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VARIATIONAL METHODS, SIZE EFFECTS AND EXTREMAL MICROGEOMETRIES FOR ELASTIC COMPOSITES WITH IMPERFECT INTERFACE

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We introduce new bounds and variational principles for the effective elasticity of anisotropic two-phase composites with imperfect bonding conditions between phases. The monotonicity of the bounds in the geometric parameters is used to predict new size effect phenomena for monodisperse and polydisperse suspensions of spheres. For isotropic elastic spheres in a more compliant isotropic matrix we exhibit critical radii for which the stress state, external to the spheres, is unaffected by their presence. Physically all size effects presented here are due to the increase in surface to volume ratio, as the sizes of the inclusions decrease. The scale at which these effects occur is determined by the parameters N_s^+, N_s^- and R_b^c. These parameters measure the relative importance of interfacial compliance and phase compliance mismatch.

1. Introduction

We examine the effect of imperfect bonding on the overall elastic properties of two-phase composites. Imperfect bonding often occurs in technologically important materials such as fiber reinforced composites. Such bonding cannot be modeled by continuous tractions and displacements across phase boundaries. An imperfect bond is characterized by a thin zone between phases. The elastic displacements may be different on either side of the zone. Such a zone is often referred to as an interphase. Two analytical approaches to modeling imperfect bonds between phases have been considered. One approach describes the interphase between the inclusion and matrix as a layer of finite thickness with elastic properties distinct from those of the matrix and inclusion. The effect of the interphase on the overall elastic behavior has been considered in Refs. 5, 15 and 16. Another approach is to model the imperfect bond by an interface across which the displacements are allowed to be discontinuous. In this model the tractions are continuous across the interface and proportional to the displacement discontinuities. The constants of proportionality characterize the stiffness of the interface. Effective properties for such "spring layer" models were studied in Refs. 1, 2 and 4. Effective elastic behavior for both imperfect bond models was compared using the Mori Tanaka approximation by Jasuik and Tong.[9] We note that all the aforementioned works studied the effects of imperfect interface either for specific composite geometries or by modeling interactions between heterogeneties using approximate formulas. Recently the classical extremum principles of minimum potential and complementary energies were extended to the case of imperfect interface by Hashin.[6] By means of a clever choice of trial fields, the variational principles were used to construct bounds on effective elastic properties. The attractive feature is that any observation deduced from the bounds is not tied to a particular composite geometry or aapproximate formula and will apply to a large class of composite systems.

The variational principles introduced in Ref. 6 form the starting point of our analysis. Hashin described the interface in terms of two tangential stiffnesses D_t, D_s, and a normal stiffness α. To fix ideas, we set the two tangential stiffnesses equal, i.e. $D_t = D_s = \beta$. However we note that the methods developed here easily extend to the case when the tangential stiffnesses are not the same. The slip coefficient β relates the tangential component of the traction to discontinuities in the tangential displacement and α relates the normal component of the traction to the jump in the normal displacement across the interface. We assume that the composite is made from two isotropic elastic materials with bulk and shear moduli specified by $\kappa_1 < \kappa_2$ and $\mu_1 < \mu_2$ in the proportions θ_1 and θ_2 respectively. In this work we adopt a rigorous approach. We derive new variational principles describing the effective elastic properties of anisotropic composites with imperfect interface. These principles are obtained with the aid of an interface comparison method previously established by the authors in the context of heat conductivity (Lipton and Vernescu[13]). The advantage of this new formulation is that solutions of the associated Euler equations involve fields that are not coupled at the two-phase boundary. Most importantly the solution operators for these problems have an explicit form or can be written in terms of solution operators for the perfect contact problem, see Theorems 2.1 and 2.3.

We apply these principles to obtain new upper and lower bounds on the effective elasticity for anisotropic particulate composites with imperfect interface. The lower bounds depend explicitly upon interfacial surface area, the interfacial slip coefficient β, the normal interfacial spring constant α and volume fraction. In addition the bound contains a scale-free tensor of parameters. This tensor corresponds to the effective elasticity of a composite with void-like inclusions having the same geometry as the original composite, see Theorem 3.1. We present an upper bound for anisotropic composites in terms of volume fraction, the two-point correlation function and the moment of inertia and surface energy tensors of the particle surfaces, see Theorem 3.4.

To illustrate our method we consider particulate suspensions of relatively stiff particles in a more compliant matrix. We find new bounds on the bulk and shear traces of the effective elastic tensor C^e and compliance $C^{e^{-1}}$ for anisotropic composites. Here the bulk and shear traces are given by:

$$\mathrm{tr}_b \mathcal{C}^e = \frac{1}{3} \mathcal{C}^e_{iijj} \tag{1.1}$$

and by

$$\mathrm{tr}_s \mathcal{C}^e = \frac{1}{5} \left(\mathcal{C}^e_{ijij} - \frac{1}{3} \mathcal{C}^e_{iijj} \right) \tag{1.2}$$

respectively. These bounds are given in Theorems 3.2, 3.3, 3.5 and 3.6. The bounds given here are shown to be strict improvements on those of Hashin,[6] see Sec. 5. A subtle computational error in the upper and lower shear modulus bounds given in Ref. 6 is found and corrected in Sec. 5.

The monotonicity of the upper and lower bounds in the geometric parameters allow for the analysis of size effect phenomena (see Sec. 6). For monodisperse suspensions of spheres at fixed volume fraction one uses the monotonicity of the lower bound given by Theorem 3.2 to show that the estimate:

$$2\mu_1 \le \mathrm{tr}_s \mathcal{C}^e \tag{1.3}$$

holds for suspensions of spheres with radii "a" bounded below by

$$3N_s^+ \le a\,, \tag{1.4}$$

see, Corollary 6.5. Here $N_s^+ = p/\Delta_s$ where $p = \frac{1}{5}(\frac{1}{\beta} + \frac{2}{3\alpha})$ and $\Delta_s = (2\mu_1)^{-1} - (2\mu_2)^{-1}$. From the upper bound given in Theorem 3.5 it follows that

$$(2\mu_1)^{-1} \le \mathrm{tr}_s(\mathcal{C}^{e-1}) \tag{1.5}$$

for

$$a \le N_s^-\,, \tag{1.6}$$

where $N_s^- = q/\Delta_s$, with $q = 5(3\beta + 2\alpha)^{-1}$, see Corollary 6.5. We note that Δ_s is a measure of the shear compliance mismatch between phases and the parameters p and q are measures of interfacial compliance. The ratios N_s^+ and N_s^- express the relative importance of the shear mismatch and the interfacial compliances. For monodisperse suspensions of spheres we establish the existence of two critical radii. The first critical radius R_b^c, concerns the bulk trace. We define the critical radius to be that for which the bulk trace equals that of the matrix material given by $3\kappa_1$. The critical radius R_b^c is given by

$$R_b^c = \frac{\alpha^{-1}}{\Delta_b}\,, \tag{1.7}$$

where $\Delta_b = (3\kappa_1)^{-1} - (3\kappa_2)^{-1}$ is a measure of the bulk compliance mismatch between phases, see Corollary 4.5. When the slip coefficient and normal spring constant are equal (i.e. $\beta = \alpha$) we have $3N_s^+ = N_s^- = R_s^c$, where

$$R_s^c = \frac{\beta^{-1}}{\Delta_s}\,. \tag{1.8}$$

For monodisperse suspensions of spheres of radius R_s^c we show in Corollary 4.6 that the effective shear trace equals that of the matrix. These results follow from a more fundamental interaction between interfacial and phase compliances. We consider a sample of composite containing spheres of critical radius R_b^c subjected to a homogeneous hydrostatic strain. We calculate the fields external to the spheres to observe that the strain state is unaffected by the presence of the spheres, see Theorem 4.1. For composites containing spheres of critical radius R_s^c subjected to homogeneous shear strain we also find that the strain state external to the spheres is unaltered by their presence, see Theorem 4.2. In this way we see that the mismatch between component elasticities together with the interfacial compliances conspire to make the particles undetectable when the composite is subject to selected homogeneous strains.

For polydisperse suspensions of spheres we apply the bounds to provide new theoretical predictions of critical mean radii. It is shown that when the average sphere radius $\langle a \rangle$ is less than N_s^-, then the shear trace of the effective compliance lies above that of the matrix material. Similarly for suspensions with mean radius less than R_b^c the bulk trace of the effective compliance lies above that of the matrix, see Theorems 6.3 and 6.4. The results given in this paper, are to the best of our knowledge the first theoretical predictions of critical radii for monodisperse and polydisperse suspensions at nondilute concentration.

More generally we consider particulate composites with no assumption on particle shape or distribution. For stiff particles in a more compliant matrix we exhibit upper bounds on the interfacial surface area to particle volume ratio for which the bulk and shear traces of the effective tensor always lie above that of the matrix material, see Theorems 6.1 and 6.2. The bounds are given in terms of the parameters N_s^+ and R_b^c.

We summarize by noting that all of the size effects may be ascribed to the increase in particle surface to volume ratio, as the sizes of the particles decrease. The scale at which these effects occur is determined by the parameters N_s^+, N_s^- and R_b^c. These parameters measure the relative importance between interfacial compliance and the mismatch between phase compliances.

To simplify reading of the text we introduce commonly used notation. Given two 3×3 matrices η and ζ we write the Hilbert–Schmidt inner product:

$$\eta : \zeta = \operatorname{tr}(\eta \zeta^{\mathrm{T}}). \tag{1.9}$$

The inner product between two vectors u and v in $\mathcal{R}^3$ is denoted by $u \cdot v$. Contraction between a fourth-order tensor $\mathcal{T}$ and a 3×3 matrix η is written:

$$(\mathcal{T}\eta)_{ij} = \mathcal{T}_{ijkl}\eta_{kl}\,, \tag{1.10}$$

where repeated indices indicate summation. Contraction between a 3×3 matrix η and a vector v in $\mathcal{R}^3$ is written:

$$(\eta v)_i = \eta_{ij} v_j. \tag{1.11}$$

Last, for vectors u and v in $\mathcal{R}^3$ we write the symmetric dyadic product:

$$(u \odot v)_{ij} = \frac{1}{2}(u_i v_j + u_j v_i). \tag{1.12}$$

2. Variational Principles for the Effective Elasticity Tensor for Composites with Imperfect Interface

2.1. *Mathematical and physical background and variational principles of Hashin*

For periodic elastic composites we may decompose the displacement field u into two parts, a periodic fluctuation $\tilde{\phi}$ and a linear part $\varepsilon_{ij}x_j$ such that $u_i = \tilde{\phi}_i + \varepsilon_{ij}x_j$. The average strain

$$\langle e_{ij}(u)\rangle = \left\langle \frac{1}{2}(u_{i,j} + u_{j,i}) \right\rangle \tag{2.1}$$

seen by an outside observer is

$$\langle e_{ij}(u)\rangle = \int_{\partial Q} (\tilde{\phi}_i + \varepsilon_{is}x_s)n_j dS = \varepsilon_{ij}\,. \tag{2.2}$$

Here Q is the unit cell occupied by the composite, ∂Q is the boundary of the cell and n is the outward directed normal. The two-phase geometry can be arbitrarily complicated. However, we do constrain the phase boundaries to be Lipschitz continuous surfaces.

The cell is composed of two isotropic elastic materials occupying regions Y_1 and Y_2 separated by an interface denoted by Γ.

The materials occupying Y_1 and Y_2 have elasticities $\mathcal{C}_1$ and $\mathcal{C}_2$ specified by:

$$\mathcal{C}_i = 3\kappa_i \mathbb{P}_I + 2\mu_i \mathbb{P}_s \qquad i = 1, 2\,, \tag{2.3}$$

where κ_i and μ_i are the bulk and shear moduli and

$$(\mathbb{P}_I)_{ijkl} = \frac{1}{3}\,\delta_{ij}\delta_{kl}, \qquad (\mathbb{P}_s)_{ijkl} = \frac{1}{2}\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \frac{2}{3}\delta_{ij}\delta_{kl}\right). \tag{2.4}$$

We introduce the space V of Q periodic functions φ given by

$$V = \{\varphi \in H^1(Y_1 \cup Y_2)^3 | \varphi \ Q\text{-periodic}\}\,. \tag{2.5}$$

For future reference we denote the normal trace of a vector field v on a surface Γ with normal n by $v_n = v \cdot n$ and the tangential projection by $v_\tau = v - v_n n$. The elastic displacement inside the composite has periodic fluctuation $\tilde{\phi}$ in V and satisfies:

$$\operatorname{div}(\mathcal{C}_i(e(\tilde{\phi}) + \varepsilon)) = 0 \ \text{ in } \ Y_i, \ \ i = 1, 2 \tag{2.6}$$

$$[\mathcal{C}(x)(e(\tilde{\phi}) + \varepsilon)]n = 0 \ \text{ on } \ \Gamma \tag{2.7}$$

and

$$(\mathcal{C}_2(e(\tilde{\phi}) + \varepsilon)_1 n)_\tau = -\beta[\tilde{\phi}_\tau]_1^2 \ \text{ on } \ \Gamma\,, \tag{2.8}$$

$$(C_2(e(\tilde{\phi})+\varepsilon)_1 n)_n = -\alpha[\tilde{\phi}_n]_1^2 \text{ on } \Gamma. \tag{2.9}$$

Here n denotes the unit normal to Γ and points into the interior of phase-1 and (2.7) is the continuity of the traction across the interface. The numerical subscripts indicate the phase in which the boundary trace is taken. Equations (2.8) and (2.9) represent the Hooke law of the interface relating the tangential component of the traction to tangential slip and the normal component of the traction to the jump in normal displacement. We emphasize that Eq. (2.9) models the effect of an interphase of finite thickness, so jumps in the normal direction are not discounted. The problem (2.6)–(2.9) is well-posed mathematically. Its variational formulation is given by:

$$\int_{Y_1\cup Y_2} C(x)(e(\tilde{\phi})+\varepsilon) : e(\delta)dx + \alpha\int_\Gamma [\tilde{\phi}_n][\delta_n]ds + \beta\int_\Gamma [\tilde{\phi}_\tau]\cdot[\delta_\tau]ds = 0 \tag{2.10}$$

for all δ in V. Here the piecewise constant elastic tensor is given by $C(x) = C_1\chi_1 + C_2\chi_2$ where χ_j are the indicator functions of Y_j, $j = 1,2$. The solution is found to exist and is unique up to a constant; this was established in Lene and Leguillon.[11] The limiting case $\beta = \alpha = \infty$ corresponds to perfect contact between phases and the case $\alpha = \infty$ corresponds to the problem addressed in Ref. 11.

The effective elasticity tensor for the composite is defined by:

$$C^e\varepsilon = \int_Q C(x)(e(\tilde{\phi})+\varepsilon)dx\,. \tag{2.11}$$

Generalizations of the Dirichlet and Thompson variational principles for composites with imperfect interface have been put forth in the work of Hashin.[6] According to these principles the effective elasticity is given by:

$$C^e\varepsilon:\varepsilon = \min_{\phi\in V}\left\{\int_Q C(x)(e(\phi)+\varepsilon):(e(\phi)+\varepsilon)dx \right.$$
$$\left. +\,\alpha\int_\Gamma([\phi_n])^2ds + \beta\int_\Gamma([\phi_\tau])^2ds\right\}. \tag{2.12}$$

Introducing the space W of Q-periodic matrix fields τ characterized by:

$$W = \left\{\tau\in L^2(Q)^{3\times3}|\ \mathrm{div}\tau = 0 \text{ in } Y_1\cup Y_2, [\tau]n = 0 \text{ on } \Gamma,\right.$$
$$\left.\int_Q \tau dx = 0,\ Q\text{-periodic}\right\}. \tag{2.13}$$

The effective compliance is given by:

$$C^{e^{-1}}\bar{\sigma}:\bar{\sigma} = \min_{\tau\in W}\left\{\int_Q C^{-1}(x)(\tau+\bar{\sigma}):(\tau+\bar{\sigma})dx + \alpha^{-1}\int_\Gamma |((\tau+\bar{\sigma})n)_n|^2ds\right.$$
$$\left. +\,\beta^{-1}\int_\Gamma |((\tau+\bar{\sigma})n)_\tau|^2ds\right\}, \tag{2.14}$$

for all constant stresses $\bar{\sigma}$.

2.2. *Interface comparison method variational principles*

We present two new variational principles describing the effective elastic tensor. Before stating the first variational principle we introduce a comparison material with elasticity tensor specified by:

$$\gamma = 3\gamma_I \mathbb{P}_I + \gamma_s \mathbb{P}_s \tag{2.15}$$

such that $\gamma < C_1$, and formulate two auxiliary elasticity problems. For $p \in L^2_{\text{per}}(Q)^{3\times 3}$ the vector potential $u^p \in H^1_{\text{per}}(Y_1 \cup Y_2)^3$ solves the comparison problem:

$$\text{div}(\gamma e(u^p)) = -\text{div}\, p \;\text{ in }\; Y_1 \cup Y_2\,, \tag{2.16}$$

$$(\gamma e(u^p) + p)_1 n = (\gamma e(u^p) + p)_2 n = 0 \;\text{ on }\; \Gamma. \tag{2.17}$$

For $v \in L^2(\Gamma)^3$ the vector potential $u^v \in H^1_{\text{per}}(Y_1 \cup Y_2)^3$ is a solution of the comparison problem:

$$\text{div}\, \gamma(e(u^v)) = 0 \;\text{ in }\; Y_1 \cup Y_2 \tag{2.18}$$

and

$$\gamma e(u^v)_1 n = \gamma e(u^v)_2 n = -v \;\text{ on }\; \Gamma. \tag{2.19}$$

We introduce the space P by

$$P = \{(p, v) \in (L^2_{\text{per}}(Q)^{3\times 3} \times L^2(\Gamma)^3)\}. \tag{2.20}$$

Here the fields p and v are referred to as bulk and surface polarizations respectively.

Introducing the linear operators M and R given by:

$$Mp = e(u^p) \text{ in } Y_1 \cup Y_2\,, \tag{2.21}$$

and

$$Rv = e(u^v) \text{ in } Y_1 \cup Y_2\,, \tag{2.22}$$

one has the new variational principle:

Theorem 2.1. *Comparison Variational Principle. For any constant* 3×3 *strain* ε *one has,*

$$\begin{aligned} C^e \varepsilon : \varepsilon - \gamma\varepsilon : \varepsilon + \left[(2\beta)^{-1} \int_\Gamma \Gamma_s(n) ds + \alpha^{-1} \int_\Gamma \Gamma_h(n) ds \right] \gamma\varepsilon : \gamma\varepsilon \\ = \max_{(p,v)\in P} \left\{ 2\underline{L}(\varepsilon, p, v) - \underline{Q}(p, v) \right\}\,, \end{aligned} \tag{2.23}$$

where

$$\underline{L}(\varepsilon, p, v) = \int_Q p : \varepsilon dx + \beta^{-1} \int_\Gamma v_\tau \cdot \gamma\varepsilon n \, ds + \alpha^{-1} \int_\Gamma v_n n \cdot \gamma\varepsilon n \, ds\,. \tag{2.24}$$

The quadratic form $\underline{Q}(p,v)$ is given by:

$$\underline{Q}(p,v) = \int_Q (C(x)-\gamma)^{-1}p : p \ dx + \beta^{-1}\int_\Gamma |v_\tau|^2 ds + \alpha^{-1}\int_\Gamma |v_n|^2 ds$$
$$+ \int_Q \gamma(Mp+Rv) : (Mp+Rv)dx. \tag{2.25}$$

Here $\Gamma_s(n)$ and $\Gamma_h(n)$ are tensor valued functions of the unit normal given by:

$$(\Gamma_s(n))_{ijk\ell} = \frac{1}{2}(n_i n_\ell \delta_{jk} + n_i n_k \delta_{j\ell} + n_j n_\ell \delta_{ik} + n_j n_k \delta_{i\ell}) - n_i n_j n_k n_\ell \tag{2.26}$$

and

$$(\Gamma_h(n))_{ijk\ell} = n_i n_j n_k n_\ell. \tag{2.27}$$

Remark. We note that the tensor $\Gamma_s(n)$ is precisely that appearing in the decomposition of a second-rank tensor with respect to an interface introduced by Hill.[7]

Proof. Our starting point is the extension of the Dirichlet variational principle for composites with imperfect interface put forth by Hashin.[6] In our context this is given by (2.12). Noting that the solution $\tilde{\phi}$ of (2.6)–(2.9) is the minimizer of (2.12) we write:

$$C^e\varepsilon : \varepsilon = \int_Q C(x)(e(\tilde{\phi})+\varepsilon) : (e(\tilde{\phi})+\varepsilon)dx + \alpha\int_\Gamma ([\tilde{\phi}_n])^2 ds + \beta\int_\Gamma ([\tilde{\phi}_\tau])^2 ds. \tag{2.28}$$

Adding and subtracting the reference energy $\gamma(e(\tilde{\phi})+\varepsilon) : (e(\tilde{\phi})+\varepsilon)$ to the right-hand side of (2.28) and rearrangement gives:

$$(C^e-\gamma)\varepsilon : \varepsilon = \int_Q (C(x)-\gamma)(e(\tilde{\phi})+\varepsilon) : (e(\tilde{\phi})+\varepsilon)dx + \int_Q \gamma e(\tilde{\phi}) : e(\tilde{\phi})dx$$
$$+ 2\int_Q \gamma e(\tilde{\phi}) : \varepsilon dx + \alpha\int_\Gamma ([\tilde{\phi}_n])^2 ds + \beta\int_\Gamma ([\tilde{\phi}_\tau])^2 ds. \tag{2.29}$$

Integrating by parts one obtains:

$$2\int_Q \gamma e(\tilde{\phi}) : \varepsilon \ dx = 2\int_\Gamma [\tilde{\phi}_n]n \cdot \gamma\varepsilon n \ ds + 2\int_\Gamma [\tilde{\phi}_\tau] \cdot \gamma\varepsilon n \ ds. \tag{2.30}$$

Applying (2.30) and completing the square in the last three terms of (2.29) gives:

$$(C^e-\gamma)\varepsilon : \varepsilon + \beta^{-1}\int_\Gamma |(\gamma\varepsilon n)_\tau|^2 ds + \alpha^{-1}\int_\Gamma |(\gamma\varepsilon n)_n|^2 ds$$
$$= \int_Q (C(x)-\gamma)(e(\tilde{\phi})+\varepsilon) : (e(\tilde{\phi})+\varepsilon)dx + \int_Q \gamma e(\tilde{\phi}) : e(\tilde{\phi})dx$$
$$+ \beta\int_\Gamma |[\tilde{\phi}_\tau] + \beta^{-1}(\gamma\varepsilon n)_\tau|^2 ds + \alpha\int_\Gamma |[\tilde{\phi}_n] + \alpha^{-1}(\gamma\varepsilon n)_n|^2 ds. \tag{2.31}$$

Introducing the bulk and surface polarizations p and v one has the elementary estimates:

$$\beta\int_\Gamma |[\tilde{\phi}_\tau]+\beta^{-1}(\gamma\varepsilon n)_\tau|^2 ds \geq \int_\Gamma \{2v_\tau\cdot([\tilde{\phi}_\tau]+\beta^{-1}(\gamma\varepsilon n)_\tau)-\beta^{-1}|v_\tau|^2\}ds\,, \quad (2.32)$$

$$\alpha\int_\Gamma |[\tilde{\phi}_n]+\alpha^{-1}(\gamma\varepsilon n)_n|^2 ds \geq \int_\Gamma \{2v_n([\tilde{\phi}_n]+\alpha^{-1}(\gamma\varepsilon n)_n)-\alpha^{-1}|v_n|^2\}ds\,, \quad (2.33)$$

and

$$\int_Q (C(x)-\gamma)(e(\tilde{\phi})+\varepsilon):(e(\tilde{\phi})+\varepsilon)dx \geq 2\int_Q p:(e(\tilde{\phi})+\varepsilon)dx-\int_Q (C(x)-\gamma)^{-1}p:pdx, \quad (2.34)$$

for any $(p,v)\in P$. Introducing the Lagrangian $\underline{\mathcal{L}}(p,v,\phi)$ defined by:

$$\begin{aligned}\underline{\mathcal{L}}(p,v,\phi) &= 2\int_Q p:\varepsilon dx+2\beta^{-1}\int_\Gamma v_\tau\cdot(\gamma\varepsilon n)_\tau ds+2\alpha^{-1}\int_\Gamma v_n(\gamma\varepsilon n)_n ds\\ &\quad -\int_Q (C(x)-\gamma)^{-1}p:pdx-\beta^{-1}\int_\Gamma |v_\tau|^2 ds-\alpha^{-1}\int_\Gamma (v_n)^2 ds\\ &\quad +2\int_Q p:e\phi dx+2\int_\Gamma v_\tau\cdot[\phi_\tau]ds+2\int_\Gamma v_n[\phi_n]ds\\ &\quad +\int_Q \gamma e(\phi):e(\phi)dx, \end{aligned} \quad (2.35)$$

and applying the estimate (2.32)–(2.34) to (2.31) gives the inequality:

$$(\mathcal{C}^e-\gamma)\varepsilon:\varepsilon+\beta^{-1}\int_\Gamma |(\gamma\varepsilon n)_\tau|^2 ds+\alpha^{-1}\int_\Gamma |(\gamma\varepsilon n)_n|^2 ds \geq \mathcal{L}(p,v,\tilde{\phi}). \quad (2.36)$$

Next we observe:

$$\begin{aligned}&(\mathcal{C}^e-\gamma)\varepsilon:\varepsilon+\beta^{-1}\int_\Gamma |(\gamma\varepsilon n)_\tau|^2 ds+\alpha^{-1}\int_\Gamma |(\gamma\varepsilon n)_n|^2 ds\\ &\quad \geq \mathcal{L}(p,v,\tilde{\phi}) \geq \inf_{\phi\in V}\mathcal{L}(p,v,\phi)=\mathcal{L}(p,v,\overset{*}{\phi})\,,\end{aligned} \quad (2.37)$$

where $\overset{*}{\phi}$ is the minimizer of

$$\inf_{\phi\in V}\left\{2\int_Q p:e(\phi)dx+2\int_\Gamma v_\tau\cdot[\phi_\tau]ds+2\int_\Gamma v_n[\phi_n]ds+\int_Q \gamma e(\phi):e(\phi)dx\right\} \quad (2.38)$$

and satisfies

$$\operatorname{div}\gamma e(\overset{*}{\phi})=-\operatorname{div}p \;\text{ in }\; Y_1\cup Y_2\,, \quad (2.39)$$

and

$$(\gamma e(\overset{*}{\phi})+p)_2 n=(\gamma e(\overset{*}{\phi})+p)_1 n=-v\,. \quad (2.40)$$

Observing that $\overset{*}{\phi}$ is linear in the data (p,v) we have $\overset{*}{\phi} = \phi^p + \phi^v$ where ϕ^p and ϕ^v solve problems (2.16)–(2.17) and (2.18)–(2.19) respectively. Recalling the definitions of the operators M and R given by (2.21) and (2.22), inequality (2.37) can be written as the inequality:

$$\begin{aligned}(C^e - \gamma)\varepsilon : \varepsilon + \beta^{-1}\int_\Gamma |(\gamma\varepsilon n)_\tau|^2 ds + \alpha^{-1}\int_\Gamma |(\gamma\varepsilon n)_n|^2 ds \\ \geq 2\underline{L}(p,v,\varepsilon) - \underline{Q}(p,v)\,. \end{aligned} \tag{2.41}$$

One observes for the choice of bulk and surface polarizations, consistent with the actual displacements inside the composite, i.e.

$$p = (C(x) - \gamma)(e(\tilde{\phi}) + \varepsilon), \tag{2.42}$$

$$v_\tau = [\tilde{\phi}_\tau] + \beta^{-1}(\gamma\varepsilon n)_\tau, \tag{2.43}$$

and

$$v_n = [\tilde{\phi}_n] + \alpha^{-1}(\gamma\varepsilon n)_n \tag{2.44}$$

that (2.41) holds with equality.

To conclude the proof we show that the left-hand side of (2.41) agrees with the left-hand side of (2.23). To see this we expand $(\gamma\varepsilon n)_\tau$ to find

$$|(\gamma\varepsilon n)_\tau|^2 = |(\gamma\varepsilon n) - (\gamma\varepsilon n\cdot n)n|^2 = \frac{1}{2}\Gamma_s(n)\gamma\varepsilon : \gamma\varepsilon\,, \tag{2.45}$$

where $\Gamma_s(n)$ is given by (2.26). Similarly we find

$$(\gamma\varepsilon n)_n^2 = \Gamma_h(n)\gamma\varepsilon : \gamma\varepsilon\,, \tag{2.46}$$

where $\Gamma_h(n)$ is given by (2.27). Substitution of (2.45) and (2.46) into the left-hand side of (2.41) gives the left-hand side of (2.23) and the theorem is proved. □

For the second new variational principle we introduce an isotropic comparison material with elasticity given by (2.15) such that $\gamma > C_2$ and formulate two auxiliary elasticity problems. For $p \in L^2_{\text{per}}(Q)^{3\times 3}$ the potential ψ^p solves:

$$\operatorname{div}\gamma e(\psi^p) = \operatorname{div}\gamma p \quad \text{in} \quad Y_1 \cup Y_2 \tag{2.47}$$

$$[\gamma e(\psi^p) - \gamma p]n = 0, \quad [\psi^p] = 0 \quad \text{on} \quad \Gamma\,. \tag{2.48}$$

For a square integrable vector field $v \in L^2(\Gamma)^3$, the potential ψ^v is a solution of

$$\operatorname{div}\gamma e(\psi^v) = 0 \quad \text{in} \quad Y_1 \cup Y_2 \tag{2.49}$$

$$[\gamma e(\psi^v)]n = 0, \quad [\psi^v] = -v \text{ on } \Gamma. \tag{2.50}$$

We introduce the linear operators N and S defined by

$$N\gamma p = e(\psi^p) \text{ in } Q, \quad Sv = e(\psi^v) \quad \text{in} \quad Y_1 \cup Y_2\,. \tag{2.51}$$

Since the measure of the two-phase interface Γ is zero it follows that Sv lies in $L^2_{\text{per}}(Q)^{3\times3}$. From standard estimates one easily sees that the operators are bounded, i.e. $N \in \mathcal{L}(L^2_{\text{per}}(Q)^{3\times3}, L^2_{\text{per}}(Q)^{3\times3})$ and $S \in \mathcal{L}(L^2_{\text{per}}(\Gamma), L^2_{\text{per}}(Q)^{3\times3})$. The operator N appears in the context of Hashin–Shtrikman variational principles for anisotropic elastic composites with perfect contact as seen in Refs. 3, 10, 12 and 14.

In the sequel we shall need explicit formulas for the operators N and S, these formulas are given as follows:

Lemma 2.2. *The linear operators N and S are given by:*

$$Nq = \sum_{k\neq 0} e^{2\pi i k\cdot x}(\gamma_s^{-1}\Gamma_s(\hat{k}) + r\Gamma_h(\hat{k}))\hat{q}(k) \tag{2.52}$$

and

$$Sv = \sum_{k\neq 0} e^{2\pi i k\cdot x}((\gamma_s^{-1}\Gamma_s(\hat{k}) + r\Gamma_h(\hat{k}))\gamma - \mathbf{I})\int_\Gamma e^{-2\pi i k\cdot y} v\odot n ds$$
$$- \int_\Gamma v \odot n ds \tag{2.53}$$

for any (q, v) in P. Here $\hat{q}(k)$ is the Fourier transform of q at wave vector k, $\hat{k} = k/|k|$, $r = 3/(3\gamma_I + 2\gamma_s)$, and $\mathbf{I}$ is the identity on symmetric 3×3 matrices.

Proof. The explicit formula for the operator N follows immediately from solution of the comparison problem (2.47), (2.48) using Fourier expansions. To obtain the desired representation for the operator S, we extend the function v defined on Γ to the space $H^1_{\text{per}}(Y_1)^3$. Denoting the extension also by v we introduce the auxiliary problem. We consider the potential w in $H^1_{\text{per}}(Q)^3$ solving:

$$\operatorname{div}\gamma(e(w)) = \operatorname{div}\gamma(\chi_1(e(v)))\,, \tag{2.54}$$

$$[\gamma e(w) - \gamma\chi_1 e(v)]n = 0, \quad [w] = 0 \text{ on } \Gamma\,. \tag{2.55}$$

Observing that $\chi_1 e(v)$ lies in $L^2_{\text{per}}(Q)^{3\times3}$ we see that (2.54), (2.55) are equivalent to the comparison problem (2.47), (2.48) with $p = \chi_1 e(v)$. Therefore:

$$e(w) = N(\gamma\chi_1 e(v))\,. \tag{2.56}$$

From (2.54) and (2.55) we observe that the function:

$$\psi = \begin{cases} w - v & \text{in } Y_1 \quad (2.57)\\ w & \text{in } Y_2 \quad (2.58)\end{cases}$$

is a solution of the problem (2.49), (2.50). Since the solution of the comparison problem (2.49), (2.50) is unique up to a constant, we may take ψ^v to be represented by (2.57) and (2.58). It follows that:

$$Sv = e(\psi^v) = e(w) - \chi_1 e(v) \quad \text{in } Y_1 \cup Y_2\,. \tag{2.59}$$

Denoting the identity operator on $L^2_{\text{per}}(Q)^{3\times 3}$ by I we have from (2.56) and (2.59)

$$Sv = (N\gamma - I)\chi_1 e(v)\,. \tag{2.60}$$

Expanding N using (2.52), Eq. (2.60) reads

$$Sv = \sum_{k\neq 0} e^{2\pi ik\cdot x}\{(\gamma_s^{-1}\Gamma_s(\hat{k}) + r\Gamma_h(\hat{k}))\gamma - \mathbf{I}\}\widehat{\chi_1 e}(v)(k) - \widehat{\chi_1 e}(v)(0)\,. \tag{2.61}$$

From the divergence theorem we have

$$\widehat{\chi_1 e}(v)(k) = \int_\Gamma e^{-2\pi ik\cdot y} v \odot n dS + 2\pi i\widehat{\chi_1 v}(k)\odot k. \tag{2.62}$$

Lastly a lengthy but straightforward computation shows

$$\{(\gamma_s^{-1}\Gamma_{1,s}(\hat{k}) + r\Gamma_h(\hat{k}))\gamma - \mathbf{I}\}(\widehat{\chi_1 v}(k)\odot k) = 0 \tag{2.63}$$

and the theorem follows from substitution of (2.62) into (2.61). □

We now give the second comparison method variational principle.

Theorem 2.3. *Dual Comparison Method Variational Principle. For any constant* 3×3 *stress* $\bar{\sigma}$ *one has:*

$$(C^{e^{-1}} - \gamma^{-1})\bar{\sigma} : \bar{\sigma} = \max_{(p,v)\in P} 2\bar{L}(p,v,\bar{\sigma}) - \bar{Q}(p,v)\,, \tag{2.64}$$

where the linear form $\bar{L}$ *is given by:*

$$\bar{L}(p,v,\bar{\sigma}) = 2\int_Q p : \bar{\sigma} dx + 2\int_\Gamma \bar{\sigma} n\cdot v ds\,, \tag{2.65}$$

and the quadratic form $\bar{Q}$ *is given by:*

$$\begin{aligned}\bar{Q}(p,v) = &\int_Q (C^{-1}(x) - \gamma^{-1})^{-1} p : p + \beta\int_\Gamma |v_\tau|^2 ds + \alpha\int_\Gamma |v_n|^2 ds\\ &+ \int_Q \gamma(Sv + N\gamma p - p - \langle Sv + N\gamma p - p\rangle)\\ &: (Sv + N\gamma p - p - \langle Sv + N\gamma p - p\rangle)dx\,.\end{aligned} \tag{2.66}$$

Here $\langle\cdot\rangle$ stands for the average of a quantity over the unit cell Q.

Proof. As before we start with the extension of the Thompson variational principle to composites with imperfect interface as introduced by Hashin.[6] In this context it is given by (2.14). We denote by $\tilde{\sigma}$ the minimizer of (2.14) and write

$$\begin{aligned}C^{e^{-1}}\bar{\sigma}\cdot\bar{\sigma} = &\int_Q C^{-1}(\tilde{\sigma}+\bar{\sigma}) : (\tilde{\sigma}+\bar{\sigma})dx + \alpha^{-1}\int_\Gamma |((\tilde{\sigma}+\bar{\sigma})n)_n|^2 ds\\ &+ \beta^{-1}\int |((\tilde{\sigma}+\bar{\sigma})n)_\tau|^2 dS\,.\end{aligned} \tag{2.67}$$

Adding and subtracting the reference energy $\gamma(\tilde{\sigma}+\bar{\sigma}):(\tilde{\sigma}+\bar{\sigma})$ to the right-hand side of (2.67) yields:

$$(\mathcal{C}^{e^{-1}}-\gamma^{-1})\bar{\sigma}:\bar{\sigma}=\int_Q(\mathcal{C}^{-1}(x)-\gamma^{-1})(\tilde{\sigma}+\bar{\sigma}):(\tilde{\sigma}+\bar{\sigma})dx+\int_Q\gamma^{-1}\tilde{\sigma}:\tilde{\sigma}dx$$
$$+\alpha^{-1}\int_\Gamma((\tilde{\sigma}+\bar{\sigma})n)_n^2dS+\beta^{-1}\int_\Gamma|((\tilde{\sigma}+\bar{\sigma})n)_\tau|^2ds\,. \quad (2.68)$$

One has the estimates:

$$\int_Q(\mathcal{C}^{-1}(x)-\gamma^{-1})(\tilde{\sigma}+\bar{\sigma}):(\tilde{\sigma}+\bar{\sigma})dx\geq 2\int_Q p:(\tilde{\sigma}+\bar{\sigma})dx$$
$$-\int_Q(\mathcal{C}^{-1}(x)-\gamma^{-1})^{-1}p:pdx, \quad (2.69)$$

$$\alpha^{-1}\int_\Gamma((\tilde{\sigma}+\bar{\sigma})n^2)_ndS\geq 2\int_\Gamma((\tilde{\sigma}+\bar{\sigma})n)_nv_nds-\alpha\int_\Gamma v_n^2ds\,, \quad (2.70)$$

and

$$\beta^{-1}\int_\Gamma|((\tilde{\sigma}+\bar{\sigma})n)_\tau|^2dS\geq 2\int_\Gamma((\tilde{\sigma}+\bar{\sigma})n)_\tau\cdot v_\tau ds-\beta\int_\Gamma|v_\tau|^2ds\,. \quad (2.71)$$

We introduce the Lagrangian $\bar{\mathcal{L}}(p,v,\sigma)$ defined by

$$\bar{\mathcal{L}}(p,v,\sigma)=2\int_Q p:\bar{\sigma}dx+2\int_\Gamma(\bar{\sigma}n)_nv_nds+2\int_\Gamma(\bar{\sigma}n)_\tau\cdot v_\tau ds$$
$$-\int_Q(\mathcal{C}^{-1}(x)-\gamma^{-1})^{-1}p:pdx-\alpha\int_\Gamma v_n^2ds-\beta\int_\Gamma|v_\tau|^2ds$$
$$+2\int_Q p:\tilde{\sigma}dx+2\int_\Gamma(\tilde{\sigma}n)_nv_nds+2\int_\Gamma(\tilde{\sigma}n)_\tau\cdot v_\tau ds$$
$$+\int_Q\gamma^{-1}\tilde{\sigma}:\tilde{\sigma}dx\,. \quad (2.72)$$

Application of the estimates to (2.68) gives:

$$(\mathcal{C}^{e^{-1}}-\gamma^{-1})\bar{\sigma}:\bar{\sigma}\geq\bar{\mathcal{L}}(p,v,\tilde{\sigma})\,. \quad (2.73)$$

Now we observe that

$$(\mathcal{C}^{e^{-1}}-\gamma^{-1})\bar{\sigma}:\bar{\sigma}\geq\bar{\mathcal{L}}(p,v,\tilde{\sigma})\geq\inf_{\sigma\in W}\bar{\mathcal{L}}(p,v,\sigma)=\bar{\mathcal{L}}(p,v,\overset{*}{\sigma}), \quad (2.74)$$

where $\overset{*}{\sigma}$ is the minimizer of

$$\inf_{\sigma\in W}\left\{2\int_Q p:\sigma dx+\int_\Gamma(\tilde{\sigma}n)_nv_nds+2\int_\Gamma(\tilde{\sigma}n)_\tau\cdot v_\tau ds+\int_Q\gamma^{-1}\sigma:\sigma dx\right\}\,. \quad (2.75)$$

Calculation shows that $\overset{*}{\sigma}$ is given by

$$\overset{*}{\sigma} = \gamma(e(\overset{*}{\psi}) - p - \langle e(\overset{*}{\psi}) - p\rangle), \tag{2.76}$$

where $\overset{*}{\psi}$ is a solution of

$$\operatorname{div} \gamma e(\overset{*}{\psi}) = \operatorname{div} \gamma p \quad \text{in} \quad Y_1 \cup Y_2 \tag{2.77}$$

$$[\gamma e(\overset{*}{\psi}) - \gamma p]n = 0, \quad [\overset{*}{\psi}] = -v \text{ on } \Gamma. \tag{2.78}$$

Noting that $\overset{*}{\psi}$ is linear in the data (p, v) we write $\overset{*}{\psi} = \psi^p + \psi^v$, where ψ^p and ψ^v are solutions of (2.47)–(2.48) and (2.49)–(2.50). Recalling the definition of the operators N and S given by (2.51), inequality (2.74) is written:

$$(C^{e^{-1}} - \gamma^{-1})\bar{\sigma} : \bar{\sigma} \geq 2\bar{L}(\bar{\tau}, p, v) - \bar{Q}(p, v). \tag{2.79}$$

For the choice of bulk and surface polarizations, consistent with the actual stress in the composite, i.e.

$$p = (C^{-1}(x) - \gamma^{-1})(\tilde{\sigma} + \bar{\sigma}), \quad v_n = \alpha^{-1}((\tilde{\sigma} + \bar{\sigma})n)_n, \tag{2.80}$$

and

$$v_\tau = \beta^{-1}((\tilde{\sigma} + \bar{\sigma})n)_\tau \tag{2.81}$$

one observes that (2.79) holds with equality and the Theorem is proved. □

3. Bounds on the Effective Elasticity for Anisotropic Composites

We apply the variational principles developed in Sec. 2.2 to obtain new upper and lower bounds on the effective elastic tensor for anisotropic composites.

3.1. *Lower bounds for particulate composites*

To fix ideas, we use the new variational principles to obtain bounds on effective elastic properties for a compliant matrix reinforced with stiffer particles, i.e. particles with elasticity C_2 and matrix with elasticity C_1. We consider a distribution of N particles none of which intersect the boundary of the domain Q. The particle region is denoted by Y_2 and the matrix by Y_1. We consider a comparison problem with the particle region Y_2 filled by a soft elastic material, (i.e. zero stiffness) and a matrix region with isotropic elasticity γ. We introduce the effective elastic tensor C^γ associated with this composite, defined by:

$$C^\gamma \varepsilon : \varepsilon = \min_{u \in V} \int_{Y_1} \gamma(e(u) + \varepsilon) : (e(u) + \varepsilon) dy \tag{3.1}$$

for all constant 3×3 strains ε.

We introduce the following tensor notation, let

$$D = \theta_2 C_1^{-1} C_2 (C_2 - C_1)^{-1},$$

$$E = (2\beta)^{-1} \int_\Gamma \Gamma_s(n) ds + \alpha^{-1} \int_\Gamma \Gamma_h(n) ds,$$

and C^* is given by (3.1) for the choice $\gamma = C_1$. The tensor inequality bounding the effective tensor from below is given by:

Theorem 3.1. *Lower Bound. For any constant* 3×3 *strain* ε:

$$C^e \varepsilon : \varepsilon \geq (C_1 - C_1 E C_1)\varepsilon : \varepsilon + \begin{pmatrix} \theta_2 \varepsilon \\ C_1 E \varepsilon \end{pmatrix}^{\mathrm{T}} \begin{pmatrix} D & \theta_2 C_1^{-1} \\ \theta_2 C_1^{-1} & E + C_1^{-1} - C_1^{-1} C^* C_1^{-1} \end{pmatrix}^{-1} \begin{pmatrix} \theta_2 \varepsilon \\ C_1 E \varepsilon \end{pmatrix} . \quad (3.2)$$

Proof. We make the choice $p = \chi_2 \mu, v = r_{ij} n_j$ in the variational principle given by Theorem 2.1. Here μ and r are constant symmetric 3×3 matrices, and n is the unit normal pointing into phase 1. The associated bound is given by:

$$C^e \varepsilon : \varepsilon - \gamma \varepsilon : \varepsilon + E \gamma \varepsilon : \gamma \varepsilon \geq \max_{\mu, r} \left\{ 2\underline{L}(\varepsilon, \chi_2 \mu, rn) - \underline{Q}(\chi_2 \mu, rn) \right\} . \quad (3.3)$$

Setting the comparison elasticity γ to C_1 the theorem follows from explicit computation of $\underline{L}$ and $\underline{Q}$. The formula for $\underline{L}$ follows directly and is given by:

$$\underline{L} = \theta_2 \mu : \varepsilon + C_1 E \mu : \varepsilon . \quad (3.4)$$

The formula for $\underline{Q}$ requires computation; we first display the formula and then proceed with its computation:

$$\underline{Q} = \theta_2 (C_2 - C_1)^{-1} \mu : \mu + E r : r + (\theta_2 C_1^{-1} \mu : \mu + 2\theta_2 C_1^{-1} \mu : r + (C_1^{-1} - C_1^1 C^* C_1^{-1}) r : r) . \quad (3.5)$$

The first three terms of $\underline{Q}$ follow directly from substitution of the polarizations $p = \chi_2 \mu, v = rn$ into the formula for $\underline{Q}$ given by (2.25). The last three terms of (3.5) follow from solution of the comparison problems (2.16)–(2.19) and evaluation of the nonlocal term in (2.25), i.e. we show:

$$\int_Q C_1 (Mp + Rv) : (Mp + Rv) dx = (\theta_2 C_1^{-1} \mu : \mu + 2\theta_2 C_1^{-1} \mu : r + (C_1^{-1} - C_1^{-1} C^* C_1^{-1}) r : r). \quad (3.6)$$

For the choice $p = \chi_2 \mu$ the solution of (2.16), (2.17), yields $M \chi_2 \mu = -C_1^{-1} \chi_2 \mu$ and therefore

$$\int_Q C_1 M(\chi_2 \mu) : M(\chi_2 \mu) dx = \theta_2 C_1^{-1} \mu : \mu . \quad (3.7)$$

Solution of (2.18), (2.19) provides the relation $R(rn) = -\mathcal{C}_1^{-1}r$ in region 2, so

$$2\int_Q \mathcal{C}_1 M(\chi_2\mu) : R(rn)dx = 2\theta_2\mathcal{C}_1^{-1}\mu : r\,. \tag{3.8}$$

Last, in region 1 we have $R(rn) = e(u^r)$ where u^r is a solution of

$$\nabla \cdot \mathcal{C}_1 e(u^r) = 0 \quad \text{in} \quad Y_1\,, \tag{3.9}$$

$$\mathcal{C}_1(e(u^r) + \mathcal{C}_1^{-1}r)\cdot n = 0 \quad \text{on} \quad \Gamma. \tag{3.10}$$

Physically we see that u^r is the periodic fluctuation in the elastic displacement for a composite made from soft elastic material in Y_2 and an elastic material with Hooke's law $\mathcal{C}_1$ in Y_1. Here the composite is subject to a constant strain $\mathcal{C}_1^{-1}r$.

Integration by parts and using (3.9), (3.10) gives:

$$\int_{Y_1} \mathcal{C}_1 R(rn) : R(rn)dx + \int_{Y_1} \mathcal{C}_1 e(u^r) : \mathcal{C}_1^{-1}r = 0\,. \tag{3.11}$$

Completing squares gives:

$$\begin{aligned}\mathcal{C}_1^{-1}\,\overset{*}{\mathcal{C}}\,\mathcal{C}_1^{-1}r : r &\equiv \int_{Y_1} \mathcal{C}_1(e(u^r) + \mathcal{C}_1^{-1}r) : (e(u^r) + \mathcal{C}_1^{-1}r)dx \\ &= \theta_1\mathcal{C}_1^{-1}r : r - \int_{Y_1} \mathcal{C}_1 R(rn) : R(rn)dx.\end{aligned} \tag{3.12}$$

Thus

$$\begin{aligned}\int_Q \mathcal{C}_1 R(rn) : R(rn)dx &= \int_{Y_2} \mathcal{C}_1 R(rn) : R(rn) + \int_{Y_1} \mathcal{C}_1 R(rn) : R(rn) \\ &= \theta_2\mathcal{C}_1^{-1}r : r + \int_{Y_1} \mathcal{C}_1 R(rn) : R(rn),\end{aligned} \tag{3.13}$$

and applying (3.12) gives

$$\int_Q \mathcal{C}_1 R(rn) : R(rn)dx = \mathcal{C}_1^{-1}r : r - \mathcal{C}_1^{-1}\,\overset{*}{\mathcal{C}}\,\mathcal{C}_1^{-1}r : r. \tag{3.14}$$

Using the identities (3.7), (3.8) and (3.14) we have established the desired formula (3.6). The Theorem follows upon maximization of (3.3) over all 3×3 symmetric matrices r and μ. □

The lower bounds given in Theorem 3.1 hold for any anisotropic particulate mixture and is a tensor inequality. On the other hand, similar methods yield lower bounds on selected traces of the effective elastic tensor. In this direction we provide lower bounds on the bulk and shear traces given by (1.1) and (1.2) respectively.

For isotropic composites the bulk and shear traces correspond to $3\kappa^e$ and $2\mu^e$ respectively, where μ^e and κ^e are the effective shear and bulk moduli for an isotropic composite.

To generate trace bounds we return to the lower bound (3.3), with $\underline{L}$ and $\underline{Q}$ given by (3.4) and (3.5). Group averaging the left- and right-hand sides of (3.3) over the isotropic group and choosing $\varepsilon = \bar{\varepsilon}$ where $\mathrm{tr}\bar{\varepsilon} = 0$ yields:

$$\left(\mathrm{tr}_s C^e - 2\mu_1 + \frac{4\mu_1^2}{5\beta}s + \frac{8\mu_1^2}{15\alpha}s\right)(\bar{\varepsilon} : \bar{\varepsilon}) \geq \max_{\bar{\mu},\bar{r}}\{\underline{L} - 2\underline{Q}\}\,, \tag{3.15}$$

where

$$\underline{L} = 2\theta_2(\bar{\mu} : \bar{\varepsilon}) + \frac{4\mu_1}{5\beta}s(\bar{r} : \bar{\varepsilon}) + \frac{8\mu_1}{15\alpha}s(\bar{r} : \bar{\varepsilon}) \tag{3.16}$$

and

$$\begin{aligned}\underline{Q} = {}& \theta_2\mu_2/\mu_1(2\mu_2 - 2\mu_1)^{-1}(\bar{\mu} : \bar{\mu}) + 2\theta_2(2\mu_1)^{-1}(\bar{r} : \bar{\mu}) \\ &+ \left(\frac{s}{5\beta} + \frac{2s}{15\alpha} + (2\mu_1)^{-1} - (2\mu_1)^{-2}2\,\overset{*}{\mu}\right)(\bar{r} : \bar{r})\,.\end{aligned} \tag{3.17}$$

Here $2\,\overset{*}{\mu} = \mathrm{tr}_s\,\overset{*}{C}$, s is interfacial surface area and $\bar{\mu}, \bar{r}$ are the trace free parts of μ and r. Optimization of (3.15) yields:

Theorem 3.2. *Lower bound for the shear trace.*

$$\mathrm{tr}_S C^e \geq 2\mu_1 - 2\mu_1((1 - (2\mu_1)^{-1}2\,\overset{*}{\mu})^{-1} + (2\mu_1\theta_2 c)^{-1})^{-1}\,, \tag{3.18}$$

where

$$c = sp/\theta_2 - \Delta_s \tag{3.19}$$

and

$$p = \frac{1}{5}\left(\frac{1}{\beta} + \frac{2}{3\alpha}\right), \quad \Delta_s = (2\mu_1)^{-1} - (2\mu_2)^{-1}\,. \tag{3.20}$$

Proceeding in a similar manner we obtain a lower bound for the bulk trace:

Theorem 3.3. *Lower bound for the bulk trace.*

$$\mathrm{tr}_b C^e \geq 3\kappa_1 - 3\kappa_1((1 - (3\kappa_1)^{-1}3\,\overset{*}{\kappa})^{-1} + (3\kappa_1\theta_2\ell)^{-1})^{-1}\,, \tag{3.21}$$

where

$$\ell = s/(3\alpha\theta_2) - \Delta_b\,, \tag{3.22}$$

and $3\,\overset{*}{\kappa} = \mathrm{tr}_b C^*$, $\Delta_b = (3\kappa_1)^{-1} - (3\kappa_2)^{-1}$.

We denote the lower bound in Theorem 3.2 by $ICLS(\overset{*}{\mu}, p)$ and investigate its behavior in the geometric parameter $\overset{*}{\mu}$ and interfacial material compliance parameter p.

Elementary estimates give

$$0 \leq \overset{*}{\mu} \leq \theta_1\mu_1 \tag{3.23}$$

and we have $p \geq 0$.

Analysis shows for fixed $\overset{*}{\mu}$ $ICLS(\overset{*}{\mu}, p)$ is monotone decreasing in p and

$$\mathrm{tr}_s C^e \geq ICLS(\overset{*}{\mu}, p) \geq ICLS(\overset{*}{\mu}, \infty) = 2\,\overset{*}{\mu}\ . \tag{3.24}$$

On the other hand, for $p \geq 0$ analysis shows that the bound is monotone increasing in $\overset{*}{\mu}$ and

$$\mathrm{tr}_s C^e \geq ICLS(\overset{*}{\mu}, p) \geq ICLS(0, p)\,, \tag{3.25}$$

where

$$ICLS(0,p) = \left(\frac{\theta_1}{2\mu_2} + \frac{\theta_2}{2\mu_2} + ps\right)^{-1}\ . \tag{3.26}$$

We investigate the behavior of the lower bound on the bulk trace given by Theorem 3.3. The lower bound is denoted by $ICLB(\overset{*}{\kappa}, \alpha^{-1})$. Elementary estimates give

$$0 \leq \overset{*}{\kappa} \leq \theta_1 \kappa_1 \tag{3.27}$$

and we note $\alpha^{-1} \geq 0$. For $\overset{*}{\kappa}$ fixed analysis shows that $ICLB$ is monotone decreasing in α^{-1} and

$$\mathrm{tr}_b C^e \geq ICLB(\overset{*}{\kappa}, \alpha^{-1}) \geq ICLB(\overset{*}{\kappa}, \infty) = 3\,\overset{*}{\kappa}\ . \tag{3.28}$$

On the other hand, for α^{-1} fixed the bound is monotone increasing in $\overset{*}{\kappa}$ and

$$\mathrm{tr}_b C^e \geq ICLB(\overset{*}{\kappa}, \alpha^{-1}) \geq ICLB(0, \alpha^{-1}) = \left(\frac{\theta_1}{3\kappa_1} + \frac{\theta_2}{3\kappa_2} + \frac{s}{3\alpha}\right)^{-1}\ . \tag{3.29}$$

We observe that $ICLB(0, \alpha^{-1})$ agrees with the bound obtained by Hashin.[6]

3.2. *Upper bounds for particulate composites*

We consider a compliant matrix with elasticity $\mathcal{C}_1$ reinforced with particle of elasticity $\mathcal{C}_2$. As before we consider a distribution of particles none of which intersect the boundary of the domain Q. We denote the region occupied by the mth particle by Y_m and its boundary by ∂Y_m. The center of mass of the mth particle is denoted by r^m and for a point x on the particle surface we introduce the normalized coordinate $y^m = x - r^m$. We now introduce the following tensors:

$$B_{ijk\ell} = \sum_m \int_{\partial Y^m} \frac{1}{4}(n_i y_k^m \delta_{j\ell} + n_j y_k^m \delta_{i\ell} + n_i y_\ell^m \delta_{jk} + n_j y_\ell^m \delta_{ik})dS\,, \tag{3.30}$$

$$M_{ijk\ell} = \sum_m \int_{\partial Y^m} \frac{1}{4}(y_i^m y_\ell^m \delta_{kj} + y_i^m y_k^m \delta_{\ell j} + y_j^m y_\ell^m \delta_{ki} + y_j^m y_k^m \delta_{\ell i})dS\,, \tag{3.31}$$

$$R_{ijk\ell} = \sum_m \int_{\partial Y^m} \frac{1}{4}(y_i^m n_j y_k^m n_\ell + y_i^m n_j y_\ell^m n_k + y_j^m n_i y_k^m n_\ell + y_j^m n_i y_\ell^m n_k)dS\,, \tag{3.32}$$

$$\mathcal{T} = 2\int_Q \chi_1 \mathcal{C}_2 N \mathcal{C}_2 \chi_1 dx - \int_Q \chi_1 \mathcal{C}_2 N \mathcal{C}_2 N \mathcal{C}_2 \chi_1 dx\,, \tag{3.33}$$

and

$$A = \theta_1\theta_2\mathcal{C}_2 - \mathcal{T}\,. \tag{3.34}$$

The tensor $\mathcal{T}$ defined by (3.34) contains two-point correlation information on the composite microstructure. To see this we introduce the two-point correlation function:

$$c(t) = \int_Q \chi_1(x)\chi_1(x+t)dx\,.$$

This function gives the probability that the ends of a rod of length and orientation described by the vector t lies in both phases. Noting that $|\hat{\chi}_1(k)|^2 = \widehat{\chi_1 * \chi_1}(k)$ we see that $\hat{c}(k) = |\hat{\chi}_1(k)|^2$ and a computation shows that $\mathcal{T}$ is written:

$$\begin{aligned}\mathcal{T} &= 2\sum_{k\neq 0} \hat{c}(k)\mathcal{C}_2((2\mu_2)^{-1}\Gamma_s(\hat{k}) + r\Gamma_h(\hat{k}))\mathcal{C}_2 \\ &\quad - \sum_{k\neq 0} \hat{c}(k)\mathcal{C}_2((2\mu_2)^{-1}\Gamma_s(\hat{k}) + r\Gamma_h(\hat{k}))\mathcal{C}_2((2\mu_2)^{-1}\Gamma_s(\hat{k}) + r\Gamma_h(\hat{k}))\mathcal{C}_2\,.\end{aligned}$$

The tensor inequality bounding the effective tensor from above is given by:

Theorem 3.4. *Upper bound. For any* 3×3 *constant stress* $\bar{\sigma}$:

$$\begin{aligned}&(\mathcal{C}^{e^{-1}} - \mathcal{C}_2^{-1})\bar{\sigma} : \bar{\sigma} \\ &\quad \geq \begin{pmatrix}\theta_1\bar{\sigma} \\ B\bar{\sigma}\end{pmatrix}^{\mathrm{T}} \begin{pmatrix}\theta_1(\mathcal{C}_1^{-1} - \mathcal{C}_2^{-1})^{-1} + A & -A \\ -A & \beta M + (\alpha-\beta)R + A\end{pmatrix}^{-1} \begin{pmatrix}\theta_1\bar{\sigma} \\ B\bar{\sigma}\end{pmatrix}.\end{aligned} \tag{3.35}$$

Proof. For any pair of 3×3 symmetric matrices we make the choice $p = \chi_1\mu$ and $v = r_{ij}y_j^m$ on ∂Y_m. Upon substitution into the variational principle given by Theorem 2.3 and choosing $\gamma = \mathcal{C}_2$ we apply (2.52) and obtain after a tedious computation the bound:

$$(\mathcal{C}^{e^{-1}} - \mathcal{C}_2^{-1})\bar{\sigma} : \bar{\sigma} \geq \max_{(\mu,r)}\{2\bar{L}(\mu,r) - \bar{Q}(\mu,r)\}\,, \tag{3.36}$$

where

$$\bar{L}(\mu,r) = \theta_1\bar{\sigma} : \mu + B\bar{\sigma} : r \tag{3.37}$$

and

$$\bar{Q}(\mu,r) = \theta_1(\mathcal{C}_1^{-1} - \mathcal{C}_2^{-1})^{-1}\mu : \mu + (\beta M + (\alpha-\beta)R)r : r + A(\mu - r) : (\mu - r)\,. \tag{3.38}$$

The theorem follows upon maximization of (3.36) over μ and r. □

The upper bounds hold for any anisotropic particulate mixture. Similar methods yield upper bounds on the bulk and shear trace for the effective tensor. Returning to (3.37) we average both sides over the isotropic group of rotations. Choosing $\varepsilon = \bar{\varepsilon}$ where $\mathrm{tr}\bar{\varepsilon} = 0$ and optimization over trace-free choices of μ and r gives the upper bound on the shear trace of the effective tensor. After a straightforward but tedious computation we obtain:

Theorem 3.5. *Upper bound on shear trace of the effective compliance.*

$$(\mathrm{tr}_s \mathcal{C}^{e^{-1}})^{-1} \leq \left((2\mu_1)^{-1} + \frac{(\theta_2^2 - g_s\theta_2\Delta_s)(1+\Delta_s(1-t_s)2\mu_2)}{g_s(1+\theta_2\Delta_s(1-t_s)2\mu_2)+\theta_1\theta_2(1-t_s)2\mu_2}\right)^{-1}, \quad (3.39)$$

where $\Delta_s = (2\mu_1)^{-1} - (2\mu_2)^{-1}, t_s = (6/5)(\kappa_2 + 2\mu_2)/(3\kappa_2 + 4\mu_2)$, and

$$\begin{aligned} g_s &= \beta \mathrm{tr}_s M + (\alpha - \beta)\mathrm{tr}_s R \\ &= (\beta/5)(\frac{7}{6}\sum_m \int_{\partial Y^m} | y^m |^2 \, dS - \frac{1}{6}\sum_m \int_{\partial Y^m} (y^m \cdot n)^2 dS) \\ &\quad + (\alpha/5)(\frac{1}{2}\sum_m \int_{\partial Y^m} | y^m |^2 \, dS + \frac{1}{6}\sum_m \int_{\partial Y^m} (y^m \cdot n)^2 dS). \end{aligned} \quad (3.40)$$

We note that application of Cauchy's inequality to the integrands in (3.40) implies that g_s is increasing in the interfacial stiffnesses α and β and $g_s \geq 0$.

Proceed in a similar fashion we obtain

Theorem 3.6. *Upper bound on bulk trace of the effective compliance.*

$$(\mathrm{tr}_b \mathcal{C}^{e^{-1}})^{-1} \leq \left((3\kappa_1)^{-1} + \frac{(\theta_2^2 - g_b\theta_2\Delta_b)(1+\Delta_b(1-t_b)3\kappa_2)}{g_b(1+\theta_2\Delta_b(1-t_b)3\kappa_2)+\theta_1\theta_2(1-t_b)3\kappa_2}\right)^{-1}, \quad (3.41)$$

where $\Delta_b = (3\kappa_1)^{-1} - (3\kappa_2)^{-1}, \;\; t_b = 3\kappa_2/(3\kappa_2 + 4\mu_2)$, and

$$\begin{aligned} g_b &= \beta \mathrm{tr}_b M + (\alpha - \beta)\mathrm{tr}_b R \\ &= (\beta/3)(\sum_m \int_{\partial Y^m} | y^m |^2 \, dS) + \frac{(\alpha-\beta)}{3}\sum_m \int_{\partial Y^m} (y^m \cdot n)^2 dS. \end{aligned} \quad (3.42)$$

Here an application of Cauchy's inequality shows that g_b is increasing in the parameters α and β and $g_b \geq 0$. We denote the upper bound in Theorem 3.5 by $ICUS(g_s)$ and note that for θ_2 fixed it is monotone increasing in g_s. In the limit as either β or α goes to ∞ one has that $g_s = \infty$ and

$$ICUS(\infty) = 2\mu_2 + \frac{\theta_1}{\frac{1}{2\mu_1 - 2\mu_2} + \frac{3\theta_2(\kappa_2+2\mu_2)}{5\mu_2(3\kappa_2+4\mu_2)}}. \quad (3.43)$$

The expression given above is precisely the upper shear modulus bound for isotropic composites with perfect interface obtained by Hashin and Shtrikman.[7] Analogously we denote the upper bound given by Theorem 3.6 by $ICUB(g_b)$. For fixed θ_2 this bound is also monotone increasing in g_b and in the limit as either β or α tends to ∞, one has:

$$ICUB(\infty) = 3\kappa_2 + \frac{\theta_1}{\frac{1}{3\kappa_1 - 3\kappa_2} + \frac{\theta_2}{3\kappa_2+4\mu_2}}. \quad (3.44)$$

This expression is the upper bulk modulus bound obtained by Hashin and Shtrikman[7] for isotropic composites with perfect interface.

4. Effective Behavior for Monodisperse Suspensions of Spheres at Critical Particle Size

In this section we consider a monodisperse suspension of relatively stiff isotropic elastic spheres with elasticity $\mathcal{C}_2$ embedded in a softer matrix with isotropic elasticity $\mathcal{C}_1$. For prescribed interfacial spring constant α, we exhibit a critical particle radius R_b^c for which the effective bulk trace $\mathrm{tr}_b\mathcal{C}^e$ of the composite is identical to that of the matrix. We show that:

$$R_b^c = \alpha^{-1}/\Delta_b \,. \tag{4.1}$$

When the interfacial spring constants α and β are equal, we exhibit a critical particle radius R_s^c for which the effective shear trace $\mathrm{tr}_s\mathcal{C}^e$ equals that of the matrix material. We show that:

$$R_s^c = \beta^{-1}/\Delta_s \,. \tag{4.2}$$

At these critical radii the effect of the interface is balanced by the greater elastic stiffness of the particles.

We begin with the obvious remark that for composites occupying the unit cube we only consider parameter values $\kappa_1, \kappa_2, \mu_1, \mu_2$ for which both R_b^c and R_s^c are less than one-half. We consider a dispersion of N spheres of common radius a with centers denoted by r^i. We assume all spheres are contained in the unit cube and do not touch. The formula for the critical radii follow from the following two theorems.

Theorem 4.1. *Given a prescribed hydrostatic strain λI, if the common radii of the particles equal R_b^c then there exists a periodic piecewise affine solution to the problem* (2.6)–(2.9) *given by:*

$$\tilde{\phi}_j + \lambda x_j = \begin{cases} \lambda x_j & \text{in the matrix,} \\ \dfrac{\kappa_1}{\kappa_2}\lambda\xi_j + \left(1 - \dfrac{\kappa_1}{\kappa_2}\right)\lambda r_j^i & \text{in the } i\text{th particle.} \end{cases} \tag{4.3}$$

Theorem 4.2. *Given a prescribed trace free constant strain ε, if the interfacial spring constants α, β satisfy $\alpha = \beta$ and if the common radii of the particles equals R_s^c then there exists a periodic piecewise affine solution to the problem* (2.6)–(2.9) given by:

$$\tilde{\phi}_j + \varepsilon_{jk} x_k = \begin{cases} \varepsilon_{jk} x_k & \text{in the matrix,} \\ \dfrac{\mu_1}{\mu_2}\varepsilon_{jk} x_k + \left(1 - \dfrac{\mu_1}{\mu_2}\right)\varepsilon_{jk} r_k^i & \text{in the } i\text{th particle.} \end{cases} \tag{4.4}$$

Remark. We point out that Theorems 4.1 and 4.2 show that the elastic field in the matrix remains undisturbed when the spheres are at critical radius!

Substitution of (4.3) and (4.4) into (2.11) yields the following:

Corollary 4.3. *For α fixed and for any value of β if one applies an average hydrostatic strain λI of the form (2.2) to a suspension of N spheres each at critical radius R_b^c, then the resulting average stress is given by*

$$3\kappa_1(\lambda I)\,. \tag{4.5}$$

Corollary 4.4. *For $\alpha = \beta$ fixed, if one applies an average derivatoric strain ε^{D} ($\mathrm{tr}\varepsilon^{\mathrm{D}} = 0$) of the form (2.2) to a suspension of N spheres each at critical radius R_s^c, then the resulting average stress is given by*

$$2\mu_1\varepsilon^D\,. \tag{4.6}$$

It follows that:

Corollary 4.5. *Critical radius for the bulk trace. For α fixed, if the common radius of the suspension equals R_b^c, then*

$$\mathrm{tr}_b C^e = 3\kappa_1\,. \tag{4.7}$$

Corollary 4.6. *Critical radius for the shear trace. For $\alpha = \beta$, if the common radius of the suspension equals R_s^c, then*

$$\mathrm{tr}_s C^e = 2\mu_1\,. \tag{4.8}$$

Proof of (4.1) and (4.2). For a prescribed average strain ε we look for a solution of (2.6)–(2.9) of the form

$$\tilde{\varphi}_j + \varepsilon_{jk}x_k = \begin{cases} \varepsilon^A_{jk}x_k & \text{in the matrix} \\ \varepsilon^B_{jk}x_k + v^i_j & \text{in the } i\text{th particle}. \end{cases} \tag{4.9}$$

From (2.2), (2.7)–(2.9) it follows that:

$$\varepsilon + \int_\Gamma [\tilde{\varphi}] \odot n dS = \theta_1\varepsilon^A + \theta_2\varepsilon^B\,, \tag{4.10}$$

$$C_1\varepsilon^A n = C_2\varepsilon^B n\,, \tag{4.11}$$

$$(C_2\varepsilon^B n)_\tau = -\beta\{(\varepsilon^B - \varepsilon^A)x + v^i\}_\tau\,, \tag{4.12}$$

and

$$(C_2\varepsilon^B n)_n = -\alpha\{(\varepsilon^B - \varepsilon^A)x + v^i\}_n\,. \tag{4.13}$$

We solve the system (4.10)–(4.13) to find the unknowns $\varepsilon^A, \varepsilon^B, v^i, i = 1, 2, \ldots, N$, and the sphere radius.

From (4.11) we may conclude that $C_1^{-1}C_2\varepsilon^B = \varepsilon^A$. On the surface of the ith sphere the unit normal is written $n = (x - r^i)/a$, thus $x = an + r^i$ on the surface and (4.12), (4.13) are written as

$$\left(\left\{3\kappa_2 + \beta a\left(1 - \frac{\kappa_2}{\kappa_1}\right)\right\}\mathbb{P}_I\varepsilon^B n\right)_\tau + \left(\left\{2\mu_2 + \beta a\left(1 - \frac{\mu_2}{\mu_1}\right)\right\}\mathbb{P}_s\varepsilon^B n\right)_\tau$$
$$= -\beta\left\{\left(\left(1 - \frac{\kappa_2}{\kappa_1}\right)\mathbb{P}_I\varepsilon^B r^i\right)_\tau + \left(\left(1 - \frac{\mu_2}{\mu_1}\right)\mathbb{P}_s\varepsilon^B r^i\right)_\tau + v^i_\tau\right\} \tag{4.14}$$

and

$$\left(\left\{3\kappa_2 + \alpha a\left(1 - \frac{\kappa_2}{\kappa_1}\right)\right\}\mathbb{P}_I\varepsilon^B n\right)_n + \left(\left\{2\mu_2 + \alpha a\left(1 - \frac{\mu_2}{\mu_1}\right)\right\}\mathbb{P}_s\varepsilon^B n\right)_n$$
$$= -\alpha\left\{\left(\left(1 - \frac{\kappa_2}{\kappa_1}\right)\mathbb{P}_I\varepsilon^B r^i\right)_n + \left(\left(1 - \frac{\mu_2}{\mu_1}\right)\mathbb{P}_s\varepsilon^B r^i\right)_n + v^i_n\right\}. \quad \square \tag{4.15}$$

We observe that (4.14) is an equation of the form

$$(Ln)_\tau = q_\tau\,, \tag{4.16}$$

where L is a constant symmetric matrix and q is a constant vector. Here (4.16) holds for all unit vectors n. It is evident that (4.16) is not satisfied by every symmetric matrix L or vector q, indeed one has

Lemma 4.7. *Equation* (4.16) *holds for all unit vectors only when* $q = 0$ *and* L *is a multiple of the identity.*

Proof. To show $q = 0$ we let $n = e^i, i = 1, 2, 3$ where e^i are eigenvectors of L associated with eigenvalues $L_i, i = 1, 2, 3$. Substitution into (4.16) gives

$$L_i e^i - q = L_i e^i - q_i e^i \tag{4.17}$$

or

$$q = q_1 e^1 = q_2 e^2 = q_3 e^3\,, \tag{4.18}$$

where q_i are the components of q. Since e^i are all pairwise orthogonal we conclude that $q_i = 0$. Thus we have

$$Ln = (Ln \cdot n)n \tag{4.19}$$

for all unit vectors n. Next we show that all eigenvalues of L are identical to conclude that L is a multiple of the identity. Choosing two unit vectors v^1, v^2 such that $v^1 \cdot v^2 \neq 0$ we observe that $Lv^1 \cdot v^2 = Lv^2 \cdot v^1$ and apply (4.19) to obtain

$$(Lv^1 \cdot v^1 - Lv^2 \cdot v^2)v^1 \cdot v^2 = 0\,, \tag{4.20}$$

hence $Lv^1 \cdot v^1 = Lv^2 \cdot v^2$. From continuity it follows that the function $Ln \cdot n$ is constant for all unit vectors and so all eigenvalues of L agree. $\square$

Application of Lemma 4.7 to Eq. (4.14) shows that there exists a constant c such that:

$$\left\{3\kappa_2 + \beta a\left(1 - \frac{\kappa_2}{\kappa_1}\right)\right\}\mathbb{P}_I\varepsilon^B = cI\,, \tag{4.21}$$

$$\left\{2\mu_2 + \beta a\left(1 - \frac{\mu_2}{\mu_1}\right)\right\}\mathbb{P}_s\varepsilon^B = 0\,, \tag{4.22}$$

and

$$\left(1 - \frac{\kappa_2}{\kappa_1}\right)\mathbb{P}_I\varepsilon^B r^i + \left(1 - \frac{\mu_2}{\mu_1}\right)\mathbb{P}_s\varepsilon^B r^i + v^i = 0\,. \tag{4.23}$$

Since $\mathbb{P}_I\varepsilon^B = (1/3\mathrm{tr}\varepsilon^B)I$, it is evident that Eq. (4.21) is satisfied for all values of β, a, κ_1 and κ_2. On the other hand, Eq. (4.15) is of the form:

$$h + q\cdot n + Ln\cdot n = 0 \tag{4.24}$$

for all unit vectors n, where h is a scalar, q is a constant vector and $\mathrm{tr}L = 0$. We adopt the frame of the eigenbasis of L and write (4.24) as:

$$h + q\cdot n + L_1 n_1^2 + L_2 n_2^2 + L_3 n_3^2 = 0\,. \tag{4.25}$$

Since $\mathrm{tr}L = 0$ we may assume $L_1 \geq L_2 \geq L_3$ with $L_1 \geq 0$ and $L_3 \leq 0$. It is evident that for $L_1 \geq 0$ and $L_3 \leq 0$, Eq. (4.25) never describes a sphere. Thus (4.24) holds for all unit vectors only if

$$L = 0, \quad q = 0 \,\mathrm{and}\, h = 0\,. \tag{4.26}$$

Applying (4.26) to (4.15) we recover (4.23) as well as the new identities given by,

$$\left(3\kappa_2 + \alpha a\left(1 - \frac{\kappa_2}{\kappa_1}\right)\right)\mathbb{P}_I\varepsilon^B = 0\,, \tag{4.27}$$

and

$$(2\mu_2 + \alpha a(1 - \frac{\mu_2}{\mu_1}))\mathbb{P}_s\varepsilon^B = 0\,. \tag{4.28}$$

We observe from (2.8) and (2.9) that:

$$[\tilde{\phi}_\tau] = -\beta^{-1}(C_2\varepsilon^B n)_\tau = -\beta^{-1}\left(C_2\varepsilon^B\frac{x - r^i}{a}\right)_\tau\,, \tag{4.29}$$

and

$$[\tilde{\phi}_n] = -\alpha^{-1}(C_2\varepsilon^B n)_n = -\alpha^{-1}\left(C_2\varepsilon^B\left(\frac{x - r^i}{a}\right)\right)_n\,. \tag{4.30}$$

Substitution of (4.29) and (4.30) into (4.10) together with a long but straightforward calculation yields:

$$\mathbb{P}_I\varepsilon^B = \left(\theta_1\frac{\kappa_2}{\kappa_1} + \theta_2 + \theta_2\frac{3\kappa_2}{a\alpha}\right)^{-1}\mathbb{P}_I\varepsilon \tag{4.31}$$

and

$$\mathbb{P}_s\varepsilon^B = \left(\theta_1\frac{\mu_1}{\mu_1} + \theta_2 + \frac{\theta_2 12\mu_2}{5a}\left(\frac{1}{2\beta} + \frac{1}{3\alpha}\right)\right)^{-1}\mathbb{P}_s\varepsilon. \tag{4.32}$$

To recover Theorem 4.1 we let $\varepsilon = \lambda I$. For this choice $\mathbb{P}_s\varepsilon = 0$ and from (4.32) it follows that $\mathbb{P}_s\varepsilon^B = 0$ and Eqs. (4.22) and (4.28) are satisfied. From (4.31) it follows that $\mathbb{P}_I\varepsilon^B \neq 0$, hence (4.27) holds only if

$$3\kappa_2 + \alpha a\left(1 - \frac{\kappa_2}{\kappa_1}\right) = 0\,. \tag{4.33}$$

Equation (4.33) provides the relation defining the critical radius. Indeed (4.33) holds provided that $a = R_b^c$. For this choice Eq. (4.31) gives:

$$\text{tr}\varepsilon^B = \frac{\kappa_1}{\kappa_2}\text{tr}\varepsilon\,. \tag{4.34}$$

As can be seen earlier we have $\varepsilon^A = C_1^{-1}C_2\varepsilon^B$ and so

$$\text{tr}\varepsilon^A = \text{tr}\varepsilon\,. \tag{4.35}$$

Theorem 4.1 follows from substitution of (4.34) into (4.23) and Eqs. (4.9) and (4.35).

Theorem 4.2 is established by setting $\varepsilon = \varepsilon^D$. For this choice $\mathbb{P}_I\varepsilon^D = 0$ and from (4.31) it follows that $\mathbb{P}_I\varepsilon^B = 0$ and Eq. (4.27) is satisfied. From (4.32) it follows that $\mathbb{P}_s\varepsilon^B \neq 0$ and so Eqs. (4.22) and (4.28) hold only if

$$2\mu_2 + \beta a\left(1 - \frac{\mu_2}{\mu_1}\right) = 0\,, \tag{4.36}$$

and

$$2\mu_2 + \alpha a\left(1 - \frac{\mu_2}{\mu_1}\right) = 0\,. \tag{4.37}$$

Equations (4.36) and (4.37) provide the relations defining the critical radius. Indeed for $\alpha = \beta$ (4.36), (4.37) hold provided that $a = R_s^c$. For this choice we find

$$\varepsilon^B = \frac{\mu_1}{\mu_2}\varepsilon^D \text{ and } \varepsilon^A = \varepsilon^D\,. \tag{4.38}$$

Theorem 4.2 follows from substitution of $(4.38)_1$ into (4.23) and Eq. (4.9).

5. Comparison with the Bounds of Hashin

Hashin[6] derives extensions of the classical variational principles for the case of imperfect contact. The interface is described by two tangential stiffnesses D_t, D_s, and a normal stiffness D_n. For isotropic composites these principles are used to obtain bounds on effective shear μ^e and bulk moduli κ^e. When $D_s = D_t = \beta$ and

$D_n = \alpha$ the bounds reported in Ref. 6 are given by:

$$HLS = \left(\frac{\theta_1}{2\mu_1} + \frac{\theta_2}{2\mu_2} + \frac{s}{5}\left(\frac{2}{\beta} + \frac{2}{3\alpha}\right)\right)^{-1} \leq 2\mu^e$$
$$\leq \theta_1 2\mu_1 + \theta_2 2\mu_2 \left(\frac{1}{1+\alpha_s}\right) = HUS\,, \tag{5.1}$$

$$HLB = \left(\frac{\theta_1}{3\kappa_1} + \frac{\theta_2}{2\kappa_2} + \frac{s}{3\alpha}\right)^{-1} \leq 3\kappa^e \leq \theta_1 3\kappa_1 + \theta_2 3\kappa_2 \left(\frac{1}{1+\alpha_b}\right) = HUB\,, \tag{5.2}$$

where

$$\alpha_s = \theta_2 2\mu_2/(\delta + g_s)\,, \tag{5.3}$$

$$\alpha_b = \theta_2 3\kappa_2/g_b \tag{5.4}$$

with s denoting interfacial surface area and

$$\delta = (\alpha/10)\sum_m \int_{\partial Y_m} (|y^m|^2 - (y^m \cdot n)^2)ds + (\beta/10)\sum_m \int_{\partial Y_m} (|y^m|^2 + (y^m \cdot n)^2)ds\,. \tag{5.5}$$

The upper and lower bounds for the shear modulus reported in Hashin[6] (i.e. HLS and HUS) are incorrect due to a computational error. The corrected upper and lower bounds are given by:

$$LS = \left(\frac{\theta_1}{2\mu_1} + \frac{\theta_2}{2\mu_2} + \frac{s}{5}\left(\frac{1}{\beta} + \frac{2}{3\alpha}\right)\right)^{-1} \leq 2\mu^e \leq \theta_1 2\mu_1 + \theta_2 2\mu_2 \left(\frac{1}{1+\tilde{\alpha}_s}\right) = US\,, \tag{5.6}$$

where

$$\tilde{\alpha}_s = \theta_2 2\mu_2/g_s\,. \tag{5.7}$$

We first show that the parameter δ appearing in (5.3) is redundant. We introduce the tensor P as given in Ref. 6 by:

$$P = \alpha J^n + \beta(J^t + J^s)\,, \tag{5.8}$$

where

$$J^n_{ijkl} = \sum_m \int_{\partial Y^m} n_i y^m_j n_k y^m_\ell ds\,, \tag{5.9}$$

$$J^t_{ijk\ell} = \sum_m \int_{\partial Y^m} t_i y^m_j t_k y^m_\ell ds \tag{5.10}$$

and

$$J^s_{ijk\ell} = \sum_m \int_{\partial Y^m} s_i y^m_j s_k y^m_\ell ds\,. \tag{5.11}$$

Here t and s form an orthonormal basis in the plane tangent to the two-phase interface. Noting that P is a symmetric map on 3×3 matrices we see for isotropic suspensions that P is isotropic and is written

$$P = g_b \mathbb{P}_I + g_s \mathbb{P}_s + \frac{5}{3} \delta \mathbb{P}_A \,, \tag{5.12}$$

where $\mathbb{P}_A$ is the projection onto antisymmetric matrices given by

$$(\mathbb{P}_A)_{ijk\ell} = \frac{1}{2} \left(\delta_{ik} \delta_{j\ell} - \delta_{i\ell} \delta_{jk} \right) \tag{5.13}$$

and

$$g_b = \frac{1}{3} P_{iijj} \,, \tag{5.14}$$

$$g_s = \frac{1}{5} \left(\frac{1}{2} \left(P_{ijij} + P_{ijji} \right) - \frac{1}{3} P_{iijj} \right) \,, \tag{5.15}$$

$$\frac{5}{3} \delta = \frac{1}{6} \left(P_{ijij} - P_{ijji} \right) \,. \tag{5.16}$$

We point out that Eq. (28) of Hashin[6] is incorrect. Indeed, in (28) of Ref. 6 the restriction of P to 3×3 symmetric matrices is written as:

$$3P^{(1)} \mathbb{P}_I + 2P^{(2)} \mathbb{P}_s \,, \tag{5.17}$$

where

$$3P^{(1)} = \frac{1}{3} P_{iijj} \tag{5.18}$$

and

$$2P^{(2)} = \frac{1}{5} \left(P_{ijij} - \frac{1}{3} P_{iijj} \right) = g_s + \delta \,. \tag{5.19}$$

We see from (5.12) and (5.15) that the trace given by $2P^{(2)}$ is in error. It should instead be computed as in (5.15). We apply the correct formula for P given by (5.12)–(5.16) and proceed as in Ref. 6 to obtain the corrected upper bound "US" given by (5.6).

A similar computational error is present in the lower bound displayed in (5.1). Introducing the tensor Q given by Hashin[6]:

$$Q = \frac{1}{\alpha} L^n + \frac{1}{\beta} (L^t + L^s) \,, \tag{5.20}$$

where

$$L^n_{ijk\ell} = \int_\Gamma n_i n_j n_k n_\ell ds \,, \tag{5.21}$$

$$L^s_{ijk\ell} = \int_\Gamma s_i n_j s_k n_\ell ds \,, \tag{5.22}$$

and

$$L^t_{ijk\ell} = \int_\Gamma t_i n_j t_k n_\ell ds \,. \tag{5.23}$$

We observe as before that Q is a symmetric map on 3×3 matrices. For isotropic suspensions, Q is isotropic and is written

$$Q = \lambda_I \mathbb{P}_I + \lambda_D \mathbb{P}_s + \lambda_A \mathbb{P}_A \,, \tag{5.24}$$

where

$$\lambda_I = \frac{1}{3} Q_{iijj} = \frac{s}{3\alpha} \,, \tag{5.25}$$

$$\lambda_D = \frac{1}{5}\left(\frac{1}{2}\Big(Q_{ijij} + Q_{ijji}\Big) - \frac{1}{3} Q_{iijj}\right) = \frac{s}{5}\left(\frac{1}{\beta} + \frac{2}{3\alpha}\right), \tag{5.26}$$

and

$$\lambda_A = \frac{1}{2}(Q_{ijij} - Q_{ijji}) = \frac{s}{\beta} \,. \tag{5.27}$$

We note that in Eq. (39) of Ref. 6 the restriction of Q to 3×3 symmetric matrices is written as

$$3Q^{(1)} \mathbb{P}_I + 2Q^{(2)} \mathbb{P}_s \,, \tag{5.28}$$

where

$$3Q^{(1)} = \frac{1}{3} Q_{iijj} \tag{5.29}$$

and

$$2Q^{(2)} = \frac{1}{5}\left(Q_{ijij} - \frac{1}{3} Q_{iijj}\right) = \frac{s}{5}\left(\frac{2}{\beta} + \frac{2}{3\alpha}\right). \tag{5.30}$$

We see from (5.24) and (5.26) that the trace given by $2Q^{(2)}$ is incorrect and should be computed as in (5.26). Applying the correct formula for Q given by (5.24)–(5.27) and proceeding as in Ref. 6 we obtain the corrected lower bound on the shear modulus "LS" given by (5.6).

We now compare the bounds developed in the previous sections to the Hashin bulk modulus bounds given in (5.2) and with the corrected shear bounds given by (5.6) and (5.7). For fixed values of volume fractions θ_1, θ_2 and g_s a straightforward calculation shows:

$$(\mathrm{tr}_s C^{e^{-1}})^{-1} \leq ICUS(g_s) \leq US(g_s) \tag{5.31}$$

and

$$\mathrm{tr}_s C^e \geq ICLS(\overset{*}{\mu}, p) \geq ICLS(0, p) = LS \,, \tag{5.32}$$

where $ICUS$ is the upper bound given in Theorem 3.5 and $ICLS$ is the lower bound given in Theorem 3.2. We remind the reader that the geometric parameters $g_s, \overset{*}{\mu}$ lie in the ranges $0 \leq g_s, 0 \leq \overset{*}{\mu} \leq \theta_1 \mu_1$ and that, $US(g_s), ICUS(g_s), ICLS(\overset{*}{\mu}, p)$ are monotone increasing in g_s and $\overset{*}{\mu}$.

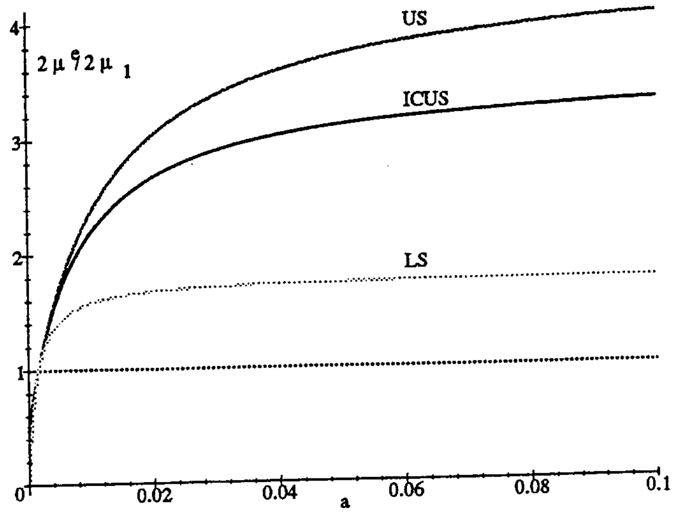

Fig. 1. Comparison between the new effective shear trace bounds ICUS, US and LS for monodisperse suspension of graphite spheres in an epoxy matrix. The volume fraction of spheres is fixed at 50% and the sphere radius is less than 0.1 m.

The new upper bound $ICUS$ together with the corrected upper and lower bounds US and LS are plotted in Fig. 1 for a monodisperse suspension of graphite spheres in an epoxy matrix. For monodisperse suspensions of spheres of radius a, the parameters $g_s = \frac{1}{5}(3\beta+2\alpha)\theta_2 a$ and $s/\theta_2 = 3/a$. The bounds on the shear trace are plotted for radii between 0 and 0.1×10^{-2} m, with sphere volume fraction fixed at 50%. The interfacial stiffnesses are chosen to be $\alpha = \beta = 1 \times 10^6$ MPa/m. Here, the shear moduli and Poisson's ratios of the spheres are $\mu_1 = 0.7 \times 10^3$ MPa and $\nu_1 = 0.33$; the moduli for the matrix are $\mu_2 = 5.5 \times 10^3$ MPa, $\nu_2 = 0.25$. Note that for sufficiently small sphere radii the bounds indicate that the shear trace drops below that of the matrix. This topic is pursued in the following section.

For fixed values of volume fractions and g_b we have:

$$(\mathrm{tr}_b C^{e^{-1}})^{-1} \le ICUB(g_b) \le HUB(g_b) \tag{5.33}$$

and

$$\mathrm{tr}_b C^e \ge ICLB(\overset{*}{\kappa}, \alpha^{-1}) \ge ICLB(0, \alpha^{-1}) = HLB\,, \tag{5.34}$$

where $ICUB$ is the upper bound given in Theorem 3.6 and $ICLB$ is the lower bound given in Theorem 3.3. Here, $g_b \ge 0, 0 \le \overset{*}{\kappa} \le \theta_1\kappa_1$ and $ICUB(g_b)$, $ICLB(\overset{*}{\kappa}, \alpha^{-1})$ are strictly monotone increasing in g_b and $\overset{*}{\kappa}$ respectively. In this way we see that the new upper and lower bounds $ICUB$ and $ICLB$ are always tighter than the upper and lower bulk trace bounds in Ref. 6.

6. Size Effects

We apply the bounds given by Theorems 3.2, 3.3, 3.5 and 3.6 and make use of their monotonic behavior in the geometric parameters g_b, g_s and surface area. The monotonicity is used to predict new size effect phenomena for elastic composites with imperfect interfaces. We consider first anisotropic particulate suspensions with no assumption on the distribution or shape of the particles. We introduce the ratios

$$N_s^+ = p/\Delta_s, \quad N_s^- = q/\Delta_s\,, \tag{6.1}$$

$$R_b^c = \alpha^{-1}/\Delta_b\,. \tag{6.2}$$

Here $p = \frac{1}{5}(\frac{1}{\beta} + \frac{2}{3\alpha})$, $q = 5(3\beta + 2\alpha)^{-1}$, $\Delta_s = (\frac{1}{2\mu_1} - \frac{1}{2\mu_2})$ is the contrast between matrix and particle shear compliances, and $\Delta_b = (\frac{1}{3k_1} - \frac{1}{3k_2})$ is the contrast between matrix and particle bulk compliances. The quantities p, q and α^{-1} are measures of the interfacial compliance.

We point out that the parameters N_s^-, N_s^+ and R_b^c estimate the relative importance of the contrast between phase compliances and the interfacial compliance. In what follows we illustrate how these parameters influence the overall elastic properties of composites with imperfect interface.

It is evident that the lower shear trace bound given by Theorem 3.2 is strictly monotone decreasing in the interfacial surface to particle volume ratio "s/θ_2". Moreover the lower bound equals $2\mu_1$ for $s/\theta_2 = (N_s^+)^{-1}$. We collect these observations and state the following theorem.

Theorem 6.1. *For suspensions of particles with elasticity C_2 in a matrix of elasticity C_1: if $s/\theta_2 < (N_s^+)^{-1}$, then*

$$\mathrm{tr}_s C^e > 2\mu_1\,. \tag{6.3}$$

Noting that the lower bulk trace bound given by Theorem 3.3 is also strictly monotone in s/θ_2 and equals $3\kappa_1$ for $s/\theta_2 = 3(R_b^c)^{-1}$, we have:

Theorem 6.2. *For suspensions of particles with elasticity C_2 in a matrix of elasticity C_1: if $s/\theta_2 < 3(R_b^c)^{-1}$, then*

$$\mathrm{tr}_b C^e > 3\kappa_1\,. \tag{6.4}$$

We consider polydisperse suspensions of spheres of C_2 material in a matrix of C_1. The volume average radius of the suspension of N spheres $Y_m, m = 1, 2, \ldots, N$, each of radius a_m is given by

$$\langle a \rangle = \theta_2^{-1} \sum_{m=1}^{N} |Y_m| a_m\,. \tag{6.5}$$

For such suspensions the geometric parameters g_s and g_b defined by (3.40) and (3.42) reduce to:

$$g_s = \frac{1}{5}(3\beta + 2\alpha)\theta_2\langle a\rangle \tag{6.6}$$

and

$$g_b = \alpha\theta_2\langle a\rangle\,. \tag{6.7}$$

For this case the upper bounds on the shear and bulk traces are given respectively by

$$ICUS(g_s) = \left((2\mu_1)^{-1} + \frac{\theta_2^2(1-\langle a\rangle/N_s^-)(1+\Delta_s(1-t_s)2\mu_2)}{\frac{3\theta_2}{5}(\beta+\frac{2\alpha}{3})\langle a\rangle(1+\theta_2\Delta_s(1-t_s)2\mu_s) + \theta_1\theta_2(1-t_s)2\mu_2}\right)^{-1} \tag{6.8}$$

and

$$ICUB(g_b) = \left((3\kappa_1)^{-1} + \frac{\theta_2^2(1-\frac{\langle a\rangle}{R_b^c})(1+\Delta_b(1-t_b)3\kappa_2)}{\alpha\theta_2\langle a\rangle(1+\theta_2\Delta_b(1-t_b)3\kappa_2) + \theta_1\theta_2(1-t_b)3\kappa_2}\right)^{-1}. \tag{6.9}$$

These bounds are strictly monotone increasing in $\langle a\rangle$ and for $\langle a\rangle = N_s^-$,

$$ICUS = 2\mu_1\,, \tag{6.10}$$

and for $\langle a\rangle = R_b^c$ we have

$$ICUB = 3\kappa_1\,. \tag{6.11}$$

Thus from monotonicity we have:

Theorem 6.3. *Size effect theorem for the shear trace. For polydisperse suspensions of spheres of elasticity C_2 in a matrix of elasticity C_1 and if $\langle a\rangle \le N_s^-$, then*

$$\mathrm{tr}_s(C^{e^{-1}}) \ge (2\mu_1)^{-1}\,. \tag{6.12}$$

Theorem 6.4. *Size effect theorem for the bulk trace. For anisotropic polydisperse suspensions of spheres of elasticity C_2 in a matrix of elasticity C_1 and if $\langle a\rangle \le R_b^c$ then:*

$$\mathrm{tr}_b(C^{e^{-1}}) \ge (3\kappa_1)^{-1}\,. \tag{6.13}$$

For monodisperse suspensions of spheres of radius a the geometric parameters g_s and g_b are given by

$$g_s = \frac{1}{5}(3\beta + 2\alpha)\theta_2 a, \quad g_b = \alpha\theta_2 a, \tag{6.14}$$

and the interfacial surface to particle volume ratio s/θ_2 is:

$$s/\theta_2 = 3/a\,. \tag{6.15}$$

We apply Theorems 6.1–6.4 to this case to obtain:

Corollary 6.5. *For anisotropic monodisperse suspensions of spheres of radius a*

$$\mathrm{tr}_s \mathcal{C}^e > 2\mu_1 \text{ if } \quad a > 3N_s^+ , \tag{6.16}$$

$$\mathrm{tr}_b \mathcal{C}^e > 3\kappa_1 \quad \text{if} \quad a > R_b^c , \tag{6.17}$$

$$\mathrm{tr}_s(\mathcal{C}^{e^{-1}}) > (2\mu_1)^{-1} \quad \text{if} \quad a < N_s^- , \tag{6.18}$$

$$\mathrm{tr}_b(\mathcal{C}^{e^{-1}}) > (3\kappa_1)^{-1} \quad \text{if} \quad a < R_b^c . \tag{6.19}$$

Here we note that Jensen's inequality gives $3N_s^+ \geq N_s^-$. Moreover, when the interfacial coefficients α and β are equal, we have $3N_s^+ = N_s^- = R_s^c$. Here the critical radius R_s^c is given by (4.2) and

$$\mathrm{tr}_s \mathcal{C}^e > 2\mu_1 \text{ if } a > R_s^c \tag{6.20}$$

and

$$\mathrm{tr}_s(\mathcal{C}^{e^{-1}}) > (2\mu_1)^{-1} \text{ if } a < R_s^c . \tag{6.21}$$

In Fig. 2 the shear trace bounds are plotted for the monodisperse suspension of graphite spheres in epoxy for small sphere radii, i.e. $a < 0.12 \times 10^{-4}$ m. Here $\alpha = \beta = 1 \times 10^6$ MPa/m. Note that the shear trace upper bounds lie far below that of the matrix shear trace. In Fig. 3 we plot the shear trace bounds near the critical radius $R_s^c = 0.0016$ m.

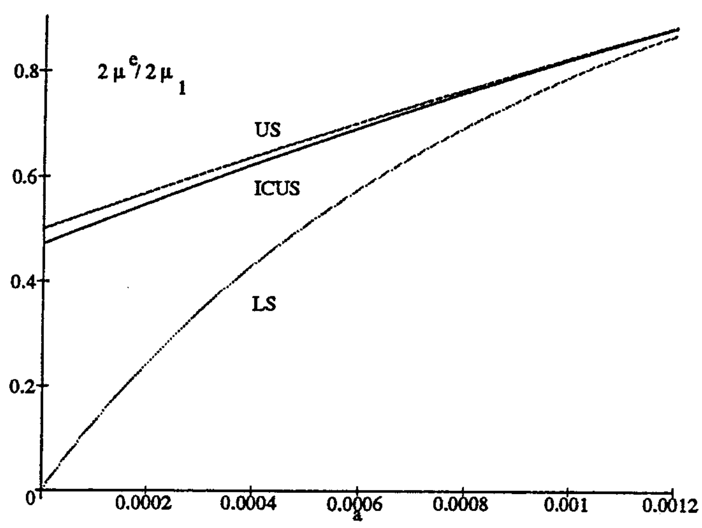

Fig. 2. Shear trace bounds ICUS, US and LS for monodisperse suspensions of graphite in an epoxy matrix for particle radius less than critical: $R_s^c = 0.0016$ m.

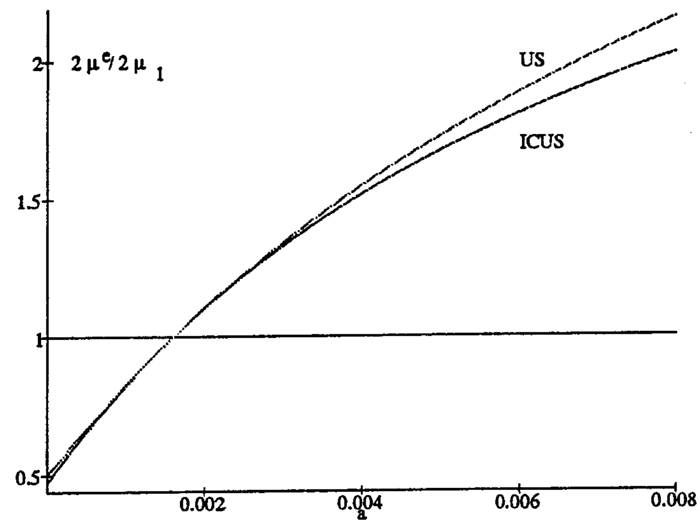

Fig. 3. Illustration of the comparison between the bounds ICUS and US in (5.31) for radius a close to the critical value $R_s^c = 0.0016$ m.

For cubic composites we observe that $\mathrm{tr}_b \mathcal{C}^e = 3\kappa^e$ and $\mathrm{tr}_b(\mathcal{C}^{e^{-1}}) = (3\kappa^e)^{-1}$, where κ^e the effective bulk modulus of the composite. In this case we deduce from Corollary 6.5 that

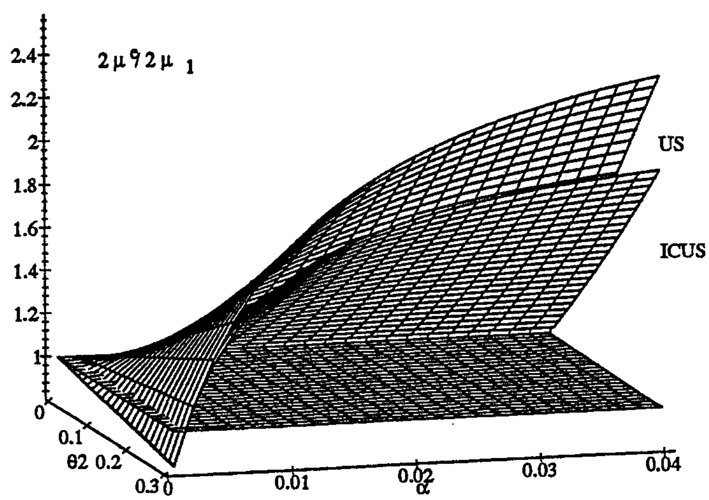

Fig. 4. Interface comparison method upper bound ICUS compared with the corrected upper bound US in (5.31) for 3-D monodisperse suspensions of graphite spheres in an epoxy matrix in terms of volume fraction θ_2 and radius a.

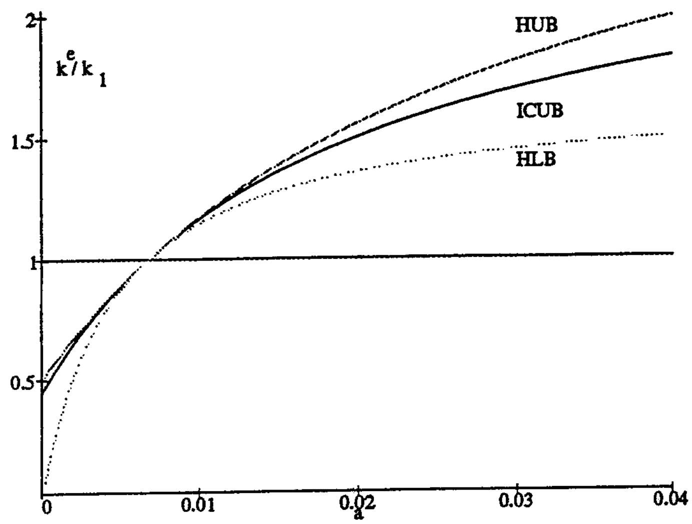

Fig. 5. Illustration of Corollary 6.6. Interface comparison method upper bound ICUB compared with Hashin's upper and lower bulk modulus bounds (HUB and HLB) (see 5.33 and 5.34) for cubic composites.

Corollary 6.6. *For cubic monodisperse suspensions of spheres:*

$$\kappa^e > \kappa_1 \ \text{if}\, a > R_b^c \tag{6.22}$$

$$\kappa^e < \kappa_1 \ \text{if}\, a < R_b^c\,, \tag{6.23}$$

and

$$\kappa^e = \kappa_1 \ \text{if}\, a = R_b^c\,. \tag{6.24}$$

We note that (6.24) also follows from Corollary 4.5.

The interface comparison upper bound ICUB is compared to Hashin's[6] upper and lower bulk moduli bound for cubic composites in Fig. c5. The suspension consists of graphite spheres in an epoxy matrix. The volume fraction of spheres is fixed at 50% and the sphere radii range from 0.04 m to infinitesimal. One sees that the bounds are tangent and touch precisely at the critical radius $R_b^c = 0.00683$ m.

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