of $\varepsilon^0$ as well as on the detailed microgeometry. However, knowledge of the volume fraction $p_1$ of the $C^{(1)}$ material suffices for the calculation of the zero moment sum rule

$$Q_0 \equiv \sum_\alpha F_\alpha = \frac{1}{V} \langle \tilde{\varepsilon}^0 | \varepsilon^0 \rangle = \frac{1}{V} \int \theta_1 \varepsilon^0 \delta C \varepsilon^0 \, \mathrm{d}V = p_1 \varepsilon^0 \delta C \varepsilon^0. \tag{3.2}$$

A knowledge of $Q_0$ is sufficient for the derivation of a simple bound on $C^{(e)}$ as follows: From the inequality (2.21) and the fact that the poles $s_\alpha$ satisfy $s_\alpha < 1$,

$$F(1) = \sum_\alpha F_\alpha \left(1 + \frac{s_\alpha}{1 - s_\alpha}\right) \leqslant \sum_\alpha F_\alpha = Q_0. \tag{3.3}$$

In the usual notation we thus get

$$\varepsilon^0 C^{(e)} \varepsilon^0 \leqslant \varepsilon^0 C^{(2)} \varepsilon^0 + p_1 \varepsilon^0 \delta C \varepsilon^0 = \varepsilon^0 \langle C \rangle_{av} \varepsilon^0, \tag{3.4}$$

which is the well known Voigt bound (HILL, 1963).

The Reuss bound (HILL, 1963), which is complementary to Voigt's bound, can be derived from a similar formalism. The main difference in derivations is that instead of parametrising the local elastic stiffness tensor with the variable $s$ (see (2.6)), we now parametrise the elastic compliance tensor of the material in a similar fashion. A detailed derivation of the Reuss bound along these lines will be given elsewhere.

Knowledge of higher order moments $Q_n$ ($n \geqslant 1$) would enable us to derive more restrictive bounds. From (2.9), (2.10), (3.1) and the definition of $\hat{G}$ we find that

$$Q_1 \equiv \sum_\alpha F_\alpha \cdot s_\alpha = \frac{1}{V} \langle \tilde{\varepsilon}^0 | \hat{G} | \varepsilon^0 \rangle = \frac{1}{V} \int \theta_1(\mathbf{r}) \theta_1(\mathbf{r}') \varepsilon^0 \delta C G(\mathbf{r}, \mathbf{r}') \delta C \varepsilon^0 \, \mathrm{d}V \, \mathrm{d}V'. \tag{3.5}$$

In general, the evaluation of $Q_1$ requires knowledge of the volume average of the two-point correlation function of the microgeometry $\theta_1(\mathbf{r}) \cdot \theta_1(\mathbf{r}')$. However, in some cases, $Q_1$ can be evaluated with less detailed information.

Once $Q_1$ is known, we can use it as an additional constraint on $F_\alpha$ and $s_\alpha$ and the bounds on $F(1)$ can be found by varying the parameters in (2.19) subject to that constraint. However, these bounds can also be obtained more easily by examining the function

$$D(s) \equiv \frac{1}{Q_0} - \frac{1}{sF(s)}, \tag{3.6}$$

which has a structure similar to that of $F(s)$ and can also be written as a sum of simple poles

$$D(s) = \sum_\alpha \frac{D_\alpha}{s - \tilde{s}_\alpha}. \tag{3.7}$$

The poles $\tilde{s}_\alpha$ are the zeroes of $F(s)$, and there is also a pole at $s = 0$, unless $F(s)$ has a pole at that point. The zero moment sum rule for $F(s)$ has been incorporated in the definition of $D(s)$ and is responsible for the fact that $D \to 0$ as $s \to \infty$. The first moment sum rule of $F(s)$ now determines the zero moment of $D(s)$, since by expanding (3.6) and (3.7) in powers of $1/s$ we obtain

$$\sum_\alpha D_\alpha = \frac{Q_1}{Q_0^2}. \tag{3.8}$$

Similarly, higher order moments of $D(s)$ can be related to the higher moments of $F(s)$. It can easily be shown that all residues $D_\alpha$ in (3.7) are negative:

$$\frac{1}{D_\alpha} = -\frac{\mathrm{d}}{\mathrm{d}s} [sF(s)]_{s=\tilde{s}_\alpha} = \sum_\beta \frac{F_\beta s_\beta}{(\tilde{s}_\alpha - s_\beta)^2} < 0, \tag{3.9}$$

where we used the inequality (2.21). The poles $\tilde{s}_\alpha$ of $D(s)$ are subject to the same restrictions as the poles of $F(s)$ (see (2.17)).

The bounds on $D(1)$ can now easily be found. For $\delta C \geqslant 0$, we obtain

$$D(1) = \sum_{\alpha} \frac{D_\alpha}{1-\tilde{s}_\alpha} \geqslant \sum_{\alpha} D_\alpha = \frac{Q_1}{Q_0^2} \tag{3.10}$$

or, returning to the function $F$,

$$F(1) \geqslant Q_0^2/(Q_0 - Q_1). \tag{3.11}$$

In order to translate these inequalities into an explicit lower bound for $C^{(e)}$ we must be able to evaluate $Q_1$ explicitly. This can be done for any choice of $\varepsilon^0$ in the case of a composite with isotropic microgeometry (whose components must also be isotropic), leading to the well known Hashin–Shtrikman lower bound (HASHIN and SHTRIKMAN (1963), HILL (1963), WALPOLE (1966a, b)). The Hashin–Shtrikman upper bound can be obtained by a similar procedure, in which the roles of $C^{(1)}$ and $C^{(2)}$ are interchanged. In that case one is lead to a different definition of $F(s)$, and because now $\delta C \leqslant 0$, the inequalities corresponding to (3.10) and (3.11) are reversed.

Explicit bounds for composites with a microgeometry of lower symmetry (but still made of isotropic components) were occasionally also derived, e.g. by HILL (1963) for the bulk modulus in the case of cubic symmetry, and by WILLIS (1977) for all the elastic moduli in the case of ellipsodial symmetry. If the two-point correlation function is known then $Q_1$ can always be evaluated from (3.5) and explicit bounds can be found. For that case, a pair of bounds was derived by WILLIS (1982), which reduce to the Hashin–Shtrikman bounds in the case of an isotropic composite. We will show below that $Q_1$ can in fact be evaluated explicitly in a whole new class of cases, *without the need to know the two-point correlation function.* This leads to new explicit bounds on elastic moduli for a variety of microgeometries with symmetries lower than isotropic.

The Hashin-Shtrikman bounds are valid only in the case of (positive or negative) semi-definite $\delta C$. When this is not the case, then the less stringent Walpole bounds (WALPOLE, 1966a, b) can be derived by a small modification of the above formalism. However, these bounds are more easily obtained from the more general formalism which will be presented in the next Section.

The function $F(s)$, as well as the moments $Q_n$, depend on our choice of $\varepsilon^0$. Thus we can isolate different parts of $C^{(e)}$ by a proper choice of $\varepsilon^0$ (see (2.8)). Thus, the choice

$$\varepsilon_{ij}^0 = \varepsilon_{ij}^{0\kappa} \equiv \frac{1}{d}\delta_{ij}, \tag{3.12}$$

where $d$ is the dimensionality of the system, selects the bulk modulus

$$\kappa^{(e)} \equiv \varepsilon^{0\kappa} C^{(e)} \varepsilon^{0\kappa} = \frac{1}{d^2} C_{iikk}^{(e)}. \tag{3.13}$$

In this context we should point out that $d = 2$ does not mean a really two-dimensional (2D) material but rather a composite in the form of parallel fibers. In such a material $\kappa^{(e)}$ is taken to denote the *transverse* bulk modulus while $C^{(e)}$ is taken to denote the *transverse* stiffness tensor. For the choice (3.12) of $\varepsilon^0$ we find from (3.2) that

$$Q_0^{(\kappa)} = p_1 \delta\kappa, \tag{3.14}$$

where the superscript $\kappa$ relates to the choice $\varepsilon^0 = \varepsilon^{0\kappa}$, and where obviously $\delta\kappa = \kappa^{(1)} - \kappa^{(2)}$. The evaluation of $Q_1$ for an arbitrary choice $\varepsilon^0$ would require, in general, a detailed knowledge of the two-point correlation function. However, if we restrict ourselves to mixtures of isotropic components, then $Q_1^{(\kappa)}$ *does not depend on the microgeometry of the composite.* The stiffness tensor of the isotropic $n$th component of the composite can be written in the form

$$C_{ijkl}^{(n)} = \kappa^{(n)}\delta_{ij}\delta_{kl} + 2\mu^{(n)}\left(I_{ijkl} - \frac{1}{d}\delta_{ij}\delta_{kl}\right) \equiv C^{(n\kappa)} + C^{(n\mu)}, \tag{3.15}$$

(for the time being, $n = 1, 2$) where $\kappa^{(n)}$, $\mu^{(n)}$ are the bulk and shear moduli (the transverse bulk and shear moduli in 2D) of that component, and

$$I_{ijkl} = \tfrac{1}{2}(\delta_{ik}\delta_{jl} + \delta_{jk}\delta_{il}) \tag{3.16}$$

is the symmetric unit tensor of rank four.

In Appendix A we show that

$$Q_1^{(\kappa)} = -p_1(1-p_1)\delta\kappa^2 E^{(\kappa)}(C^{(2)}), \tag{3.17a}$$

$$E^{(\kappa)}(C^{(2)}) \equiv 1\Big/\left(\kappa^{(2)} + 2\frac{d-1}{d}\mu^{(2)}\right) \tag{3.17b}$$

for *any* mixture of *isotropic* components. When the sum rules of (3.14) and (3.17) are used in (3.11), and in the analogous inequality that arises when $C^{(1)}$ and $C^{(2)}$ are interchanged, the bounds that are obtained for $\kappa^{(e)}$

$$\frac{p_1}{1+(1-p_1)(\kappa^{(1)}-\kappa^{(2)})\Big/\left(\kappa^{(2)}+2\frac{d-1}{d}\mu^{(2)}\right)} \leqslant \frac{\kappa^{(e)}-\kappa^{(2)}}{\kappa^{(1)}-\kappa^{(2)}}$$

$$\leqslant \frac{p_1}{1+(1-p_1)(\kappa^{(1)}-\kappa^{(2)})\Big/\left(\kappa^{(1)}+2\frac{d-1}{d}\mu^{(1)}\right)} \tag{3.18}$$

have the form of the Hashin–Shtrikman bounds. However, by our method of derivation, it is clear that they really apply *irrespective of the symmetry of the microgeometry.* As far as we know, this fact has not been recognized before (see, e.g. HILL, 1963; WALPOLE, 1966a, b, 1969).

A different choice of $\varepsilon^0$, namely

$$\varepsilon_{ij}^0 = \varepsilon_{ij}^{0\mu} \equiv I_{ij12}, \tag{3.19}$$

will lead us to the following expressions for the moment sum rules

$$Q_0^{(\mu)} = p_1\delta\mu, \tag{3.20}$$

$$Q_1^{(\mu)} = -p_1(1-p_1)\delta\mu^2 E^{(\mu)}(C^{(2)}), \tag{3.21a}$$

$$E^{(\mu)}(C^{(2)}) \equiv 2(\kappa^{(2)}+2\mu^{(2)})\Big/\left[(d+2)\mu^{(2)}\left(\kappa^{(2)}+2\frac{d-1}{d}\mu^{(2)}\right)\right]. \tag{3.21b}$$

The expression (3.21) is valid only for an *isotropic* mixture of *isotropic* components (see Appendix A). Thus the bounds for $\mu^{(e)}$ of such a composite are found to be

$$\frac{p_1}{1+2(1-p_1)(\mu^{(1)}-\mu^{(2)})(\kappa^{(2)}+2\mu^{(2)})\Big/\left[(d+2)\mu^{(2)}\left(\kappa^{(2)}+2\frac{d-1}{d}\mu^{(2)}\right)\right]} \leqslant \frac{\mu^{(e)}-\mu^{(2)}}{\mu^{(1)}-\mu^{(2)}}$$

$$\leqslant \frac{p_1}{1+2(1-p_1)(\mu^{(1)}-\mu^{(2)})(\kappa^{(1)}+2\mu^{(1)})\Big/\left[(d+2)\mu^{(1)}\left(\kappa^{(1)}+2\frac{d-1}{d}\mu^{(1)}\right)\right]}, \tag{3.22}$$

which are the usual Hashin–Shtrikman bounds. However, by an appropriate redefinition of $\mu^{(e)}$, the validity of these bounds can also be extended to arbitrary microgeometries. For example, if the microgeometry of the composite has cubic (square in 2D) symmetry, then its effective stiffness tensor has the form

$$C^{(e)}_{ijkl} = \kappa^{(e)}\delta_{ij}\delta_{kl} + 2\mu^{(e)}(I_{ijkl}-\delta_{ijkl}) + 2M^{(e)}\left(\delta_{ijkl}-\frac{1}{d}\delta_{ij}\delta_{kl}\right) \equiv C^{(e\kappa)} + C^{(e\mu)} + C^{(eM)}, \tag{3.23}$$

where $\mu^{(e)}$ and $M^{(e)}$ are two (different) shear moduli, which coincide in the isotropic case, and

$$\delta_{ijkl} = \begin{cases} 1, & \text{for } i=j=k=l, \\ 0, & \text{otherwise.} \end{cases} \tag{3.24}$$

In this case two different first moments can be defined: $Q_1^{(\mu)}$ for $\varepsilon^0 = \varepsilon^{0\mu}$ (see (3.19)) and $Q_1^{(M)}$ for the choice

$$\varepsilon^0 = \varepsilon^{0M} \equiv \tfrac{1}{2}(I_{ij11} - I_{ij22}). \tag{3.25}$$

In Appendix A we show that for such a mixture of *isotropic* materials $Q_1^{(\mu)}$ in (3.21a) should be replaced by linear combinations of $Q_1^{(\mu)}$ and $Q_1^{(M)}$, i.e.

$$(Q_1^{(\mu)} + Q_1^{(M)})/2 = -p_1(1-p_1)\delta\mu^2 E^{(\mu)}(C^{(2)}), \quad \text{in 2D}, \tag{3.26}$$

$$(3Q_1^{(\mu)} + 2Q_1^{(M)})/5 = -p_1(1-p_1)\delta\mu^2 E^{(\mu)}(C^{(2)}), \quad \text{in 3D}. \tag{3.27}$$

Thus, the bounds (3.22) remain valid if we replace $\mu^{(e)}$ by the linear combination $(\mu^{(e)} + M^{(e)})/2$ in 2D, or by $(3\mu^{(e)} + 2M^{(e)})/5$ in 3D.

In Appendix A we show that (3.26) remains valid even for 2D composites with an arbitrary (i.e. lower than square) symmetry. Consequently, the bounds obtained from (3.22) by the replacement $\mu^{(e)} \to (\mu^{(e)} + M^{(e)})/2$ in 2D remain valid irrespective of the symmetry. We also show there that in a 3D composite with arbitrary (i.e. lower than cubic) symmetry, a sum rule similar to (3.21a) holds for a certain weighted average of six different $Q_1$'s. Consequently, in 3D composites of arbitrary symmetry a pair of bounds is shown to hold whose form is obtained from (3.22) by replacing $\mu^{(e)}$ by the similarly weighted average of six different shear moduli.

From all that we have said above, it is clear that while the forms of the bounds (3.18) and (3.22) are not new, we have succeeded in extending their ranges of validity considerably. These bounds were originally discovered and derived for an isotropic

mixture of isotropic materials, using different approaches, by HASHIN and SHTRIKMAN (1963) and by HILL (1963) for 3D, and by HASHIN (1965) for 2D.

Finally we would like to mention that when higher order moments $Q_n$ ($n \geqslant 2$) are known the entire procedure can be repeated. We can define a new function of $s$ which is obtained from $D(s)$ in the same way that $D(s)$ was obtained from $F(s)$ in (3.6), and which thus has the same pole structure. The bounds arising from that function, incorporating the additional information, will naturally be more stringent than the Hashin–Shtrikman bounds.

## 4. EXTENSION OF THE THEORY

In order to improve the Hashin–Shtrikman-type bounds it is usually necessary to have more detailed information on the microstructure of the composite. One way to obtain such information is through a knowledge of the corresponding effective elastic constant of *another* composite, which is made of different components but has the same or a sufficiently similar microgeometry (e.g. both composites are produced by the same technological process). If an elastic constant of both composites is represented by *the same function* $F(s)$, taken at different values of the argument $s$, say $s = 1$ and $s = s_+$, then knowledge of the effective elastic constant of the $s_+$ material can be expressed in the form

$$F(s_+) = \sum_\alpha \frac{F_\alpha}{s_+ - s_\alpha}, \tag{4.1}$$

which becomes an additional constraint in the derivation of bounds on $F(1)$.

However, the form (2.6) is not flexible enough for such treatment. The argument $s$ does not affect the $C^{(2)}$ tensor and restricts the values of the $C^{(1)'}$ tensor to lie on a straight line in the space of elastic constants (see (2.7)). We shall, therefore, replace (2.6) by a more general form. We will also extend the formalism to handle composites made of more than two components.

We will represent the microgeometry of a multi-component composite, by a set of step functions $\{\theta_n\}$, where $\theta_n$ defines the microgeometry of the $n$th component as in (2.2). Sometimes we will find it convenient to split $C^{(n)}$ into a sum of several symmetric positive definite tensors

$$C^{(n)} = \sum_\gamma C^{(n\gamma)}, \tag{4.2}$$

as we did in the isotropic case in (3.15) and in the cubic case in (3.23), where $\gamma$ took the values $\kappa$, $\mu$, $M$. We again define a new stiffness tensor, that depends on the continuous parameter $s$

$$C(\mathbf{r}; s) = C^{(0)} + \sum_{n\gamma} v_{n\gamma}\theta_n(\mathbf{r})(C^{(n\gamma)} - C^{(0\gamma)}) \equiv C^{(0)} + \sum_{n\gamma} v_{n\gamma}\theta_n \delta C^{(n\gamma)}, \tag{4.3}$$

$$v_{n\gamma} \equiv 1/[(s-1)/b_{n\gamma} + 1], \tag{4.4}$$

where $C^{(0)}$ is an arbitrary (symmetric positive definite) tensor, and $b_{n\gamma}$ are constants. For $s = 1$, also $v_{n\gamma} = 1$ and this expression reduces to the actual $C(\mathbf{r})$. However, when $s$

varies the stiffness tensor in each component moves along a line in the space of elastic constants which, in general, will not be straight. The effective elastic constants now have an analytic structure as functions of $s$ which is similar to (2.19), namely

$$F(s) \equiv \varepsilon^0(C^{(e)} - C^{(0)})\varepsilon^0 = \sum_\alpha \frac{F_\alpha}{s - s_\alpha}. \tag{4.5}$$

However, as things stand now, both $s_\alpha$ and $F_\alpha$ will, in general, be complex numbers. In order to facilitate their manipulation, we therefore restrict the choice of $C^{(0)}$ so that *all* $\delta C^{(n\gamma)} \geqslant 0$ (or *all* $\delta C^{(n\gamma)} \leqslant 0$) and we restrict the constants $b_{n\gamma}$ to be real numbers satisfying $b_{n\gamma} \geqslant 1$ ($0 < b_{n\gamma} \leqslant 1$). In Appendix B we prove that, under these restrictions, all $F_\alpha$ and $s_\alpha$ are real numbers and that

$$s_\alpha \leqslant 0, \quad \text{when } \textit{all } \delta C^{(n\gamma)} \geqslant 0, \tag{4.6a}$$

$$0 \leqslant s_\alpha < 1, \quad \text{when } \textit{all } \delta C^{(n\gamma)} \leqslant 0, \tag{4.6b}$$

$$F_\alpha \cdot s_\alpha \leqslant 0, \quad \text{in both cases.} \tag{4.6c}$$

In Appendix B we also give explicit expressions for the moments $Q_0$ and $Q_1$ of $F(s)$ for the representation of (4.3) in the most general case. We also show that in the case of an isotropic mixture of isotropic components and for the choice of boundary conditions $\varepsilon^0 = \varepsilon^{0\gamma}$ ($\gamma = \kappa$ or $\mu$) we obtain

$$Q_0^{(\gamma)} = \sum_n p_n b_{n\gamma} \delta\gamma^{(n)}, \tag{4.7}$$

$$Q_1^{(\gamma)} = -\sum_{mn} p_m(\delta_{mn} - p_n)\delta\gamma^{(m)}\delta\gamma^{(n)}E^{(\gamma)}(C^{(0)})b_{n\gamma}b_{m\gamma} - \sum_n p_n \delta\gamma^{(n)} b_{n\gamma}(1 - b_{n\gamma}), \tag{4.8}$$

where $\delta\gamma^{(n)} = \gamma^{(n)} - \gamma^{(0)}$, $p_n$ is the volume fraction of the $n$th component and $E^{(\gamma)}(C^{(0)})$ is defined by (3.17b) or (3.21b) with $(\kappa^{(2)}, \mu^{(2)})$ replaced by $(\kappa^{(0)}, \mu^{(0)})$.

Using this more general formalism, we are now able to derive Walpole's bounds, i.e. we can treat the case of non-definite $C^{(1)} - C^{(2)}$, by a suitable choice of $C^{(0)}$ and $b_{n\gamma}$. In the isotropic case, for instance, we choose $(\kappa^{(0)}, \mu^{(0)}) = (\min(\kappa^{(1)}, \kappa^{(2)}), \min(\mu^{(1)}, \mu^{(2)}))$ or $(\kappa^{(0)}, \mu^{(0)}) = (\max(\kappa^{(1)}, \kappa^{(2)}), \max(\mu^{(1)}, \mu^{(2)}))$ and all $b_{n\gamma} = 1$. For a such a choice all $\delta C^{(n\gamma)} \geqslant 0$ (all $\delta C^{(n\gamma)} \leqslant 0$) and we can derive the bounds for $F(1)$ in the same way as in the previous Section. The resulting bounds are similar to (3.18) and (3.22) but with $(\kappa^{(1)}, \mu^{(1)})$ replaced by $(\max(\kappa^{(1)}, \kappa^{(2)}), \max(\mu^{(1)}, \mu^{(2)}))$ and $(\kappa^{(2)}, \mu^{(2)})$ replaced by $(\min(\kappa^{(1)}, \kappa^{(2)}), \min(\mu^{(1)}, \mu^{(2)}))$. These bounds can be extended to cases of non-isotropic mixtures of isotropic materials, as was done in the previous Section.

In principle, this approach can be implemented even for a composite whose components are non-isotropic: We must choose $C^{(0)}$ to be as large as possible (as small as possible) under the restrictions $\delta C^{(1)} \geqslant 0$, $\delta C^{(2)} \geqslant 0$ ($\delta C^{(1)} \leqslant 0$, $\delta C^{(2)} \leqslant 0$) and we must take all $b_{n\gamma} = 1$. The bounds on $C^{(e)}$ are again obtained from (3.11) (and its complementary inequality) in terms of $Q_0$ and $Q_1$. The main problem is of course, how to calculate $Q_1$. Unfortunately, there is no prescription for such a calculation in the general case other than (3.5).

Another advantage of the parametric representation (4.3), (4.4) is that it enables us to represent different composites (but having the same microgeometry) by the same function $F(s)$ at different values of $s$, as was explained earlier. As an example we shall

consider two mixtures of isotropic components. The first one is composed of $C^{(1)} = (\kappa^{(1)}, \mu^{(1)})$ and $C^{(2)} = (\kappa^{(2)}, \mu^{(2)})$, and the second one is composed of $C_+^{(1)} = (\kappa_+^{(1)}, \mu_+^{(1)})$ and $C_+^{(2)} = C^{(2)}$. We will restrict ourselves to the case $C^{(1)} - C^{(2)} \geqslant 0$. We start by choosing $C^{(0)} = C^{(2)}$ in (4.3) and requiring that for some value $s = s_+ \neq 1$ the $C^{(1)}$ material be replaced by $C_+^{(1)}$. Thus the parameters in (4.3) must be chosen so as to satisfy

$$\kappa_+^{(1)} = \kappa^{(2)} + (\kappa^{(1)} - \kappa^{(2)})/[(s_+ - 1)/b_{1\kappa} + 1], \tag{4.9a}$$

$$\mu_+^{(1)} = \mu^{(2)} + (\mu^{(1)} - \mu^{(2)})/[(s_+ - 1)/b_{1\mu} + 1]. \tag{4.9b}$$

If we treat $s_+$ as given, then the $b$'s in (4.9) are determined by

$$b_{1\kappa} = (\kappa_+^{(1)} - \kappa^{(2)})(s_+ - 1)/(\kappa^{(1)} - \kappa^{(2)}), \tag{4.10a}$$

$$b_{1\mu} = (\mu_+^{(1)} - \mu^{(2)})(s_+ - 1)/(\mu^{(1)} - \mu^{(2)}). \tag{4.10b}$$

Since both $b_{1\kappa}$ and $b_{1\mu}$ must be greater than 1 in this case, these equations can be used only when the following inequalities are simultaneously valid

$$(\kappa_+^{(1)} - \kappa^{(1)})(\mu_+^{(1)} - \mu^{(1)}) \geqslant 0, \tag{4.11}$$

$$\kappa_+^{(1)} > \kappa^{(2)}, \quad \mu_+^{(1)} > \mu^{(2)}, \tag{4.12}$$

$$s_+ \geqslant 1 + \max\left(\frac{\kappa^{(1)} - \kappa_+^{(1)}}{\kappa_+^{(1)} - \kappa^{(2)}}, \frac{\mu^{(1)} - \mu_+^{(1)}}{\mu_+^{(1)} - \mu^{(2)}}\right), \quad \text{for } \kappa^{(1)} - \kappa_+^{(1)} > 0, \tag{4.13a}$$

$$0 \leqslant s_+ \leqslant 1 + \min\left(\frac{\kappa^{(1)} - \kappa_+^{(1)}}{\kappa_+^{(1)} - \kappa^{(2)}}, \frac{\mu^{(1)} - \mu_+^{(1)}}{\mu_+^{(1)} - \mu^{(2)}}\right), \quad \text{for } \kappa^{(1)} - \kappa_+^{(1)} < 0. \tag{4.13b}$$

Inequalities (4.11) and (4.12) limit the range of applicability of this particular implementation of the formalism. Different choices of $C^{(0)}$ (e.g. $C^{(0)} = C^{(1)}$) allow us to circumvent the limitations arising from (4.12) (or a similar pair of inequalities), but an inequality similar to (4.11) will always appear. Thus there will be pairs of composites for which representation of an elastic modulus by a single function $F(s)$ is impossible. Nevertheless, improved bounds can be derived even in those cases by a two-stage procedure which will be explained in the next Section.

Finally we would like to stress that suitable choices of $C^{(0)}$ enable us to treat very general cases, including the case of non-definite $C^{(1)} - C^{(2)}$.

## 5. Improved bounds from additional information

In this Section we shall exploit the additional constraint (4.1) to derive improved bounds on the effective elastic constants. We shall discuss only the case when all the $\delta C^{(n\gamma)}$ appearing in (4.3) are positive semi-definite. In that case all the poles $s_\alpha \leqslant 0$ and their residues $F_\alpha > 0$. The case of all $\delta C^{(n\gamma)} \leqslant 0$ in (4.3) is completely analogous.

Although the entire calculation can be performed using the function $F(s)$, it is convenient to introduce a new related function

$$H(s) \equiv F(s)/(F(s) + \varepsilon^0 C^{(0)} \varepsilon^0) = 1 - \varepsilon^0 C^{(0)} \varepsilon^0 / \varepsilon^0 C^{(e)} \varepsilon^0. \tag{5.1}$$

Clearly, the function $H(s)$ includes the same information about the system as does $F(s)$.

Furthermore, we will now show that it also has a similar analytic structure, namely,

$$H(s) = \sum_{\alpha} \frac{H_\alpha}{s - h_\alpha}, \tag{5.2}$$

where the poles $h_\alpha$ and the residues $H_\alpha$ are all real and satisfy

$$s' \leqslant h_\alpha \leqslant 0, \qquad H_\alpha > 0. \tag{5.3}$$

These poles and residues also satisfy certain sum rules. In order to demonstrate these properties, we note first that the poles $h_\alpha$ must be zeros of the denominator $F(s) + \varepsilon^0 C^{(0)} \varepsilon^0$ and that, due to the structure of $F(s)$, these zeros must lie on the negative real axis. At these zeros, both $F(s)$ and its derivative are negative, and thus the residues $H_\alpha$ must be positive. The lower bound $s'$ on $h_\alpha$ arises by noting that even for negative $s$, as long as $C(\mathbf{r}; s)$ of (4.3) is positive semi-definite in all components, $C^{(e)}$ will also be positive semi-definite, and consequently $F(s) + \varepsilon^0 C^{(0)} \varepsilon^0 > 0$. This will certainly be true for any $s$ which is sufficiently negative. If we denote by $s'$ the most negative value of $s$ for which $C(\mathbf{r}; s)$ ceases to be positive semi-definite, then clearly all zeros of $F(s) + \varepsilon^0 C^{(0)} \varepsilon^0$ must lie to its right. By expanding (5.2) and (5.1) in powers of $1/s$ and equating the coefficients, we obtain the following moment sum rules for $H(s)$ in terms of corresponding quantities of $F(s)$

$$Q_{0H} \equiv \sum_{\alpha} H_\alpha = Q_0 / \varepsilon^0 C^{(0)} \varepsilon^0, \tag{5.4a}$$

$$Q_{1H} \equiv \sum_{\alpha} H_\alpha h_\alpha = Q_1 / \varepsilon^0 C^{(0)} \varepsilon^0 - Q_{0H}^2, \quad \text{etc.} \tag{5.4b}$$

We can now manufacture bounds on $H(s)$ in much the same way as we did for $F(s)$, and these can of course be translated into bounds on either $F(s)$ or $\varepsilon^0 C^{(e)} \varepsilon^0$. We note that since $H(1)$ is a monotonically increasing function of $F(1)$, therefore the upper (lower) bound on $H(1)$ leads to an upper (lower) bound on $F(1)$.

One way to obtain bounds on $F(1)$ and $H(1)$ is by a direct variation of the free parameters in (4.5) and (5.2). However, we can obtain bounds more easily by the following procedure. We first define two auxiliary functions

$$B(s) \equiv \frac{1}{Q_1} - \frac{1}{s^2 F(s) - s Q_0}, \tag{5.5a}$$

$$Y(s) \equiv \frac{s - s'}{s(Q_{1H} - s' Q_{0H})} - \frac{1}{s[(s - s')H(s) - Q_{0H}]}. \tag{5.5b}$$

Both of these functions have an analytic structure similar to that of $F(s)$ and $H(s)$:

$$B(s) = \sum_{\alpha} \frac{B_\alpha}{s - b_\alpha}, \tag{5.6a}$$

$$Y(s) = \sum_{\alpha} \frac{Y_\alpha}{s - y_\alpha}, \tag{5.6b}$$

where the poles $b_\alpha$, $y_\alpha$ and the residues $B_\alpha$, $Y_\alpha$ satisfy

$$b_\alpha < 0, \qquad y_\alpha < 0, \tag{5.7a}$$

$$B_\alpha > 0, \qquad Y_\alpha < 0. \tag{5.7b}$$

Clearly, the poles $b_\alpha$ of $B(s)$ are the zeros of $s(sF(s)-Q_0)$, and they always include a pole at $s = 0$. Similarly, the poles $y_\alpha$ of $Y(s)$ are the zeros of $s[(s-s')H(s)-Q_{0H}]$, and they too always include a pole at $s = 0$. It may be noted that $s'$, which was defined in (5.3), constitutes a lower bound also on $b_\alpha$ and $y_\alpha$ (and, in fact, also on the poles $s_\alpha$ of $F(s)$), but this property will not be used here. The residues $B_\alpha$, $Y_\alpha$ can be evaluated in terms of derivatives of $F$ and $H$ at appropriate values of $s$ as follows

$$\frac{1}{B_\alpha} = -b_\alpha \frac{\mathrm{d}}{\mathrm{d}s}[sF(s)-Q_0]_{s=b_\alpha} = b_\alpha \sum_\beta \frac{F_\beta s_\beta}{(b_\alpha - s_\beta)^2} > 0, \tag{5.8a}$$

$$\frac{1}{Y_\alpha} = -y_\alpha \frac{\mathrm{d}}{\mathrm{d}s}[(s-s')H(s)-Q_{0H}]|_{s=y_\alpha} = -y_\alpha \sum_\beta \frac{H_\beta(s'-h_\beta)}{(y_\alpha - h_\beta)^2} < 0, \tag{5.8b}$$

thus demonstrating the inequalities (5.7b). It is now easy to convince ourselves that the combinations

$$sB(s) = \sum_\alpha \frac{sB_\alpha}{s-b_\alpha} = \sum_\alpha B_\alpha + \sum_\alpha \frac{B_\alpha b_\alpha}{s-b_\alpha}, \tag{5.9a}$$

$$sY(s) = \sum_\alpha \frac{sY_\alpha}{s-y_\alpha} = \sum_\alpha Y_\alpha + \sum_\alpha \frac{Y_\alpha y_\alpha}{s-y_\alpha}, \tag{5.9b}$$

are a monotonic increasing function and a monotonic decreasing function, respectively. If the value of $F(s)$ is known at some point $s_+$ (see (4.1)), and, consequently, $H(s_+)$, $B(s_+)$, and $Y(s_+)$ are also known, then in the case of $s_+ > 1$ the two inequalities

$$B(1) \leqslant s_+ B(s_+), \tag{5.10a}$$

$$Y(1) \geqslant s_+ Y(s_+), \tag{5.10b}$$

provide us with an upper bound and a lower bound on $F(1)$ that include the information on $F(s_+)$. These bounds are always more stringent than the Hashin–Shtrikman bounds. Moreover, a simple variational calculation shows that they are the best bounds that can be obtained by making use of the extra information on $F(s_+)$ (see Appendix C). In the case of $s_+ < 1$, the inequalities (5.10) get their signs reversed, so that the upper and lower bounds on $F(1)$ and on $\varepsilon^0 C^{(e)} \varepsilon^0$ switch roles.

We should keep in mind that $Q_0$, $Q_1$, $Q_{0H}$, $Q_{1H}$ and $s'$ which appear in (5.4) and (5.5) depend on our choice of $b_{ny}$ in (4.4) (see, e.g. (4.7) and (4.8)), which themselves depend on $s_+$ (see (4.10)). Thus the improved bounds on the effective elastic constants depend on $s_+$ and they should be optimized (made as narrow as possible) by a suitable choice of $s_+$. The range of values of $s_+$ is determined by inequalities of the type (4.13).

The extent to which the new bounds are an improvement over the Hashin–Shtrikman bounds can be demonstrated by the following numerical example: Suppose we have a 50%–50% isotropic mixture of isotropic components $C^{(1)} = (\kappa^{(1)}, (\mu^{(1)}) = (10, 4)$ and $C^{(2)} = (1, 2)$. The Hashin–Shtrikman bounds (3.18) and (3.22) on the

effective elastic constants of such a composite are

$$3.02 \leqslant \kappa^{(e)} \leqslant 4.48, \tag{5.11a}$$

$$2.78 \leqslant \mu^{(e)} \leqslant 2.89. \tag{5.11b}$$

Suppose we know that $C_+^{(e)} = (\kappa_+^{(e)}, \mu_+^{(e)}) = (7, 4.5)$ for a mixture with the same microgeometry but with the $C^{(1)}$ material replaced by $C_+^{(1)} = (30, 10)$. Using this information in (5.10) we obtain the following new bounds

$$3.41 \leqslant \kappa^{(e)} \leqslant 3.99, \tag{5.12a}$$

$$2.82 \leqslant \mu^{(e)} \leqslant 2.86, \tag{5.12b}$$

which are clearly much narrower than (5.11). The optimization of these bounds with respect to $s_+$ (which can vary in the range defined by (4.13b)) was accomplished numerically. The minimal upper bound was obtained for $s_+ \to 0$ and the maximal lower bound was obtained when $s_+$ was equal to its maximum allowed value.

The extent to which we can improve over the Hashin–Shtrikman bounds depends strongly on the differences between the elastic constants of the two materials. This fact can be demonstrated by the following example: Let us examine again a 50%–50% mixture of isotropic materials in which $C^{(2)} = (\kappa^{(2)}, \mu^{(2)}) = (2, 0.75)$ and $C^{(1)} = (x\kappa^{(2)}, x\mu^{(2)})$ where $x$ is a continuous variable which enables the properties of the $C^{(1)}$ material to be varied continuously. Suppose we know that for $x = x_+ \equiv 8$, i.e. for $C_+^{(1)} = (16, 6)$, the effective bulk modulus is $\kappa_+^{(e)} = 5$. Figure 1 depicts the dependence on $x$ of the improved bounds. The upper and lower bounds coincide for $x = x_+$, and begin to

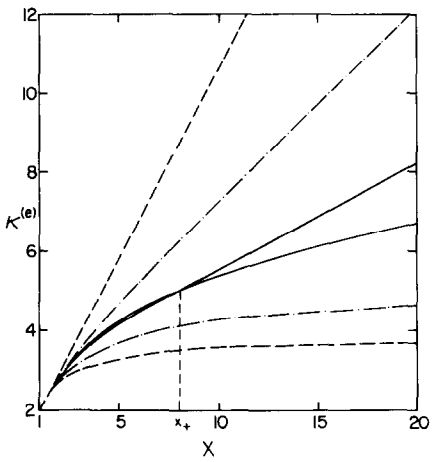


FIG. 1. Improved upper and lower bounds on the effective bulk modulus $\kappa^{(e)}$ of a two-component composite made of isotropic components $C^{(2)} = (\kappa^{(2)}, \mu^{(2)}) = (2, 0.75)$, $C^{(1)} = (x\kappa^{(2)}, x\mu^{(2)})$, as a function of $x$. The dashed lines are the Voigt (upper) and the Reuss (lower) bounds. The dot–dashed lines are the two Hashin–Shtrikman bounds. The solid lines are our improved bounds, obtained by including the additional information that for $x = x_+ \equiv 8$, i.e. for $C^{(1)} = C_+^{(1)} = (16, 6)$, the effective bulk modulus is $\kappa_+^{(e)} = 5$. The improvement is most striking for $x$ near $x_+$, but it is quite considerable even far away from that point. Note that all the bounds in this figure apply in the case of a two-component composite with an *arbitrary* microgeometry.

diverge when $x$ becomes larger or smaller than $x_+$. However, even when $x$ is far away from $x_+$ the bounds are still considerably narrower than the Hashin–Shtrikman bounds.

The bounds (5.10) can be used even in the case when $F(s_+)$ is not known exactly: If we only know that $F(s_+)$ is somewhere in the range $[F'_+, F''_+]$ then by inserting the appropriate end of that range into (5.10) we can still derive an improved bound (provided that the range of $F(s_+)$ itself is narrower than the Hashin–Shtrikman bounds, otherwise no additional information is actually supplied).

In the previous Section we mentioned cases in which we cannot represent two different composites by the same function $F(s)$. Nevertheless, we can derive an improved bound even in those cases by a two stage procedure which is demonstrated by the following example: If we have two isotropic composites made of isotropic components in which the $C^{(2)}$ material is the same but $C^{(1)}$ is different from $C_+^{(1)}$, and if $\kappa^{(2)} < \kappa^{(1)} < \kappa_+^{(1)}$, $\mu^{(2)} < \mu_+^{(1)} < \mu^{(1)}$, then the inequality (4.11) is not satisfied and both composites cannot be represented by the same function $F(s)$. However we can introduce a hypothetical third mixture in which $C_*^{(1)} = (\kappa^{(1)}, \mu_+^{(1)})$. The knowledge of $C_+^{(e)}$ can now be used to first derive improved bounds on the $C^{(2)}$, $C_*^{(1)}$ composite, and those bounds can be used in the next stage to derive improved bounds on the $C^{(2)}$, $C^{(1)}$ composite. Note that at each stage the inequality (4.11) holds.

## 5. DISCUSSION

We have presented a new approach to the calculation of rigorous bounds on the effective elastic constants of a composite material. Using this approach, we rederived some of the known bounds and extended the range of their validity to lower symmetries. An interesting open question is whether these extensions could also have been obtained from the correlation-function-dependent bounds of WILLIS (1982). We also showed that it is possible to improve these bounds in a straightforward manner by including additional information about the material in the form of higher order, geometric correlation functions. We then showed that our formalism can also be used to include other types of information about the system in order to improve the bounds on the effective elastic constants. Specifically, we showed that knowledge of the effective elastic constants of one composite can be used to improve the bounds on the elastic constants of another composite with the same microgeometry. The bounds thus obtained can be considerably narrower than the Hashin–Shtrikman bounds. Other types of information, including both equalities and inequalities, can also be incorporated in the derivation of exact bounds.

It should be kept in mind that the improved bounds which were derived in the previous Section are the best possible for a *given parametric representation* (4.3) *and* (4.4) of $C(\mathbf{r}; s)$. We used a particularly simple representation, but even then we still had to optimize the bounds *numerically* with respect to the value of the one parameter $s_+$. Although the optimum values of $s_+$ turned out empirically (i.e. by the numerical experiments) to be very simple (i.e. either $s_+ = 0$ or $s_+ =$ the maximum possible value) for the case we considered, we were not able to derive a generally applicable analytical result for the optimal value of $s_+$. Thus this remains an open problem. Moreover, there

is a possibility that a more general parametric representation of $C(\mathbf{r};s)$ would lead to even more stringent bounds on $C^{(e)}$ from the same information.

The representation of $C^{(e)}$ as a sum of simple poles can also be applied in the case of viscoelastic materials, i.e. when the elastic tensor is complex. It seems, however, that one must then use a representation that is more general than (4.3) in order to endow $C(\mathbf{r};s)$ with enough freedom to move around in the space of *complex* elastic constants when $s$ is varied.

In summary, we believe that the new approach to the derivation of rigorous bounds on the elastic moduli of a composite material which was presented here is a very fruitful and promising one. While some of its uses and implications for various situations, where only partial information is available about the microstructure of the composite, have been explored, further work along these lines will surely lead to more new and interesting results.

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## APPENDIX A

In this Appendix we analyze several cases in which explicit expressions for the first moment $Q_1$ sum rule (see (3.5)) can be obtained without detailed knowledge of the microgeometry of the sample.

The tensor Green's function $G$ in (3.5) depends on the shape of the sample as well as on the elastic stiffness tensor $C^{(2)}$ (WU and MCCULLOUGH, 1977). The boundary conditions used to derive $G$ (i.e. undisplaced boundaries) imply that the integral $\int G(\mathbf{r},\mathbf{r}')\,dV'$ vanishes whenever the integration is performed over the entire sample volume. Thus subtraction of a constant from $\theta_1(\mathbf{r})\theta_1(\mathbf{r}')$ in (3.5) will not change the result of the integration, and we can rewrite (3.5) in the following form

$$Q_1 = \frac{1}{V}\int (\theta_1(\mathbf{r})\theta_1(\mathbf{r}') - p_1^2)\varepsilon^0 \delta C G(\mathbf{r},\mathbf{r}')\delta C \varepsilon^0 \, dV \, dV'. \tag{A.1}$$

The constant $p_1^2$ was subtracted from the geometric correlation function $\theta_1(\mathbf{r})\theta_1(\mathbf{r}')$ in order to make the average correlation decay to zero for large separation $|\mathbf{r}-\mathbf{r}'|$, and thus have a nonsingular Fourier transform. In the infinite volume limit the tensor Green's function becomes translationally invariant, i.e. $G(\mathbf{r},\mathbf{r}') = G(\mathbf{r}-\mathbf{r}')$, and we may perform one volume integration trivially in (A.1) to obtain

$$Q_1 = \varepsilon^0 \delta C \int f_{11}(\mathbf{r}) G(\mathbf{r}) \, dV \; \delta C \varepsilon^0, \tag{A.2}$$

where

$$f_{11}(\mathbf{r}-\mathbf{r}') \equiv \langle \theta_1(\mathbf{r})\theta_1(\mathbf{r}')\rangle_{\mathrm{av}} - p_1^2. \tag{A.3}$$

When $C^{(2)} = (\kappa^{(2)}, \mu^{(2)})$ is an isotropic material, the infinite-volume Green's function $G(\mathbf{r}; C^{(2)})$ has a relatively simple form. Its Fourier transform $\tilde{G}(\mathbf{k})$ (see WU and MCCULLOUGH, 1977) depends only on the direction of $\mathbf{k}$

$$\tilde{G}_{mnij}(\mathbf{k}) = \frac{\lambda^{(2)}+\mu^{(2)}}{\mu^{(2)}(\lambda^{(2)}+2\mu^{(2)})}\frac{k_m k_n k_i k_j}{k^4} - \frac{1}{4\mu^{(2)}k^2}(\delta_{mi}k_n k_j + \delta_{ni}k_m k_j + \delta_{mj}k_n k_i + \delta_{nj}k_m k_i), \tag{A.4}$$

where $\lambda^{(2)}$ and $\mu^{(2)}$ are the Lamé constants of the $C^{(2)}$ material. The bulk modulus $\kappa$ is related to the Lamé constants by

$$\kappa = \lambda + \frac{2}{d}\mu. \tag{A.5}$$

If the sample has an isotropic microgeometry, i.e. $f_{11}(\mathbf{r}) = f_{11}(r)$ and its Fourier transform $\tilde{f}_{11}(\mathbf{k}) = \tilde{f}_{11}(k)$ then, by combining this property with the fact that $\tilde{G}$ depends only on the unit vector $\hat{k} \equiv \mathbf{k}/k$, we can rewrite (A.2) in the form

$$Q_1 = \varepsilon^0 \delta C f_{11}(0) \frac{1}{\Omega_d} \int \tilde{G}(\hat{k})\, \mathrm{d}\Omega_d\, \delta C \varepsilon^0, \tag{A.6}$$

where $\Omega_d$ is the $d$-dimensional solid angle in $k$-space, and $f_{11}(0) = p_1 - p_1^2$. The average of $\tilde{G}(\hat{k})$ over all directions

$$R_{mnij} \equiv \frac{1}{\Omega_d} \int \tilde{G}_{mnij}(\hat{k})\, \mathrm{d}\Omega_d \tag{A.7}$$

can be easily evaluated once we realize that the integrals which appear are of only two types, and that they can be found without actually performing any integration (see (A.4)).

$$\frac{1}{\Omega_d} \int \frac{k_i k_j}{k^2}\, \mathrm{d}\Omega_d = \frac{1}{d} \delta_{ij}, \tag{A.8}$$

$$\frac{1}{\Omega_d} \int \frac{k_m k_n k_i k_j}{k^4}\, \mathrm{d}\Omega_d = \frac{1}{d(d+2)} (\delta_{ij}\delta_{mn} + 2I_{ijmn}). \tag{A.9}$$

The result (A.8) is obtained from the following consideration: Under rotations, the integral must behave like an invariant second rank symmetric tensor. Thus the only possibility is

$$\frac{1}{\Omega_d} \int \frac{k_i k_j}{k^2}\, \mathrm{d}\Omega_d = \text{const.} \cdot \delta_{ij}. \tag{A.10}$$

The value of the constant multiplier is determined by considering the trace of the tensor

$$\frac{1}{\Omega_d} \int \frac{k_i k_i}{k^2}\, \mathrm{d}\Omega_d = \frac{1}{\Omega_d} \int \mathrm{d}\Omega_d = 1 = \text{const.} \cdot \delta_{ii} = \text{const.} \cdot d. \tag{A.11}$$

By analogous reasoning, the integral of (A.9) must be an invariant fourth rank tensor symmetric under any permutation of its indices. Thus it must be proportional to $\delta_{ij}\delta_{mn} + \delta_{mi}\delta_{nj} + \delta_{mj}\delta_{ni} = \delta_{ij}\delta_{mn} + 2I_{ijmn}$, with a coefficient whose value is easily found by considering the trace of the tensor. Using these results we finally obtain

$$R_{mnij} = \frac{d\kappa^{(2)} + (d-2)\mu^{(2)}}{d(d+2)\mu^{(2)}[d\kappa^{(2)} + 2(d-1)\mu^{(2)}]} \delta_{mn}\delta_{ij} - \frac{d\kappa^{(2)} + 2d\mu^{(2)}}{(d+2)\mu^{(2)}[d\kappa^{(2)} + 2(d-1)\mu^{(2)}]} I_{mnij}. \tag{A.12}$$

Thus $Q_1$ can be calculated from the expression

$$Q_1 = p_1(1-p_1)\varepsilon^0_{kl}\delta C_{klmn} R_{mnij} \delta C_{ijrs} \varepsilon^0_{rs} \tag{A.13}$$

for an isotropic mixture of components one of which ($C^{(2)}$) is isotropic. Note that this expression can be used also in the case of nonisotropic $C^{(1)}$. When both components are isotropic we may use either $\varepsilon^0 = \varepsilon^{0\kappa}$ (see (3.12)) or $\varepsilon^0 = \varepsilon^{0\mu}$ (see (3.19)) to derive the expressions (3.17) or (3.21), respectively, for $Q_1$ and thereafter the bounds on $\kappa^{(e)}$ and $\mu^{(e)}$ ((3.18) and (3.22)).

However the validity of these bounds can in fact be extended beyond the special case of isotropic mixtures. First we note that when both materials are isotropic and $\varepsilon^0 = \varepsilon^{0\kappa}$ then the expression

$$\varepsilon^0 \delta C \tilde{G}(\hat{k}) \delta C \varepsilon^0 = -\delta\kappa^2 E^{(\kappa)}. \tag{A.14}$$

($E^{(\kappa)}$ was defined in (3.17b)) is a *constant* (i.e., independent of $\hat{k}$) and therefore $\varepsilon^0 \delta C G(\mathbf{r}) \delta C \varepsilon^0$ is proportional to a delta-function. Thus the integration (A.2) is trivial and (3.17) and the bound (3.18) on $\kappa^{(e)}$ are universally valid *regardless of the microgeometry of the composite*!

There is no such simple rule for an arbitrary shear modulus in an arbitrary composite. However, we can find simple rules for a particular linear combination of the shear moduli. In 2D we can use $\varepsilon^0 = \varepsilon^{0\mu}$ and $\varepsilon^0 = \varepsilon^{0\mathrm{M}}$ as defined by (3.19) and (3.25) to define two different functions

$F(s)$ and two different first moments $Q_1^{(\mu)}$ and $Q_1^{(\mathrm{M})}$, respectively. The expression

$$\tfrac{1}{2}(\varepsilon^{0\mu}\delta C\tilde{G}(\hat{k})\delta C\varepsilon^{0\mu}+\varepsilon^{0\mathrm{M}}\delta C\tilde{G}(\hat{k})\delta C\varepsilon^{0\mathrm{M}}) = -\delta\mu^2 E^{(\mu)} \tag{A.15}$$

($E^{(\mu)}$ was defined in (3.21b)) is a *constant* (i.e. independent of $\hat{k}$) and therefore an expression identical to the r.h.s. of (3.21a) is valid for the quantity $\frac{1}{2}(Q_1^{(\mu)}+Q_1^{(\mathrm{M})})$ irrespective of the symmetry of $f(\mathbf{r})$. Thus the bounds (3.22) are valid regardless of the microgeometry of the composite if $\mu^{(\mathrm{e})}$ which appears in (3.22) is replaced by the "average shear modulus" $\frac{1}{2}(\mu^{(\mathrm{e})}+M^{(\mathrm{e})})$.

A similar procedure can be followed for shear moduli in the 3D case, but now we have to consider six different shears $\varepsilon^{0\mu}(12)\equiv I_{ij12}$, $\varepsilon^{0\mathrm{M}}(12)\equiv\frac{1}{2}(I_{ij11}-I_{ij22})$, $\varepsilon^{0\mu}(23)$, $\varepsilon^{0\mathrm{M}}(23)$, $\varepsilon^{0\mu}(31)$, $\varepsilon^{0\mathrm{M}}(31)$. In order to obtain a constant term similar to (A.15) we must now use the linear combination

$$[2(Q_1^{(\mathrm{M}(12))}+Q_1^{(\mathrm{M}(23))}+Q_1^{(\mathrm{M}(13))})+3(Q_1^{(\mu(12))}+Q_1^{(\mu(23))}+Q_1^{(\mu(13))})]/15.$$

We thus conclude that (3.22) is valid for a composite with an *arbitrary microgeometry* only if we replace $\mu^{(\mathrm{e})}$ by

$$[2(M^{(\mathrm{e})}(12)+M^{(\mathrm{e})}(23)+M^{(\mathrm{e})}(13))+3(\mu^{(\mathrm{e})}(12)+\mu^{(\mathrm{e})}(23)+\mu^{(\mathrm{e})}(13))]/15,$$

where $M^{(\mathrm{e})}(12)\equiv\varepsilon^{0\mathrm{M}}(12)C^{(\mathrm{e})}\varepsilon^{0\mathrm{M}}(12)$, etc. For a microgeometry with cubic symmetry this expression reduces to the form $(2M^{(\mathrm{e})}+3\mu^{(\mathrm{e})})/5$.

## Appendix B

In this Appendix we analyse the analytic properties of $F(s)$ in the case of the parametric representation (4.3) and (4.4).

Let $s=s_\alpha$ be a pole of $F(s)$. For that value of $s$, there exists a resonance solution $\varepsilon^{(\alpha)}$, i.e. the equation

$$\partial_j\left[\left(C_{ijkl}^{(0)}+\sum_{n\gamma}v_{n\gamma}(s_\alpha)\theta_n\delta C_{ijkl}^{(n\gamma)}\right)\varepsilon_{kl}^{(\alpha)}\right]=0 \tag{B.1}$$

has a non-zero solution for which the corresponding displacement vector $\mathbf{u}^{(\alpha)}$ vanishes at the boundaries of the sample. In that case the total elastic energy of the sample must vanish (cf. Appendix A in I):

$$\int\varepsilon^{(\alpha)*}\left(C^{(0)}+\sum_{n\gamma}v_{n\gamma}(s_\alpha)\theta_n\delta C^{(n\gamma)}\right)\varepsilon^{(\alpha)}\,\mathrm{d}V=0. \tag{B.2}$$

If we restrict ourselves to case of positive $b_{n\gamma}$ in (4.4) and *all* $\delta C^{(n\gamma)}\geqslant 0$ (or *all* $\delta C^{(n\gamma)}\leqslant 0$) then by considering the imaginary part of (B.2), namely

$$\sum_{n\gamma}\frac{|v_{n\gamma}(s_\alpha)|^2\,\mathrm{Im}\,s_\alpha}{b_{n\gamma}}\int\theta_n\varepsilon^{(\alpha)*}\delta C^{(n\gamma)}\varepsilon^{(\alpha)}\,\mathrm{d}V=0, \tag{B.3}$$

we arrive to the conclusion that $\mathrm{Im}\,s_\alpha=0$. Rewriting (B.2) in the form

$$\int\varepsilon^{(\alpha)*}\sum_{n\gamma}[v_{n\gamma}(s_\alpha)\theta_nC^{(n\gamma)}+(1-v_{n\gamma}(s_\alpha))\theta_nC^{(0)}]\varepsilon^{(\alpha)}\,\mathrm{d}V=0, \tag{B.4}$$

we note that there can be no poles in the region $s_\alpha>1$ (because $0<v_{n\gamma}(s_\alpha)<1$). Moreover, from (B.2) and (B.4) we find that all $s_\alpha\leqslant 0$ in the case where all $\delta C^{(n\gamma)}\geqslant 0$ and $b_{n\gamma}\geqslant 1$, and that all $0\leqslant s_\alpha<1$ in the case where all $\delta C^{(n\gamma)}\leqslant 0$ and $0<b_{n\gamma}\leqslant 1$.

The parameters $F_\alpha$ and $s_\alpha$ in (4.5) satisfy moment sum rules analogous to (3.1). The $n$th moment can be found by calculating the $(n+1)$th derivative of $F(s)$ with respect to $w\equiv 1/s$:

$$Q_n\equiv\sum_\alpha F_\alpha s_\alpha^n=\frac{1}{(n+1)!}\frac{\mathrm{d}^{n+1}F}{\mathrm{d}w^{n+1}}\bigg|_{w=0}. \tag{B.5}$$

This follows from an expansion of $F(s)$ in (4.5) in powers of $w$. Thus, in order to evaluate the $n = 0$ and $n = 1$ moments we must find the first and second derivatives of $F$ at $w = 0$. We do this by using the representation for $F(s)$:

$$F(s) = \frac{1}{V}\int \varepsilon C\varepsilon \, \mathrm{d}V - \varepsilon^0 C^{(0)}\varepsilon^0. \tag{B.6}$$

The first variation of that expression is

$$\Delta F = \frac{2}{V}\int \Delta\varepsilon C\varepsilon \, \mathrm{d}V + \frac{1}{V}\int \varepsilon \Delta C \varepsilon \, \mathrm{d}V. \tag{B.7}$$

The first integral in (B.7) can be shown to vanish by transforming it to a surface integral and using the fact that $\Delta\mathbf{u} \equiv 0$ on the surface. Using (4.3) to evaluate $\Delta C$ in terms of the variation of $v_{n\gamma}$, we finally get

$$\Delta F = \frac{1}{V}\sum_{n\gamma} \Delta v_{n\gamma} \int \theta_n \varepsilon \delta C^{(n\gamma)} \varepsilon \, \mathrm{d}V. \tag{B.8}$$

In the limit $w = 0$ ($v_{n\gamma} = 0$) the sample becomes homogeneous, i.e. $\varepsilon = \varepsilon^0$ and it is now straightforward to obtain $Q_0$ from (B.5), (B.8) and (4.4):

$$Q_0 = \frac{1}{V}\int \varepsilon^0 \sum_{n\gamma} \theta_n b_{n\gamma} \delta C^{(n\gamma)} \varepsilon^0 \, \mathrm{d}V = \sum_{n\gamma} p_n b_{n\gamma} \varepsilon^0 \delta C^{(n\gamma)} \varepsilon^0. \tag{B.9}$$

We also note that if *all* $\delta C^{(n\gamma)} \geqslant 0$ ($\delta C^{(n\gamma)} \leqslant 0$) in (B.8) then all the partial derivatives $\partial F/\partial v_{n\gamma}$ are positive (negative). Under the restrictions on the form of $v_{n\gamma}$, the derivatives $\mathrm{d}v_{n\gamma}/\mathrm{d}w$ are always positive for real $w$, therefore $\mathrm{d}F/\mathrm{d}w > 0$. From this it follows that all $F_\alpha > 0$ ($F_\alpha < 0$).

The second variation of $F$ is obtained from (B.8):

$$\tfrac{1}{2}\Delta^2 F = \frac{1}{V}\sum_{n\gamma} \Delta v_{n\gamma} \int \theta_n \Delta\varepsilon \delta C^{(n\gamma)} \varepsilon \, \mathrm{d}V. \tag{B.10}$$

We cannot evaluate this expression in the general case. However, for $w = 0$ ($v_{n\gamma} = 0$, $\varepsilon = \varepsilon^0$) the variation $\Delta\varepsilon$ must satisfy the inhomogeneous differential equation

$$\partial_j C^{(0)}_{ijkl} \Delta\varepsilon_{kl} = -\partial_j \left( \sum_{n\gamma} \theta_n \Delta v_{n\gamma} \delta C^{(n\gamma)}_{ijkl} \varepsilon^0_{kl} \right), \tag{B.11}$$

while the displacement vector $\Delta\mathbf{u}$, which corresponds to $\Delta\varepsilon$, vanishes at the boundaries. The solution of (B.11) and the homogeneous boundary condition on $\Delta\mathbf{u}$ can be written with the help of the tensor Green's function $G$ (see Wu and McCullough, 1977), and compare with (2.12)) corresponding to $C^{(0)}$.

$$\Delta\varepsilon_{kl}(\mathbf{r}) = \int \sum_{n\gamma} \theta_n(\mathbf{r}') \Delta v_{n\gamma} G_{klmn}(\mathbf{r}, \mathbf{r}'; C^{(0)}) \delta C_{mnrt} \varepsilon^0_{rt} \, \mathrm{d}V'. \tag{B.12}$$

Substituting this result in (B.10) we get

$$\tfrac{1}{2}\Delta^2 F|_{v_{n\gamma}=0} = \frac{1}{V}\int\int \sum_{nm\gamma\beta} \theta_n(\mathbf{r}') \theta_m(\mathbf{r}) \Delta v_{n\gamma} \Delta v_{m\beta} \varepsilon^0_{rt} \delta C^{(m\beta)}_{rtij} G_{ijkl}(\mathbf{r}, \mathbf{r}') \delta C^{(n\gamma)}_{klps} \varepsilon^0_{ps} \, \mathrm{d}V \, \mathrm{d}V'. \tag{B.13}$$

Note that integrals of this type have already been evaluated in Appendix A. Following the same line of reasoning, we rewrite (B.13) in the form

$$\tfrac{1}{2}\Delta^2 F|_{v_{n\gamma}=0} = \varepsilon^0 \left( \sum_{nm\gamma\beta} \Delta v_{m\beta} \Delta v_{n\gamma} \delta C^{(m\beta)} \int f_{mn}(\mathbf{r}) G(\mathbf{r}) \, \mathrm{d}V \, \delta C^{(n\gamma)} \right) \varepsilon^0, \tag{B.14}$$

where

$$f_{mn}(\mathbf{r} - \mathbf{r}') \equiv \langle \theta_m(\mathbf{r}) \theta_n(\mathbf{r}') \rangle_{\mathrm{av}} - p_m p_n. \tag{B.15}$$

Expressions (A.2) and (A.3) are special cases of (B.14) and (B.15), respectively. With the help of these results it is now a straightforward matter to calculate $d^2F/dw^2$ at $w = 0$, and this leads to

$$Q_1 = \sum_{mn\beta\gamma} \left[ \varepsilon^0 \delta C^{(m\beta)} \int f_{mn}(\mathbf{r}) G(\mathbf{r})\, d\mathbf{r}\ \delta C^{(n\gamma)} \varepsilon^0 b_{m\beta} b_{n\gamma} \right] - \sum_{n\gamma} [p_n \varepsilon^0 \delta C^{(n\gamma)} \varepsilon^0 b_{n\gamma}(1 - b_{n\gamma})]. \quad \text{(B.16)}$$

As in the case of a two-component composite (see Appendix A) the integrals in this equation can always be evaluated for an isotropic mixture of isotropic components. Thus for $\varepsilon^0 = \varepsilon^{0\gamma}$ where $\gamma = \kappa$ or $\gamma = \mu$ we obtain

$$Q_1^{(\gamma)} = -\sum_{mn} p_m(\delta_{mn} - p_n) \delta\gamma^{(m)} \delta\gamma^{(n)} E^{(\gamma)}(C^0) b_{n\gamma} b_{m\gamma} - \sum_n p_n \delta\gamma^{(n)} b_{n\gamma}(1 - b_{n\gamma}), \quad \text{(B.17)}$$

where $\delta\gamma^{(n)} \equiv \gamma^{(n)} - \gamma^{(0)}$, $E^{(\gamma)}(C^{(0)})$ is defined by (3.17b) or (3.21b) with $\kappa^{(2)}, \mu^{(2)}$ replaced by $\kappa^{(0)}, \mu^{(0)}$, and the term $p_m(\delta_{mn} - p_n)$ is just the value of $f_{mn}(0)$. This result for $Q_1$ can be extended to the case of nonisotropic microgeometries, similarly to the way in which it was done for a two-component composite in Appendix A.

## APPENDIX C

In this Appendix we show that the improved bounds (5.10) are the best bounds that can be obtained under the circumstances, i.e. when the available information consists of $Q_0$, $Q_1$ and $F(s_+)$ for a given parametrization of $F(s)$ (i.e. a given choice of $v_{n\gamma}(s)$). We will limit our discussion to the bound (5.10a) for the case $s_+ > 1$. Other cases, i.e. the bound (5.10b) and the two bounds for the case $s_+ < 1$, can be treated similarly.

The zero and first moment sum rules for $F(s)$ have been incorporated in the definition of $B(s)$, and they are responsible for the fact that $B \to 0$ for $s \to \infty$. The zero and higher order sum rules on $B(s)$ thus depend on the second and higher order sum rules on $F(s)$. Since these higher moments are unknown, they do not provide any constraints on $B(s)$. Thus we only have to include the constraint arising from knowledge of $B(s_+)$. Since $B(s)$ always has a pole at $s = 0$ (see (5.5a)), the easiest way to use this constraint is by forming the combination $B(1) - s_+ B(s_+)$, in which the pole at zero has been eliminated. The unconstrained variation of this quantity is then easily shown to be

$$\Delta(B(1) - s_+ B(s_+)) = \sum_{\alpha \neq 0} \frac{\Delta B_\alpha b_\alpha (s_+ - 1)}{(1 - b_\alpha)(s_+ - b_\alpha)} + \sum_{\alpha \neq 0} \frac{\Delta b_\alpha B_\alpha (s_+ - 1)(s_+ - b_\alpha^2)}{(1 - b_\alpha)^2 (s_+ - b_\alpha)^2}, \quad \text{(C.1)}$$

where the sum over $\alpha$ does not include the pole $b_0 \equiv 0$. Having assumed $s_+ > 1$, the coefficient of $\Delta B_\alpha$ is always negative. By taking all $\Delta B_\alpha$ negative, $B(1)$ is thereby increased. An upper bound is thus obtained if we let all $B_\alpha$ assume their lowest possible values, i.e. $B_\alpha = 0$ for $\alpha \neq 0$. We thus obtain the upper bound

$$B(1) - s_+ B(s_+) \leqslant 0, \quad \text{(C.2)}$$

which is identical to (5.10a) and is clearly the best possible bound under the circumstances.