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ON STATIONARY DIFFUSION IN HETEROGENEOUS MEDIA

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Abstract

This work concerns steady state diffusion in a medium containing a random distribution of sinks. The physical properties of the medium are described in terms of geometrical correlation functions, for example, two or three point correlations. Variational principles of classical type are developed and configuration dependent trial fields are substituted into them. Restrictions on certain integrals of the two and three point correlation functions then follow from the avoidance of mathematical contradictions. The consequences of the restrictions and the relationship between the classical variational principles and variational principles of Hashin-Shtrikman type are discussed.

1 Introduction

The main focus of this work is the problem of determining the overall sink strength of an inhomogeneous "lossy" material. In the steady state, the problem is described mathematically by the equation

$$\Delta c - k^2(x)c + K = 0, \tag{1.1}$$

where c represents the concentration of some diffusing species, K is its generation rate (which may depend on position) and the loss term $-k^2(x)c$ models a continuous distribution of sinks. The sink strength k^2 varies on a microscopic length scale characterized by a length scale a, say. The problem is then to find an 'overall sink strength' $\tilde{k}^2$, so that, when K varies slowly relative to the microscale, some 'local average' $\tilde{c}$ of c satisfies a 'homogenized' version of (1.1):

$$\Delta\tilde{c} - \tilde{k}^2\tilde{c} + K = 0. \tag{1.2}$$

If $k^2(x)$ is bounded, it has been proved that $\tilde{k}^2$ exists and is equal to the mean value, $\overline{k^2}$, in the homogenization limit $a \to 0$ (Papanicolaou (1980). We are interested also in the limit

$a \to 0$ while $\max(ka)$ remains finite, however. We are not aware of any homogenization theorem in this case. A number of methods have been proposed for estimating $\tilde{k}^2$ including the simple self-consistent scheme of Brailsford and Bullough (1981) and methods based on variational principles developed by Reck and Prager (1965), and Talbot and Willis (1984a, b). Under the assumption that (1.1) indeed can be homogenized to the form (1.2), Talbot and Willis (1984b) gave a definition of $\tilde{k}^2$ analogous to the definition of overall elastic moduli given by Hill (1963). In this work $\tilde{k}^2$ is defined in terms of certain energy principles.

Talbot and Willis (1984b) considered a variational characterization of (1.1) for a finite body with boundary condition $\partial c/\partial n = 0$ where n is the outward normal to the surface of the body. This meant that the trial fields that were substituted into the variational principle also had to satisfy the boundary condition. The trial fields involved a Green function and in turn this meant that a finite body Green function was used. As the shape of the body was unspecified, the finite-body Green function was, in general, unknown and this meant that a careful limiting process had to be undertaken in order to obtain results using the *infinite-body* Green function. In this paper energy principles are considered which are free of this restriction. Configuration-dependent trial fields which involve the infinite-body Green function can now be employed directly and, by comparing the resulting bound on the energy with that obtained using a constant trial field, restrictions on certain integrals involving two and three point correlation functions can be derived, analogous to those obtained by Milton (1981). The bounds involve both two and three point correlation functions and the restrictions imply a range of values which the bounds can assume. This range is also compared with bounds of Hashin-Shtrikman type.

In what follows, a random, statistically uniform medium of infinite extent is considered. The medium has two phases, with labels 1 and 2. It occupies d-dimensional space $\mathbf{R}^d$. Phase 1 (which need not be connected) occupies Ω_1 and phase 2 occupies its complement, Ω_2. The characteristic function of Ω_1 is f_1. Thus,

$$\begin{aligned} f_1(x) &= 1 \quad \text{if} \quad x \in \Omega_1 \\ &= 0 \quad \text{otherwise.} \end{aligned} \tag{1.3}$$

The statistical properties of the medium follow from the set of multipoint probabilities, or moments of f_1, Vanmarcke, (1983)

$$\begin{aligned} \eta_1 &= \langle f_1(0) \rangle, \\ \eta_r(z_1, z_2, \cdots z_{r-1}) &= \langle f_1(0) f_1(z_1) f_1(z_2) \cdots f_1(z_{r-1}) \rangle, \quad r = 2, 3, \cdots, \end{aligned} \tag{1.4}$$

where each $z_k \in \mathbf{R}^d$. The angled brackets signify ensemble averaging. Such multipoint probabilities are symmetric in their arguments. One point could be taken at the origin, because of the assumed statistical uniformity.

It is, in fact, convenient to work with η_1 and the multipoint moments

$$M_r(z_1, z_2, \cdots z_{r-1}) = \langle f_1'(0) f_1'(z_1) f_1'(z_2) \cdots f_1'(z_{r-1}) \rangle, \tag{1.5}$$

where

$$f_1'(z) = f_1(z) - \eta_1. \tag{1.6}$$

The sink strength $k^2(x)$ in (1.1) is taken to have the form

$$k^2(x) = k_1^2 f_1(x) + k_2^2(1 - f_1(x)), \tag{1.7}$$

where k_1^2 and k_2^2 are constants.

The plan of the remainder of this work is as follows. In the next two sections, variational principles and general bounds are derived. Bounds of Hashin-Shtrikman type are then considered. The bounds are then compared and some results presented.

2 Variational principles

Two variational principles associated with equation (1.1) will be considered. In each case, although the domain Ω over which the problem is defined will be taken to be finite, the field $k(x)$ will be considered to be defined over the whole of $\mathbf{R}^d$. Thus, the given domain Ω could be regarded as a "test specimen" cut from a much larger piece of material. It is convenient, to avoid the need for explicit volume averaging, to select units so that Ω has unit volume. Then, integrals over Ω coincide with volume averages.

The first variational problem to be considered is

$$V^*(K) = \sup_{c \in H_1(\Omega)} \int_\Omega \left[Kc - \frac{1}{2}[(\nabla c)^2 + k^2 c^2] \right] \mathrm{d}x. \tag{2.1}$$

A closely related principle is

$$V(\overline{c}) = \inf_{c \in S} \frac{1}{2} \int_\Omega \left[(\nabla c)^2 + k^2 c^2 \right] \mathrm{d}x, \tag{2.2}$$

where S is the set

$$S = \left\{ c \in H_1(\Omega) : \int_\Omega c \, \mathrm{d}x = \overline{c} \right\}. \tag{2.3}$$

The functions V and V^* are convex duals; the principle (2.1) is the more useful for present purposes because the field $c(x)$ is subject to no constraint.

The second variational principle that will be considered is

$$W(\overline{c}) = \sup_{(q,s) \in T} \int_\Omega \left[\overline{c}s - \frac{1}{2}(q^2 + s^2/k^2) \right] \mathrm{d}x, \tag{2.4}$$

where T is the set of fields

$$\begin{aligned} T = \Big\{ (q,s) \quad : \quad & q \in [L_2(\Omega)]^3, \ s \in L_2(\Omega), \\ & \textstyle\int_\Omega \ [\nabla \phi \cdot q + \phi \, s] \mathrm{d}x = 0 \quad \text{for all} \quad \phi \in H_1^0(\Omega), \int_\Omega \phi \, \mathrm{d}x = 0 \Big\}. \end{aligned} \tag{2.5}$$

This is closely related to the principle

$$W^*(\overline{s}) = \inf_{(q,s) \in T} \frac{1}{2} \int_\Omega [q^2 + s^2/k^2] \mathrm{d}x, \tag{2.6}$$

with the additional restriction that $\int_\Omega s\,\mathrm{d}x = \overline{s}$. Note that (2.2) and (2.6) are not *quite* natural duals, in the sense of Toland and Willis (1989): the principle dual to (2.2) has the additional restriction that $q \cdot n = 0$ on $\partial\Omega$. Likewise, the natural dual to (2.4) is like (2.1), except for the additional restriction that $c = \overline{c}$ on $\partial\Omega$. It follows that

$$V(\overline{c}) \leq W(\overline{c}) \quad \text{and} \quad W^*(\overline{s}) \leq V^*(\overline{s}). \tag{2.7}$$

3 Implications of the variational principles

For a two-phase medium as defined by (1.6), substitute into the integral in (2.1) the configuration-dependent trial field

$$c(x) = c_0 - \lambda \int G(x,y) f_1'(y)\mathrm{d}y, \tag{3.1}$$

where c_0 and λ are constants and $G(x,y)$ is any function defined for all $x \in \Omega$ and all $y \in \mathbf{R}^d$. Even though x is restricted to lie in Ω, the integral with respect to y is over all space. Since (1.6) is expressible in the form

$$k^2(x) = \overline{k^2} + [k^2] f_1'(x); \quad \overline{k^2} = \eta_1 k_1^2 + (1-\eta_1)k_2^2, \;\; [k^2] = k_1^2 - k_2^2, \tag{3.2}$$

this yields the inequality

$$\begin{aligned} V^*(K) \;\; \geq \;\; & K\Big(c_0 - \lambda \int \overline{G}(y) f_1'(y)\mathrm{d}y\Big) - \frac{1}{2}\lambda^2 \int_\Omega \mathrm{d}x \int \mathrm{d}y \nabla G(x,y) f_1'(y) \cdot \int \mathrm{d}z \nabla G(x,z) f_1'(z) \\ - \;\; & \frac{1}{2}\overline{k^2}\Big[c_0^2 - 2\lambda c_0 \int \overline{G}(y) f_1'(y)\mathrm{d}y + \lambda^2 \int_\Omega \mathrm{d}x \int \mathrm{d}y\, G(x,y) f_1'(y) \int \mathrm{d}z\, G(x,z) f_1'(z)\Big] \\ - \;\; & \frac{1}{2}[k^2] \int_\Omega \mathrm{d}x\, f_1'(x)\Big[c_0^2 - 2\lambda c_0 \int \mathrm{d}y\, G(x,y) f_1'(y) \\ & \qquad + \lambda^2 \int \mathrm{d}y\, G(x,y) f_1'(y) \int \mathrm{d}z\, G(x,z) f_1'(z)\Big], \end{aligned} \tag{3.3}$$

where

$$\overline{G}(y) = \int_\Omega G(x,y)\mathrm{d}x. \tag{3.4}$$

Ensemble averaging (3.3) gives

$$\langle V^*(K)\rangle \geq K\, c_0 - \frac{1}{2}\lambda^2 L_2 - \frac{1}{2}\overline{k^2}(c_0^2 + \lambda^2 J_2) + \frac{1}{2}[k^2](2\lambda c_0 I_2 - \lambda^2 I_3), \tag{3.5}$$

where

$$\begin{aligned} L_2 &= \int_\Omega \mathrm{d}x \int \mathrm{d}y \nabla G(x,y) \cdot \int \mathrm{d}z \nabla G(x,z) M_2(z-y), \\ I_2 &= \int_\Omega \mathrm{d}x \int \mathrm{d}y\, G(x,y) M_2(y-x), \quad J_2 = \int_\Omega \mathrm{d}x \int \mathrm{d}y\, G(x,y) \int \mathrm{d}z\, G(x,z) M_2(z-y), \\ I_3 &= \int_\Omega \mathrm{d}x \int \mathrm{d}y\, G(x,y) \int \mathrm{d}z\, G(x,z) M_3(y-x, z-x). \end{aligned} \tag{3.6}$$

Two particular bounds will now be deduced from (3.5). First, choose $\lambda = 0$ and then optimize with respect to c_0. This gives

$$\langle V^*(K)\rangle \geq \frac{1}{2}(\overline{k^2})^{-1}K^2. \tag{3.7}$$

Next, optimize (3.5) with respect to c_0 and λ:

$$\langle V^*(K)\rangle \geq \frac{1}{2}\left(\frac{L_2 + \overline{k^2}J_2 + [k^2]I_3}{\overline{k^2}(L_2 + \overline{k^2}J_2 + [k^2]I_3) - [k^2]^2 I_2^2}\right)K^2. \tag{3.8}$$

The bound (3.8) has to be at least as good as (3.7); it follows, therefore, that

$$L_2 + \overline{k^2}J_2 + [k^2]I_3 - [k^2]^2 I_2^2/\overline{k^2} \geq 0 \tag{3.9}$$

for every choice of k_1 and k_2, and for every kernel function G.

Now substitute into the right side of (2.4) the trial field

$$q = -\lambda\int \nabla G(x,y)f_1'(y)\mathrm{d}y, \quad s = s_0 - \lambda\int \Delta G(x,y)f_1'(y)\mathrm{d}y, \tag{3.10}$$

where s_0 and λ are constants. This gives, upon ensemble averaging,

$$\langle W(\overline{c})\rangle \geq \overline{c}\,s_0 - \frac{1}{2}\lambda^2 L_2 - \frac{1}{2}\overline{k^{-2}}(s_0^2 + \lambda^2 K_2) + \frac{1}{2}[k^{-2}](2\lambda s_0 N_2 - \lambda^2 N_3), \tag{3.11}$$

where

$$\overline{k^{-2}} = \eta_1 k_1^{-2} + (1-\eta_1)k_2^{-2}, \quad [k^{-2}] = k_1^{-2} - k_2^{-2} \tag{3.12}$$

and

$$\begin{aligned}
N_2 &= \int_\Omega \mathrm{d}x \int \mathrm{d}y \Delta G(x,y) M_2(y-x),\\
K_2 &= \int \mathrm{d}x \int \mathrm{d}y \Delta G(x,y) \int \mathrm{d}z \Delta G(x,z) M_2(z-y),\\
N_3 &= \int \mathrm{d}x \int \mathrm{d}y \Delta G(x,y) \int \mathrm{d}z \Delta G(x,z) M_3(y-x, z-x).
\end{aligned} \tag{3.13}$$

Choosing $\lambda = 0$ and then optimizing with respect to s_0 gives

$$\langle W(\overline{c})\rangle \geq \frac{1}{2}(\overline{k^{-2}})^{-1}\overline{c}^2, \tag{3.14}$$

whereas optimizing with respect to s_0 and λ gives

$$\langle W(\overline{c})\rangle \geq \frac{1}{2}\left(\frac{L_2 + \overline{k^{-2}}K_2 + [k^{-2}]N_3}{\overline{k^{-2}}(L_2 + \overline{k^{-2}}K_2 + [k^{-2}]N_3) - [k^{-2}]^2 N_2^2}\right)\overline{c}^2. \tag{3.15}$$

The observation that the bound (3.15) has to be at least as good as (3.14) leads to an inequality similar to (3.9). It has, in fact, exactly the same information content and so is not given explicitly.

Since the inequality (3.9) holds for all k_1 and k_2, it implies (and is implied by)

$$L_2 \geq 0, \quad J_2 \geq 0 \tag{3.16}$$

and

$$\eta_1^2 J_2 + \eta_1 I_3 \geq I_2^2, \quad \eta_2^2 J_2 - \eta_2 I_3 \geq I_2^2, \tag{3.17}$$

where $\eta_2 = 1 - \eta_1$. Relations (3.16) are both elementary: with the definition

$$\chi(x) = \int G(x,y) f_1'(y) \mathrm{d}y, \tag{3.18}$$

they state, respectively, that

$$\langle |\nabla \chi|^2 \rangle \geq 0 \quad \text{and} \quad \langle \chi^2 \rangle \geq 0. \tag{3.19}$$

4 Hashin-Shtrikman structure

The Hashin-Shtrikman variational principle corresponding to (2.1) is found by introducing a comparison material with sink strength k_0^2, with $k_0^2 \geq k^2$, and a polarization field π. It then follows from the equality

$$\frac{1}{2}(k^2 - k_0^2)^{-1}\pi^2 = \inf_c \left\{ \pi c - \frac{1}{2}(k^2 - k_0^2)c^2 \right\}, \tag{4.1}$$

that for any π and c,

$$\frac{1}{2}k^2c^2 \leq \frac{1}{2}k_0^2c^2 + \pi c - \frac{1}{2}(k^2 - k_0^2)^{-1}\pi^2. \tag{4.2}$$

Substituting (4.2) into (2.1) leads to the inequality

$$V^*(K) \geq \sup_{c \in H_1(\Omega)} \int_\Omega \left[Kc - \frac{1}{2}\left((\nabla c)^2 + k_0^2 c^2 + 2\pi c - (k^2 - k_0^2)^{-1}\pi^2 \right) \right] \mathrm{d}x, \tag{4.3}$$

for any field π.

To derive the alternative principle from (2.4), introduce a field ν and k_0^2 such that $k_0^2 \leq k^2$. Then similar manipulations starting from the equality

$$\frac{1}{2}(k^{-2} - k_0^{-2})^{-1}\nu^2 = \inf_s \left\{ \nu s - \frac{1}{2}(k^{-2} - k_0^{-2})s^2 \right\}, \tag{4.4}$$

lead to the inequality

$$W(\overline{c}) \geq \sup_{(q,s) \in T} \int_\Omega \left[\overline{c}s - \frac{1}{2}\left(q^2 + \frac{s^2}{k_0^2} + 2\nu s - (k^{-2} - k_0^{-2})^{-1}\nu^2 \right) \right] \mathrm{d}x, \tag{4.5}$$

for any field ν. The inequalities (4.3) and (4.5) are the Hashin-Shtrikman variational principles associated with (2.1) and (2.4). The best bounds are obtained by optimizing the right sides over π and ν. The derivation here follows that of Talbot and Willis (1986) in the context of a nonlinear problem. It is equivalent to the derivation in Talbot and Willis (1984b), although that paper subsequently considered spherical inclusions embedded in a matrix.

Next, starting from (4.3), further progress is made by restricting π to be piecewise constant, so that

$$\pi(x) = \pi_1 f_1(x) + \pi_2(1 - f_1(x)), \tag{4.6}$$

where π_1, π_2 are constants, and taking as a trial field

$$c(x) = c_0 - (\pi_1 - \pi_2) \int G_0(x - y) f_1'(y) \mathrm{d}y. \tag{4.7}$$

The kernel G_0 in (4.7) is the infinite-body Green function, satisfying

$$\Delta G_0(x) - k_0^2 G_0(x) + \delta(x) = 0. \tag{4.8}$$

On substituting (4.7) into (4.3) and taking the ensemble average, the inequality

$$\begin{aligned}\langle V^*(K)\rangle \;\; &\geq \;\; K c_0 - \frac{1}{2} k_0^2 c_0^2 - \overline{\pi} c_0 - \frac{1}{2}(\pi_1 - \pi_2)^2 \left[L_2^{(0)} + k_0^2 J_2^{(0)} - 2 I_2^{(0)}\right] \\ &+ \;\; \frac{1}{2}\eta_1 (k_1^2 - k_0^2)^{-1}\pi_1^2 + \frac{1}{2}\eta_2 (k_2^2 - k_0^2)^{-1}\pi_2^2\end{aligned} \tag{4.9}$$

is obtained, where $\overline{\pi} = \eta_1 \pi_1 + \eta_2 \pi_2$ and the superscript 0 indicates that L_2, J_2 and I_2 are found using G_0 satisfying (4.8). The optimum value of c_0 follows as

$$c_0 = \frac{1}{k_0^2}\left(K - \overline{\pi}\right), \tag{4.10}$$

and (4.9) becomes

$$\begin{aligned}\langle V^*(K)\rangle \;\; &\geq \;\; \frac{1}{2k_0^2}\left(K - \overline{\pi}\right)^2 - \frac{1}{2}(\pi_1 - \pi_2)^2 \left[L_2^{(0)} + k_0^2 J_2^{(0)} - 2 I_2^{(0)}\right] \\ &+ \;\; \frac{1}{2}\eta_1 (k_1^2 - k_0^2)^{-1}\pi_1^2 + \frac{1}{2}\eta_2 (k_2^2 - k_0^2)^{-1}\pi_2^2.\end{aligned} \tag{4.11}$$

Next, by using (4.8) to obtain a representation for G_0 in terms of itself, it is easy to show that $L_2^{(0)} = -k_0^2 J_2^{(0)} + I_2^{(0)}$ so that (4.11) becomes

$$\begin{aligned}\langle V^*(K)\rangle \;\; &\geq \;\; \frac{1}{2k_0^2}\left(K - \overline{\pi}\right)^2 + \frac{1}{2}(\pi_1 - \pi_2)^2 I_2^{(0)} \\ &+ \;\; \frac{1}{2}\eta_1 (k_1^2 - k_0^2)^{-1}\pi_1^2 + \frac{1}{2}\eta_2 (k_2^2 - k_0^2)^{-1}\pi_2^2.\end{aligned} \tag{4.12}$$

If now $k_1^2 \geq k_2^2$, the Hashin-Shtrikman bound follows by taking $k_0^2 = k_1^2, \pi_1 = 0$ and maximizing the right side of (4.12) over π_2. The result is

$$\langle V^*(K)\rangle \geq \frac{1}{2} K^2 \frac{\eta_2 - [k^2] I_2^{(1)}}{\eta_2 \overline{k^2} - k_1^2 [k^2] I_2^{(1)}}. \tag{4.13}$$

A lower bound for $\langle W(\overline{c})\rangle$ is found by choosing ν so that

$$\nu(x) = \nu_1 f_1(x) + \nu_2(1 - f_1(x)), \tag{4.14}$$

where ν_1 and ν_2 are constants, and taking as trial fields

$$\begin{aligned} q &= k_0^2(\nu_1-\nu_2)\int \nabla G_0(x-y) f_1'(y)\mathrm{d}y, \\ s &= s_0 + k_0^2(\nu_1-\nu_2)\int \Delta G_0(x-y) f_1'(y)\mathrm{d}y. \end{aligned} \tag{4.15}$$

Substituting into (4.5) and optimizing over s_0 leads to the bound

$$\begin{aligned} \langle W(\bar{c})\rangle &\geq \frac{1}{2}k_0^2(\bar{c}-\bar{\nu})^2 - \frac{1}{2}k_0^4(\nu_1-\nu_2)^2\left[L_2^{(0)} + \frac{1}{k_0^2}K_2^{(0)} + \frac{2}{k_0^2}N_2^{(0)}\right] \\ &+ \frac{1}{2}\eta_1(k_1^{-2}-k_0^{-2})^{-1}\nu_1^2 + \frac{1}{2}\eta_2(k_2^{-2}-k_0^{-2})^{-1}\nu_2^2. \end{aligned} \tag{4.16}$$

The integrals $L_2^{(0)}, K_2^{(0)}$ and $N_2^{(0)}$ are easily written in terms of $I_2^{(0)}$ using equation (4.8) and on setting $-k_0^2 = k_2^2$, $\nu_2 = 0$ and optimizing the right side of (4.16) over ν_1, a little algebra produces the bound

$$\langle W(\bar{c})\rangle \geq \frac{1}{2}\bar{c}^2\frac{\eta_1\overline{k^2} + k_2^2[k^2]I_2^{(2)}}{\eta_1 + [k^2]I_2^{(2)}}. \tag{4.17}$$

5 Results and discussion

As remarked in the Introduction we are unaware of a rigorous proof of the existence of an overall sink strength $\tilde{k}^2$. However it seems reasonable to *define* $\tilde{k}_V^2$ and $\tilde{k}_W^2$ by

$$\langle V(K)\rangle = \frac{1}{2}\frac{K^2}{\tilde{k}_V^2}, \quad \langle W(\bar{c})\rangle = \frac{1}{2}\tilde{k}_W^2\bar{c}^2. \tag{5.1}$$

The first inequality in (2.7) implies that

$$\tilde{k}_V^2 \leq \tilde{k}_W^2. \tag{5.2}$$

If equation (1.1) really does homogenize, then $\tilde{k}_V^2 = \tilde{k}_W^2 = \tilde{k}^2$. Only the inequality (5.2) is employed here, however. It now follows from (4.13) and (4.17) that

$$\tilde{k}_V^2 \leq HS_V^+ = \frac{\eta_2\overline{k^2} - k_1^2[k^2]I_2^{(1)}}{\eta_2 - [k^2]I_2^{(1)}} \tag{5.3}$$

and

$$\tilde{k}_W^2 \geq HS_W^- = \frac{\eta_1\overline{k^2} + k_2^2[k^2]I_2^{(2)}}{\eta_1 + [k^2]I_2^{(2)}}, \tag{5.4}$$

which define HS_V^+ and HS_W^-.

In order to see the relationship between the bound (3.8) and (5.3) and (5.4), first specialise to the case where G in (3.8) is the solution G_0 to (4.8). Then from the right hand inequality in (3.17)

$$\eta_2 I_3^{(0)} \leq \eta_2^2 J_2^{(0)} - {I_2^{(0)}}^2, \tag{5.5}$$

and on replacing I_3 by $\eta_2 J_2^{(0)} - {I_2^{(0)}}^2/\eta_2$ and choosing $k_0^2 = k_1^2$, the bound HS_V^+ is recovered. If the right side of the other inequality, in the form

$$\eta_1 I_3^{(0)} \geq {I_2^{(0)}}^2 - \eta_1^2 J_2^{(0)}, \tag{5.6}$$

is used in a similar way, the right side of (3.8) is no longer strictly a bound. However, in this case, with $k_0^2 = k_2^2$, HS_W^- is recovered. Thus, as $I_3^{(0)}$ ranges over all its allowed values, the upper bound on $\tilde{k}_V^2$ induced by (3.8) ranges from the lower bound (5.4) on $\tilde{k}_W^2$ to the upper bound (5.3) on $\tilde{k}_V^2$. If only two point information is available, the "worst case" of (3.8) must be chosen, namely HS_V^+. This Hashin-Shtrikman bound is thus obtained directly from the classical principle (2.1) coupled with simple reasoning comparing the three-point bound with the elementary bound (3.7).

With G replaced by G_0 it is possible to rewrite (3.15) in terms of $L_2^{(0)}, J_2^{(0)}$ and $I_2^{(0)}$. In this case, the best lower bound on $\tilde{k}_W^2$, obtained by using the right side of (5.5) and $k_0^2 = k_1^2$, coincides with HS_V^+ and the worst lower bound, found by using the right side of (5.6) with $k_0^2 = k_2^2$ coincides with HS_W^-. It follows that the situation here is analogous to the relationship between bounds on the conductivity of an isotropic two-phase composite obtained using the classical energy principles and the Hashin-Shtrikman bounds. It is well known, see Milton (1981), that as the value of a parameter containing information about the three point statistics of the medium is varied, an upper bound derived from the classical minimum energy principle can take all values between the lower and upper Hashin-Shtrikman bounds.

In order to evaluate the bounds (5.3) and (5.4) it is necessary to have information about the parameter I_2. First, note that when $G(x,y) = G_0(x-y)$, the solution to (4.8), the integrals over Ω in (3.6) are trivial and

$$I_2^{(0)} = \int G_0(x) M_2(x) \mathrm{d}x, \qquad J_2^{(0)} = \int \mathrm{d}x G_0(x) \int \mathrm{d}y G_0(y) M_2(x-y), \tag{5.7}$$

are obtained. It is convenient to introduce the two-point correlation

$$\rho_2 = M_2(x)/M_2(0) = M_2(x)/(\eta_1\eta_2), \tag{5.8}$$

and the parameter

$$i_2(k_0) = k_0^2 I_2^{(0)}/(\eta_1\eta_2). \tag{5.9}$$

Next, since $J_2^{(0)} \geq 0$ (see (3.16)) for any $G(x,y)$, this inequality holds in particular when $G(x,y) = \phi(x-y)$ for any function ϕ. The expression for J then takes the form shown in (5.7) for $J_2^{(0)}$, except that G_0 is replaced by ϕ. This implies the inequality $\hat{\rho}_2 \geq 0$ for the Fourier transform $\hat{\rho}_2$ of ρ_2. It now follows that

$$i_2(k_0) = k_0^2 \int \hat{G}_0(-\xi)\hat{\rho}_2(\xi)\mathrm{d}\xi = k_0^2 \int \frac{\hat{\rho}_2(\xi)}{|\xi|^2 + k_0^2}\mathrm{d}\xi \leq \int \hat{\rho}_2(\xi)\mathrm{d}\xi = \rho_2(0) = 1. \tag{5.10}$$

Hence,

$$0 \leq i_2(k_0) \leq 1. \tag{5.11}$$

It is easy to see that when $i_2 = 0$ both the bounds reproduce the simple upper bound $\overline{k^2}$ induced by (3.7) and when $i_2 = 1$ the simple lower bound $\overline{k^{-2}}$ from (3.14) is recovered. Some sample results are now presented for the simple correlation function

$$\rho_2(x) = e^{-r/a}, \tag{5.12}$$

where $r = |x|$ and $d = 3$. This is chosen for convenience only and is not meant to model any particular composite. In this case $G_0(x) = e^{-k_0 r}/(4\pi r)$ and

$$i_2(k_0) = \frac{(k_0 a)^2}{(1 + k_0 a)^2}. \tag{5.13}$$

For illustration, the bounds (5.3) and (5.4) are plotted against $i_2(k_2)$, for $\eta_1 = \eta_2 = 0.5$, in Figure 1 for $k_1^2/k_2^2 = 2$ and in Figure 2 for $k_1^2/k_2^2 = 10$. The bounds are normalized by k_2^2. The plots were obtained by choosing values of $k_2 a$ between zero (when $i_2(k_2) = 0$) and infinity (when $i_2(k_2) = 1$). It can be seen that the bounds remain reasonably close, even for $k_1^2/k_2^2 = 10$. Also, they improve significantly on the simple upper and lower bounds $\overline{k^2}$ and $\overline{k^{-2}}$, which are obtained from $i_2(k_2) = 0$ and $i_2(k_2) = 1$, respectively.

It is worth noting that the bounds (5.3) and (5.4) are valid for any statistically uniform microstructure and any dimension d. The bounds obtained by Talbot and Willis (1984b) were for a distribution of spheres with sink strength k_1^2 embedded in a matrix with sink strength k_2^2 and the polarization π in the spheres was allowed to vary. It follows that the bounds obtained here will not in general be as restrictive for that type of microstructure.

Finally, the restriction $\hat{\rho}(\xi) \geq 0$ imposes a necessary condition on realizable two point correlation functions. It implies that Λ, defined by

$$\Lambda = \hat{\rho}(0) = \int \rho_2(x)\mathrm{d}x, \tag{5.14}$$

should be positive. Now, it is common to use approximations, such as (5.12), for the two-point correlation functions of a random medium. Another that has been used in a variety of applications is the well-stirred approximation for the pair distribution function of a random array of spheres. In the context of bounding the effective sink strength of a medium containing voids produced by irradiation Talbot and Willis (1980) found that a lower bound (which mathematically had to be finite) tended to infinity at a void fraction around 0.2 when this approximation was used. Later, Willis (1980) and Talbot and Willis (1982) demonstrated that use of the well-stirred approximation predicted the growth rather than attenuation of long waves through a matrix containing spherical inclusions, at volume fractions greater than 1/8. Their method of calculation relied upon an approximation, however. Thus, although *something* was wrong, this calculation did not directly discredit the well-stirred approximation. In either problem, however, all results behaved as they should, when the Percus-Yevick pair distribution function (Percus and Yevick 1957) was used. In the wave propagation problem it was precisely the *sign* of Λ that determined whether growth or attenuation was predicted. The observation that $\Lambda \geq 0$ thus guarantees attenuation, as expected physically, for any realizable two-point statistics. The well-stirred approximation cannot be valid at volume fractions greater than 1/8 because, at such volume fractions, the value of Λ calculated from it is negative.

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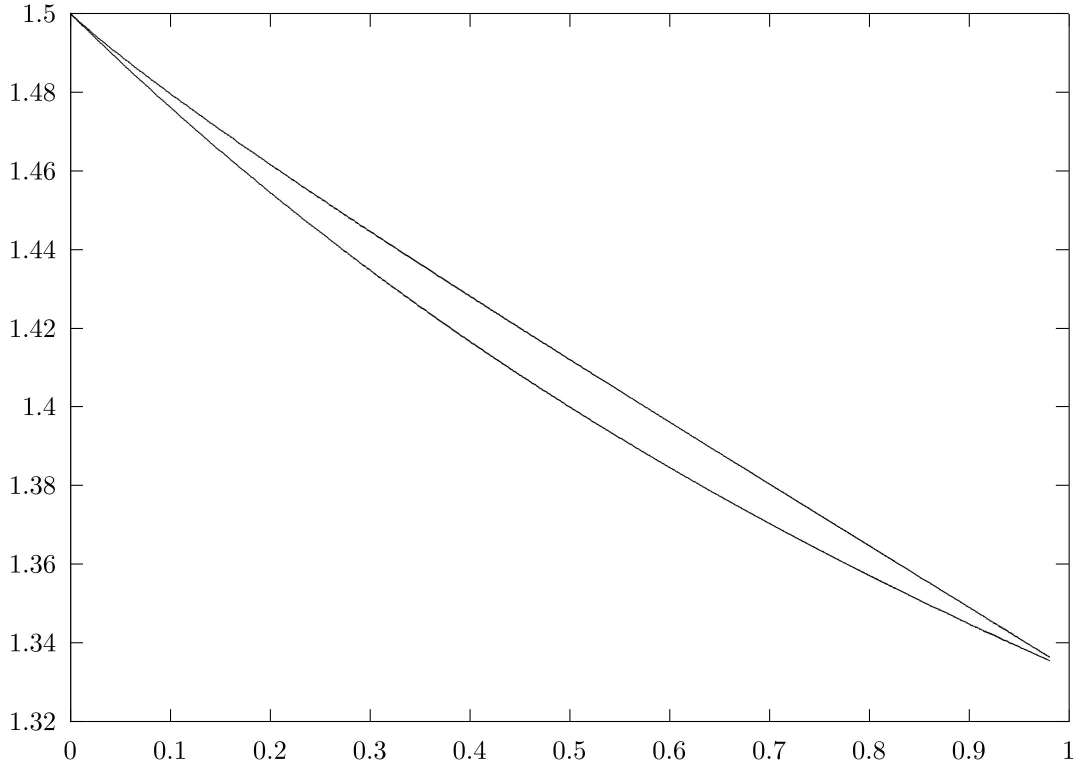
Figure Captions

Fig. 1. Plots of the bounds for the case $k_1^2/k_2^2 = 2$ when $\eta_1 = \eta_2 = 0.5$. The top line is HS_V^+ and the bottom line is HS_W^-.

Fig. 2. Plots of the bounds for the case $k_1^2/k_2^2 = 10$ when $\eta_1 = \eta_2 = 0.5$.



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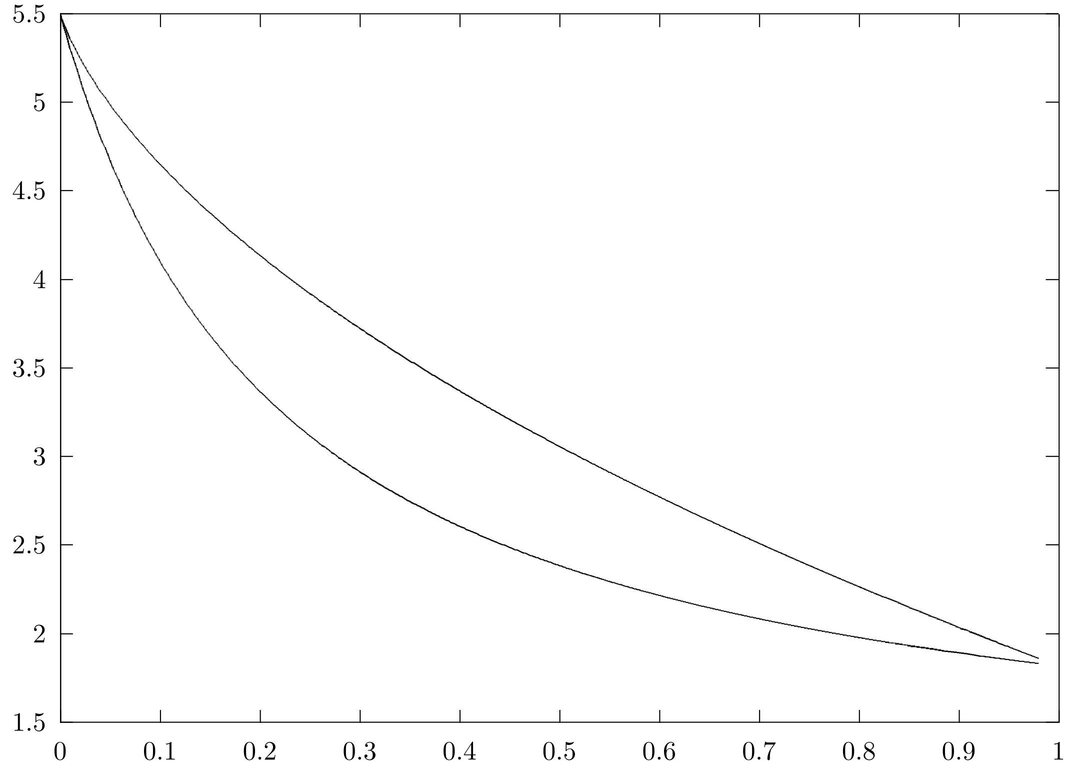
1.44
1.42
1.4
1.38
1.36
1.34
1.32
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1



5.5
5
4.5
4
3.5
3
2.5
2
1.5
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1