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INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS
MIRAMARE - P.O.B. 586 - 34100 TRIESTE (ITALY) - TELEPHONES: 224281/2/3/4/5/6 - CABLE: CENTRATOM

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OVERALL PROPERTIES OF COMPOSITE MATERIALS

J.R. Willis
University of Bath
U.K.

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a single spherical inclusion of type r in a homogeneous material whose moduli are the overall moduli of the composite itself, namely $\bar{L}$. We note that $\tau_r = (L_r - L_0)e_r$, that is, τ_r is the polarization relative to the matrix. Therefore, if we define $\tau_r^* = (L_r - \bar{L})e_r$, so that τ_r^* is the polarization relative to a comparison material $\bar{L}$, we can read off τ_r^* from the results of §4 and then set $\tau_r = (L_r - L_0)(L_r - \bar{L})^{-1}\tau_r^*$. In particular, if the composite is isotropic, with isotropic phases, τ_r^* is given by (4.11) with $L^{(2)}$ replaced by L_r and $L^{(1)}$ by $\bar{L}$. But then, from (5.1),

$$\bar{\sigma}_{kk} = 3\bar{\kappa}\,\bar{e}_{kk}\,, \quad \bar{\sigma}'_{ij} = 2\bar{\mu}\,\bar{e}'_{ij}\,,$$

where

$$\left.\begin{aligned}
\bar{\kappa} &= \kappa^{(0)} + \sum_{r=1}^{N} c_r\left(\frac{\kappa^r-\kappa^{(0)}}{\kappa^r-\bar{\kappa}}\right)\frac{1}{3}\left\{\frac{1}{3(\kappa^r-\bar{\kappa})}+\frac{1}{3\bar{\kappa}+4\bar{\mu}}\right\}^{-1},\\
\bar{\mu} &= \mu^{(0)} + \sum_{r=1}^{N} c_r\left(\frac{\mu^r-\mu^{(0)}}{\mu^r-\bar{\mu}}\right)\frac{1}{2}\left\{\frac{1}{2(\mu^r-\bar{\mu})}+\frac{3(\bar{\kappa}+2\bar{\mu})}{5\bar{\mu}(3\bar{\kappa}+4\bar{\mu})}\right\}^{-1}.
\end{aligned}\right\} \quad (5.2)$$

Equations (5.2) can be solved to give the so-called "self-consistent" estimates of the overall moduli. In the absence of a matrix, we could formulate the problem directly, using $\bar{L}$ as the moduli of a 'comparison' material, and obtain

$$\bar{L}\bar{e} = \bar{L}\bar{e} + \sum_{r=1}^{N} c_r\tau_r\,,$$

implying that

$$\sum_{r=1}^{N} c_r\tau_r = 0\,. \quad (5.3)$$

For a polycrystal, we may have just <u>one</u> anisotropic material, but the grains might have all orientations. For this case, we might consider the limit of several L^r, anisotropic, at concentrations c_r. If the polycrystal is isotropic overall,

$$\tau_r = (P^r + \bar{A})^{-1}\bar{e}\,, \quad (5.4)$$

where $P^r = (L^r - \bar{L})^{-1}$ and

Overall Properties of Composite Materials

A brief outline will be given of some of the basic ideas underlying the study of the elastic behaviour of composites. A composite material will be taken as consisting of several phases (or, for a polycrystal, grains of one anisotropic material at a variety of orientations), which are perfectly bonded at all interfaces. Thus, across any interface, displacements and tractions will be taken as continuous; it is admitted, of course, that perfect interfacial bonding will not always be realized in practice. A composite material so defined is therefore a particular kind of inhomogeneous body, but the variations in properties will be taken on a microscopic scale only so that, macroscopically, the body appears uniform. The distribution of the phases could be known explicitly (e.g. it could be periodic, as in a laminated material) or it could be random, as in a polycrystal. The basic question to be answered is, given the mechanical behaviour of each constituent phase, how does the body behave overall? That is, given some boundary conditions on the body, what homogeneous body would best reproduce the deformation in the actual inhomogeneous body? This problem will be approached in the present work by considering boundary conditions that would produce in a homogeneous body just uniform states of stress and strain. The object will be to select the homogeneous body so that these uniform states of stress and strain coincide with the mean values of stress and strain in the actual body. The problem will be [posed] only for elastic bodies; the important subjects of the plasticity some of the notions that will be developed actually would serve as building blocks for considering such behaviour.

1. Mean value theorems.

Let the stress in a body be denoted by σ_{ij}. The body is inhomogeneous but elastic, with strain energy function $W(e_{ij})$, so that $\sigma_{ij} = \frac{\partial W}{\partial e_{ij}}$. (In fact, later, W will be taken to have the linear elastic form $W = \frac{1}{2} L_{ijkl} e_{ij} e_{kl}$, but this is not crucial at the moment)

Suppose the body occupies the region V and that displacements

$$u_i = \bar{u}_{i,j} x_j \qquad (1.1)$$

are applied to its surface ∂V, where $\bar{u}_{i,j}$ is just a set of nine constants. The mean distortion in the body is then given as

$$\frac{1}{V}\int_V u_{i,j}\, dx$$

(using V also to denote the volume). This transforms by Gauss' thm. to $\frac{1}{V}\int_{\partial V} u_i n_j\, dS = \frac{1}{V}\int_{\partial V} \bar{u}_{i,k} x_k n_j\, dS = \frac{1}{V}\int_V \bar{u}_{i,k} \delta_{kj}\, dV = \bar{u}_{i,j}$,

by using the boundary condition (1.1) and Gauss again. Thus, $\bar{u}_{i,j}$ actually is the mean distortion of the body, independently of constitutive properties.

Let us now define the mean stress by

$$\bar{\sigma}_{ij} = \frac{1}{V}\int_V \sigma_{ij}\, dV \qquad (1.2)$$

The boundary condition (1.1) would induce the stress $\bar{\sigma}_{ij}$ in a homogeneous body with stress-strain relation

$$\bar{\sigma}_{ij} = f_{ij}(\bar{e}_{kl}), \qquad (1.3)$$

so long as $f_{ij}(\bar{e}_{kl})$ took the value (1.2) for each choice of $\bar{e}_{kl}$. Thus, (1.3) defines the appropriate homogeneous body, but, to find the f_{ij}, the boundary value problem for the inhomogeneous body has to be solved.

We now consider the energy contained in the body, by allowing $\bar{u}_{ij}$ to depend upon time t. Slow changes are considered, so the eqns. of statics remain valid. The rate of increase of energy of the body is then

$$\dot{E} = \int_V \sigma_{ij}\, \dot{u}_{i,j}\, dV \qquad (1.4)$$

and so, by Gauss, using $\sigma_{ij,j} = 0$,

$$\dot{E} = \int_{\partial V} \sigma_{ij}\, \dot{u}_i\, n_j\, dS = \int_{\partial V} \sigma_{ij}\, \dot{\bar{u}}_{i,k}\, x_k\, n_j\, dS .$$

Employing Gauss again now gives

$$\dot{E} = \int_V \sigma_{ij}\, \dot{\bar{u}}_{i,j}\, dV = V \bar{\sigma}_{ij}\, \dot{\bar{u}}_{ij} \qquad (1.5)$$

Thus, if we define

$$\bar{W}(\bar{e}_{ij}) = \int_0^{\bar{e}_{ij}} \bar{\sigma}_{ij}\, d\bar{e}_{ij} = \int_0^t \bar{\sigma}_{ij}\, \dot{\bar{u}}_{i,j}\, dt , \qquad (1.6)$$

$$\bar{\sigma}_{ij} = \frac{\partial \bar{W}}{\partial \bar{e}_{ij}} \qquad (1.7)$$

and, since

$$\dot{\bar{W}} = \frac{1}{V}\int_V \sigma_{ij}\dot{u}_{i,j}\,dV = \frac{1}{V}\int_V \dot{W}\,dV,$$

also

$$\bar{W}(\bar{e}) = \frac{1}{V}\int_V W(e)\,dV \qquad (1.8)$$

Thus, the ~~homogeneous body~~ stress-strain law (1.3) for the homogeneous body takes the form (1.7), where, by (1.8), $\bar{W}$ is actually the mean energy density of the inhomogeneous body.

The same result can be established when traction boundary conditions

$$\sigma_{ij} n_j = \bar{\sigma}_{ij} n_j$$

are applied on ∂V; the manipulations are similar and will not be given.

Remark: If x_i are taken as Lagrangian coordinates and σ_{ij} is taken as the nominal (or first Piola-Kirchhoff) stress, so that $\sigma_{ij} = \partial W/\partial(\partial u_i/\partial x_j)$, as in R. J. Knops' lecture notes, all of the above holds. Thus, the statements are true in the context of finite elasticity, as well as infinitesimal, though subsequent calculations will be restricted to infinitesimal elasticity.

2. The Classical Energy Theorems.

Suppose $\sigma_{ij} n_j$ is given on a part S_T of the surface ∂V and u_i is given on the complementary part, S_u. [Conditions could also be mixed, but then more notation is required.]

Minimum energy principle.

For a linearly elastic body (possibly inhomogeneous), with energy density

$$W(e) = \tfrac{1}{2} L_{ijkl}(x) e_{ij} e_{kl}, \qquad (2.1)$$

which we take as positive definite for all symmetric e_{ij}, the actual displacement u_i is that which minimizes the functional

$$\mathcal{F}(u) = \int_V W(e)\,dV - \int_V f_i u_i\,dV - \int_{S_T} T_i u_i\,dS, \qquad (2.2)$$

where f_i are the prescribed body forces and T_i the prescribed tractions, given on S_T. u must be chosen, of course, to be consistent with the displacement conditions on S_u.

To verify the principle, we consider

$$\mathcal{F}(u^*) - \mathcal{F}(u) = \int_V \{W(e^*) - W(e) - f_i(u_i^* - u_i)\}\,dV - \int_{S_T} T_i(u_i^* - u_i)\,dS,$$

where u is the solution and u^* any other field. We have

$$W(e^*) - W(e) = \tfrac{1}{2} L_{ijkl}(e^*_{ij} - e_{ij})(e^*_{kl} - e_{kl}) + L_{ijkl} e^*_{ij} e_{kl} - L_{ijkl} e_{ij} e_{kl}$$

and so

$$\mathcal{F}(u^*) - \mathcal{F}(u) = \int_V W(e^* - e)\,dV + \int_V \{\sigma_{ij}(e^*_{ij} - e_{ij}) - f_i(u_i^* - u_i)\}\,dV - \int_{\partial V} \sigma_{ij} n_j (u_i^* - u_i)\,dS,$$

Application of Gauss to the integral over ∂V and use of $\sigma_{ij,j} + f_i = 0$ now shows that

$$F(u^*) - F(u) = \int_V W(e^* - e)\, dV \geq 0\,, \qquad (2.3)$$

since W is positive definite.

Complementary energy principle

Define $\quad W_c(\sigma) = \sigma_{ij} e_{ij} - W(e)\,, \qquad (2.4)$

for a linearly elastic body $W_c(\sigma)$ is numerically equal to $W(e) = \frac{1}{2}\sigma_{ij} e_{ij}$. The complementary energy principle states that, among all stress fields σ_{ij} satisfying $\sigma_{ij,j} + f_i = 0$ in V and $\sigma_{ij} n_j = T_i$ on ∂V, the actual solution σ_{ij} is that which maximizes the functional

$$\mathcal{G}(\sigma) = \int_{S_u} \sigma_{ij} n_j u_i\, dS - \int_V W_c(\sigma)\, dV\,, \qquad (2.5)$$

where the u_i are given over S_u. It can be proved, as above, that

$$\mathcal{G}(\sigma^*) - \mathcal{G}(\sigma) = -\int_V W_c(\sigma^* - \sigma)\, dV$$

and the principle follows

An application of the principles.

Suppose that u is prescribed over the whole of ∂V. Then, by the minimum energy principle,

$$\int_V W(e)\, dV \leq \int_V W(e^*)\, dV\,,$$

for any choice of u^* consistent with the b. conds.

Let us now take $u_i = \bar{u}_{ij} x_j$ on ∂V. An admissible u^* is then $u^* = \bar{u}_{ij} x_j$ throughout V, and ... deduce that

$$\int_V W(e)\,dV \le \int_V W(\bar{e}_{ij})\,dV \qquad (2.6)$$

Now $$\int_V W(e)\,dV = V\,\overline{W}(\bar{e}) = V.\tfrac{1}{2}\bar{L}_{ijkl}\,\bar{e}_{ij}\,\bar{e}_{kl}, \qquad (2.7)$$

where $\bar{L}_{ijkl}$ is the tensor of "overall" moduli, i.e. the moduli of the homogeneous body. (we used here (1.8)).

Hence,

$$\bar{L}_{ijkl}\,\bar{e}_{ij}\,\bar{e}_{kl} \le \left(\frac{1}{V}\int_V L_{ijkl}\,dV\right)\bar{e}_{ij}\,\bar{e}_{kl},$$

so $\left(\bar{L}_{ijkl} - \frac{1}{V}\int_V L_{ijkl}\,dV\right)$ is negative (semi) definite.

Suppose now the constituent phases are isotropic, and the geometry of the composite is such that $\bar{L}$ is isotropic. Then we can write L, $\bar{L}$ in the forms

$$L_{ijkl} = \left(\kappa - \tfrac{2}{3}\mu\right)\delta_{ij}\delta_{kl} + \mu\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right),$$

and $\bar{L}_{ijkl}$ similarly, so that

$$L_{ijkl}\,e_{ij}\,e_{kl} = \kappa(e_{kk})^2 + \mu\,e'_{ij}\,e'_{ij}, \qquad (2.7)$$

where $e'_{ij} = e_{ij} - \frac{1}{3}\delta_{ij}\,e_{kk}$. It follows then that

$$\left.\begin{aligned}\bar{\kappa} - \frac{1}{V}\int_V \kappa\,dV &\le 0,\\ \bar{\mu} - \frac{1}{V}\int_V \mu\,dV &\le 0,\end{aligned}\right\} \qquad (2.8)$$

so that $\bar{\kappa}$ and $\bar{\mu}$ are bounded from above. The bounds are called the ...

Let us now define the tensor of elastic compliances M_{ijkl}, so that

$$e_{ij} = M_{ijkl}\,\sigma_{kl} \qquad (2.9)$$

Correspondingly,

$$\bar{e}_{ij} = \bar{M}_{ijkl}\,\bar{\sigma}_{kl} \qquad (2.10)$$

so that $\bar{M}$ is the inverse of $\bar{L}$ and, since $W_c(\sigma)$ is numerically equal to $W(e)$,

$$\bar{W}(\bar{e}) = \tfrac{1}{2}\bar{M}_{ijkl}\,\bar{\sigma}_{ij}\,\bar{\sigma}_{kl} \quad , \text{ in numerical value.} \qquad (2.11)$$

If, now, we apply tractions $\sigma_{ij}n_j = \bar{\sigma}_{ij}n_j$ over all of ∂V, the complementary energy principle states that

$$-\bar{W}(\bar{e}) \geq -\frac{1}{V}\int W_c(\sigma^*)\,dV \; ,$$

for any admissible σ^* and, in particular, for $\sigma^* = \bar{\sigma}$. It follows that

$$\left(\bar{M}_{ijkl} - \frac{1}{V}\int_V M_{ijkl}\right)\bar{\sigma}_{ij}\,\bar{\sigma}_{kl} \leq 0 . \qquad (2.12)$$

For an isotropic $\bar{M}$,

$$\bar{M}_{ijkl} = \frac{1}{3\bar{\kappa}}\delta_{ij}\delta_{kl} + \frac{1}{2\bar{\mu}}\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - \tfrac{2}{3}\delta_{ij}\delta_{kl}\right) \qquad (2.13)$$

so that, when the phases are isotropic and the composite is also isotropic,

$$\left.\begin{aligned} \frac{1}{\bar{\kappa}} - \frac{1}{V}\int_V \frac{dV}{\kappa} &\leq 0, \\ \frac{1}{\bar{\mu}} - \frac{1}{V}\int_V \frac{dV}{\mu} &\leq 0 . \end{aligned}\right\} \qquad (2.14)$$

Thus, combining (2.8) and (2.14),

$$\left.\begin{aligned}\left(\frac{1}{V}\int_V \frac{dV}{\kappa}\right)^{-1} \;&\le\; \bar{\kappa} \;\le\; \frac{1}{V}\int_V \kappa\, dV, \\ \left(\frac{1}{V}\int_V \frac{dV}{\mu}\right)^{-1} \;&\le\; \bar{\mu} \;\le\; \frac{1}{V}\int_V \mu\, dV.\end{aligned}\right\} \qquad (2.15)$$

The lower bounds are associated with the name Reuss [but I may have the names Reuss + Voigt interchanged – I forget!]. The bounds (2.15) were first proved by R Hill, around 1951.

Some more refined bounds will be developed later.

3. An integral equation.

Let L^0_{ijkl} be the elastic moduli of a "comparison" material, that is homogeneous. We define the "polarization stress" by the formula

$$\sigma_{ij} = L_{ijkl}\, e_{kl} = L^0_{ijkl}\, e_{kl} + \tau_{ij}, \qquad (3.1)$$

so that

$$\tau_{ij} = \left(L_{ijkl} - L^0_{ijkl}\right) e_{kl}; \qquad (3.2)$$

σ_{ij}, e_{ij} are the stress and strain in the inhomogeneous body.
Suppose, for the moment, that τ_{ij} is known. Then, since $\sigma_{ij,j} = 0$, we have

$$L^0_{ijkl}\, u_{k,lj} + \tau_{ij,j} = 0 \quad \text{in } V. \qquad (3.3)$$

If, for example, displacements are prescribed on ∂V, we define $G_{ij}(x, x')$ as the Greens function for the displacement boundary value problem for the "comparison" material. Then, we have

$$u_i(x) = \int_V G_{ik}(x, x')\, \tau_{kl,l}(x')\, dx' + u_i^0(x), \qquad (3.4)$$

and u_i^0 is the displacement that would be produced in the comparison material by the imposed surface displacements, if no body-force were applied. Hence, by Gauss,

$$u_{i,j}(x) = -\frac{\partial}{\partial x_j}\int_V \frac{\partial G_{ik}(x,x')}{\partial x'_l}\left\{\tau_{kl}(x') - \bar{\tau}_{kl}\right\} dx'$$

$$+ \int_{\partial V} \frac{\partial G_{ik}(x,x')}{\partial x_j}\left\{\tau_{kl}(x') - \bar{\tau}_{kl}\right\} n_l \, dS + u^{0}_{i,j}(x), \quad (3.5)$$

for any constant $\bar{\tau}_{kl}$, and, in particular, when $\bar{\tau}_{kl}$ is the mean value of $\tau_{kl}(x)$:

$$\bar{\tau}_{kl} = \frac{1}{V}\int_V \tau_{kl}(x)\, dx \quad (3.6)$$

Having established (3.5), we return to (3.2), which we now write in the form

$$e_{kl} = P_{klij}\, \tau_{ij}, \quad (3.7)$$

so that P is the inverse to $L - L^{0}$:

$$P = \left(L - L^{0}\right)^{-1}, \quad (3.8)$$

on the space of symmetric second-order tensors. Eqns (3.5) and (3.7) now give, symbolically,

$$P\tau = A\tau + e^{0}, \quad (3.9)$$

where e^{0}_{ij} is the strain corresponding to $u^{0}_{i,j}$ and A is the operator:

$$A_{ijkl}\tau_{kl} = \frac{1}{2}\left[-\frac{\partial}{\partial x_j}\int_V \frac{\partial G_{ik}(x,x')}{\partial x'_l}\left\{\tau_{kl}(x') - \bar{\tau}_{kl}\right\} dx' + \int_{\partial V} \frac{\partial G_{ik}(x,x')}{\partial x_j}\left\{\tau_{kl}(x') - \bar{\tau}_{kl}\right\} n_l \, dS\right.$$

$$\left. \ldots \right]$$

The integral equation (3.9) of course requires knowledge of the Green's function G. We now derive G, for an *infinite body*.

First, note the plane wave decomposition

$$\delta(\underline{x}) = -\frac{1}{8\pi^2}\int_{|\underline{\xi}|=1} \delta''(\underline{\xi}.\underline{x})\,d\omega \qquad (3.11)$$

[*Proof*. $\nabla^2\left\{\int_{|\underline{\xi}|=1} \delta(\underline{\xi}.\underline{x})\,d\omega\right\} = \nabla^2\left\{\frac{2\pi}{|\underline{x}|}\right\} = -8\pi^2\delta(\underline{x})$.]

Now the Green's function $G(x)$ satisfies the equation

$$L^0(\nabla)G + I\,\delta(\underline{x}) = 0 \qquad (3.12)$$

in matrix notation, where $\left(L^0(\xi)\right)_{ik} = L^0_{ijkl}\,\xi_j\,\xi_l$. Let us consider now the problem

$$L^0(\nabla)G_\xi + I\,\delta''(\underline{\xi}.\underline{x}) = 0. \qquad (3.13)$$

Clearly G_ξ depends only on $(\underline{\xi}.x)$, so $L^0(\nabla)G_\xi = L^0(\xi)G_\xi''(\xi.\underline{x})$

Hence,

$$L^0(\xi)\,G_\xi(\underline{\xi}.\underline{x}) + I\,\delta(\underline{\xi}.\underline{x}) = 0,$$

or

$$G_\xi(\underline{\xi}.\underline{x}) = -\left(L^0(\xi)\right)^{-1}\delta(\underline{\xi}.\underline{x}) \qquad (3.14)$$

Now using (3.11), we deduce

$$G(\underline{x}) = -\frac{1}{8\pi^2}\int_{|\underline{\xi}|=1} G_\xi(\underline{\xi}.\underline{x})\,d\omega = +\frac{1}{8\pi^2}\int_{|\underline{\xi}|=1}\left(L^0(\xi)\right)^{-1}\delta(\underline{\xi}.\underline{x})\,d\omega. \qquad (3.15)$$

Hence, reverting to the eqn (3.9), with A defined by (3.10), we would get, for an *infinite* region at least, expressions like

$$\int G(x-x')f(x')\,dx' = +\frac{1}{8\pi^2}\int\left(L^0(\xi)\right)^{-1}d\omega\int\delta[\xi.(x-x')]\,f(x')\,dx' \qquad (3.16)$$

The last integral is just an integral of f over the plane $\underline{\xi}.\underline{x}' = \underline{\xi}.\underline{x}$. The Radon transform of f is defined as

$$\check{f}(p,\xi) = \int f(x)\delta(\xi.x - p)\,dx \quad ; \tag{3.17}$$

Then,

$$\int G(x-x')f(x')\,dx' = +\frac{1}{8\pi^2}\int_{|\xi|=1} (L^0(\xi))^{-1}\,d\omega\,\check{f}(\xi.x,\xi). \tag{3.18}$$

All this, of course, is assuming that (3.17) is defined.

We have now that

$$(\Lambda\tau)_{ij} = \frac{1}{2}\left\{+\frac{1}{8\pi^2}\int_{|\xi|=1}\xi_j\xi_l(L^0(\xi))^{-1}_{ik}(\tau_{kl}-\bar{\tau}_{kl})^{\check{}\,\prime\prime}\,d\omega + (i\leftrightarrow j)\right\}, \tag{3.19}$$

for an infinite body, where $\check{f}''(p,\xi) = \dfrac{\partial^2\check{f}(p,\xi)}{\partial p^2}$, since the volume integral term in (3.10) contains two derivatives.

4. Dilute suspension theory.

Suppose now our composite comprises a homogeneous matrix with moduli $L^{(1)}$, say, with a dilute suspension of inclusions, with moduli $L^{(2)}$, and concentration c_2. We shall now consider the calculation of $\bar{L}$, to order (c_2). First, we have, in the inclusions, the mean strain e_2, say, and in the matrix, e_1, so that

$$\bar{e} = c_1 e_1 + c_2 e_2, \tag{4.1}$$

where $\bar{e}$ is the prescribed mean strain $(c_1 + c_2 = 1)$.

Also, suppose the mean stress in the inclusions is σ_2 and σ_1 in the matrix. Then, Also, of course,

$$\bar{\tau} = \bar{L}\,\bar{e}. \qquad (4.3)$$

We can eliminate e_1 from (4.1) and (4.2), so that

$$\bar{\sigma} = L^{(1)}\left(\bar{e} - c_2 e_2\right) + c_2 L^{(2)} e_2 = L^{(1)}\bar{e} + c_2\left(L^{(2)} - L^{(1)}\right)e_2. \qquad (4.4)$$

Suppose now we choose the <u>matrix</u> as the comparison material. The polarization stress τ is now

$$\tau = \left(L^{(2)} - L^{(1)}\right)e \quad , \text{ in inclusions}$$
$$= 0 \quad , \text{ in matrix},$$

and hence

$$\bar{\tau} = c_2\left(L^{(2)} - L^{(1)}\right)e_2 \qquad (4.5)$$

Thus, exactly,

$$\bar{\sigma} = L^{(1)}\bar{e} + \bar{\tau} \qquad (4.6)$$

The integral equation (3.9) now applies for $x \in$ any inclusion and, since the inclusions are <u>dilute</u>, the integral over V can be replaced by the integral over the inclusion, V_i say, being considered (thus we are neglecting interactions). We have, therefore,

$$(P\tau)_{ij} = \frac{1}{2}\left\{+\frac{1}{8\pi^2}\int_{|\xi|=1} \xi_j \xi_l \left(L^0(\xi)\right)^{-1}_{ik} \check{\tau}''_{kl}(\xi.x, \xi) + (i \leftrightarrow j)\right\} + \bar{e}_{ij}, \quad (4.7)$$

~~disregarding $\bar{\tau}$~~

where now $\check{\tau} = \int_{V_i} \tau(x)\,\delta(\xi.x - p)\,dx$,

and disregarding $\bar{\tau}$ as it would produce a term of order c_2^2.

Now consider the case when V_i is a <u>sphere</u> of radius 'a'. We have clearly

<u>if</u> τ is constant over V_i, $\check{\tau} = \pi(a^2 - p^2)$, $|p| < a$

and we see that (4.7) can be satisfied with τ_{kl} constant over V_i, so long as τ_{kl} satisfies the linear algebraic equations

$$P_{ijkl}\,\tau_{kl} + \left\{ \frac{1}{8\pi} \int_{|\xi|=1} \xi_j \xi_l \left(L^0(\xi)\right)^{-1}_{ik} d\omega \;+\; (i \leftrightarrow j) \right\} \tau_{kl} = \bar{e}_{ij} \,, \qquad (4.8)$$

where $P = \left(L^{(2)} - L^{(1)}\right)^{-1}$. Eqn (4.6) then implies $\bar{\sigma} = L^{(1)}\bar{e} + c_2 \tau + O(c_2^2)$

[Ellipsoids have the same property, that τ'' is constant, but the constant of course depends on the ratios of the axes.]

Example: Isotropic phases

For an isotropic body, with

$$L_{ijkl} = \lambda \delta_{ij}\delta_{kl} + \mu(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}),$$

the equilib. eqns are

$$L(\nabla)u \equiv (\lambda+\mu)\nabla \operatorname{div} u + \mu \nabla^2 u = 0 ,$$

so
$$L_{ik}(\xi) = (\lambda+\mu)\xi_i \xi_k + \mu \xi_j \xi_j \delta_{ik} \,, \qquad (4.9)$$

and
$$\left(L^{-1}(\xi)\right)_{ik} = \frac{1}{\mu|\xi|^2}\left[\delta_{ik} - \left(\frac{\lambda+\mu}{\lambda+2\mu}\right)\frac{\xi_i \xi_k}{|\xi|^2}\right] \quad ; \quad |\xi|^2 = \xi_j \xi_j \qquad (4.10)$$

There are essentially only two different terms in (4.8), one for the hydrostatic part of τ, and one for the shear part. Thus, we need to evaluate only the cases $\tau_{kl} = \tau\,\delta_{kl}$ and $\tau_{kl} = \tau_{12}$, $(k,l) = 1,2$. The result is:

$$\left.\begin{aligned} &\frac{1}{3(K^{(2)}-K^{(1)})}\,\tau_{ii} + \frac{1}{3\left(K^{(1)}+\frac{4}{3}\mu^{(1)}\right)}\,\tau_{ii} = \bar{e}_{ii} \,, \\ &\frac{1}{\quad}\,\tau' + \left(3\left(K^{(1)}+2\mu^{(1)}\right)\right)\tau' = \bar{e}' \end{aligned}\right\} \qquad (4.11)$$

where $\tau'_{ij} = \tau_{ij} - \frac{1}{3}\delta_{ij}\tau_{kk}$, and similarly for $\bar e'_{ij}$.

The overall stress-strain law (4.6) thus becomes, approximately,

$$\bar\sigma_{kk} = 3\bar K \bar e_{kk}, \qquad \bar\sigma'_{ij} = 2\bar\mu\, \bar e'_{ij}, \tag{4.12}$$

where

$$\bar K = K^{(1)} + \frac{c_2}{3}\left\{\frac{1}{3(K^{(2)}-K^{(1)})} + \frac{1}{(3K^{(1)}+4\mu^{(1)})}\right\}^{-1}, \tag{4.13}$$

$$\bar\mu = \mu^{(1)} + \frac{c_2}{2}\left\{\frac{1}{2(\mu^{(2)}-\mu^{(1)})} + \frac{3(K^{(1)}+2\mu^{(1)})}{5\mu^{(1)}(3K^{(1)}+4\mu^{(1)})}\right\}^{-1}. \tag{4.14}$$

Similar calculations could be performed for ellipsoids, and for anisotropic phases. If we had several different types of inclusion, each would contribute a term of the form given above.

5. The self-consistent method

Suppose now we have a matrix, with elastic moduli L^0_{ijkl}, and N different kinds of inclusions, with elastic moduli L^r_{ijkl}, $r = 1 \dots N$, and concentrations c_r. We can define $c_0 = 1 - \sum_{r=1}^{N} c_r$. We have now, if the mean stress in the rth phase is σ_r, and the mean strain e_r,

$$\bar e = c_0 e_0 + \sum_{r=1}^{N} c_r e_r,$$

$$\bar\sigma = c_0 L^0 e_0 + \sum_{r=1}^{N} c_r L^r e_r,$$

and $\bar\sigma = \bar L \bar e$.

Hence, eliminating e_0,

$$\bar\sigma = L^0 \bar e + \sum_{r=1}^{N} c_r (L^r - L^0) e_r = L_0 \bar e + \sum_{r=1}^{N} c_r \tau_r \tag{5.1}$$

generalizing (4.4), where τ_r is the mean polarization stress in the rth phase.

Now suppose that c_r are not small. We might attempt to all a single spherical inclusion of type r in a homogeneous material whose moduli are the overall moduli of the composite itself, namely, $\bar{L}$. Then if the composite is isotropic, with isotropic phases, τ_r is given by (4.11). with $L^{(2)}$ replaced by L^r and $L^{(1)}$ by $\bar{L}$. But then, from (5.1),

$$\bar{\sigma}_{kk} = 3\bar{K}\,\bar{e}_{kk}, \quad \bar{\sigma}'_{ij} = 2\bar{\mu}\,\bar{e}'_{ij},$$

Where

$$\left.\begin{aligned} \bar{K} &= K^{(0)} + \sum_{r=1}^{N} c_r \cdot \frac{1}{3}\left\{ \frac{1}{3(K^r-\bar{K})} + \frac{1}{3\bar{K}+4\bar{\mu}} \right\}^{-1}, \\ \bar{\mu} &= \mu^{(0)} + \sum_{r=1}^{N} c_r \cdot \frac{1}{2}\left\{ \frac{1}{2(\mu^r-\bar{\mu})} + \frac{3(\bar{K}+2\bar{\mu})}{5\bar{\mu}(3\bar{K}+4\bar{\mu})} \right\}^{-1} \end{aligned}\right\} \quad (5.2)$$

Equations (5.2) may be solved for $\bar{K}, \bar{\mu}$, to give the so-called "self-consistent" estimate of the overall moduli. In the absence of a matrix, $K^{(0)}$ and $\mu^{(0)}$ would simply be absent.

For a polycrystal, we may have just <u>one</u> anisotropic material, but the grains might have all orientations. For this case, we might consider the limit of several L^r, anisotropic, at concentrations c_r. By similar reasoning to the above, we would get

$$P^{(r)}\tau_r + \bar{A}\,\tau_r = \bar{e}, \quad (5.3)$$

$$\text{or} \quad \tau_r = \left(P^{(r)} + \bar{A}\right)^{-1}\bar{e}, \quad (5.4)$$

Where $P^{(r)} = \left(L^r - \bar{L}\right)^{-1}$ and (5.5)

$$\bar{A}_{ijkl} = \left(\frac{1}{\bar{K}+\frac{4}{3}\bar{\mu}}\right)\delta_{ij}\delta_{kl} + \frac{3(\bar{K}+2\bar{\mu})}{10\bar{\mu}(3\bar{K}+4\bar{\mu})}\left\{\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}-\frac{2}{3}\delta_{ij}\delta_{kl}\right\} \quad (5.6)$$

(cf. (4.11)).

6. More refined bounds

We shall now derive a variational principle, due to Hashin & Shtrikman, J. Mech. Phys. Solids, 1962. We re-capitulate the integral equation (3.9):

$$P\tau = A\tau + e^{\circ}, \qquad (3.9)$$

and note that

$$\varepsilon = A\tau$$

is the strain that would be produced by τ, if zero boundary displacements were imposed. It is easy to show that A is self-adjoint, for, if

$$(\tau_1, A\tau_2) = \int_V \tau^{(1)}_{ij} A_{ijkl} \tau^{(2)}_{kl} \, dV,$$

we have

$$(\tau_1, A\tau_2) = \int_V \tau^{(1)}_{ij} \varepsilon^{(2)}_{ij} \, dV = \int_V \tau^{(1)}_{ij} u^{(2)}_{i,j} \, dV$$

$$= -\int_V \tau^{(1)}_{ij,j} u^{(2)}_{i} \, dV \quad , \text{ by Gauss, since } u^{(2)} = 0 \text{ on } \partial V.$$

But $\tau^{(1)}_{ij,j} = -L^{\circ}_{ijkl} u^{(1)}_{k,lj}$, so

$$(\tau_1, A\tau_2) = \int_V u^{(1)}_{k,lj} L^{\circ}_{ijkl} u^{(2)}_{i} \, dV = -\int_V u^{(1)}_{k,l} L^{\circ}_{ijkl} u^{(2)}_{i,j} \, dV = (\tau_2, A\tau_1), \quad (6.1)$$

by reversing the reasoning, using $L^{\circ}_{ijkl} = L^{\circ}_{klij}$. We also have $P_{ijkl} = P_{klij}$ and so, immediately, eqn. (3.9) is self-adjoint and

$$\delta\left\{ (\tau, P\tau) - (\tau, A\tau) - 2(\tau, e^{\circ}) \right\} = 0 \qquad (6.2)$$

Next, we note that, when τ is the actual polarization,

$$(\tau, P\tau) - (\tau, A\tau) - 2(\tau, e^{\circ}) = -(\tau, e^{\circ}) = -\int_V \tau_{ij} u^{\circ}_{i,j} \, dV$$

$$= \int ((L^{\circ}_{ijkl} - L_{ijkl}) u_{k,l} \, u^{\circ}_{i,j} \, dV$$

But $L^0_{ijkl} u^0_{i,j} = \sigma^0_{kl}$, say, and $\sigma^0_{kl,l} = 0$.

Also, $L_{ijkl} u_{k,l} = \sigma_{ij}$, and $\sigma_{ij,j} = 0$

Hence,

$$-(\tau, e^0) = \int_{\partial V} \sigma^0_{ij} u_i n_j \, dS - \int_{\partial V} \sigma_{ij} u^0_i n_j \, dS .$$

But $u_i = u^0_i$ on ∂V and so we see that

$$(\tau, P\tau) - (\tau, A\tau) - 2(\tau, e^0) = 2(E_0 - E) , \qquad (6.3)$$

where $E_0 = \frac{1}{2}(e^0, L^0 e^0)$ and $E = \frac{1}{2}(e, Le)$.

Next, if we put $\tau_1 = \tau_2$ in (6.1), we see that

$$(\tau_1, A\tau_1) \leq 0 \quad \text{for all } \tau_1$$

Therefore, if $(\tau_1, P\tau_1) \geq 0$ for all τ_1, i.e. if P is positive (semi-) definite $\Leftrightarrow$ <u>if $L - L^0$ is positive semi definite</u> at each point, we have

$$(\tau, P\tau) - (\tau, A\tau) \geq 0$$

and our variational principle (6.2) is actually a <u>minimum</u> principle:

$$2(E_0 - E) \leq (\tau, P\tau) - (\tau, A\tau) - 2(\tau, e^0) , \text{ for any } \tau . \quad (6.4)$$

We can also show that, if <u>$L - L^0$ is negative (semi) definite at each point</u>, then $(\tau, P\tau) - (\tau, A\tau) \leq 0$ and

$$2(E_0 - E) \geq (\tau, P\tau) - (\tau, A\tau) - 2(\tau, e^0) \quad \text{for any } \tau . \quad (6.5)$$

The results (6.4), (6.5) give further possibilities for bounding the overall moduli, by choosing different L^0.

To prove the result (6.5), we introduce, dually to τ, a strain polarization $\eta = M_0 \tau \Leftrightarrow \tau = L_0 \eta$. If we define $e = A\tau$, so that e is associated with a displacement that vanishes on ∂V, we have $\sigma = L_0 e + \tau$, $e = M_0 \sigma - \eta$, but not $\sigma = Le$, $e = M\sigma$, because τ is any polarization, not necessarily the correct one for a solution of the inhomogeneous problem.

We now have

$$(\tau, A\tau) = (\tau, e) = (L_0\eta, e) = (\eta, L_0 e) = (\eta, \sigma - L_0\eta)$$

$$= (M_0\sigma - e, \sigma) - (\eta, L_0\eta)$$

$$= (\sigma, M_0\sigma) - (\eta, L_0\eta), \quad \text{since} \quad (e, \sigma) = (\sigma, e) = 0 \quad \text{as}$$

$\sigma_{ij,j} = 0$ and the displacement vanishes on ∂V.

Hence,

$$(\tau, P\tau) - (\tau, A\tau) = (L_0\eta, (L - L_0)^{-1} L_0\eta) + (\eta, L_0\eta) - (\sigma, M_0\sigma).$$

But $\quad (L - L_0)^{-1} L_0 = (L - L_0)^{-1}(L_0 - L + L) = (L - L_0)^{-1} L - I.$

Also, $\quad (L - L_0) M_0 = L M_0 - I = L(M_0 - M),$

so $\quad (L - L_0)^{-1} L = M_0 (M_0 - M)^{-1}.$

Thus,

$$(\tau, P\tau) - (\tau, A\tau) = (L_0\eta, M_0(M_0 - M)^{-1}\eta) - (L_0\eta, \eta) + (\eta, L_0\eta) - (\sigma, M_0\sigma)$$

$$= (\eta, (M_0 - M)^{-1}\eta) - (\sigma, M_0\sigma) \leq 0$$

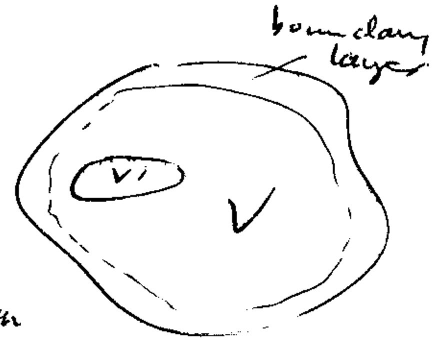
if $(M_0 - M)^{-1}$ is negative semi definite, which is equivalent to $(L - L_0)$ negative semi definite.]

Now to exploit the principles.

To evaluate the term $(\tau, P\tau)$, we assume that the composite is "homogeneous", in the sense that

$$\frac{1}{V'}\int_{V'} \tau_{ij} P_{ijkl} \tau_{kl}\, dx$$

is independent of the choice of V', provided only that V' has dimensions large compared with a typical microstructural dimension d. In that case, we see that, if V' is allowed to range over V, all possible configurations of the composite are sampled, and



$$\frac{1}{V}(\tau, P\tau) = \mathcal{E}(\tau, P\tau) = \mathcal{E}\left(\tau_{ij}(x) P_{ijkl} \tau_{kl}(x)\right)$$

the *expectation value* being taken at some chosen point x, and evaluated over all possible realizations of the composite.

Similarly, to evaluate $(\tau, A\tau)$, we note first that the integral over ∂V in the definition (3.10) of $A\tau$ is (probably!) significant only in a "boundary layer" whose thickness is of order d, and so will not contribute to the mean value $\frac{1}{V}(\tau, A\tau)$. Then, again appealing to "statistical homogeneity",

$$\frac{1}{V}(\tau, A\tau) = \mathcal{E}(\tau, A\tau) = \mathcal{E}\left\{\tau_{ij}(x)\left(A_{ijkl}\tau_{kl}\right)(x)\right\}. \qquad (6.6)$$

Thus, disregarding the integral over ∂V,

$$\frac{1}{V}(\tau, A\tau) = \mathcal{E}\left\{-\frac{1}{2}\int_V \frac{\partial^2 G_{ik}(x,x')}{\partial x_j\, \partial x_l'}\, \tau_{ij}(x)\left(\tau_{kl}(x') - \bar{\tau}_{kl}\right) dx' + (i \leftrightarrow j)\right\},$$

so $$\frac{1}{V}(\tau, A\tau) = \left\{-\frac{1}{2}\int \frac{\partial^2 G_{ik}(x,x')}{\cdots}\, \mathcal{E}\left\{\tau_{ij}(x)\left(\tau_{kl}(x') - \bar{\tau}_{kl}\right)\right\} dx' + (i \leftrightarrow j)\right\} \qquad (6.7)$$

(2)

The integrand in (6.7) has an r^{-3} singularity and is to be interpreted in the sense of distributions, when it agrees with (3.10).

Eqn. (6.7) can be written symmetrically, by noting that $\mathcal{E}(\tau-\bar{\tau})=0$, so that

$$\frac{1}{V}\left(\tau, A\tau\right) = \left\{-\frac{1}{2}\int_V \frac{\partial^2 G_{ik}(x,x')}{\partial x_j\,\partial x_l'}\ \mathcal{E}\left\{\left(\tau_{ij}(x)\,\tau_{kl}(x') - \bar{\tau}_{ij}\bar{\tau}_{kl}\right)\right\}dx + (i\Leftrightarrow j)\right\}. \quad (6.8)$$

We may observe finally that the infinite body Greens function behaves like $|x-x'|^{-3}$ for large $|x-x'|$ and so the integral in (6.8) would be defined if $V\to\infty$ and G were replaced by the infinite body Green's function, $G(x-x')$. [The integral converges only because, when $x-x'$ is large, the expectation value in (6.8) tends to zero. Strictly, of course, some estimates should be given for the various terms that are being dropped, but there is no time here!]

Hence, finally, we have

$$\frac{2}{V}\left(E_0 - E\right) \underset{(\le)}{\ge} \mathcal{E}\left(P_{ijkl}\left(\tau_{ij}(x)\tau_{kl}(x)\right)\right)$$
$$+\frac{1}{2}\left[\int\!\!\int G_{ik,jl}(x-x')\ \mathcal{E}\left\{\left(\tau_{ij}(x)\,\tau_{kl}(x') - \bar{\tau}_{ij}\,\bar{\tau}_{kl}\right)\right\}dx' + (i\Leftrightarrow j)\right]$$
$$-2\,\bar{\tau}_{ij}\,e^0_{ij}\,, \quad (6.9)$$

if $L-L^0$ is negative (positive) semi-definite everywhere.

We can now generate bounds for E by choosing a particular form for $\tau(x)$. The simplest choice is to let

We have, then,

$$\bar{\tau}_{ij} = \mathcal{E}\left(\tau_{ij}(x)\right) = \sum_r c_r \tau_{ij}^{(r)} \qquad (6.10)$$

and

$$\mathcal{E}\left\{P_{ijkl}(x)\left(\tau_{ij}(x)\tau_{kl}(x)\right)\right\} = \sum_r c_r \tau_{ij}^{(r)} \tau_{kl}^{(r)} P_{ijkl}^{(r)} \qquad (6.11)$$

where c_r is the concentration of the r^{th} phase $\left(\sum_r c_r = 1\right)$, and $P_{ijkl}^{(r)} = \left(L_{ijkl}^{(r)} - L_{ijkl}^{o}\right)^{-1}$.

Also,

$$\mathcal{E}\left(\tau_{ij}(x)\tau_{kl}(x') - \bar{\tau}_{ij}\bar{\tau}_{kl}\right) = \sum_r \sum_s \tau_{ij}^{(r)}\tau_{kl}^{(s)}\left[P_{rs}(x,x') - c_r c_s\right], \qquad (6.12)$$

where the two-point correlation function $P_{rs}(x, x')$ represents the probability that phase r will be found at x and phase s will be found at x'. At large separations $(|x-x'| \to \infty)$, the properties at r and at s will be assumed independent, so that

$$P_{rs}(x, x') - c_r c_s \to 0 \quad \text{as} \quad |x-x'| \to \infty. \qquad (6.13)$$

Finally, for statistical homogeneity, P_{rs} depends upon x, x' only through $(x-x')$. Therefore, we obtain

$$\frac{2}{V}\left(E_o - E\right) \underset{(\leq)}{\geq} \sum_r c_r P_{ijkl}^{(r)} \tau_{ij}^{(r)}\tau_{kl}^{(r)}$$

$$+ \frac{1}{2}\left[\sum_r \sum_s \tau_{ij}^{(r)}\tau_{kl}^{(s)} \int G_{ikjl}(x-x')\left[P_{rs}(x-x') - c_r c_s\right] dx' + (i \leftrightarrow j)\right] - 2\sum_r c_r \tau_{ij}^{(r)} e_{ij}^{o} \qquad (6.14)$$

The inequality (6.14) yields bounds, as soon as the integral has been evaluated. A particularly simple example, derived rather differently by Hashin and Shtrikman, J. Mech. Phys. Solids, ~1964, is obtained when the correlation function $\rho_{rs}(x-x')$ is <u>isotropic</u>

$$\rho_{rs}(x-x') = \rho_{\sigma}(|x-x'|) \tag{6.15}$$

(it is not necessary to assume anything else - eg, higher-order correlation functions might be anisotropic).

We already know, from Section ~~4~~, that if $\tau_{ij} = \text{const}, r<a$ and $\tau_{ij} = 0, r>a$,

$$\int G_{ik,jl}(x-x')\,\tau_{\cancel{kl}}(x')\,dx' = A_{ijkl}\,\tau_{kl}, \quad |x|<a,$$

where

$$A_{ijkl} = -\frac{1}{4\pi}\int_{|\xi|=1} \xi_j \xi_l \left(L^0(\xi)\right)^{-1}_{ik}\,dw.$$

Therefore, in particular, when $x=0$,

$$\int_{|x'|<a} G_{i\cancel{k},jl}(x')\,dx' = A_{ijkl},$$

independently of 'a', and so,

$$\int_{a<|x'|<b} G_{ik,jl}(x')\,dx' = 0 \tag{*}$$

for all a, b; $a<b$. Therefore, for any function $f(|x'|)$, we have

$$\int G_{ik,jl}(x')\,f(|x'|)\,dx' = A_{ikjl}\,f(0).$$

Applying (6.16) to the integral in (6.14) now gives

$$\frac{2}{V}\left(E_0 - E\right) \underset{(\leq)}{\geq} \sum_r c_r \rho^{(r)}_{ijkl}\, \tau^{(r)}_{ij} \tau^{(r)}_{kl}$$

$$+ \frac{1}{2}\left[\sum_r \sum_s \tau^{(r)}_{ij} \tau^{(s)}_{kl} A_{ijkl} \left(c_r \delta_{rs} - c_r c_s \right) + (i \leftrightarrow j) \right]$$

$$- 2 \sum_r c_r \tau^{(r)}_{ij} e^0_{ij} \qquad (6.17)$$

since, plainly,

$$\left. \begin{aligned} P_{rs}(0) &= c_r \quad \text{if} \quad r = s \\ &= 0 \quad \text{if} \quad r \neq s \end{aligned} \right\} \qquad (6.18)$$

Thus, defining W_0 as E_0/V, and $\bar{W}$ as E/V (as previously), we have

$$2(W_0 - W) \underset{(\leq)}{\geq} \sum_r c_r \tau^{(r)}_{ij} \tau^{(r)}_{kl} \left[\rho^{(r)}_{ijkl} + A_{ijkl} \right]$$

$$- \sum_r \sum_s c_r c_s \tau^{(r)}_{ij} \tau^{(s)}_{kl} A_{ijkl} - 2 \sum_r c_r \tau^{(r)}_{ij} e^0_{ij}. \quad (6.19)$$

The right side of (6.19) may now be extremized by choosing the $\tau^{(r)}_{ij}$ to satisfy

$$c_r\left[\left(\rho^{(r)}_{ijkl} + A_{ijkl}\right)\tau^{(r)}_{kl} - \sum_s c_s A_{ijkl} \tau^{(s)}_{kl} - e^0_{ij}\right] = 0 \qquad (6.20)$$

When (6.20) is satisfied, the right side of (6.19) reduces to give

$$2(W_0 - W) \underset{(\leq)}{\geq} - \sum_r c_r \tau^{(r)}_{ij} e^0_{ij}, \qquad (6.21)$$

and $\sum c_r \tau^{(r)}$ is easily found from (6.20) to have the value

$$\sum_r c_r \tau_{ij}^{(r)} = \bar{\tau}_{ij} ,$$

where, symbolically,

$$\bar{\tau} = \left[I - \sum_r c_r (P^r + A)^{-1} A \right]^{-1} \sum_s c_s (P^s + A)^{-1} e^0 . \tag{6.22}$$

Substituting the value of A from Section 4 (in effect, from eqn. (4.11)) gives the required bounds, when the 'comparison material' is chosen so that $L - L^0$ is positive or negative semi-definite. For a composite with isotropic phases, the best choices are $K^0 = \mathrm{Max}$ or $\mathrm{Min}\ (K^r)$, $\mu^0 = \mathrm{Max}$ or $\mathrm{Min}\ (\mu^r)$. The bounds were first produced, with some restrictions, by Hashin and Shtrikman, and generalized by L.J. Walpole (J. Mech. Phys. Solids, 1966) to the form given above. The variational principle (6.14) has not been given before, however, and is applicable to more general situations.

The above is not the only approach that can be followed to obtain bounds, and reference should be made to the work of Dederichs and Zeller (Physica Status Solidi, 1972). They applied formal perturbation theory to obtain estimates of moduli, and, by substitution into the classical variational principles (§ 2), obtained bounds involving n-point correlation functions. (6.14), however, is probably the best that can be done with 2-point correlation functions alone. E Kröner has very recently been working upon extending the work of Dederichs - Zeller (not yet published).

It should, perhaps, be mentioned that the bounds are of (and reasonably accurate) method of estimating moduli, however, is the self-consistent method (§5). This is also used for some nonlinear problems, treated incrementally; a recent application is that of J.W. Hutchinson (Proc. R. Soc. 1975), to the nonlinear creep of a polycrystal, for instance.