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Spectral representation of the electrical properties of layered materials

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Abstract

We present a spectral representation for the effective conductivity of two homogeneous layers joined at a rough interface. This spectral representation is closely related to the Bergman–Milton spectral representation for bulk composites, and is easily extended to multilayered materials. By comparing the layered system to a reference layered system that has a flat interface, we form a surface spectral density that captures all the effects of surface structure on the effective conductivity of the layered sample, and is independent of the conductivities of the two layers. Because of the anisotropy of the layered system there are two surface spectral densities, one for the case where the applied field is parallel to the interface, and one for the case where the applied field is perpendicular to the interface. We discuss the relationship between these two spectral representations and present sum rules that are directly related to the degree of surface roughness. We present numerical calculations of the surface spectral density for Gaussian random surfaces which have been extensively used to study light scattering from rough surfaces.

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1. Introduction

The effective transport properties of any two component composite can be expressed in terms of the spectral representation of Bergman and Milton [1,2], which was subsequently placed on a more rigorous footing by Golden and Papanicolaou [3]. In this paper we study a composite formed by two

parallel slabs joined at a rough interface, as sketched in Fig. 1(a). For this geometry the contribution of the surface roughness to the spectral density is very small. We have developed a spectral representation, closely based on the Bergman–Milton representation, that isolates the effect of surface roughness.

The paper is divided into four sections. In Section 2 we develop the spectral representation for the surface and introduce a sum rule for the surface spectral density. In Section 3 we present numerical simulations for the surface spectral

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density of Gaussian random surfaces and discuss the results. In Section 4 we give general conclusions and discuss future work.

2. Spectral representation

The numerical simulations we will present in Section 3 are for a two-dimensional model. Thus, for convenience, we will develop the theory in two dimensions but it can be extended to three dimensions. We choose the z direction to be perpendicular to the average surface with the x -axis along the average surface, as sketched in Fig. 1(a). The surface profile is described by a function $\xi(u)$ such that $\langle \xi(u) \rangle = \int_0^L \xi(u) du/L = 0$. We consider electrically isotropic materials and assign conductivities σ_1 and σ_2 (or equivalently, resistivities ρ_1 and ρ_2) to the bottom and top layer, respectively. Because of the layered geometry the composite is anisotropic and the effective conductivity depends on the direction of the applied field. We consider two cases: when the applied field is in the x -direction (parallel to the average interface), and when the applied field is in the z -direction (perpendicular to the average interface). Using the spectral representation of Bergman and Milton we define characteristic functions for each of these two cases:

$$F(s) = 1 - \frac{\sigma_{\text{eff}}^{xx}}{\sigma_2}, \quad (1)$$

where $s = \sigma_2/(\sigma_2 - \sigma_1)$, and

$$H(t) = 1 - \frac{\rho_{\text{eff}}^{zz}}{\rho_2}, \quad (2)$$

where $t = \rho_2/(\rho_2 - \rho_1)$. As usual, these two characteristic functions can be expressed as

$$F(s) = \sum \frac{f_n}{s - s_n} \quad (3)$$

and

$$H(t) = \sum \frac{h_n}{t - t_n}, \quad (4)$$

where the poles s_n and t_n are confined to the real interval $0 \leq u < 1$, and the residues f_n and h_n are also positive real numbers. The sum over simple poles could also include an integral over a continuous distribution of poles, or spectral densities, $f(u)$ and $h(u)$. The spectral density is completely determined by the geometry of the composite and thus has the appealing property of separating the role of the geometry from material properties of the components in calculations of the effective properties of the composite.

The moments of the spectral density are determined from correlation functions of the composite. The zeroth moment, μ_0 , is particularly simple

$$\mu_0 = \int_0^1 f(u) du = \int_0^1 h(u) du = p_1, \quad (5)$$

where p_1 is the area fraction of component one.

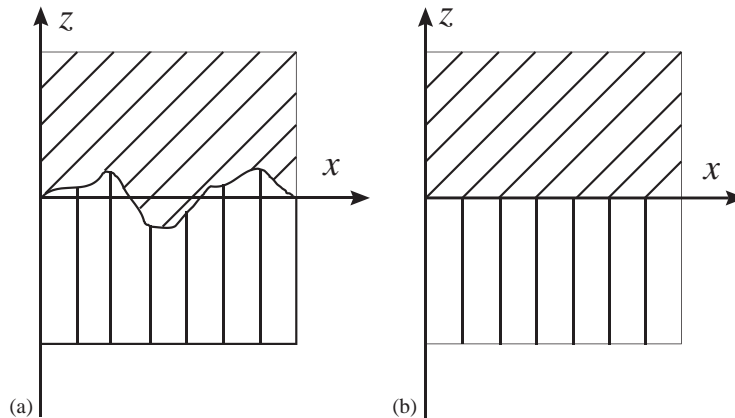


Fig. 1. A schematic representation of (a) the rough interface between two parallel slabs and (b) the reference system which has a flat interface along the x -axis. The area fractions of each component is the same for both configurations.

In general, the function $F(s)$ has a simple pole at $s = 0$ with residue f_0 equal to the effective conductivity of the composite when $\sigma_1 = 1$ and $\sigma_2 = 0$. That is, it describes the effective conductivity of the percolation problem and thus the pole is only present when component one percolates. In the slab geometry considered in this work, both materials percolate from left to right and the pole at $s = 0$ dominates $F(s)$. To display the small effect of surface roughness we compare the composite to a reference configuration formed by two slabs with a flat interface at the x -axis, as in Fig. 1(b). For the reference configuration the characteristic function is

$$F^0(s) = \frac{p_1}{s}, \quad (6)$$

where, by construction, the area fraction of component one is the same for both the rough and the reference configuration. We now define a characteristic function for the surface as

$$\begin{aligned} \bar{F}(s) &= F(s) - F^0(s) \\ &= \sum \frac{f_n}{s - s_n} - \frac{1}{s} \sum f_n \\ &= \frac{1}{s} \sum \frac{s_n f_n}{s - s_n} \\ &= \frac{1}{s} \sum \frac{g_n^\sigma}{s - s_n}, \end{aligned} \quad (7)$$

where we used the sum rule from Eq. (5) in the second step. In the last step we have introduced $g_n^\sigma = s_n f_n$. For a continuous distribution of poles we call this the surface spectral density $g^\sigma(u) = uf(u)$. The superscript σ indicates that this is the surface spectral density related to $\sigma_{\text{eff}}^{\text{xx}}/\sigma_2$.

If both σ_1 and σ_2 are real and positive then $\bar{F}(s) > 0$, indicating that the conductance of two slabs connected in parallel will always be decreased by surface roughness. The reference system has an equivalent circuit of two conductors connected in parallel. The most natural way to model the surface conductance is as an additional conductor in parallel with the two conductors of the reference system. This additional element has a negative conductance that goes to zero in the limit of a flat interface. Alternatively, we can consider an equivalent circuit of a surface conductance connected in series with the conductance of the

reference configuration. In this case the surface conductance is positive and becomes infinite in the limit of a flat surface.

When the applied field is perpendicular to the average surface we can define a different surface characteristic function in an analogous fashion. The reference configuration with a flat interface has a characteristic function

$$H^0(t) = \frac{p_1}{t} \quad (8)$$

and the surface characteristic function is

$$\begin{aligned} \bar{H}(t) &= H(t) - H^0(t) \\ &= \frac{1}{t} \sum \frac{g_n^\rho}{t - t_n}, \end{aligned} \quad (9)$$

where $g_n^\rho = t_n h_n$. For a continuous distribution of poles we have a surface spectral density $g^\rho(u) = uh(u)$. The superscript ρ indicates that this is the surface spectral density related to $\rho_{\text{eff}}^{\text{zz}}/\rho_2$.

If both ρ_1 and ρ_2 are real and positive then $\bar{H}(t) > 0$, indicating that the resistance of two slabs connected in series is always decreased by surface roughness. The reference system has an equivalent circuit of two resistors connected in series. The most natural way to model the surface resistance is as an additional resistor in series with the two resistors of the reference system. This additional element has a negative resistance that goes to zero in the limit of a flat interface. Alternatively, we can consider an equivalent circuit of a surface resistance acting as a shunt connected in parallel to the reference configuration. In this case, the surface resistance becomes infinite in the limit of a flat surface.

From the definition of $g^\sigma(u)$ the weight of the surface spectral density is just the first moment of the spectral density $f(u)$. This first moment can be calculated using the operator form of the spectral representation [4,5]. We introduce an indicator function of the position variable \mathbf{r} which takes values

$$\theta_3(\mathbf{r}) = \begin{cases} +1, & 0 < z < \xi(x), \\ -1, & \xi(x) < z < 0, \\ 0, & \text{otherwise.} \end{cases} \quad (10)$$

Following a procedure similar to that developed in Ref. [5] it is possible to derive [6] an expression for

the weight of the surface spectral function

$$\begin{aligned} \int_0^1 g^\sigma(u) du &= \lim_{s \rightarrow \infty} s^2(F(s) - F^0(s)) \\ &= \frac{1}{A} \int d\mathbf{r} \int d\mathbf{r}' \theta_3(\mathbf{r}) \theta_3(\mathbf{r}') \\ &\quad \times \frac{\partial}{\partial x} \frac{\partial}{\partial x'} G_0(\mathbf{r}, \mathbf{r}'), \end{aligned} \quad (11)$$

where $G_0(\mathbf{r}, \mathbf{r}')$ is the Green's function of Laplace's equation for the homogeneous system, with the appropriate boundary conditions, and A is the area of the two-dimensional system. It can also be shown [6] that both surface spectral densities have the same weight, which we label μ_1 :

$$\int_0^1 g^\sigma(u) du = \int_0^1 g^\rho(u) du = \mu_1. \quad (12)$$

Clearly this weight is proportional to $p_3 = \int d\mathbf{r} |\theta_3(\mathbf{r})|/A$, the fraction of material in the surface region.

3. Numerical simulations

3.1. Method

We calculated the surface spectral densities, $g^\sigma(u)$ and $g^\rho(u)$ for an ensemble of two-dimensional random resistor networks following the method of Day and Thorpe [7]. For the surface profile we used Gaussian random functions, $\{\xi(x)\}$, which have been extensively used to study light scattering from rough surfaces [8,9]. They are characterized by two parameters, an amplitude δ and a correlation length a . These functions satisfy

$$\langle \xi(x) \rangle = 0, \quad (13)$$

$$\langle \xi(x) \xi(x') \rangle = \delta^2 \exp(-|x - x'|^2/a^2) \quad (14)$$

and all higher moments can be expressed as cumulants of the first two. Clearly, δ/a is a measure of the surface roughness and δ/L_z is proportional to p_3 , the area fraction of the interface region. This surface profile is symmetric under the interchange of σ_1 and σ_2 so we expect the surface spectral density to be symmetric about $u = \frac{1}{2}$. From Eqs. (11) and (14) we expect that μ_1 ,

the weight of the surface spectral density, scales as (δ/L_z) multiplied by some function of (δ/a) .

For the Gaussian random surface it is possible to develop a perturbation theory [6] for the effective conductivity (or resistivity). We expand the solution of the Laplace equation boundary value problem for the electric potential in terms of the surface profile function of the randomly rough interface. Solutions for the electric potentials above and below the random interface are matched at the interface so as to satisfy the interface boundary conditions to second order in the surface profile function. The average current is obtained from the gradient of the electric potential and an averaging over the surface profile functions. The perturbation theory is valid when the surface is smooth and the surface region is a small fraction of the total system. To leading order in δ/a and δ/L_z we find that both spectral densities have a single pole with the same residue at s (or t) = $\frac{1}{2}$

$$\bar{F}(s) = \frac{1}{\sqrt{\pi}} \frac{1}{s(s - 1/2)} \frac{\delta}{L_z} \frac{\delta}{a} \quad (15)$$

and

$$\bar{H}(t) = \frac{1}{\sqrt{\pi}} \frac{1}{t(t - 1/2)} \frac{\delta}{L_z} \frac{\delta}{a} \quad (16)$$

From this we obtain $\mu_1 = (1/\sqrt{\pi})(\delta/L_z)(\delta/a)$ for both spectral densities, as required by Eq. (12). This has the postulated functional dependence on (δ/L_z) and (δ/a) .

The simulations were done on square lattices with nearest-neighbor bonds. Sites are labelled (1) if $z < \xi(x)$ or (2) if $\xi(x) < z$. Bonds connecting two sites with label (i) have resistance ρ_i , where $i = 1$ or 2. Bonds crossing the interface have resistance $\rho_{12} = (\rho_1 + \rho_2)/2$. The data presented are for lattices with $L_x = L_z = 128$ and for 500 realizations of the random surface for each choice of δ and a .

The surface spectral density $g^\sigma(u)$ is obtained by calculating the effective conductance [10] between perfectly conducting bus bars at the left and right edge of the lattice, for values of ρ_1 and ρ_2 such that $s = \sigma_2/(\sigma_2 - \sigma_1) = \rho_1/(\rho_1 - \rho_2)$ scans just above the real axis between 0 and 1. For each realization we remove the pole at the origin by solving the

percolation problem directly. The surface spectral density $g^\sigma(u)$ is obtained in an analogous fashion by calculating the effective resistance between perfectly conducting bus bars at the top and bottom edges of the lattice.

3.2. Results

The results of the simulations are presented in Figs. 2–5. Fig. 2 is a plot of the surface spectral density, $g^\sigma(u)$, for a fixed roughness amplitude $\delta =$

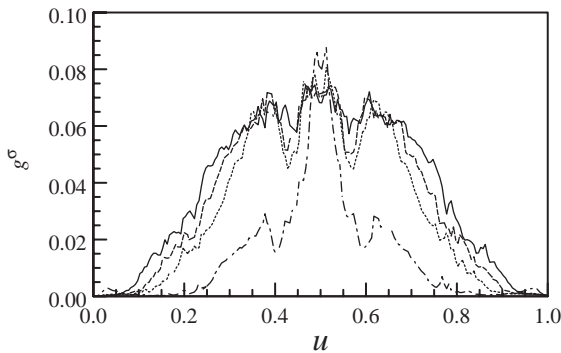


Fig. 2. The spectral density $g^\sigma(u)$ for the case where the applied field is parallel to the interface. Curves are shown for a fixed roughness amplitude $\delta = 12$, and four values of the correlation length: $a = 9$ (solid curve); $a = 12$ (broken curve); $a = 16$ (dotted curve); and $a = 48$ (dash-dot curve).

12 and four different correlation lengths, a , to give $\delta/a = 1.33, 1.00, 0.75,$ and 0.25 . The most obvious feature is the sharp peak at $u = \frac{1}{2}$ that develops as δ/a decreases, in agreement with the perturbation theory. Also note that μ_1 , the total weight of the surface spectral density, increases sublinearly with (δ/a) .

In Fig. 3 we show the normalized surface spectral density, $g^\sigma(u)/\int_0^1 g^\sigma(u) du$, for four different values of (δ/a) . In each frame there are two curves corresponding to different choices of δ and a with the same ratio δ/a . For a given value of δ/a we expect the effects of the discrete nature of the lattice to decrease with increasing δ . We see that the two curves are quite similar, indicating that we are approaching the limit where a discrete lattice is a good approximation for a continuum system. The peaks are symmetric about $u = \frac{1}{2}$, as expected. In general, for $\delta/a > 1$ (very rough surfaces) the spectral density is a broad flat function that extends over the entire interval $0 < u < 1$. As δ/a decreases (the surface becomes smoother) the spectral density narrows and a pronounced three peak structure develops. For very small values of δ/a the central peak at $u = \frac{1}{2}$ dominates, as predicted by the perturbation theory.

Fig. 4 is similar to Fig. 3 except the curves show $g^\rho(u)/\int_0^1 g^\rho(u) du$, the normalized spectral density

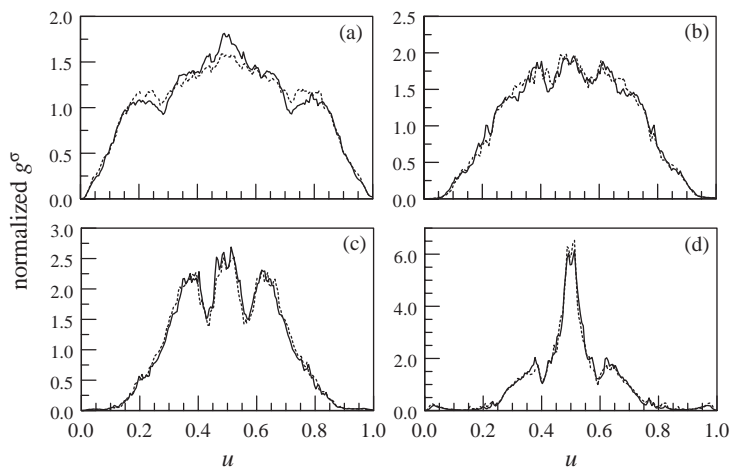


Fig. 3. The normalized surface spectral density $g^\sigma(u)/\int_0^1 g^\sigma(u) du$ for the case where the applied field is parallel to the interface for four different values of δ/a : (a) $(\delta, a) = (8, 2)$ and $(16, 4)$; (b) $(\delta, a) = (8, 6)$ and $(12, 9)$; (c) $(\delta, a) = (9, 12)$ and $(12, 16)$; and (d) $(\delta, a) = (12, 48)$ and $(16, 64)$. In each figure the solid curves are for the lower resolution data.

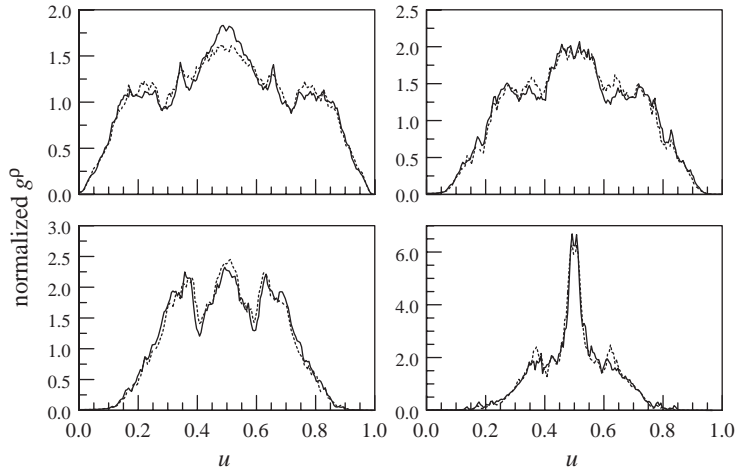


Fig. 4. As in Fig. 3 but for the case where the applied field is perpendicular to the interface.

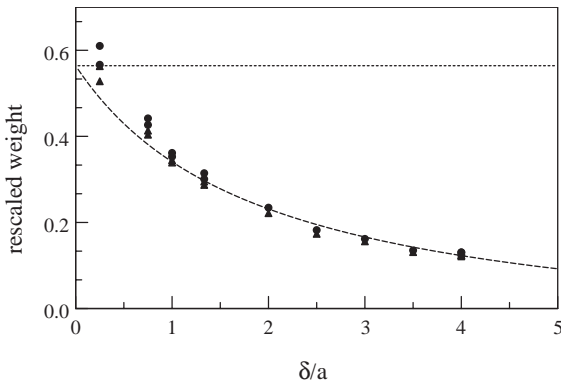


Fig. 5. The rescaled weight of the surface spectral density, $\int_0^1 g(u) du / (\delta^2 / (L_z a))$, as a function of δ/a . The solid circles are for g^σ , when the applied field is parallel to the interface. The solid triangles are for g^ρ , when the applied field is perpendicular to the interface. The dotted line is $1/\sqrt{\pi}$ and the dashed line is the Pade fit to the data shown.

when the applied field is perpendicular to the average surface. The general features of the two sets of curves are very similar and for $\delta/a = 0.25$ the two spectral densities, g^σ and g^ρ are almost identical. This should be expected because the two spectral functions are identical in the limit $\delta/a \rightarrow 0$. It is not clear why the similarity persists for larger values of δ/a .

The sum rule presented in Eq. (12) is valid for the continuum, or for lattice systems where the two spectral densities are calculated on a lattice

and its dual. In these simulations, where we use a lattice to approximate the continuum, we expect the sum rule to be approximately true. In Fig. 5 we plot $\mu_1 / (\delta^2 / (L_z a))$, the weight of the surface spectral density divided by a quantity proportional to p_3 , as a function of δ/a . For each value of δ/a there are four data points corresponding to values of $\int_0^1 g^\sigma(u) du$ and $\int_0^1 g^\rho(u) du$ and two choices of δ and a . For a continuum system all four data points should coincide. This equality is not exact for the lattice simulations but the general trend of the data is clear. A fit to the data is given by the Pade form $\mu_1 / (\delta^2 / (L_z a)) = (1 - 0.8260(\delta/a)) / (\sqrt{\pi} + 0.9179(\delta/a))$. This form is forced to give the $1/\sqrt{\pi}$ perturbation limit at $\delta/a = 0$. The mismatch near $\delta/a \approx 0$ is probably due to effects from the discrete lattice. For a given $\delta/a < 1$ the points with larger δ and a values are closest to the Pade line as they represent better approximations to the continuum model.

4. Conclusions

We have developed a spectral representation for the effective electrical properties of a layered system that successfully captures the small effect of surface roughness. We have presented numerical simulations of surface spectral density for a system of two layers with the interface described

by a Gaussian random surface and shown that this spectral density is consistent with a sum rule and a perturbation theory that are derived elsewhere. In future work we will be studying multilayered systems for which the reference configuration will have the same spectral density as the reference configuration in this work, but for which the surface spectral density will be modified by interactions between the layers.

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