

Interchangeability and Bounds on the Effective Conductivity of the Square Lattice

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Received July 13, 1989; final May 11, 1990

The effective conductivity σ^* of an infinitely interchangeable two-component random medium is considered. This class of media includes cell materials in the continuum and the bond lattice on \mathbb{Z}^d , where the cells or bonds are randomly assigned the conductivities σ_1 and σ_2 ($\sigma_1, \sigma_2 \neq 0$) with probabilities p_1 and $p_2 = 1 - p_1$. A rigorous basis for the very old and widely used low volume fraction expansion of σ^* is established, by proving that σ^* is an analytic function of p_2 in a suitable domain containing $[0, 1]$. In the case of the bond lattice in $d=2$, rigorous fourth-order upper and lower bounds on σ^* valid for all p_2, σ_1 , and σ_2 are derived. The four perturbation coefficients entering into the bounds are obtained from the first-order volume fraction coefficient using the method of infinite interchangeability.

KEY WORDS: Effective conductivity; random resistor network; composites; cell materials; perturbation expansions; bounds.

1. INTRODUCTION

Consider the effective conductivity σ^* of a two-component random medium with constituent conductivities σ_1 and σ_2 in the volume fractions p_1 and $p_2 = 1 - p_1$. We have in mind either a cell material⁽¹⁾ in the continuum \mathbb{R}^d , where all space is divided into cells which are then randomly assigned the conductivities σ_1 and σ_2 with probabilities p_1 and p_2 , or the bond lattice in \mathbb{Z}^d , where the bond conductivities are randomly assigned in a similar fashion.⁽²⁾ Due to the difficulty of calculating σ^* for such systems, much effort has been devoted to developing various schemes for obtaining approximate information about σ^* . One of the principal approaches has

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been to develop series expansions for σ^* , which have been primarily of two different types. The first type is an expansion of σ^* in powers of p_1 (or p_2), the idea of which goes back to Maxwell himself.⁽³⁾ We shall refer to this type of expansion as a volume fraction expansion. The second type involves perturbing σ^* about a homogeneous medium $\sigma_1 = \sigma_2$,^(4,5) i.e., expansion in powers of $(1 - \sigma_2/\sigma_1)$, which we shall refer to as a perturbation expansion. The coefficients in this expansion can be expressed in terms of the correlation functions of the microstructure, which in practice are quite difficult to compute.

While truncating the above expansions provides good approximate formulas for σ^* when the expansion parameter is small, this procedure typically does not provide accurate information in more delicate regimes, such as near percolation ($\sigma_2 \ll \sigma_1$, $p_2 \approx p_c = \text{percolation threshold}$), particularly when only a few of the coefficients in the expansion are available. However, one approach which has received a great deal of attention is the derivation of bounds on σ^* which incorporate the perturbation expansion coefficients.^(1,5-8) The methods that have been developed provide a way of converting uncontrolled, truncated expansions into rigorous information about σ^* , in the form of upper and lower bounds that are valid for all σ_1 , σ_2 , and p_2 . The bounds become tighter when more perturbation coefficients are known, and converge to the actual σ^* if all are known. Of course, near percolation, the upper and lower bounds typically will be relatively far apart.

The purpose of the present work is twofold, the first of which we now describe. While the validity of the perturbation expansion is well established^(5,8) due to the analyticity of σ^* in the variable $(1 - \sigma_2/\sigma_1)$ in a suitable domain containing 0, the corresponding question for the volume fraction expansion has remained open. Recently, however, it was proved⁽⁹⁾ for the bond lattice that for any σ_1 and σ_2 in an appropriate domain with $\sigma_1, \sigma_2 \neq 0$, $\sigma^*(p)$ is analytic in an open neighborhood containing $[0, 1]$ in the complex p -plane, with, say $p = p_2$. Here this result is extended to infinitely interchangeable media,^(10,11) a class which includes both the cell materials in the continuum and the bond lattice, thus providing for the first time in the present context a rigorous basis for the widely used volume fraction expansion. The proof is based on an integral representation for σ^* ,^(5,8) which holds for general stationary random media. The key fact about infinitely interchangeable media which allows the proof to go through is that the perturbation coefficients are polynomials in p . It should be remarked that the analyticity of $\sigma^*(p)$ is presumably not true in general. For example, $\sigma^*(p)$ for a periodic array of spheres of volume fraction p embedded in a host material is believed to be analytic at $p = 0$ only in the variable $p^{1/3}$, so that $\sigma^*(p)$ has a branch cut there (see, e.g., ref. 12). In this

case, while the integral representation holds, the perturbation coefficients are not polynomials in p .

The second purpose of this work is to derive bounds on $\sigma^*(p)$ for the bond lattice in two dimensions. These bounds incorporate perturbation coefficients up to fourth order. What is novel is that we use the method of infinite interchangeability^(10,11) to compute the perturbation coefficients from a single volume fraction expansion coefficient. In particular, the first three perturbation coefficients are computed from the first-order volume fraction coefficient, and then the fourth is obtained from the first three using Keller's interchange equality,^(13,13) which holds only in $d=2$. It should be noted that this method works just as easily for any infinitely interchangeable medium in $d=2$ whose first-order volume fraction coefficient is known.

The bounds are obtained from the perturbation coefficients using the integral representation method,^(5,8,15) and in particular, by iterated fractional linear transformations,⁽¹⁶⁾ which are particularly useful for higher-order bounds. The bounds we obtain are similar to the fourth-order bounds of Milton⁽¹⁷⁾ for the continuum, except that since we are on the bond lattice, our coefficients are computed explicitly, whereas theirs are given in terms of geometrical parameters, which must subsequently be calculated for any particular class of media. With $\sigma_1 = 1$ and $\sigma_2 \leq \sigma_1$, we have plotted the bounds for various σ_2 , and find that for $0.5 < \sigma_2 \leq 1$, our bounds are extremely tight for all p and essentially provide a formula for $\sigma^*(p)$. Even for $\sigma_2 = 0.1$ the bounds are reasonably tight. In the case of $\sigma_2 = 0$ the lower bound collapses to 0, but the upper bound is nontrivial, although it does not give much information in the regime $p \approx p_c = 1/2$.

We note that Bergman and Kantor⁽¹⁸⁾ have computed the perturbation coefficients up to third order analytically and to eight-order numerically for the bond lattice in $d=3$, in an effort to obtain information about the percolation regime via the truncated perturbation series, with a limited degree of success. Part of our motivation for the work here is our belief that, as mentioned before, it is useful to convert these uncontrolled series into rigorous information about $\sigma^*(p)$, which we have done analytically in the $d=2$ case to fourth order.

We close this section by remarking that the methods employed in this paper have already been applied to other situations, such as investigating the transport properties of polycrystals.⁽¹⁹⁾ We expect that these methods should be applicable to other properties of lattices, such as elasticity, and even to some nonlinear problems.

2. FORMULATION

We formulate the effective conductivity problem for general stationary random media in the continuum,^(20,8) which is the natural setting for the integral representation. Subsequently, we shall consider the special case of infinitely interchangeable media, and discuss how the lattice case fits into the general framework.

Let (Ω, P) be a probability space, and let $\sigma(x, \omega)$ be a stationary stochastic process in $x \in \mathbb{R}^d$ and $\omega \in \Omega$. The space Ω represents the set of all realizations of the random medium, and P is a probability measure on Ω which is compatible with the stationarity, i.e., it is invariant under the translation group $\tau_y: \Omega \rightarrow \Omega$ defined by

$$\tau_y \omega(x) = \omega(x + y), \quad \forall x, y \in \mathbb{R}^d, \quad \omega \in \Omega \quad (2.1)$$

We consider two-component media, so that $\sigma(x, \omega)$ takes two values σ_1 and σ_2 , and can be written as

$$\sigma(x, \omega) = \sigma_1 \chi_1(x, \omega) + \sigma_2 \chi_2(x, \omega) \quad (2.2)$$

where the characteristic function $\chi_j(x, \omega)$ equals one for all realizations ω which have medium j at x , $j = 1, 2$, and equals zero otherwise. Let $E(x, \omega)$ and $J(x, \omega)$ be the stationary random electric and current fields satisfying

$$J(x, \omega) = \sigma(x, \omega) E(x, \omega) \quad (2.3)$$

$$\nabla \cdot J(x, \omega) = 0 \quad (2.4)$$

$$\nabla \times E(x, \omega) = 0 \quad (2.5)$$

$$\int_{\Omega} P(d\omega) E(x, \omega) = e_k \quad (2.6)$$

where e_k is a unit vector in the k th direction. In (2.4) and (2.5) the differential operators $\partial/\partial x_i$ are replaced by the infinitesimal generators D_i of the unitary group T_x acting on $L^2(\Omega, P)$ defined by

$$(T_x f)(\omega) = f(\tau_x \omega) \quad (2.7)$$

where

$$f(\tau_x \omega) = f(x, \omega) \quad (2.8)$$

for any $f \in L^2(\Omega, P)$, which is a stationary process on \mathbb{R}^d and Ω .^(20,8) By stationarity, we may focus attention at $x = 0$, and subsequently we shall drop the x notation.

The effective conductivity tensor σ_{jk}^* may now be defined as

$$\sigma_{jk}^* = \int_{\Omega} P(d\omega) \sigma(\omega) E_j^k(\omega) \tag{2.9}$$

where E_j^k is the j th component of E^k satisfying (2.3)–(2.6). We shall only be interested in isotropic random media, i.e., when $\sigma_{jk}^* = \sigma^* \delta_{jk}$ (although our methods are not restricted to such media), and we then pick out one diagonal coefficient and focus on it,

$$\sigma^* = \sigma_{kk}^* = \int_{\Omega} P(d\omega) [\sigma_1 \chi_1(\omega) + \sigma_2 \chi_2(\omega)] E_k^k(\omega) \tag{2.10}$$

Since (2.3), (2.4), and (2.10) are all linear in $\sigma(\omega)$, σ^* depends only on the ratio σ_2/σ_1 , that is, σ^* is homogeneous of degree one in the σ_i . Thus, it suffices to let

$$\sigma_1 = 1, \quad \sigma_2 = z \tag{2.11}$$

and we then define

$$m(z) = \sigma^* = \int_{\Omega} P(d\omega) [\chi_1(\omega) + z\chi_2(\omega)] E_k^k(\omega) \tag{2.12}$$

It has been proven^(5,8) that $m(z)$ is analytic off the negative real axis $(-\infty, 0]$ in the z -plane. Furthermore, from the symmetric form of the definition of σ^* ,^(5,8)

$$m(z) = \int_{\Omega} P(d\omega) (\chi_1 + z\chi_2) E^k \cdot \overline{E^k} \tag{2.13}$$

where the overbar denotes complex conjugation, m maps the upper half-plane to the upper half-plane, i.e.,

$$\text{Im } m(z) > 0 \quad \text{when } \text{Im } z > 0 \tag{2.14}$$

It is useful to introduce the new function

$$F(w) = 1 - m(z), \quad w = 1/(1 - z) \tag{2.15}$$

which is analytic off $[0, 1]$ in the w -plane. In ref. 8 it was proved that $F(w)$ has the integral representation

$$F(w) = \int_0^1 \frac{d\mu(t)}{w - t}, \quad w \notin [0, 1] \tag{2.16}$$

where μ is a positive Borel measure on $[0, 1]$. This representation can be proved either as a consequence of the Herglotz theorem in analytic function theory⁽²¹⁾ or as a consequence of the spectral theorem applied to the operator representation of $F(w)$ arising from (2.4),

$$F(w) = \int_{\Omega} P(d\omega) \chi_2(\omega) [(w + \Gamma\chi_2)^{-1} e_k] \cdot e_k \tag{2.17}$$

where $\Gamma = \nabla(-\mathcal{A})^{-1}\nabla \cdot$, with the differential operators again replaced by the generators of translations on Ω . In the Hilbert space $L^2(\Omega, P)$ with weight χ_2 in the inner product, $\Gamma\chi_2$ is a bounded self-adjoint operator with norm less than or equal to one. The formula (2.16) is the spectral representation of the resolvent $(w + \Gamma\chi_2)^{-1}$, where μ is the spectral measure of the family of projections of $\Gamma\chi_2$.

We now wish to introduce a special class of stationary random media, namely, infinitely interchangeable media.^(10,11) To describe the idea, let us consider a specific example. Let all of \mathbb{R}^3 be covered with randomly positioned, nonoverlapping spherical cells with sizes ranging to the infinitesimal. Each cell is assigned the conductivity σ_1 or σ_2 with probabilities $1 - p$ and p , respectively, so that the resulting medium has a volume fraction $1 - p$ of σ_1 , and p of σ_2 . Suppose that we consider such a material in which the volume fractions of σ_1 and σ_2 are $1/3$ and $2/3$, respectively. The part covered by σ_2 can be thought of as a material which itself is composed of two materials of conductivities z_2 and z_3 that also have been assigned at random, occupy a volume fraction of $1/3$ each (of the whole material), and happen to coincide with σ_2 , i.e., $z_2 = z_3 = \sigma_2$. For consistency we set $z_1 = \sigma_1$. We then have a mixture of three materials z_1, z_2 , and z_3 , whose effective conductivity we call $s = s(z_1, z_2, z_3)$. This new material is a composite in its own right, and its effective conductivity function s can have any value of z_1, z_2 , and z_3 as arguments. Clearly, $\sigma^*(\sigma_1, \sigma_2, p = 2/3) = s(\sigma_1, \sigma_2, \sigma_3)$. Furthermore, by the random nature of the construction, it is also clear that s is a symmetric function of its variables, i.e., the conductivity s of the mixture of z_1, z_2 , and z_3 will not change if, for example, all the cells containing z_1 are reassigned to contain z_2 , and vice versa.

In view of this example, we give the following definition.^(10,11)

Definition. A family of composites with effective conductivities $\sigma^*(\sigma_1, \sigma_2, p)$, $0 \leq p \leq 1$, is said to be infinitely interchangeable iff $\sigma^*(\sigma_1, \sigma_2, p)$ is a continuous function of its three arguments σ_1, σ_2 , and p , and for each integer n there exists a function $s_n(z_1, \dots, z_n)$ such that:

- (i) s_n is an infinitely differentiable function around $z_1 = \dots = z_n = 1$.

- (ii) s_n is homogeneous of degree 1, i.e., $s_n(\lambda z_1, \dots, \lambda z_n) = \lambda s_n(z_1, \dots, z_n)$.
- (iii) s_n is symmetric in its variables, i.e.,

$$s_n(z_1, z_2, \dots, z_n) = s_n(z_{i_1}, z_{i_2}, \dots, z_{i_n})$$

for any permutation (i_1, i_2, \dots, i_n) of the indices i, \dots, n .

- (iv) For each integer $k \leq n$ we have

$$\sigma^*(\sigma_1, \sigma_2, (n-k)/n) = s_n(\sigma_1, \dots, \sigma_1, \sigma_2, \dots, \sigma_2)$$

where on the right-hand side there are k of the σ_1 and $(n-k)$ of the σ_2 .

It should be noted that all examples of symmetric cell materials in the literature are infinitely interchangeable.^(10,11)

As in the more general stationary random case, it suffices to consider $\sigma_1 = 1$ and $\sigma_2 = z$. In this case, we define again

$$m(z, p) = \sigma^*(\sigma_1 = 1, \sigma_2 = z, p) \tag{2.18}$$

Via (iv), we have

$$m(z, (n-k)/n) = s_n(1, \dots, 1, z, \dots, z) \tag{2.19}$$

with $(n-k)$ of the z 's on the right-hand side.

We finally consider the bond lattice in \mathbb{Z}^d , which is a special case of both the above classes of media. Each bond is randomly assigned the conductivity σ_1 or σ_2 with probability $1-p$ and p , respectively. The space Ω from the stationary random setup can be identified with $\{\sigma_1, \sigma_2\}^{d\mathbb{Z}^d}$. Here the unitary translation group of (2.7) is generated by composition of the operators $T_i^+ = T_{+e_i}$ and $T_i^- = T_{-e_i}$, where e_i is a unit vector in the i th direction. We can then define the forward and backward difference operators

$$D_i^+ = T_i^+ - I \tag{2.20}$$

$$D_i^- = I - T_i^-, \quad i = 1, \dots, d \tag{2.21}$$

which act on $L^2(\Omega, P)$, where I is the identity operator. The stationary random current field is $J(\omega) = (J_1(\omega), \dots, J_d(\omega))$, where $J_i(\omega)$ is the current in the bond emanating from the origin in the positive i th direction. The electric field $E(\omega) = (E_1(\omega), \dots, E_d(\omega))$ can be defined by

$$J_i(\omega) = \sigma_i(\omega) E_i(\omega), \quad i = 1, \dots, d \tag{2.22}$$

where

$$\sigma_i(\omega) = \sigma_1 \chi_1^i(\omega) + \sigma_2 \chi_2^i(\omega), \quad i = 1, \dots, d \quad (2.23)$$

is the conductivity in the bond emanating from the origin in the positive i th direction, and χ_1^i and χ_2^i are the characteristic functions of medium 1 and medium 2, respectively, in that bond. For simplicity we shall drop the “ i ” superscript from subsequent χ_1 and χ_2 . Equations (2.4) and (2.5) then take the form

$$\sum_{i=1}^d D_i^- J_i(\omega) = 0 \quad (2.24)$$

$$D_i^+ E_j(\omega) - D_j^+ E_i(\omega) = 0 \quad (2.25)$$

while (2.6) still holds.

The effective conductivity σ^* is still defined in the same way as above, and all of the results stated, including the representation formula, still hold in the present context. It should be remarked that the operator Γ in the lattice case is replaced by

$$\Gamma = \nabla^+ (-\Delta)^{-1} \nabla^-. \quad (2.26)$$

where $\nabla^\pm = (D_1^\pm, \dots, D_d^\pm)$ and $(-\Delta)^{-1}$ is the inverse of the lattice Laplacian [see (3.9)].

Clearly the bond lattice satisfies properties (i)–(iv) of an infinitely interchangeable medium. [The continuity of $\sigma^*(\sigma_1, \sigma_2, p)$ in its three variables is only a technical aspect of the definition, but can be proven for the bond lattice in a variety of ways, and is certainly obvious physically. The proof can be reduced to showing that $\sigma^*(\sigma_1, \sigma_2, p)$ is monotonic in p for real σ_1 and σ_2 .^(10,11)] Thus, the bond lattice is infinitely interchangeable.

3. PERTURBATION AND VOLUME FRACTION EXPANSIONS

In this section we first exhibit the perturbation expansion and its coefficients for general stationary random media. Subsequently we establish the validity of the volume fraction expansion for infinitely interchangeable media by proving analyticity of $m(z, p)$ in p .

The perturbation expansion around a homogeneous medium ($\sigma_1 = \sigma_2$, $w = \infty$) is obtained as follows. For $|w| > 1$, (2.16) can be expanded to yield

$$F(w) = \frac{\mu_0}{w} + \frac{\mu_1}{w^2} + \frac{\mu_2}{w^3} + \dots \quad (3.1)$$

where the μ_j are the moments of μ ,

$$\mu_j = \int_0^1 t^j d\mu(t) \geq 0 \tag{3.2}$$

Equivalently, (3.1) can be written as a Taylor series for $m(z)$,

$$m(z) = 1 + \sum_{i=1}^{\infty} a_i (z-1)^i \tag{3.3}$$

where

$$a_i = \left. \frac{d^i m}{dz^i} \right|_{z=1} = (-1)^{i-1} \mu_{i-1} \tag{3.4}$$

By equating the $|w| > 1$ expansion of (2.17) to (3.1), we obtain

$$\mu_j = (-1)^j \int_{\Omega} P(d\omega) [\chi_2 (\Gamma \chi_2)^j e_k] \cdot e_k \tag{3.5}$$

for any $k = 1, 2$.

Clearly, for any medium,

$$\mu_0 = p_2 = p \tag{3.6}$$

the volume fraction of $\sigma_2 = 1$.

In the case of the bond lattice, (3.5) for $j = 1$ and $j = 2$ becomes

$$\mu_1 = - \int_{\Omega} P(d\omega) \chi_2 D_k^+ (-\Delta)^{-1} D_k^- \chi_2 \tag{3.7}$$

$$\mu_2 = \sum_{i=1}^d \int_{\Omega} P(d\omega) \chi_2 D_k^+ (-\Delta)^{-1} D_i^- \chi_2 D_i^+ (-\Delta)^{-1} D_k^- \chi_2 \tag{3.8}$$

where the operator $(-\Delta)^{-1}$ is expressed as discrete convolution with the lattice Green's function,

$$(-\Delta)^{-1} f(x) = \sum_{y \in \mathbb{Z}^d} g(x, y) f(y) \tag{3.9}$$

where $g(x, y)$ solves

$$\Delta g(x, y) = \begin{cases} -1 & x = y \\ 0 & x \neq y \end{cases} \tag{3.10}$$

While (3.7) is fairly easy to calculate, the calculation of (3.8) becomes somewhat involved.⁽¹⁸⁾ For $j \geq 3$, μ_j apparently cannot be calculated

explicitly, except, of course, μ_3 in $d=2$, which we calculate in the next section.

We now wish to establish the validity of the volume fraction expansion for infinitely interchangeable media,

$$m(z, p) = 1 + \sum_{i=1}^{\infty} \alpha_i(z) p^i \tag{3.11}$$

Fix $\varepsilon > 0$, and let

$$D_\varepsilon = \{z \mid |z - 1| < 1 - \varepsilon\} \tag{3.12}$$

To establish (3.11), we shall prove that for any $\varepsilon > 0$ and any $z \in D_\varepsilon$, $m(z, p)$ is analytic in p in a suitable domain in the p -plane. The idea of the proof is to produce an open neighborhood of $[0, 1]$ in the p -plane for which the $a_i = a_i(p)$ in (3.3) are sufficiently bounded so as to obtain uniform convergence of (3.3). One of the key steps in the proof is contained in the following result.

Lemma 3.1. For infinitely interchangeable media, the coefficient $a_i(p) = (-1)^{i-1} \mu_{i-1}$ in (3.4) is a polynomial in p of degree less than or equal to i .

Proof. From (2.19) it follows that

$$\frac{d^i m}{dz^i} \left(1, \frac{n-k}{n} \right) = \sum_{j=1}^i \sum_{q \in Q_j} s_n^q \tag{3.13}$$

where Q_j is the set of all strings of length i in the indices $k+1, \dots, n$ that contain exactly j different indices, and

$$s_n^q = \frac{\partial^i s_n}{\partial z_{q_1} \dots \partial z_{q_i}} (1, \dots, 1) \tag{3.14}$$

Let Q_j^0 be the set of all strings of length i that contain exactly the indices $1, \dots, j$. Since s_n is a symmetric function, we have

$$\sum_{q \in Q_j} s_n^q = \binom{n-k}{j} \sum_{q \in Q_j^0} s_n^q = (n-k) \dots (n-k-j+1) b_j^i(n) \tag{3.15}$$

where

$$b_j^i(n) = \frac{1}{j!} \sum_{q \in Q_j^0} s_n^q \tag{3.16}$$

We then have from (3.13)

$$\begin{aligned} \frac{d^i m}{dz^i} \left(1, \frac{n-k}{n} \right) &= (n-k) b_1^i(n) + (n-k)(n-k-1) b_2^i(n) \\ &+ \dots + (n-k) \dots (n-k-i+1) b_i^i(n) \end{aligned} \quad (3.17)$$

Notice that $b_j^i(n)$ does not depend on k .

Let us now take i sequences of integers $k_n^l, l=1, \dots, i$, such that $\lim_{n \rightarrow \infty} (n-k_n^l)/n = l/i$, and substitute $k = k_n^l$ into (3.17). We obtain

$$\begin{aligned} \frac{d^i m}{dz^i} \left(1, \frac{n-k_n^l}{n} \right) &= \frac{n-k_n^l}{n} [n b_1^i(n)] \\ &+ \dots + \frac{(n-k_n^l) \dots (n-k_n^l-i+1)}{n^i} [n^i b_i^i(n)] \end{aligned} \quad (3.18)$$

Equation (3.18) for $1 \leq l \leq i$ is a system of linear equations for the i quantities $n^j b_j^i(n)$ ($1 \leq j \leq i$). As n tends to infinity, the matrix of coefficients converges to a nonsingular Vandermonde matrix. So, for large n , we can solve for the unknown using Cramer's rule,

$$n^j b_j^i = \Delta_n^j / \Delta_n \quad (3.19)$$

The determinant Δ_n tends to a nonzero limit. Since the left-hand side of (3.18) also has a limit, so does the determinant Δ_n^j . (The continuity in p of the derivatives of m with respect to z follows from the continuity of m itself in both variables and Cauchy's theorem in z .) It follows that the limits

$$\lim_{n \rightarrow \infty} n^j b_j^i(n) = c_j \quad (3.20)$$

exist. Finally, taking the limit as $n \rightarrow \infty$ with $(n-k)/n \rightarrow p$ in (3.17) yields

$$\frac{d^i m}{dz^i} (1, p) = c_1 p + \dots + c_i p^i \quad (3.21)$$

We are now ready to state the following result.

Theorem 3.1. For any infinitely interchangeable medium, and for any $\varepsilon > 0$, there exists an open neighborhood V_ε in the complex p plane such that $[0, 1] \subset V_\varepsilon$ and $m(z, p)$ is analytic in $D_\varepsilon \times V_\varepsilon$, with D_ε given in (3.12).

Proof. Fix $\varepsilon > 0$. Since for $p \in [0, 1]$, $\mu_0(p) = p$ and $\mu_j(p) \geq \mu_{j+1}(p)$ for all n [via (3.2)],

$$|a_i(p)| \leq 1, \quad p \in [0, 1] \quad (3.22)$$

Now we must extend what we can of (3.22) into the complex plane. Consider $S = \{p \in \mathbb{C} \mid p \notin [0, 1]\}$. Conformally map S onto the unit disk U in the ζ plane, so that $p = \infty$ gets mapped to $\zeta = 0$, and $[0, 1]$ gets mapped to the unit circle $|\zeta| = 1$. Since $a_i(p)$ is an i th-order polynomial in p , $a_i(\zeta)$ has at worst an i th-order pole at $\zeta = 0$. Thus $\zeta^i a_i(\zeta)$ is analytic in U . Since $|a_i(\zeta)| \leq 1$ for $|\zeta| = 1$, by the maximum modulus principle,

$$|a_i(\zeta)| \leq \frac{1}{|\zeta|^i}, \quad \zeta \in U \tag{3.23}$$

Thus, for any small $\delta' > 0$, there is a small $\delta > \delta' > 0$ such that in the annulus A_δ defined by $1 \geq |\zeta| > 1 - \delta'$,

$$|a_i(\zeta)| \leq (1 + \delta)^i, \quad \zeta \in A_\delta \tag{3.24}$$

For any $z \in D_\varepsilon$, we can choose δ and δ' such that

$$|(1 + \delta)(z - 1)| < k < 1 \tag{3.25}$$

Now let V_ε be the set in the p plane that maps to A_δ . Then for $p \in V_\varepsilon$ and $z \in D_\varepsilon$, $|a_i(p)(z - 1)^i| < k^i < 1$. Then (3.3) converges uniformly in $D_\varepsilon \times V_\varepsilon$, which proves the theorem.

As a consequence of Theorem 3.1, we have the following result.

Corollary 3.1. For any infinitely interchangeable medium, and for any $\varepsilon > 0$, there exists a $\delta > 0$ such that for any $p \in B_\delta = \{|p| < \delta\}$ and $z \in D_\varepsilon = \{|z - 1| < 1 - \varepsilon\}$, the Taylor series in p , or volume fraction expansion,

$$m(z, p) = 1 + \sum_{i=1}^{\infty} \alpha_i(z) p^i \tag{3.26}$$

converges uniformly. Furthermore, for any of the above p and z , the double Taylor series in p and $(z - 1)$

$$m(z, p) = 1 + \sum_{i,j=1}^{\infty} \beta_{ij} p^i (z - 1)^j \tag{3.27}$$

converges uniformly.

The first-order coefficient α_1 in (3.26) has been calculated for the bond lattice in any dimension,^(22,23)

$$\alpha_1 = \frac{d(z - 1)}{(d - 1) + z} \tag{3.28}$$

In addition, in ref. 23, expressions for α_2 in $d = 2$ are derived and are evaluated numerically.

4. PERTURBATION COEFFICIENTS FOR THE BOND LATTICE

Here we use the property of infinite interchangeability to compute the perturbation coefficients $a_i = a_i(p)$ in (3.3) to fourth order for the bond lattice. The first three are computed in any dimension and the fourth is computed in $d=2$.

In (3.6) it was remarked that for any medium, $a_1(p) = p$. To explain our procedure, we begin by rederiving this elementary result. From (2.19) and the symmetry of s_n , we have

$$a_1\left(\frac{n-k}{n}\right) = \frac{dm}{dz}\left(1, \frac{n-k}{n}\right) = (n-k)s_n^1 \tag{4.1}$$

where

$$s_n^1 = \frac{\partial s_n}{\partial z_1}(1, \dots, 1)$$

To compute a_1 , it then suffices to compute s_n^1 . To do this, we note that

$$s_n(z, \dots, z) = z \tag{4.2}$$

Thus,

$$ns_n^1 = 1 \tag{4.3}$$

or $s_n^1 = 1/n$. Now (4.1) and (4.3) give $a_1[(n-k)/n] = (n-k)/n$ and, by continuity, $a_1(p) = p$, for all volume fractions p .

Let us now compute the second-order coefficient a_2 . Two differentiations of (2.19) yield

$$\frac{d^2m}{dz^2}\left(1, \frac{n-k}{n}\right) = (n-k)s_n^{11} + (n-k)(n-k-1)s_n^{12} \tag{4.4}$$

where

$$s_n^{11} = \frac{\partial^2 s_n}{\partial z_1^2}(1, \dots, 1) \quad \text{and} \quad s_n^{12} = \frac{\partial^2 s_n}{\partial z_1 \partial z_2}(1, \dots, 1)$$

We wish to determine s_n^{11} and s_n^{12} . To do this, we differentiate (4.2) twice to obtain

$$ns_n^{11} + n(n-1)s_n^{12} = 0 \tag{4.5}$$

which gives us one relation between the two unknowns. Now call

$$A_2(v) = \frac{d^2m}{dz^2}(1, v) \tag{4.6}$$

We clearly have

$$s_n^{11} = A_2(1/n) \quad (4.7)$$

and from (4.5) it then follows that

$$s_n^{12} = -A_2(1/n)/(n-1) \quad (4.8)$$

Substitution of (4.7) and (4.8) into (4.4) gives

$$\frac{d^2m}{dz^2} \left(1, \frac{n-k}{n} \right) = nA_2 \left(\frac{1}{n} \right) \left[\frac{n-k}{n} - \frac{(n-k)(n-k-1)}{n(n-1)} \right] \quad (4.9)$$

Finally, given any volume fraction p , we take a limit in (4.9) as $n \rightarrow \infty$, $k \rightarrow \infty$, and $(n-k)/n \rightarrow p$. Now, from (4.6) and (3.26) we clearly have

$$\lim_{n \rightarrow \infty} nA_2 \left(\frac{1}{n} \right) = \frac{d^2\alpha_1}{dz^2} (1) \quad (4.10)$$

and so the limiting form of (4.9) is

$$\frac{d^2m}{dz^2} (1, p) = \frac{d^2\alpha_1}{dz^2} (1) p(1-p) \quad (4.11)$$

For the d -dimensional bond lattice, we have from (3.28) that

$$\frac{d^2\alpha_1}{dz^2} (1) = -\frac{2}{d}$$

so that

$$a_2(p) = \frac{1}{2} \frac{d^2m}{dz^2} (1, p) = \frac{-p(1-p)}{d} \quad (4.12)$$

The above procedure can be generalized^(10,11) to allow for the computation of any perturbation coefficient from appropriate information about the dilute limit. Specifically, knowledge of the first r coefficients $\alpha_1, \dots, \alpha_r$ of the volume fraction expansion yields the $2r+1$ perturbation coefficients a_1, \dots, a_{2r+1} . To do this, it is necessary to use as many "homogeneity relations" like (4.5) as possible. If derivatives of order i are being considered, then $[(i+1)/2]$ independent homogeneity relations can be obtained. This is illustrated in the following computation of the coefficient of a_3 .

From (2.19), we have

$$\begin{aligned} \frac{d^3m}{dz^3} \left(1, \frac{n-k}{n} \right) &= (n-k)s_n^{111} + 3(n-k)(n-k-1)s_n^{112} \\ &+ (n-k)(n-k-1)(n-k-2)s_n^{123} \end{aligned} \quad (4.13)$$

We thus need to obtain the unknowns s_n^{111} , s_n^{112} , and s_n^{123} . By homogeneity, we have the equations

$$s_n(z, \dots, z) = z \quad (4.14)$$

and

$$s_n(z, \dots, z, 1) = z s_n(1, \dots, 1, 1/z) \quad (4.15)$$

where in (4.15) there are $n-1$ of the z 's on the left and $n-1$ of the 1 's on the right. Three differentiations of Eqs. (4.14) and (4.15) give the following two independent equations for the unknowns:

$$s_n^{111} + 3(n-1)s_n^{112} + (n-1)(n-2)s_n^{113} = 0 \quad (4.16)$$

$$n s_n^{111} + 3(n-1)(n-2)s_n^{112} + (n-1)(n-2)(n-3)s_n^{123} = -3s_n^{11} \quad (4.17)$$

No other independent equations can be obtained by homogeneity. To complete the system (4.16), (4.17), we use again information from the dilute limit. Let us define

$$A_3(v) = \frac{d^3m}{dz^3} \left(1, v \right) \quad (4.18)$$

It is clear that

$$s_n^{111} = \frac{d^3m}{dz^3} \left(1, \frac{1}{n} \right) = A_3 \left(\frac{1}{n} \right) \quad (4.19)$$

The system (4.16), (4.17), (4.19) can now be solved, and we obtain, using (4.7),

$$\begin{aligned} s_n^{111} &= A_3(1/n) \\ s_n^{112} &= -\frac{A_2(1/n) + A_3(1/n)}{n-1} \\ s_n^{123} &= \frac{3A_2(1/n) + 2A_3(1/n)}{(n-1)(n-2)} \end{aligned} \quad (4.20)$$

Putting these results into (4.13), we obtain

$$\begin{aligned} \frac{d^3 m}{dz^3} \left(1, \frac{n-k}{n} \right) &= \left[\frac{n-k}{n} - \frac{3(n-k)(n-k-1)}{n(n-1)} \right. \\ &+ 2 \frac{(n-k)(n-k-1)(n-k-2)}{n(n-1)(n-2)} \left. \right] n A_3 \left(\frac{1}{n} \right) \\ &+ \left[- \frac{3(n-k)(n-k-1)}{n(n-1)} + \frac{3(n-k)(n-k-1)(n-k-2)}{n(n-1)(n-2)} \right] n A_2 \left(\frac{1}{n} \right) \end{aligned} \quad (4.21)$$

Now, given a certain volume fraction p , we let $n, k \rightarrow \infty$ in such a way that $(n-k)/n \rightarrow p$. To do this, we notice that from (4.18) and (3.26) we have

$$\lim_{n \rightarrow \infty} n A_3 \left(\frac{1}{n} \right) = \frac{d^3 \alpha_1}{dz^3} (1) \quad (4.22)$$

From (4.10) and (4.22) we see that the limiting form of Eq. (4.22) is

$$\frac{d^3 m}{dz^3} (1, p) = [p - 3p^3 + 2p^3] \frac{d^3 \alpha_1}{dz^3} (1) - 3[p^2 - p^3] \frac{d^2 \alpha_1}{dz^2} (1) \quad (4.23)$$

For the d -dimensional bond lattice, we have from (3.28)

$$\frac{d^2 \alpha_1}{dz^2} (1) = \frac{-2}{d} \quad (4.24)$$

and

$$\frac{d^3 \alpha_1}{dz^3} (1) = \frac{6}{d^2} \quad (4.25)$$

Therefore, after minor manipulations, we obtain

$$a_3(p) = \frac{1}{d^2} p(1-p)[1 + (d-2)p] \quad (4.26)$$

In the two-dimensional case ($d=2$) the even-order coefficients can be easily obtained from the previous ones^(7,24) by using Keller's interchange equality,^(13,14)

$$m(z, p) m(1/z, p) = z \quad (4.27)$$

For instance, differentiating (4.27) four times yields

$$a_4 = \frac{a_3}{2}(2a_1 - 3) - \frac{a_2}{2}(a_2 - a_1 + 3) + \frac{1}{2}a_1(a_1 - 1) \tag{4.28}$$

Putting the results we have already obtained into (4.28), we obtain for the two-dimensional square lattice

$$a_4(p) = \frac{1}{8}p(1 - p)(p^2 - p - 1) \tag{4.29}$$

5. FOURTH-ORDER BOUNDS ON $m(z, p)$ FOR THE SQUARE LATTICE

In this last section we use the perturbation coefficients found in the previous section to obtain rigorous, fourth-order upper and lower bounds on $m(z, p)$ valid for all $z \leq 1$ and $p \in [0, 1]$. We shall only sketch the method here, as all of the details of this well-established procedure can be found in ref. 16. For simplicity, only real z 's are considered, although complex z 's can be handled as well by a similar procedure.

Let $G(w)$ be defined by

$$G(w) = \int_0^1 \frac{d\mu(t)}{w - t}, \quad w > 1 \tag{5.1}$$

with $\mu \in M_a$,

$$M_a = \left\{ \text{positive Borel measures on } [0, 1] \left| \int_0^1 d\mu = a \right. \right\} \tag{5.2}$$

Note that M_a is a compact, convex set whose extreme points are Dirac point measures $a\delta_{t^*}(dt)$ concentrated at any $t^* \in [0, 1]$. Further note that the assumption that $\mu \in M_a$ is equivalent to knowing the first-order term in the perturbation expansion of $G(w)$ about $w = \infty$,

$$G(w) = a/w + \dots, \quad w > 1 \tag{5.3}$$

For fixed w , the extreme values of $G(w)$ under (5.3) are obtained by evaluating (5.1) with the above extremal measures. In particular, the minimum of $G(w)$ is

$$G(w) \geq a/w \tag{5.4}$$

In the present situation, $F(w)$ in (3.1) for the $d=2$ square lattice is known to fourth order,

$$F(w) = \frac{\mu_0}{w} + \frac{\mu_1}{w^2} + \frac{\mu_2}{w^3} + \frac{\mu_3}{w^4} + \dots \tag{5.5}$$

where

$$\begin{aligned} \mu_0 &= p, & \mu_1 &= \frac{p(1-p)}{2}, & \mu_2 &= \frac{p(1-p)}{4} \\ \mu_3 &= \frac{p(1-p)[1+p(1-p)]}{8} \end{aligned} \quad (5.6)$$

In order to use (5.4), we successively transform $F(w)$ three times using fractional linear transformations which preserve functions of type (5.1), to obtain a function which is known only to first order. First we let

$$F_1 = \frac{1}{\mu_0} - \frac{1}{wF} \quad (5.7)$$

which has the expansion

$$F_1 = \frac{v_0}{w} + \frac{v_1}{w^2} + \frac{v_2}{w^3} + \dots \quad (5.8)$$

$$v_0 = \frac{\mu_1}{\mu_0^2} \quad (5.9)$$

$$v_1 = \frac{\mu_2}{\mu_0^2} - \frac{\mu_1^2}{\mu_0^3} \quad (5.10)$$

$$v_2 = \frac{\mu_3}{\mu_0^2} - \frac{2\mu_1\mu_2}{\mu_0^3} + \frac{\mu_1^3}{\mu_0^4} \quad (5.11)$$

where the v_i are the moments of a positive measure on $[0, 1]$. Subsequently, we consider

$$F_2 = \frac{1}{v_0} - \frac{1}{wF_1} \quad (5.12)$$

which has the expansion

$$F_2 = \frac{\eta_0}{w} + \frac{\eta_1}{w^2} + \dots \quad (5.13)$$

$$\eta_0 = \frac{v_1}{v_0^2}, \quad \eta_1 = \frac{v_2}{v_0^2} - \frac{v_1^2}{v_0^3} \quad (5.14)$$

where the η_i are moments of a positive measure on $[0, 1]$. Finally, we consider

$$F_3 = \frac{1}{\eta_0} - \frac{1}{wF_2} = \frac{\eta_1/\eta_0^2}{w} + \dots \quad (5.15)$$

Applying (5.4) to (5.15) yields an upper bound on $m(z, p)$,

$$m(z, p) \leq 1 - \frac{1}{w/\mu_0 - [1/\nu_0 - \eta_0/(w - \eta_1/\eta_0)]^{-1}} \tag{5.16}$$

where $w = 1/(1 - z)$.

To obtain a lower bound on $m(z, p)$, we apply the same procedure to

$$E(w) = 1 - \frac{1}{m} = \frac{1 - wF}{w(1 - F)} \tag{5.17}$$

which has the expression

$$E(w) = \frac{\theta_0}{w} + \frac{\theta_1}{w^2} + \frac{\theta_2}{w^3} + \frac{\theta_3}{w^4} + \dots \tag{5.18}$$

$$\begin{aligned} \theta_1 &= 1 - \mu_0 \\ \theta_1 &= \mu_0 - \mu_0^2 - \mu_1 \\ \theta_2 &= \mu_1 - \mu_2 - 2\mu_0\mu_1 + \mu_0^2 - \mu_0^3 \\ \theta_3 &= \mu_2 - \mu_3 - (2\mu_0\mu_2 + \mu_1^2) + 2\mu_0\mu_1 - 3\mu_0^2\mu_1 \end{aligned} \tag{5.19}$$

Whereas (5.16) is of the form

$$m(z, p) \leq g(1, z, \mu_0, \mu_1, \mu_2, \mu_3) \tag{5.20}$$

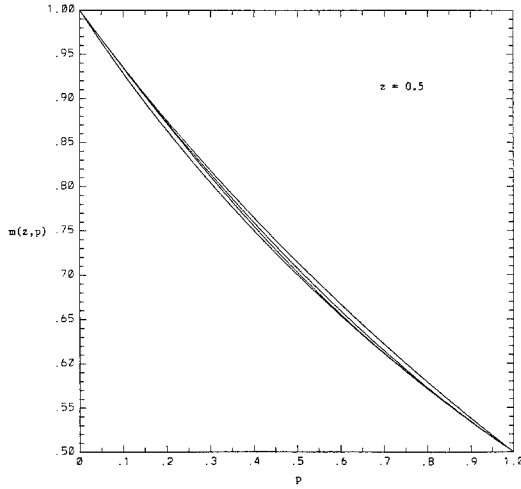
the corresponding lower bound can be written as

$$\frac{1}{m(z, p)} \leq g\left(\frac{1}{z}, 1, \theta_0, \theta_1, \theta_2, \theta_3\right) \tag{5.21}$$

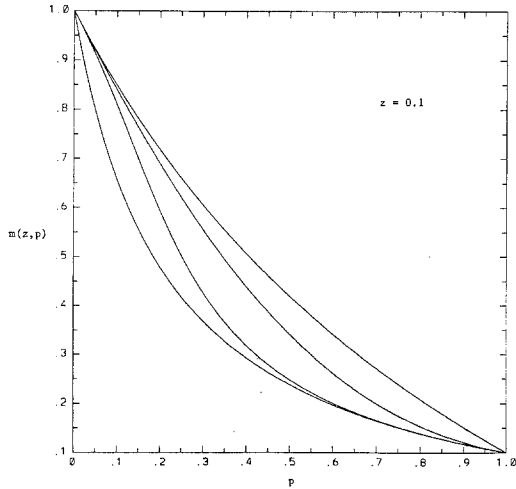
The bounds are plotted for various values of z in Fig. 1. For comparison we have included the second-order (Hashin-Shtrikman) bounds,

$$z + \frac{1 - p}{1/(1 - z) + p/2} \leq m(z, p) \leq 1 + \frac{p}{1/(z - 1) + (1 - p)/2} \tag{5.22}$$

where $0 \leq z \leq 1$ (see, e.g., ref. 8). In the case of $z = 0$ we have also plotted some numerical data of Kirkpatrick⁽²²⁾ along with his effective medium theory solution.

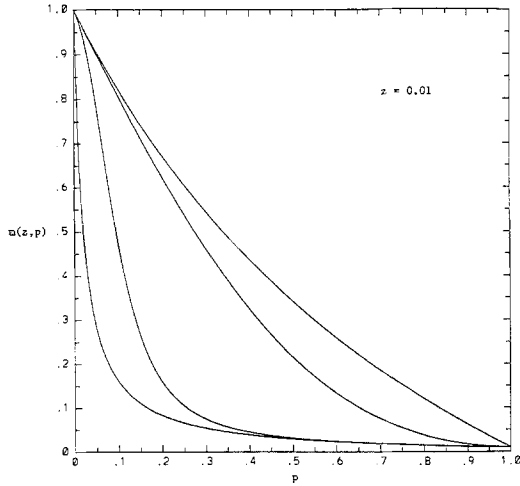


(a)

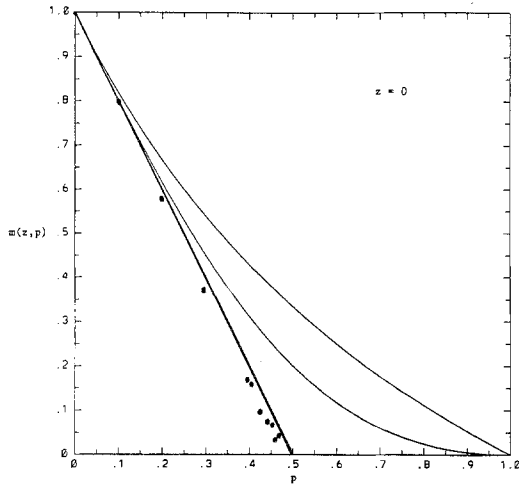


(b)

Fig. 1. Bounds on the effective conductivity $m(z, p)$ of the two-dimensional square lattice for (a) $z = 0.5$, (b) $z = 0.1$, (c) $z = 0.01$, and (d) $z = 0$. The outer set of bounds incorporate only second-order perturbation coefficients (Hashin-Shtrikman bounds). The inner set of bounds are the new fourth-order bounds. In the case $z = 0$ only the two upper bounds are included. Also in this case we have plotted the straight-line effective-medium theory solution of Kirkpatrick⁽²²⁾ along with some data from his numerical simulations.



(c)



(d)

Fig. 1. (Continued)

ACKNOWLEDGMENTS

The work of O.B. was supported in part by ARO through grant DAAL 03-88-F-0110, and the work of K.G. by AFOSR through grant AFOSR-90-0203.

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