Complex effective permittivity of a lossy composite material

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In recent work, boundary integral equations and finite elements were used to study the (real) effective permittivity for two-component dense composite materials in the quasistatic limit. In the present work, this approach is extended to investigate in detail the role of losses. We consider the special but important case of the axisymmetric configuration consisting of infinite circular cylinders (assumed to be parallel to the z axis and of permittivity ϵ_1) organized into a crystalline arrangement (simple square lattice) within a homogeneous background medium of permittivity $\epsilon_2=1$. The intersections of the cylinders with the $x-y$ plane form a periodic two-dimensional structure. We carried out simulations taking $\epsilon_1=3-0.03i$ or $\epsilon_1=30-0.3i$ and $\epsilon_2=1$. The concentration dependence of the loss tangent of the composite material can be fitted very well, at low and intermediate concentrations of inhomogeneities, with a power law. In the case at hand, it is found that the exponent parameter depends significantly on the ratio of the real part of the permittivity of the components. We argue, moreover, that the numerical results discussed here are consistent with the Bergman and Milton theory [D. J. Bergman, Phys. Rep. **43**, 377 (1978) and G. W. Milton, J. Appl. Phys. **52**, 5286 (1981)]

I. INTRODUCTION

Major new applications for aeronautics, space, and telecommunications technology using composite materials are springing up at an ever-increasing pace. The continued innovations in materials such as fiber-reinforced resins, polymer blends, and multilayered media include the recent impressive performance levels of microwave-absorbing composite devices, i.e., photonic band structures.[1] At the same time the understanding of more fundamental problems, i.e., propagation of electromagnetic waves in periodic and random dielectric structures, is of great importance to optimize these materials for a given application and has been a long standing issue.[2–5] One of the more important, yet less frequently discussed, problems in the study of the dielectric properties of dense composite materials, composed of two or more components, is the role of losses. By comparison, a great deal more attention has been given to evaluate the (real) effective dielectric constant, in the quasistatic limit, of two-component composites made of a constituent of real permittivity ϵ_1 embedded in crystalline fashion in a homogeneous three-dimensional matrix of real permittivity ϵ_2.[2–5] Most reported calculations for effective dielectric constant considered simple geometries such as spheres and ellipsoids. Some phenomenological mixture equations have been presented because of their potentially useful applications, the fit differing from material to material.[2–8] The characterization of lossy composite materials is challenging, both experimentally and theoretically, and is of considerable interest for technological applications. To obtain reliable results for the complex permittivity, one must have theoretically and computationally demanding methods that accurately account for the details of the geometric microstructure of the material. Our own interest in this problem arose from our studies, based on the resolution of boundary integral equations, of the effective dielectric constant of composite nonlossy materials in the quasistatic limit.[2] We showed that the interplay between the geometrical shape and the arrangement in space of the inclusions gives rise to a large variety of behaviors. In this article we add to these works by investigating the behavior of losses in detail. For our simulations we use the method of finite elements (FE) which deals with the details of the geometry of the composite in a particularly efficient manner. A secondary purpose is to test whether the prediction of analytical theories can be reproduced in these simulations. More specifically, the Bergman–Milton analysis is discussed in order to establish a perspective from which to assess our simulation results and thus help to decide whether our numerical method is able to correctly describe the effective permittivity of lossy composite materials.

The remainder of the article is organized in the following way. Section II provides a brief summary of the overall theoretical framework of the problem at hand. We also introduce some of the details of our model and of our simulation. Section III presents numerical evaluations of the effective complex permittivity of a simple lossy two-component composite material using the FE scheme and compares our findings with the literature. Section IV summarizes the content of the article and gives conclusions.

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II. BACKGROUND

In this section we briefly summarize the context of the problem at hand. To establish notation and terminology, we present a number of relevant definitions and the principle of our numerical approach.

A considerable amount of reliable work has already been achieved in the area of electric properties of heterogeneous media. The reader can find good introductions to the theory in some review articles[5–8] and a list of most relevant references in Ref. 3. Dielectric mixture theory dates back the pioneering researches in the late nineteenth century by such famous physicists as Maxwell and Lorentz, the (mean-field) results of which could prove to be a useful benchmark to assist those who wish to test computer simulations under the assumption that one of the constituents had much smaller volume than the other (dilute limit). However, depending on the geometric shape of the components and the topological arrangement in the two-component material, these results have varying degrees of success at high inclusion concentration. Of course, any valid model of the effective dielectric constant of mixtures must correctly describe the properties not only at low concentrations, but at high concentrations as well. In fact it turns out that an exact solution of the problem is possible only for a few specific and geometrically well-defined systems. One can also notice that the problem of the theoretical determination of the effective dielectric constant of composite materials was also approached on the basis of rigorous variational techniques which lead to upper and lower bounds for this quantity and also by an analytic-simulation method based on multipolar expansion of the fields around each inclusion arising from the presence of the other inclusions.[9–11]

The complex effective dielectric constant, relative to free space, $\epsilon=\epsilon'-i\epsilon''$ carries information about the average polarization in the heterogeneous medium and is defined as the ratio of the average displacement field vector and the applied electric field. We may write this quantity as $E^2\epsilon=(1/\Omega)\int_\Omega \hat{\epsilon}|\nabla V|^2\, d\Omega$, where $\hat{\epsilon}$ is the local permittivity value, $E=(1/\Omega)\int_\Omega E d\Omega$ denotes the average field which depends on the applied potentials, E is the local electric field in the cell, and Ω is the volume of the medium. The absolute complex dielectric constant is obtained by multiplying ϵ by the permittivity of free space ϵ_0. Here the imaginary part ϵ'' is due to the absorption since scattering attenuation is negligible. The ratio of ϵ'' to ϵ' is referred to as the loss tangent of the material, i.e., tan δ where δ is the loss angle of the material. The complex effective permittivity will depend on the permittivity of each constituent in the mixture, their volume fractions, and eventually on the spatial arrangement in the mixture.

For simplicity we consider a composite medium made up of two nonmagnetic (we take $\mu_1=\mu_2=1$) materials with dielectric constants ϵ_1 and ϵ_2. We denote by f the concentration occupied by the inclusions of permittivity ϵ_1 periodically placed within the host material with permittivity ϵ_2. When considering the propagation of an electromagnetic wave in heterogeneous media, two length scales are of importance. The first scale is the wavelength λ of the wave probing the medium. The second one is the typical size ξ of the inhomogeneities. When the two conditions $\mathbf{k}_1\xi\ll 1$ and $\mathbf{k}_2\xi\ll 1$ are met, where we have set $\mathbf{k_i}=(\epsilon_i\mu_i)^{1/2}(2\pi/\lambda)$, $i=1,2$, the wave cannot resolve the individual scatterers and thus the material appears homogeneous to the probing wave. In this quasistatic limit, the system can be described by an effective (average) dielectric constant ϵ which is a linear homogeneous function of ϵ_1 and ϵ_2. In the numerical experiments of Sec. III the component 1 is a lossy material, i.e., $\epsilon_1=\epsilon_1'-i\epsilon_1''$, with $\epsilon_1'\gg\epsilon_1''$ and the component 2 is a pure dielectric, i.e., $\epsilon_2=\epsilon_2'$.

Bergman and Milton presented independently an analytical treatment for computing the effective dielectric constant (or conductivity) of a two-component mixture as a function of the ratio of the dielectric constants (or conductivities) of those components.[12,13] For our problem, the results of Bergman and Milton suggest that if the geometry of the composite is known, the effective permittivity ϵ of the mixture can be written as a function of a simple complex variable $\epsilon/\epsilon_1=1+\Sigma_{j=1}^{N}[A_j/(\mathbf{s}_j-s)]$, where $s=1/[1-(\epsilon_2/\epsilon_1)]$, N is the number of simple poles determined by the geometry of the material, A_j and $\mathbf{s}_j$ denoting respectively the residue and pole in the real interval $[0,1]$ of the jth term in the summation. Since the above expression of ϵ is an analytic function of $[0,1]$ in the s plane, ϵ can be implicitely determined by the positions of its zeros and poles in the complex plane. As discussed in Refs. 10–12, an important feature of the Bergman–Milton analysis is that the location of the poles is under the dependence of the geometry of the structure only and is invariant for all possible values of ϵ_1 and ϵ_2. Liu and Shen have compared the Bergman–Milton simple pole analysis and the Fourier series expansion technique to compute the effective dielectric constant of two-component two-dimensional composite materials.[14] They found that both methods give comparable results in predicting ϵ even when the dielectric constants of the components are complex (this can be seen in Table IV of Ref. 14). However, it is important to observe that most of the numerical experiments of these authors concern the case of weak contrast ratios between the dielectric constants of the background medium and the inclusions.

Now we describe the simulation procedure as follows. The calculation of the effective dielectric constant of composite materials from the known properties of the pure, homogeneous components is an electrostatics problem which involve the resolution of partial differential equations and taking into account boundary conditions defined on domains with given geometries. Here we give only some of the most important details of the model investigated and the essential features of the simulation. More details can be found in Ref. 2. We assume a system as displayed in Fig. 1, where an arbitrarily shaped homogeneous inclusion with permittivity ϵ_1 is embedded in a homogeneous matrix with permittivity ϵ_2. The solution of Laplace's equation is computed with the field calculation package FLUX2D,[15] using the FE method. Its implementation consists in dividing the domain S into triangular finite elements and for each element, the calculation is carried out by interpolation of the potential V and its normal derivative $\partial V/\partial n$ with the corresponding nodal values $V=\Sigma_j\lambda_j V_j$ and $\partial V/\partial n=\Sigma_j\lambda_j(\partial V/\partial n)_j$, where λ_j denotes

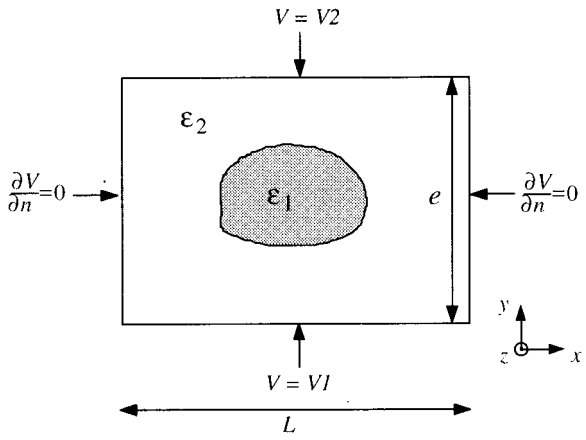


FIG. 1. Schematic drawing of the two-dimensional composite medium made of arbitrarily shaped homogeneous inclusions with permittivity ϵ_1 embedded in a homogeneous matrix with permittivity ϵ_2. On the boundary of each cell (with sides of length L and e) the potential is kept constant (V_1 or V_2) on the top and bottom faces and its normal derivative $\partial V/\partial n$ is equal to zero along the vertical walls.

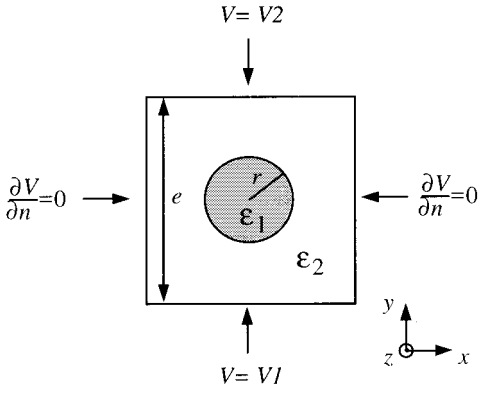


FIG. 2. Notation relating to the configuration of the two-component periodic composite material (simple square lattice) investigated in this study. The geometry is symmetrical in both the x and y directions. The unit cell consists of a square of side $e=1$ containing an infinite circular cylinder, of radius r, along the z direction and of permittivity ϵ_1. The dielectric constant of the remaining space is ϵ_2.

interpolating functions.[16] The field and potential distributions are obtained from the boundary conditions using the Galerkin method[16,17] and solving the matrix equations resulting from the discretization procedure by standard numerical techniques, e.g., the Gauss method.[18] Having computed the potential and its normal derivative on each node of the mesh, the electrostatic energy

$$\delta W_e(k)=\frac{1}{2}\int_{S_k}\varepsilon_k'\left[\left(\frac{\partial V}{\partial x}\right)^2+\left(\frac{\partial V}{\partial y}\right)^2\right]dx\,dy$$

for each triangular element is evaluated, where ϵ_k' and S_k denote the real part of the permittivity and the surface of the kth triangular element, respectively. Thus, the total energy of the entire composite can be written by summation over the n_k elements such as $W_e=\sum_{k=1}^{n_k}\delta W_e(k)$. In the problem at hand, we consider a portion of the composite material which is the filler of a parallel-plate capacitor. In this manner we obtain the real part of the complex effective permittivity from the electrostatic energy stored in such a capacitor, i.e., $W_e=\frac{1}{2}\epsilon'(S_d/e)(V2-V1)^2$ when a given potential difference is applied across the plates (see Fig. 1), $S_d=Ld$ stands for the surface of the plates with side of length L (for the two-dimensional structures considered below we take $d=1$ unit of length). Now, we must relate the dielectric losses to the imaginary part of ϵ. To do so, the dielectric losses are evaluated on each element of the mesh as

$$\delta\rho(k)=\frac{1}{2}\int_{S_k}\omega\epsilon_k'\tan\delta_k\left[\left(\frac{\partial V}{\partial x}\right)^2+\left(\frac{\partial V}{\partial y}\right)^2\right]dx\,dy,$$

where $\tan\delta_k$ is the loss tangent of the kth element and ω is the angular frequency of the electric field. The total losses of the entire composite are then obtained by summation over the n_k elements such as $\rho=\sum_{k=1}^{n_k}\delta\rho(k)$ and finally its connection with the imaginary part of the complex effective permittivity is $\rho=\frac{1}{2}\epsilon''(S_d/e)\omega(V2-V1)^2$. With these considerations in mind, we now turn to the presentation of the results of our numerical experiments.

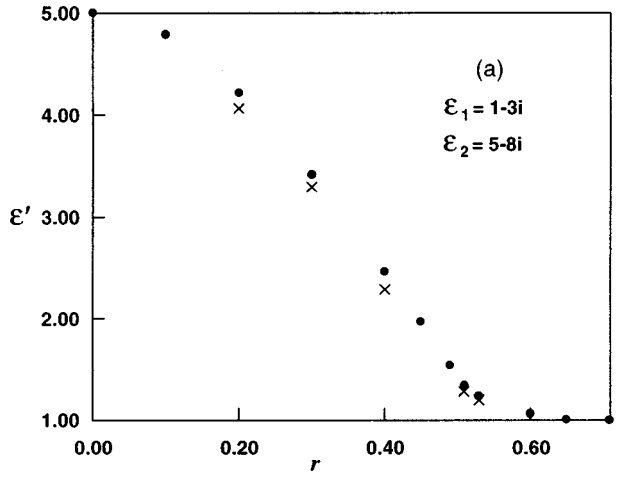
III. NUMERICAL RESULTS AND DISCUSSION

Let us consider the details of Fig. 2 with Cartesian spatial coordinates x, y, and z. This figure represents the axisymmetric unit cell of the two-component structure under study. It has been chosen for purpose of comparison with the results of Liu and Shen.[14] The structure consists of an infinite circular cylinder (ϵ_1, $\mu_1=1$) of radius r and with generator parallel to the z axis embedded in a host matrix (ϵ_2, $\mu_2=1$), i.e., the intersections of the cylinders with the $x-y$ plane form a periodic two-dimensional structure (simple square lattice of side $e=1$). This symmetrical structure in both the x and y directions renders the two-component composite material to be both translationally and rotationally invariant. We denote by f the fractional occupancy of constituent 1: $f=\pi r^2$ below the percolation threshold ($0\leqslant r\leqslant\frac{1}{2}$, nonoverlapping cylinders) and

$$f=\pi r^2-4r^2\arccos\left(\frac{1}{2r}\right)+\sqrt{4r^2-1}$$

beyond the percolation threshold ($\frac{1}{2}\leqslant r\leqslant 1/\sqrt{2}$, overlapping cylinders). The $r\geqslant\frac{1}{2}$ case is special because the symmetry of the structure provides that we can exclude in our numerical calculations the four sections of the component of permittivity ϵ_1 exceeding from the unit square cell.

To facilitate the comparison with the results of Ref. 14, we first use the following set of parameters: $\epsilon_1=1-3i$ and $\epsilon_2=5-8i$. In Fig. 3 we show a comparison of the effective permittivity computed for the two-pole approximation (Bergman–Milton theory) and the FE method versus the radius of the circular cylinder. Figures 3(a) and 3(b) are for the real part and imaginary part of ϵ, respectively. As can be recognized from these figures, in the range of r investigated, the effective complex permittivity obtained from the



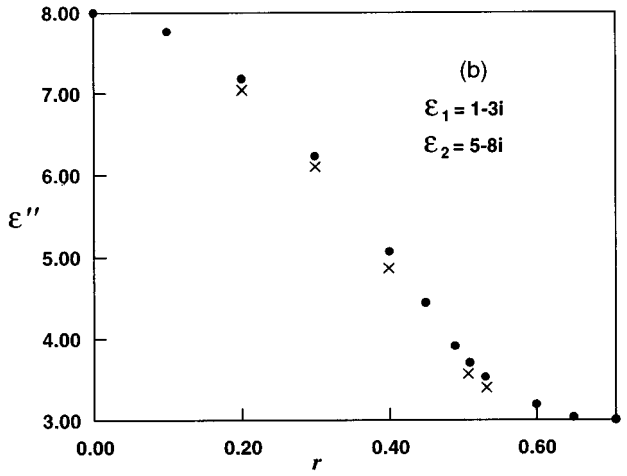
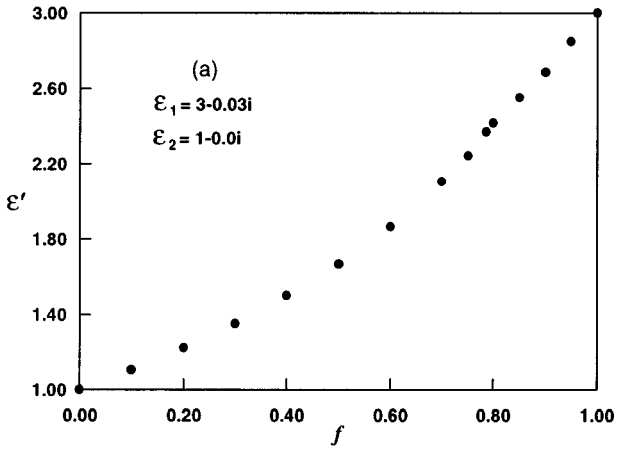


FIG. 3. (a) Comparison of the real part of the complex effective dielectric constant evaluated by the finite-element method (●) and the two-pole Bergman–Milton approximation (×, Ref. 14) as a function of the radius of the circular cylinder r. The dielectric constants of the two-component composite material are $\epsilon_1=1-3i$ and $\epsilon_2=5-8i$. (b) Comparison of the imaginary part of the complex effective dielectric constant evaluated by the finite-element method (●) and the two-pole Bergman–Milton approximation (×, Ref. 14) as a function of the radius of the circular cylinder r. The dielectric constants of the two-component composite material are $\epsilon_1=1-3i$ and $\epsilon_2=5-8i$.



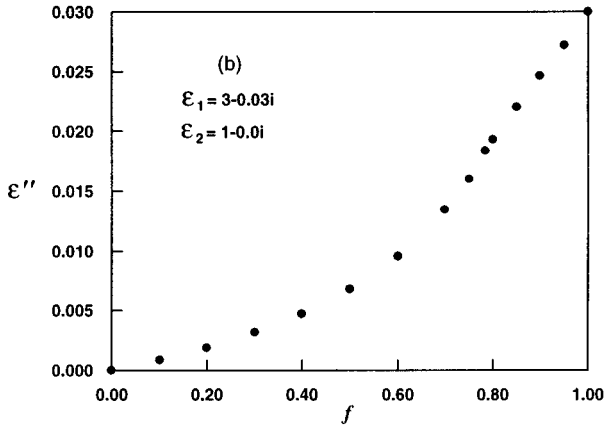
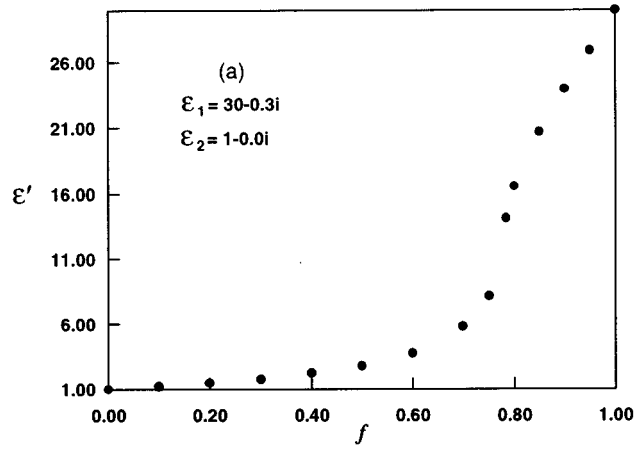


FIG. 4. (a) The real part of the effective permittivity is shown as a function of the area fraction f of the inclusion phase. Inclusions (permittivity $\epsilon_1=3-0.03i$) are placed in a host matrix material of permittivity $\epsilon_2=1$. (b) The imaginary part of the effective permittivity is shown as a function of the area fraction f of the inclusion phase. Inclusions (permittivity $\epsilon_1=3-0.03i$) are placed in a host matrix material of permittivity $\epsilon_2=1$.

Bergman–Milton theory are quantitatively very similar to the ones presented in this work. The only thing that changes is that the values of ϵ' and ϵ'' are slightly higher than those given by the two-pole approximation. These results confirm the usefulness of the FE algorithm as an efficient tool for computing the complex effective permittivity of lossy composite media. Liu and Shen's method is based on a Fourier expansion technique.[14] As discussed in Ref. 14 this computation technique is costly in computer time even in the two-dimensional case considered here. By contrast, our method is not time consuming. The CPU time for calculating the permittivity of a typical two-dimensional configuration is of the order of a few seconds. As an aside, we note that $\epsilon\cong\epsilon_1$ when r is close to 0.65, for both cases.

Having checked the FE method for the computation of the complex effective permittivity of a lossy composite medium, we now investigate what the relative importance of ϵ_1 and ϵ_2 is. Under many physically interesting conditions (e.g., polymer carbon-black composites), the dielectric constant of the matrix material is much smaller than the dielectric constant of the inhomogeneities. In our simulations we take $\epsilon_1=3-0.03i$ or $\epsilon_1=30-0.3i$ and $\epsilon_2=1$, i.e., $\tan\delta_1=10^{-2}$ and $\tan\delta_2=0$. The differences in physical behavior can only arise from a difference in the value of the ratio ϵ_1'/ϵ_2' since the physical structure and losses of both components are invariant in this study. In Figs. 4 and 5 we present the results for the real and imaginary parts of ϵ versus the concentration f for the two values of ϵ_1 investigated. Naturally, for the area fraction value $f=1$, the complex effective permittivity ϵ equals the inclusion permittivity ϵ_1. From these figures it is of interest to observe that for $\epsilon_1=30-0.3i$ the real and imaginary parts of the permittivity increase smoothly but display a sharp increase at a concentration close to 0.7. Note, however, that this phenomenon is absent for $\epsilon_1=3-0.03i$.



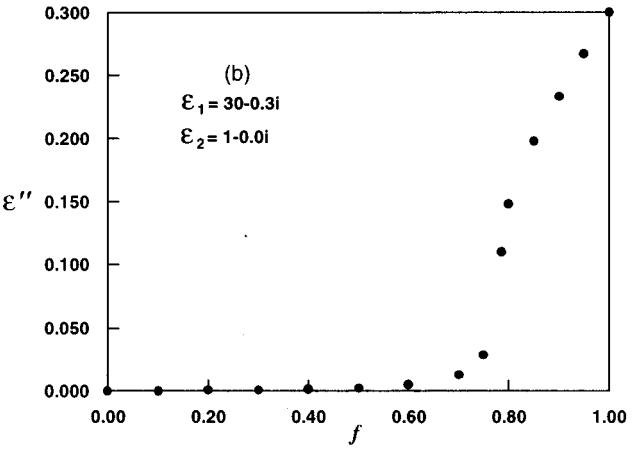


FIG. 5. (a) The real part of the effective permittivity is shown as a function of the area fraction f of the inclusion phase. Inclusions (permittivity $\epsilon_1=30-0.3i$) are placed in a host matrix material of permittivity $\epsilon_2=1$. (b) The imaginary part of the effective permittivity is shown as a function of the area fraction f of the inclusion phase. Inclusions (permittivity $\epsilon_1=30-0.3i$) are placed in a host matrix material of permittivity $\epsilon_2=1$.

An important insight can be obtained by examining the concentration dependence of the tangent of losses of the composite samples in the logarithmic plot shown in Fig. 6. Most interestingly we find that the electric loss factor increases, at low and intermediate concentrations of inhomogeneities, with increasing concentration of inhomogeneities as a power law, $\tan\delta\sim f^a$, with an exponent parameter $a\approx1.07$ for $\epsilon_1=3-0.03i$ and $a\approx1.31$ for $\epsilon_1=30-0.3i$. Thus, we find that the exponent parameter a is not constant but depends on the actual values of ϵ'_1; in fact it will depend, here, on the ratio of the real part of the permittivity of the two components. This behavior changes strongly when we go to a higher concentration of inhomogeneities. We make the additional observation that the dielectric losses are higher (by a factor of 8) for $\epsilon_1=3-0.03i$ compared to the case $\epsilon_1=30-0.3i$, while at high concentration they tend to be close for both cases. It is also seen in Fig. 6 that the two plots of $\tan\delta$ vs f intersect at a concentration close to $f^*=\pi/4$ where the cylinders touch.

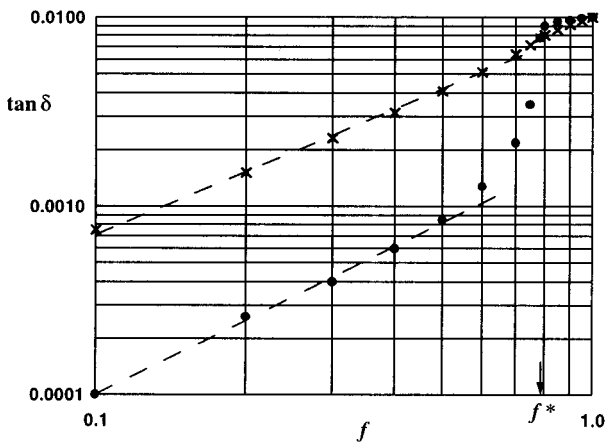


FIG. 6. Behavior of $\tan\delta=\epsilon''/\epsilon'$ as a function of the area fraction f of the inclusion phase. Inclusions (permittivity ϵ_1) are placed in a host matrix material of permittivity $\epsilon_2=1$. Crosses (solid circles) correspond to $\epsilon_1=3-0.03i$ ($\epsilon_1=30-0.3i$). The solid line is a fit according a power law $\tan\delta\sim f^a$.

IV. CONCLUSIONS

In summary, we have continued our comprehensive analysis of the dielectric properties of composite materials. In contrast to our earlier article in which we mainly concentrated on the investigation of the real effective permittivity of composite materials, we have described here a numerical scheme to compute the complex effective permittivity of a lossy composite material in the quasistatic limit. Our computational tool for studying this problem, i.e., the FE method, provides an accurate approach to evaluate this quantity. The present analysis has been carried out in a circular cylinder geometry. Similar consideration can be readily applied to other axisymmetric geometries. One goal of our investigation was to test if the Bergman–Milton theory is able to give a correct description of the effective complex dielectric constant of a lossy two-component composite medium. In this respect, we would like to point out that numerical experiments discussed here are consistent with the Bergman and Milton theory. The conclusions reached in this article, being strictly valid for the case of periodic two-dimensional configurations, are also guidelines in general context, i.e., periodic composites with deliberate introduction of imperfections. It is hoped that this work will stimulate experimental interest in these problems. For reasons of mathematical analogy, these results are also valid for the magnetic permeability of two-component composite materials. We conclude this study reminding the reader that all the simulation results presented here must be regarded as simply preliminary attempts to identify the major features of the modeling of the effective permittivity of real composite materials. In particular the present work should be extended in several directions. First, and in the light of this work and results presented in Refs. 3, 19–22 it would be of great interest to study the effect of frequency of the applied field on the effective permittivity in order to check explicitly theories aiming at elucidating the mechanism of dielectric relaxation, e.g., the scaling behaviors proposed by Jonscher.[19] An even more formidable chal-

lenge will be to investigate the random spatial arrangement of the aggregate topology. Work is in progress to meet these next challenges and full details of the above calculations will be given elsewhere.

ACKNOWLEDGMENTS

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