



The spectral function of a composite from reflectance data

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Abstract

In the Bergman–Milton spectral representation for the effective dielectric constant of a composite all relevant geometric information is captured in a spectral function that is independent of material properties. We present numerical simulations of the reflectance of a model two component composite, where both components have temperature-dependant dielectric resonances, and show that the spectral function can be extracted from the data. The same spectral function is obtained from simulation data corresponding to different temperatures but the resolution depends on the width of the resonance line and is greatest at low-temperatures. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

The effective dielectric constant of a composite depends on both the geometry of the composite and the dielectric constants of the components. In the Bergman–Milton [1–6] spectral representation all relevant geometric information is captured in a spectral function which is independent of the material properties. For a two-component composite where the components have dielectric constants ε_A , and ε_B , the effective dielectric constant, ε_{eff} , is expressed as

$$\varepsilon_{\text{eff}}(\varepsilon_A, s_A) = \varepsilon_A \left(1 - \int_0^1 \frac{h_A(x) dx}{s_A - x} \right), \quad (1)$$

where

$$s_A = \frac{1}{1 - \varepsilon_B/\varepsilon_A}, \quad (2)$$

and $h_A(x)$ is the spectral function. The subscript A denotes we are treating A as the host material. A physical interpretation of the spectral function is the density of poles of the effective dielectric constant [7–9] which lie on the real interval $x \in [0, 1]$ in the complex s_A plane.

Given the spectral function, $h_A(x)$, for a particular geometry then Eq. (1) determines ε_{eff} for any choice of ε_A and ε_B . For example, if the frequency or temperature dependence of ε_A and ε_B are known we have ε_{eff} as a function of frequency or temperature. Alternatively, if we could extract $h_A(x)$ from one set of data for a composite, for example the dielectric function measured in one frequency range at one temperature, it could be used to predict effective properties at other frequencies or temperatures. The interest of this problem is increased when one realizes that the same spectral function determines other material properties of the composite such as the thermal conductivity or the magnetic permeability. In this paper we will discuss the type of experimental data needed to determine the spectral function, and demonstrate the method using numerical simulations on a model system.

The spectral function $h_A(x)$ will have a delta function at $x = 0$ with weight σ_B equal to the normalized

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conductivity of the percolation problem when A is insulating. We define a reduced spectral function, $g(x)$, via $h_A(x) = g(x)/x + \sigma_B \delta(x)$, which is always finite. We also define the spectral function $h_B(x) = g(1-x)/x + \sigma_A \delta(x)$ where B is considered the host [10].

2. The inverse problem

Given experimental data for $\varepsilon_A(\omega)$, $\varepsilon_B(\omega)$, and for $\varepsilon_{\text{eff}}(\omega)$ of a composite, for some frequency interval then determining the spectral function is equivalent to solving the integral equation [10,11],

$$\frac{\varepsilon_{\text{eff}}(\omega)}{\varepsilon_A(\omega)} = 1 - \int_0^1 \frac{h_A(x) dx}{s_A(\omega) - x}. \quad (3)$$

Inverse problems like this are frequently ill conditioned and there is also the problem of information loss because the transform is a smoothing process. As we have shown in previous work [10], known constraints on the moments of the spectral function are sufficient to produce a stable solution and information loss can be minimized by taking the experimental data very close to the cut $x \in [0,1]$. This is achieved by taking data at a dielectric resonance where the ratio $\varepsilon_A(\omega)/\varepsilon_B(\omega)$ is negative [10,11]. For most materials the experimentally accessible quantity at these frequencies is the reflectance, $R(\omega) = |(\sqrt{\varepsilon} - 1)/(\sqrt{\varepsilon} + 1)|^2$, so we have developed a method for extracting the spectral function directly from reflectance data by minimizing the function $\chi^2 = \chi_R^2 + \chi_{\text{constr.}}^2$ where χ_R^2 is defined as

$$\chi_R^2 = \sum_k (R_k - R(\omega_k, \varepsilon_A, \varepsilon_B, h))^2. \quad (4)$$

Here the R_k are experimental values for the reflectance taken at a set of frequencies ω_k . The function $R(\omega_k, \varepsilon_A, \varepsilon_B, h)$ depends on the known dielectric functions of the pure materials, $\varepsilon_A(\omega_k)$, and $\varepsilon_B(\omega_k)$ and the unknown spectral function, h , which is adjusted to minimize χ^2 . The function $\chi_{\text{constr.}}^2$ imposes known constraints on the first two moments of the spectral function. With the moments defined as $\mu_0^A = \int_0^1 h_A(x) dx$, $\mu_0^B = \int_0^1 h_B(x) dx$ and $\mu_1 = \int_0^1 x h_A(x) dx = \int_0^1 x h_B(x) dx$ the constraints are [7,10]

$$\mu_0^A + \mu_0^B = 1 \quad (5)$$

and

$$\mu_1 = p_w \mu_0^A \mu_0^B. \quad (6)$$

The parameter p_w is determined from the weak scattering limit when the difference between ε_A and ε_B is small. For an isotropic continuum composite in d dimensions, with any inclusion geometry, $p_w = 1/d$, and for the 2d square net with random site substitution, the model system

studied in this paper, $p_w = 1 - 1/\pi$. Note that imposing these constraints on the spectral function does not require any prior knowledge of the fill fractions of the two components; They can be determined from the fit via $p_B = \mu_0^A$ and $p_A = \mu_0^B$.

The non-linear minimization is implemented using a Levenberg–Marquardt algorithm written to ensure that h is non-negative. We start with a completely un-physical initial guess for the spectral function and iterate until χ^2 stops decreasing. The details of the method can be found in Ref. [10].

3. Numerical tests on a model system

To test this approach we need a system for which we can extract the spectral function from two independent sets of data and compare the results. In Fig. 1 we show far-infrared reflectance measurements for compacted powders of pure RbCl and pure KCl at three temperatures (5, 83 and 274 K) and note that each material exhibits a reststrahlen band which broadens as the temperature increases. Thus reflectance data of a RbCl : KCl composite taken at different temperatures could give independent calculations of the spectral function and would be a good test of the method described in Section 2.

We determine the dielectric functions of the pure materials from the reflectance data using a multiple oscillator fit and calculate $s_A(\omega)$, defined by Eq. (2). We plot the result in Fig. 2 to show that the $s_A(\omega)$ track passes near the cut $x \in [0,1]$ with the low-temperature $s_A(\omega)$ track closer to the cut than the high-temperature track because the resonance lines broaden at higher temperatures. This may result in the loss of some structure of the spectral function obtained from the high-temperature data. Because both components of the composite have

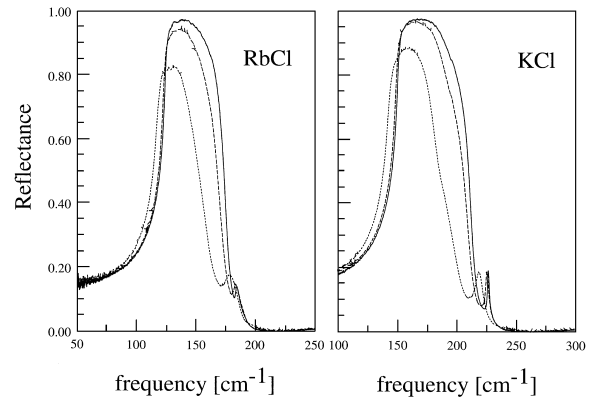


Fig. 1. The reflectance of pure RbCl and pure KCl, each at three temperatures: 5 K (solid curve), 83 K (dashed curve) and 274 K (dotted curve). The reflectance scale is the same for both figures.

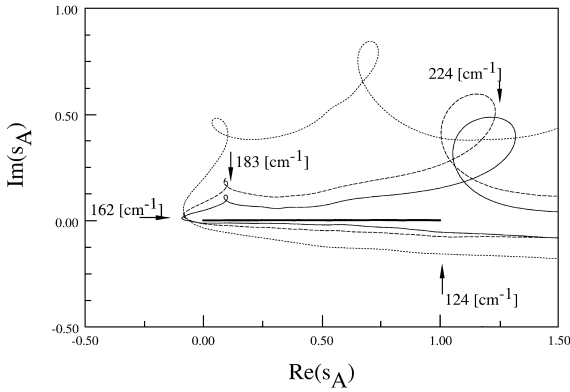


Fig. 2. The complex variable $s_A(\omega)$ at three different temperatures: 5 K (solid curve), 83 K (dashed curve) and 274 K (dotted curve).

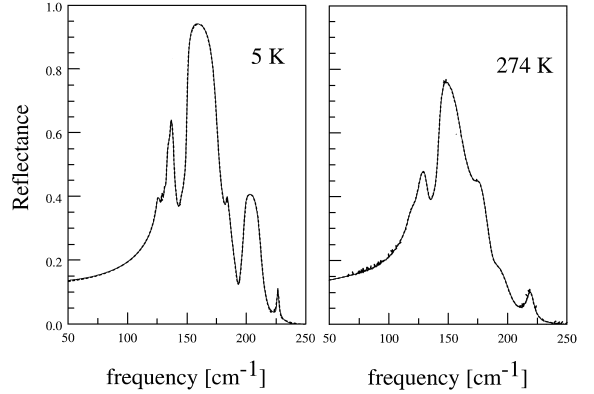


Fig. 3. The reflectance of a 2d square net with random site disorder at $p_A = 0.3$. The A component has the dielectric function of RbCl, B has that of KCl. The left panel is the reflectance at 5 K, the right at 274 K. The solid curves are the simulation data and the dotted curves are the fit to the data.

a resonance, the $s_A(\omega)$ track forms a loop, passing both above and below the cut. It should be possible to extract the spectral function from a single pass by the cut so a set of reflectance measurements for a RbCl : KCl composite covering the frequency range 50–300 cm^{-1} at three different temperatures would allow for six independent calculations of the spectral function.

Initially, we demonstrate the inverse method using data generated by numerical simulations on a 2d square network with site disorder. Sites are randomly chosen to be A with probability p_A and B with probability $1 - p_A$. We assign the dielectric function of RbCl to A sites and of KCl to B sites. The dielectric function of the composite is calculated using a $Y - \Delta$ transformation [12] where each bond has an appropriate complex impedance and we calculate the equivalent impedance of the network. From this we determine the effective dielectric function of the composite and hence the reflectance $R(\omega) = |\sqrt{\varepsilon} - 1| / (\sqrt{\varepsilon} + 1)^2$. We do this at 4000 frequencies spanning both resonances and extract the spectral function from the model data using the algorithm outlined in Section 2. There is some noise in the data and in previous work [10] we have shown that the method is stable to the introduction of additional noise.

In Fig. 3 we present the simulated reflectance of our model system with $p_A = 0.3$ at 5 and 274 K, and the reflectance obtained when fitting the spectral function. At each temperature we extract the spectral function separately from the data below 162 cm^{-1} and the data above 162 cm^{-1} . In Fig. 4 we present a representative sample of the reduced spectral functions $g(x) = xh_A(x)$ obtained from the fits, and as calculated directly [8]. For all six data sets we obtain the fill fraction of A as 0.30 to within about 1%. The weights of the delta functions vary from $\sigma_B = 0$ and $\sigma_A = 0.20$ for the low-frequency data at 5 K to $\sigma_B = 0$ and $\sigma_A = 0.08$ for the high-frequency data

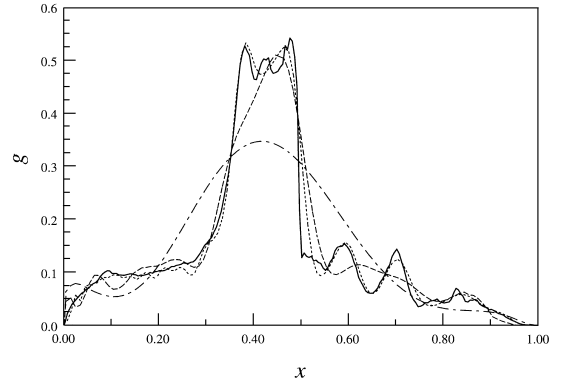


Fig. 4. The reduced spectral function for the 2d square net with random site disorder at $p_A = 0.3$. The solid curve is from a direct calculation [8], the dotted curve from the 5 K data below 162 cm^{-1} , the broken curve from the 5 K data above 162 cm^{-1} , and the dash-dot curve from the 274 K data above 162 cm^{-1} .

at 274 K. Direct simulations of the percolation problem give $\sigma_B = 0$ and $\sigma_A = 0.18$. The results clearly demonstrate that it is possible to extract the spectral function from the reflectance data with the most accurate realization of the spectral function obtained from the low-frequency data at 5 K where $s_A(\omega)$ is closest to the cut. The $s_A(\omega)$ track for the high-frequency interval at 274 K is sufficiently far from the cut that all detailed structure is lost but the central peak is still apparent. We have thus validated this method for extracting the spectral function from reflectance data and will apply it to studies of real composite materials, noting the importance of low-temperature data to obtain all the structure.

Acknowledgements

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