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A NOTE ON THE SHEAR MODULUS OF A BINARY ELASTIC MIXTURE

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The Hashin-Shtrikman and Walpole bounds on the effective shear modulus of a binary elastic mixture are revisited. A simple method of derivation is given as a generalization of the approach, recently proposed by one of the authors in the absorption and scalar conductivity problems for a two-phase medium.

Keywords: two-phase random media, effective shear modulus, variational estimates, Hashin-Shtrikman and Walpole bounds

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The aim of this note is to present and discuss a simple derivation of the well-known two-point estimates on the effective shear modulus of a binary elastic mixture, due to Hashin and Shtrikman [1] and Walpole [2]. As a matter of fact, this is a continuation of the recent paper [3], where a similar analysis is performed for bounding the effective bulk modulus of the mixture. In the case of a shear modulus, however, a number of technical difficulties arise, which makes the analysis much more involving. The basic idea, as in [3], is a generalization of the approach, used by one of the authors in the absorption and scalar conductivity cases [4].

Let us recall first how the problem is posed, see, e.g. [5, 6]. Assume that the mixture is statistically homogeneous and isotropic. Let

$$\chi_i(x) = \begin{cases} 1, & \text{if } x \in \Omega_i, \\ 0, & \text{otherwise}, \end{cases}$$

be the characteristic function of the region Ω_i, occupied by one of the constituents, labelled 'i', $i = 1, 2$, so that $\chi_1(x) + \chi_2(x) = 1$. Hereafter, all quantities, pertaining to the region Ω_1 or Ω_2, are supplied with the subscript '1' or '2', respectively.

The statistical properties of the medium follow from the set of multipoint moments of one of the functions $\chi_i(x)$, say $\chi_2(x)$ for definiteness, or, which is the same, by the volume fraction $\eta_2 = \langle\chi_2(x)\rangle$ of the phase '2', and the multipoint moments

$$M_2(x) = \langle\chi_2'(0)\chi_2'(x)\rangle\,,\ M_3(x,y) = \langle\chi_2'(0)\chi_2'(x)\chi_2'(y)\rangle\,,\dots\,, \tag{1}$$

with $\chi_2'(x) = \chi_2(x) - \eta_2$ being the fluctuating part of the field $\chi_2(x)$, see, e.g., [5, 6]. The angled brackets $\langle\cdot\rangle$ hereafter denote ensemble averaging. One point could be taken at the origin because of the assumed statistical homogeneity, as already done in (1).

Recall that for a statistically isotropic binary medium under study one has

$$M_2(0) = \left\langle\chi_2'^2(0)\right\rangle = \eta_1\eta_2, \quad M_3(0) = \left\langle\chi_2'^3(0)\right\rangle = \eta_1\eta_2(\eta_1 - \eta_2). \tag{2}$$

Assuming also the constituents isotropic, the fourth-rank tensor of elastic moduli of the medium, $\mathbf{L}(x)$, is a random field of the familiar form

$$\begin{aligned} \mathbf{L}(x) &= 3k(x)\mathbf{J}' + 2\mu(x)\mathbf{J}'', \\ k(x) &= k_1\chi_1(x) + k_2\chi_2(x) = \langle k\rangle + [k]\chi_2'(x), \\ \mu(x) &= \mu_1\chi_1(x) + \mu_2\chi_2(x) = \langle\mu\rangle + [\mu]\chi_2'(x), \end{aligned} \tag{3}$$

where k and μ stand, as usual, for the bulk and shear modulus, respectively. The square brackets denote the jumps of the appropriate quantities, say, $[k] = k_2 - k_1$, $[\mu] = \mu_2 - \mu_1$, etc. In Eq. (3), $\mathbf{J}'$ and $\mathbf{J}''$ are the basic isotropic fourth-rank tensors with the Cartesian components

$$J'_{ijkl} = \frac{1}{3}\delta_{ij}\delta_{kl}; \quad J''_{ijkl} = \frac{1}{2}\Big(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \frac{2}{3}\delta_{ij}\delta_{kl}\Big). \tag{4}$$

The displacement field $u(x)$ in the medium, at the absence of body forces, is governed by the well-known equations

$$\begin{aligned} \nabla\cdot\boldsymbol{\sigma}(x) &= 0, \\ \boldsymbol{\sigma}(x) = \mathbf{L}(x) : \boldsymbol{\varepsilon}(x) &= k(x)\theta(x)\mathbf{I} + 2\mu(x)\mathbf{d}(x), \\ \boldsymbol{\varepsilon} = \frac{1}{2}(\nabla u + u\nabla), &\quad \mathbf{d}(x) = \boldsymbol{\varepsilon}(x) - \frac{1}{3}\theta(x)\mathbf{I}, \end{aligned} \tag{5}$$

where $\boldsymbol{\sigma}$ denotes the stress tensor, $\boldsymbol{\varepsilon}$ is the small strain tensor, generated by the displacement field $u(x)$, $\mathbf{d}$ is the strain deviator, and $\theta = \operatorname{tr}\boldsymbol{\varepsilon}$ is the volumetric strain. The colon designates contraction with respect to two pairs of indices and $\mathbf{I}$ is the unit second-rank tensor.

The system (5) is supplied with the condition

$$\langle\boldsymbol{\varepsilon}(x)\rangle = \mathbf{E}, \tag{6}$$

prescribing the macroscopic strain tensor $\mathbf{E}$, imposed upon the medium.

Recall [5] that the random problem (5), (6) is equivalent to the variational principle of classical type

$$\begin{aligned} W[\varepsilon(x)] &= \langle \varepsilon(x) : \mathbf{L}(x) : \varepsilon(x) \rangle \to \min, \\ \min W &= \mathbf{E} : \mathbf{L}^* : \mathbf{E}. \end{aligned} \tag{7}$$

The energy functional W is considered over the class of random fields $u(x)$ that generate strain fields $\varepsilon(x)$, complying with the condition (6). In Eq. (7), $\mathbf{L}^*$ is the tensor of effective elastic moduli for the medium which, in the isotropic case under study, has the form

$$\mathbf{L}^* = 3k^*\mathbf{J}' + 2\mu^*\mathbf{J}'', \tag{8}$$

where k^* and μ^* are the effective bulk and shear modulus of the mixture, respectively.

Consider, guided by [3] and [4], the class of trial fields for the variational principle (7):

$$\begin{aligned} \mathcal{K}^{(1)} = \Big\{ \widetilde{u}(x) \;\Big|\; \widetilde{u}(x) &= \mathbf{E} \cdot x \\ &+ \alpha \mathbf{E} : \int \Big(\nabla G(x-y) \otimes \mathbf{I} + \kappa \nabla\nabla\nabla F(x-y) \Big) \chi_2'(y)\, d^3y \Big\}, \end{aligned} \tag{9}$$

having assumed now that $\mathbf{E}$ is deviatoric, $\operatorname{tr} \mathbf{E} = 0$. Since the solution, $u(x)$, of the problem (5), (6) linearly depends on $\mathbf{E}$, we can assume that $\operatorname{tr} \mathbf{E} \cdot \mathbf{E} = \mathbf{E} : \mathbf{E} = 1$. In the class of trial fields (9) α and κ are adjustable scalar parameters and the kernels there read

$$G(x) = \frac{1}{4\pi|x|}, \quad F(x) = \frac{|x|}{4\pi}. \tag{10}$$

Hereafter the integrals are over the whole $\mathbb{R}^3$ if the integration domain is not explicitly indicated.

It is noted that the class of trial fields (9) has been first employed by McCoy [7], when deriving the Beran's type bound [8] for the shear modulus. The only difference is that we have allowed the multiplier κ in (9) to be adjustable as well (an idea already used by Milton and Phan-Thien [9]). In the final stage of our procedure, the appropriate optimization will bring forth the "best" value $\kappa_{\text{opt}} = -1/\big(4(1-\nu_2)\big)$, see Eq. (23) below. This means that the integrand in the right-hand side of (9) would *exactly* coincide with the Green tensor of one of the constituents. Hence the original McCoy's class of trial fields [7] will show up eventually. (See also the discussion in [10].)

It is to be also noted that in [3], when studying in a similar way the effective bulk modulus, we have chosen $\mathbf{E}$ spherical. This assumption considerably simplified the analysis (in particular, there was no need to introduce the second term in the integrand of the right-hand side of (9), containing the triple gradient). In this case the result is the three-point bound on the bulk modulus, proposed by Beran and Molyneux [11].

The energy functional W, when restricted over $\mathcal{K}^{(1)}$, becomes a quadratic function of α and κ:

$$W[\widetilde{u}(x)] = A + 2B\alpha + C\alpha^2, \quad A = 2\langle\mu\rangle, \quad B = 2[\mu](\mathsf{Z}_1 + \kappa\mathsf{U}_1),$$

$$C = \langle\mu\rangle\left(\mathsf{Z}_2 + \frac{1}{3}(1 - 8\kappa - 8\kappa^2)\mathsf{T}_2 + 4\kappa\mathsf{V}_2 + 2\kappa^2\mathsf{U}_2\right) \tag{11}$$

$$+ [\mu]\left(\mathsf{Z}_3 + \frac{1}{3}(1 - 8\kappa - 8\kappa^2)\mathsf{T}_3 + 4\kappa\mathsf{V}_3 + 2\kappa^2\mathsf{U}_3\right) + (1 + 2\kappa)^2\Big(\langle k\rangle\,\mathsf{T}_2 + [k]\mathsf{T}_3\Big),$$

with the dimensionless statistical parameters for the medium, defined as follows:

$$\mathsf{Z}_1 = (\mathbf{E}\cdot\mathbf{E}) : \int \nabla\nabla G(y)\, M_2(y)\, d^3y,$$

$$\mathsf{Z}_2 = (\mathbf{E}\cdot\mathbf{E}) : \iint \nabla\nabla G(y_1)\cdot\nabla\nabla G(y_2) M_2(y_1 - y_2)\, d^3y_1 d^3y_2,$$

$$\mathsf{Z}_3 = (\mathbf{E}\cdot\mathbf{E}) : \iint \nabla\nabla G(y_1)\cdot\nabla\nabla G(y_2) M_3(y_1, y_2)\, d^3y_1 d^3y_2,$$

$$\mathsf{U}_1 = \mathbf{E} : \int \nabla\nabla\nabla\nabla F(y)\, M_2(y)\, d^3y : \mathbf{E},$$

$$\mathsf{U}_2 = \mathbf{E} : \iint \nabla\nabla\nabla\nabla F(y_1) : \nabla\nabla\nabla\nabla F(y_2)\, M_2(y_1 - y_2)\, d^3y_1 d^3y_2 : \mathbf{E},$$

$$\mathsf{U}_3 = \mathbf{E} : \iint \nabla\nabla\nabla\nabla F(y_1) : \nabla\nabla\nabla\nabla F(y_2)\, M_3(y_1, y_2)\, d^3y_1 d^3y_2 : \mathbf{E}, \tag{12}$$

$$\mathsf{V}_2 = \mathbf{E} : \iint \nabla\nabla G(y_1)\cdot\nabla\nabla\nabla\nabla F(y_2)\, M_2(y_1 - y_2)\, d^3y_1 d^3y_2 : \mathbf{E},$$

$$\mathsf{V}_3 = \mathbf{E} : \iint \nabla\nabla G(y_1)\cdot\nabla\nabla\nabla\nabla F(y_2)\, M_3(y_1, y_2)\, d^3y_1 d^3y_2 : \mathbf{E},$$

$$\mathsf{T}_2 = \mathbf{E} : \iint \nabla\nabla G(y_1)\otimes\nabla\nabla G(y_2)\, M_2(y_1 - y_2)\, d^3y_1 d^3y_2 : \mathbf{E},$$

$$\mathsf{T}_3 = \mathbf{E} : \iint \nabla\nabla G(y_1)\otimes\nabla\nabla G(y_2)\, M_3(y_1, y_2)\, d^3y_1 d^3y_2 : \mathbf{E}.$$

Moreover, the parameters Z_1 and Z_2 can be easily evaluated in the statistically isotropic case under study. Indeed, $M_2(y) = M_2(|y|)$ then and the appropriate integrals in the definitions of Z_1 and Z_2 are isotropic second-rank tensors, thus proportional to $\mathbf{I}$:

$$\int \nabla\nabla G(y)\, M_2(y)\, d^3y = c_1\mathbf{I},$$

$$\iint \nabla\nabla G(y_1)\cdot\nabla\nabla G(y_2) M_2(y_1 - y_2)\, d^3y_1 d^3y_2 = c_2\mathbf{I},$$

with certain constants c_1 and c_2. To find the latter it suffices to make a contraction in the last two formulae, integrate by parts and recall that $G(x)$ is just the Green function for the Laplacian. This procedure yields

$$c_1 = -\frac{1}{3}\, M_2(0) = -\frac{1}{3}\, \eta_1\eta_2, \quad c_2 = \frac{1}{3} M_2(0) = \frac{1}{3}\eta_1\eta_2,$$

and therefore

$$\mathsf{Z}_1 = -\frac{1}{3}\, \eta_1\eta_2, \quad \mathsf{Z}_2 = \frac{1}{3}\, \eta_1\eta_2 \tag{13}$$

(recall that we have assumed $\mathbf{E} : \mathbf{E} = 1$).

Note that in the statistically isotropic case under study $c_2 = \frac{1}{3} P_2$, where

$$P_2 = \iint \nabla\nabla G(y_1) : \nabla\nabla G(y_2)\, M_2(y_1 - y_2)\, d^3y_1 d^3y_2$$

is the two-point statistical parameter that appeared in the appropriate bounds on the conductivity coefficient [4] and the bulk modulus [3] of the mixture. The above simple reasoning is just the evaluation of this parameter done, e.g. in [3] (see Eq. (12) there). Hence

$$\mathsf{Z}_1 = -\frac{1}{3}\, P_2, \quad \mathsf{Z}_2 = \frac{1}{3}\, P_2, \quad P_2 = \eta_1\eta_2.$$

As we shall demonstrate below, the rest of the two-point statistical parameters in (12) are also proportional to P_2.

Due to the statistical isotropy of the medium the integrals

$$
\begin{aligned}
&\int \nabla\nabla\nabla\nabla F(y)\, M_2(y)\, d^3y = c_3\mathbf{H}, \\
&\iint \nabla\nabla G(y_1) \otimes \nabla\nabla G(y_2)\, M_2(y_1 - y_2)\, d^3y_1 d^3y_2 = c_4\mathbf{H}, \\
&\iint \nabla\nabla G(y_1) \cdot \nabla\nabla\nabla\nabla F(y_2)\, M_2(y_1 - y_2)\, d^3y_1 d^3y_2 = c_5\mathbf{H}, \\
&\iint \nabla\nabla\nabla\nabla F(y_1) : \nabla\nabla\nabla\nabla F(y_2)\, M_2(y_1 - y_2)\, d^3y_1 d^3y_2 = c_6\mathbf{H}
\end{aligned}
\tag{14}
$$

are fourth-rank fully symmetric isotropic tensors, thus proportional to the tensor $\mathbf{H}$, whose Cartesian components read

$$H_{ijkl} = \delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk},$$

with certain constants c_3 to c_6. (The fact that the integrals in (14) represent fully symmetric tensors is easily seen if appropriate integration by parts is performed.) Making a full contraction in (14) and integrating by parts, we find the needed constants to be

$$c_3 = -\frac{2}{15} M_2(0), \quad c_4 = \frac{1}{15} M_2(0), \quad c_5 = \frac{2}{15} M_2(0), \quad c_6 = \frac{4}{15} M_2(0),$$

so that the parameters U_1, T_2, V_2 and U_2 thus become simply:

$$\begin{aligned} \mathsf{U}_1 &= -\frac{4}{15}\,\eta_1\eta_2, \quad \mathsf{T}_2 = \frac{2}{15}\,\eta_1\eta_2, \\ \mathsf{V}_2 &= \frac{4}{15}\,\eta_1\eta_2, \quad \mathsf{U}_2 = \frac{8}{15}\,\eta_1\eta_2, \end{aligned} \tag{15}$$

taking into account Eq. (2) as well.

The variational principle (7), together with (11), implies

$$2\mu^* \le W[\tilde{u}(x)] = A + 2B\alpha + C\alpha^2, \quad \forall\alpha,\ \forall\kappa. \tag{16}$$

In particular, at $\alpha = 0$, one has

$$\mu^* \le \langle\mu\rangle \tag{17}$$

which, obviously, is the elementary (Voigt) bound on μ^*.

Next, optimizing the right-hand side of (16) with respect to α, one gets another estimate on μ^*:

$$2\mu^* \le A - \frac{B^2}{C},$$

i.e.

$$\mu^* \le \langle\mu\rangle - \frac{2[\mu]^2(\mathsf{Z}_1 + \kappa\mathsf{U}_1)^2}{C}, \tag{18}$$

having taken into account the expressions for A and B, see (11).

In (18) we have fixed the constant κ. The next stage is to optimize it with respect to κ. The resulting bound will be then just the Milton–Phan-Thien's one [9] on the effective shear modulus μ^*. If κ has the special value $\kappa = -1/\big(4(1-\nu_2)\big)$, see (23) below (so that the integrand in (9) is just the appropriate Green tensor), then (18) is the McCoy's bound on μ^*. This is obviously a three-point estimate since for its evaluation three-point statistical information — the correlations $M_3(y_1, y_2)$ — is needed in the three-point parameters Z_3, T_3, V_3 and U_3, see (12).

The main problem in specifying the bound (18) are the three-point parameters Z_3, T_3, V_3 and U_3, whose evaluation for special and realistic random constitution is clearly a nontrivial problem. Note that the first of these parameters, Z_3, is

$$\mathsf{Z}_3 = \frac{1}{3}\,P_3, \quad P_3 = \iint \nabla\nabla G(y_1) : \nabla\nabla G(y_2) M_3(y_1, y_2)\, d^3y_1 d^3y_2,$$

where P_3 is the three-point parameter that showed up in conductivity and bulk modulus bounding procedures [3], [4]. This parameter is simply connected to the so-called ζ-parameter of Torquato and Milton [12–14], see also [3, Eqs. (18), (19)].

In the variational reasoning of [3], [4] we have excluded the parameter P_3, using the fact that the appropriate three-point bounds should be more restrictive than the elementary ones, whatever the properties of the constituents. This fact led us to an inequality between P_3 and P_2. Here we shall employ the same procedure; though a certain additional three-point parameter (a linear combination of Z_3, T_3, V_3 and U_3) will show up, we shall obtain two inequalities for the two such parameters as a consequence of the fact that now we can vary more material properties, namely, the bulk and shear moduli of the constituents.

Indeed, the bound (18) should be at least as good as the elementary bound (17) (since the energy functional is minimized over a broader class of trial fields). This implies that

$$C > 0, \quad AC - B^2 \geq 0, \tag{19}$$

because $\mu^* \geq 0$. Since $A = 2\langle\mu\rangle > 0$, one has

$$C \geq B^2/A > 0,$$

which means that the second inequality in (19) is the stronger one. Using the expressions for A, B and C from (11), we can write the latter in the form

$$\begin{aligned}
&\langle\mu\rangle \left(\mathsf{Z}_2 + \frac{1}{3}(1 - 8\kappa - 8\kappa^2)\mathsf{T}_2 + 4\kappa\mathsf{V}_2 + 2\kappa^2\mathsf{U}_2 \right) \\
&+ [\mu] \left(\mathsf{Z}_3 + \frac{1}{3}(1 - 8\kappa - 8\kappa^2)\mathsf{T}_3 + 4\kappa\mathsf{V}_3 + 2\kappa^2\mathsf{U}_3 \right) \\
&+ (1 + 2\kappa)^2 \Big(\langle k\rangle\, \mathsf{T}_2 + [k]\mathsf{T}_3 \Big) - \frac{2[\mu]^2(\mathsf{Z}_1 + \kappa\mathsf{U}_1)^2}{\langle\mu\rangle} \geq 0.
\end{aligned} \tag{20}$$

The inequality (20) should hold for every "realistic" choice of the elastic moduli of the constituents (i.e. for which the appropriate elastic energy is positive-definite). This implies

$$-\eta_2\mathsf{T}_2 \leq \mathsf{T}_3 \leq \eta_1\mathsf{T}_2, \tag{21a}$$

$$\begin{aligned}
&\frac{2(\mathsf{Z}_1 + \kappa\mathsf{U}_1)^2}{\eta_2} - \eta_2 \left(\mathsf{Z}_2 + \frac{1}{3}(1 - 8\kappa - 8\kappa^2)\mathsf{T}_2 + 4\kappa\mathsf{V}_2 + 2\kappa^2\mathsf{U}_2 \right) \\
&\leq \mathsf{Z}_3 + \frac{1}{3}(1 - 8\kappa - 8\kappa^2)\mathsf{T}_3 + 4\kappa\mathsf{V}_3 + 2\kappa^2\mathsf{U}_3 \\
&\leq \eta_1 \left(\mathsf{Z}_2 + \frac{1}{3}(1 - 8\kappa - 8\kappa^2)\mathsf{T}_2 + 4\kappa\mathsf{V}_2 + 2\kappa^2\mathsf{U}_2 \right) - \frac{2(\mathsf{Z}_1 + \kappa\mathsf{U}_1)^2}{\eta_1}.
\end{aligned} \tag{21b}$$

Hence we have indeed *two* sets of inequalities for the three-point parameters that enter the bound (18). (And this is a consequence, let us underline once again, of the fact that *two* material properties have been varied *independently* — the bulk and shear moduli of the constituents.) Following the idea of [4], we can exclude the "bad" three-point quantities

$$\mathsf{T}_3 \quad \text{and} \quad \mathsf{Z}_3 + \frac{1}{3}(1 - 8\kappa - 8\kappa^2)\mathsf{T}_3 + 4\kappa\mathsf{V}_3 + 2\kappa^2\mathsf{U}_3$$

from this bound, by means of (21), thus replacing them by the two-point quantities already evaluated. Depending on the signs of $[\mu] = \mu_2 - \mu_1$ and $[k] = k_2 - k_1$, we should use to this end the upper or lower bounds (21).

For example, in the case $[\mu] \geq 0$ (i.e. $\mu_2 \geq \mu_1$) and $[k] \geq 0$ (i.e. $k_2 \geq k_1$), the upper bounds (21) are to be used, which results in the estimate

$$\mu^* \leq \langle \mu \rangle - 2[\mu]^2 \eta_1 \eta_2 \frac{16\kappa^2 + 40\kappa + 25}{p\kappa^2 + q\kappa + r}, \tag{22}$$

$$p = 160\mu_2 + 120k_2 - 32[\mu]\eta_2,$$
$$q = 160\mu_2 + 120k_2 - 80[\mu]\eta_2,$$
$$r = 85\mu_2 + 30k_2 - 50[\mu]\eta_2.$$

Optimizing this bound with respect to κ, we find[1]

$$\kappa_{\rm opt} = -\frac{\mu_2 + 3k_2}{2(4\mu_2 + 3k_2)} = -\frac{1}{4(1-\nu_2)}, \quad \nu_2 = \frac{3k_2 - 2\mu_2}{2\mu_2 + 6k_2}, \tag{23}$$

so that ν_2 is the Poisson ratio of the phase '2'. The best bound on μ^* thus becomes

$$\mu^* \leq \langle \mu \rangle - \frac{\eta_1 \eta_2 [\mu]^2}{\eta_1 \mu_2 + \eta_2 \mu_1 + \dfrac{\mu_2(8\mu_2 + 9k_2)}{6(2\mu_2 + k_2)}}, \quad \text{if } \mu_2 \geq \mu_1 \text{ and } k_2 \geq k_1. \tag{24a}$$

The calculations in the rest of the cases are fully similar, so that only the final results will be given:

$$\mu^* \leq \langle \mu \rangle - \frac{\eta_1 \eta_2 [\mu]^2}{\eta_1 \mu_2 + \eta_2 \mu_1 + \dfrac{\mu_1(8\mu_1 + 9k_1)}{6(2\mu_1 + k_1)}}, \quad \text{if } \mu_2 \leq \mu_1 \text{ and } k_2 \leq k_1,$$

$$\mu^* \leq \langle \mu \rangle - \frac{\eta_1 \eta_2 [\mu]^2}{\eta_1 \mu_2 + \eta_2 \mu_1 + \dfrac{\mu_1(8\mu_1 + 9k_2)}{6(2\mu_1 + k_2)}}, \quad \text{if } \mu_2 \leq \mu_1 \text{ and } k_2 \geq k_1, \tag{24b}$$

$$\mu^* \leq \langle \mu \rangle - \frac{\eta_1 \eta_2 [\mu]^2}{\eta_1 \mu_2 + \eta_2 \mu_1 + \dfrac{\mu_2(8\mu_2 + 9k_1)}{6(2\mu_2 + k_1)}}, \quad \text{if } \mu_2 \geq \mu_1 \text{ and } k_2 \leq k_1.$$

In the so-called "well-ordered" case, when $(k_2 - k_1)(\mu_2 - \mu_1) > 0$, (24a) and the first of the estimates (24b) coincide with the Hashin-Shtrikman bound on μ^*, see [1]. The general "non-ordered" case was considered by Walpole [2]. It is easily seen that our bounds (24) are just the Walpole bounds [2, 15].

The derivation of the lower bound, corresponding to (24), is fully similar. In this case we write the elastic energy (7) as a functional of the stress tensor field:

$$W[\boldsymbol{\sigma}(x)] = \langle \boldsymbol{\sigma}(x) : \mathbf{L}^{-1}(x) : \boldsymbol{\sigma}(x) \rangle \to \min,$$
$$\min W = \boldsymbol{\Sigma} : \mathbf{L}^{*-1} : \boldsymbol{\Sigma}. \tag{25}$$

[1] The right-hand side of (22) has one more extremum point, $\kappa = -5/4$, but it corresponds to its maximum value and hence is of no interest for us.

The functional W is considered over the class of trial fields, such that

$$\nabla \cdot \boldsymbol{\sigma}(x) = 0, \quad \langle \boldsymbol{\sigma}(x) \rangle = \boldsymbol{\Sigma}, \tag{26}$$

with a prescribed macrostress tensor $\boldsymbol{\Sigma}$, imposed upon the mixture.

The natural counterpart of the class (9) of trial stress fields for the functional W in (25) now reads

$$\begin{aligned}\mathcal{N}^{(1)} = \Bigg\{ \widetilde{\boldsymbol{\sigma}}(x) \mid \widetilde{\boldsymbol{\sigma}}(x) = \boldsymbol{\Sigma} + \alpha \Bigg[& \chi_2'(y)\, \boldsymbol{\Sigma} \\ & - (1+2\kappa)\, \mathbf{I} \left(\boldsymbol{\Sigma} : \int \nabla\nabla G(x-y) \chi_2'(y)\, d^3y \right) \\ & + \kappa \boldsymbol{\Sigma} : \int \nabla\nabla\nabla\nabla F(x-y) \chi_2'(y)\, d^3y \\ & + 2\kappa \operatorname{def} \left(\boldsymbol{\Sigma} \cdot \int \nabla\nabla G(x-y) \chi_2'(y)\, d^3y \right) \Bigg] \Bigg\},\end{aligned} \tag{27}$$

with deviatoric $\boldsymbol{\Sigma}$, $\operatorname{tr} \boldsymbol{\Sigma} = 0$, and adjustable scalar parameters α and κ; $G(x)$ and $F(x)$ are the functions, defined in (10). In (27) 'def' denotes symmetrization of a second-rank tensor, i.e. $\operatorname{def} \mathbf{T} = \frac{1}{2}(\mathbf{T} + \mathbf{T}^*)$, see [10] for discussion.

The energy functional W, when restricted over $\mathcal{N}^{(1)}$, becomes a quadratic function of α:

$$W[\widetilde{\boldsymbol{\sigma}}(x)] = A + 2B\alpha + C\alpha^2, \ A = \frac{1}{2} \langle \gamma \rangle\, \boldsymbol{\Sigma} : \boldsymbol{\Sigma},$$

$$B = \frac{1}{2} [\gamma] \Big(\eta_1 \eta_2 \boldsymbol{\Sigma} : \boldsymbol{\Sigma} + 2\mathsf{Z}_1 + \kappa \mathsf{U}_1 \Big),$$

$$\begin{aligned} C = \frac{1}{2} \Bigg[& \left(\eta_1\eta_2(\eta_1\gamma_1 + \eta_2\gamma_2) + \left(\frac{4}{3} + \frac{8}{15}\kappa \right) [\gamma](\eta_2 - \eta_1)\eta_1^2 \right) \boldsymbol{\Sigma} : \boldsymbol{\Sigma} \\ & + 2(\eta_1\gamma_2 + \eta_2\gamma_1)(2\mathsf{Z}_1 + \kappa\mathsf{U}_1) + \langle \gamma \rangle \left(2\mathsf{Z}_2 + \frac{1}{3}(2 - 8\kappa - 4\kappa^2)\mathsf{T}_2 + 4\kappa \mathsf{V}_2 + \kappa^2 \mathsf{U}_2 \right) \\ & + [\gamma] \left(2\mathsf{Z}_3 + \frac{1}{3}(2 - 8\kappa - 4\kappa^2)\mathsf{T}_3 + 4\kappa\mathsf{V}_3 + \kappa^2\mathsf{U}_3 \right) \Bigg] + \frac{(1+4\kappa)^2}{9} \Big(\langle \beta \rangle\, \mathsf{T}_2 + [\beta]\mathsf{T}_3 \Big), \end{aligned} \tag{28}$$

where

$$\gamma(x) = \frac{1}{\mu(x)}, \quad \beta(x) = \frac{1}{k(x)}$$

are the respective compliances of the mixture, and $[\gamma] = \gamma_2 - \gamma_1$, $[\beta] = \beta_2 - \beta_1$ are their jumps. Note that the *same* two- and three-point statistical parameters (12), that already showed up in (11), enter (28) as well; the only difference is that the tensor $\mathbf{E}$ in their definitions (12) is to be replaced by the tensor $\boldsymbol{\Sigma}$.

The elementary (Reuss) lower bound on μ^* now follows from (28) at $\alpha = 0$:

$$\frac{1}{\mu^*} \leq \left\langle \frac{1}{\mu(x)} \right\rangle = \frac{\eta_1}{\mu_1} + \frac{\eta_2}{\mu_2}\,.$$

Almost literally, the arguments that have led us to the inequality (20) are to be repeated now — that is the estimate that results from (28) upon minimizing with respect to α should be always more restrictive than the elementary Reuss' one. The final result is another set of inequalities, similar to (21), namely,

$$-\eta_2 \mathsf{T}_2 \leq \mathsf{T}_3 \leq \eta_1 \mathsf{T}_2, \tag{29a}$$

$$\begin{aligned}
&\frac{(\eta_1\eta_2\boldsymbol{\Sigma}:\boldsymbol{\Sigma} + 2\mathsf{Z}_1 + \kappa\mathsf{U}_1)^2}{\eta_2\boldsymbol{\Sigma}:\boldsymbol{\Sigma}} - \eta_2^2\left[\eta_1 + \left(\frac{4}{3} + \frac{8}{15}\kappa\right)(\eta_1 - \eta_2\right]\boldsymbol{\Sigma}:\boldsymbol{\Sigma} \\
&-2\eta_1(2\mathsf{Z}_1 + \kappa\mathsf{U}_1) - \eta_2\left(2\mathsf{Z}_2 + \frac{1}{3}(2 - 8\kappa - 4\kappa^2)\mathsf{T}_2 + 4\kappa\mathsf{V}_2 + \kappa^2\mathsf{U}_2\right) \\
&\leq 2\mathsf{Z}_3 + \frac{1}{3}(2 - 8\kappa - 4\kappa^2)\mathsf{T}_3 + 4\kappa\mathsf{V}_3 + \kappa^2\mathsf{U}_3 \qquad \text{(29b)} \\
&\leq -\frac{(\eta_1\eta_2\boldsymbol{\Sigma}:\boldsymbol{\Sigma} + 2\mathsf{Z}_1 + \kappa\mathsf{U}_1)^2}{\eta_1\boldsymbol{\Sigma}:\boldsymbol{\Sigma}} + \eta_2\left[\eta_1^2 - \left(\frac{4}{3} + \frac{8}{15}\kappa\right)(\eta_1 - \eta_2)\eta_2\right]\boldsymbol{\Sigma}:\boldsymbol{\Sigma} \\
&+2\eta_2\Big(2\mathsf{Z}_1 + \kappa\mathsf{U}_1\Big) + \eta_1\left(2\mathsf{Z}_2 + \frac{1}{3}(2 - 8\kappa - 4\kappa^2)\mathsf{T}_2 + 4\kappa\mathsf{V}_2 + \kappa^2\mathsf{U}_2\right).
\end{aligned}$$

Next we employ (29) in the estimates that follow from (28) in order to exclude the three-point parameters. The details are tedious and fully similar to those, already performed when deriving the bounds (24). The final result reads

$$\begin{aligned}
&\mu^* \geq \langle\mu\rangle - \frac{\eta_1\eta_2[\mu]^2}{\eta_1\mu_2 + \eta_2\mu_1 + \dfrac{\mu_2(8\mu_2 + 9k_2)}{6(2\mu_2 + k_2)}}\,, \quad \text{if } \mu_2 \leq \mu_1 \text{ and } k_2 \leq k_1, \\
&\mu^* \geq \langle\mu\rangle - \frac{\eta_1\eta_2[\mu]^2}{\eta_1\mu_2 + \eta_2\mu_1 + \dfrac{\mu_1(8\mu_1 + 9k_1)}{6(2\mu_1 + k_1)}}\,, \quad \text{if } \mu_2 \geq \mu_1 \text{ and } k_2 \geq k_1, \\
&\mu^* \geq \langle\mu\rangle - \frac{\eta_1\eta_2[\mu]^2}{\eta_1\mu_2 + \eta_2\mu_1 + \dfrac{\mu_2(8\mu_2 + 9k_1)}{6(2\mu_2 + k_1)}}\,, \quad \text{if } \mu_2 \leq \mu_1 \text{ and } k_2 \geq k_1, \\
&\mu^* \geq \langle\mu\rangle - \frac{\eta_1\eta_2[\mu]^2}{\eta_1\mu_2 + \eta_2\mu_1 + \dfrac{\mu_1(8\mu_1 + 9k_2)}{6(2\mu_1 + k_2)}}\,, \quad \text{if } \mu_2 \geq \mu_1 \text{ and } k_2 \leq k_1.
\end{aligned} \tag{30}$$

The inequalities (24), combined with (30), are just the Walpole bounds on the effective shear modulus of a binary mixture, see [2] and also [15], which are a direct generalization of the Hashin-Shtrikman result, with condition of "well-orderness" removed. Here we have shown how these classical estimates show up simply and naturally within the frame of the general method recently developed by one of the authors [4] in the absorption and scalar conductivity contexts.

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