# Effective dielectric constant of random composite materials

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The randomness in the structure of two-component dense composite materials influences the scalar effective dielectric constant, in the quasistatic limit. A numerical analysis of this property is developed in this paper. The computer-simulation models used are based on both the finite element method and the boundary integral equation method for two- and three-dimensional structures, respectively. Owing to possible anisotropy the orientation of spatially fixed inhomogeneities of permittivity $\varepsilon_1$, embedded in a matrix of permittivity $\varepsilon_2$, affects the effective permittivity of the composite material sample. The primary goal of this paper is to analyze this orientation dependence. Second, the effect of the components geometry on the dielectric properties of the medium is studied. Third the effect of inhomogeneities randomly distributed within a matrix is investigated. Changing these three parameters provides a diverse array of behaviors useful to understand the dielectric properties of random composite materials. Finally, the data obtained from this numerical simulation are compared to the results of previous analytical work.

## I. INTRODUCTION

The concept of randomness permeates much of the current literature on the dielectric properties of heterogeneous dense materials. Understanding the transport properties of classical waves in random media remains an unrealized ambition even if the efforts of many scientists in the last decades have resulted in a considerable amount of valuable information. In fact, the scientific community is only learning how to deal with these complex systems now that ample and subtle data are provided to them. A considerable body of knowledge on the electromagnetic properties of condensed matter systems has been acquired from both experimental and theoretical studies. Previous investigations have indicated that the phenomena studied in this paper have complex ramifications. For example, in black carbon filled-polymer composites, the carbon black aggregates tend to localize in the amorphous regions of the polymer and induce interfacial aspects. Isolating the matter crucial to the physics from its irrelevant surroundings is the central task of the subject. The polarization of these materials in an external field depends on the disorder and the intrinsic dielectric characteristics of components. The challenge in the physics of disordered materials lies in relating the microscopic characteristics of the internal structure to the macroscopic property of interest, e.g., permittivity, conductivity. However, relating the parameters obtained from a constantly evolving investigation area to the geometrical details of the material is not usually straightforward; consequently, *a priori* assumptions have to be made.

A previous article from our team presented an *ab initio* treatment which, assuming periodical embedding of a constituent of permittivity $\varepsilon_1$ in a homogeneous three-dimensional matrix of permittivity $\varepsilon_2$, allows the evaluation of the scalar effective constant of the macroscopic sample in the quasistatic limit; the electromagnetic wave cannot see the individual scattering centers.[1] The effective permittivity carries information about the average polarization in the heterogeneous medium. The geometrical shape and volume fraction of each component are studied in this paper. The method to obtain numerical data by using an algorithm based upon the solution of boundary integral equation (BIE) is also given. The treatment put forward in Ref. 1 was limited to the periodic lattices of inhomogeneities. The present paper extends this treatment to three aspects of the dielectric characterization of two-component composite materials containing inclusions of permittivity $\varepsilon_1$ randomly distributed within a homogeneous matrix of permittivity $\varepsilon_2$. The three topics studied are: (a) the influence of inhomogeneities orientation, (b) the effects of component geometry, and (c) the consequences of the random distribution of inhomogeneities within the matrix. As already mentioned, the computer-simulation model, based on the BIE method developed in a previous work, was applied to three-dimensional systems. Two-dimensional configurations were also considered to make the computational requirements more reasonable. Within this approach, the finite elements (FE) method was used to obtain the potential distribution in the composite material and to derive the effective dielectric constant. In this last case, it is of the utmost importance that the present method makes it possible to determine the domain of applicability of existing theories.

The paper is organized as follows. Section II of this paper summarizes previous works dealing with the calcula-

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tion of effective permittivity of random two-component composite materials, and the technique used to compute these parameters. The numerical evaluation carried out on different types of composite media is developed in Sec. III. Moreover, the results obtained are compared to those provided by literature. Section IV draws conclusions and develops some possible extensions to our approach.

## II. FORMULATION OF THE PROBLEM

### A. Background

The main features of the analysis developed to evaluate the scalar effective permittivity of heterogeneous materials are presented in this section. A set of relevant definitions is first given to establish notation and terminology. Abundant theoretical and computational descriptions of the effective dielectric constant of two-component periodic materials do exist, but testing real composite data requires use of a physical model. The randomness and connectedness properties of its internal structure must be characterized in detail. Historically this has been a difficult task as totally different descriptions of randomness can lead to almost identical results, and consequently to serious errors in interpreting experimental results.

Predicting the effective dielectric properties of any composite material is scientifically and practically of the utmost importance, but to date there is no comprehensive and universally accepted theory to account for its whole aspects. Quoting Hashin and Shtrikman on this point is interesting (Ref. 2, page 3130): ''The indeterminacy of the effective permittivity is an inherent property of the physical situation, resulting from the fact that generally nothing is known about spatial distribution of the components except that the material is macroscopically homogeneous and isotropic.''[2] It is worthwhile to refer to the detailed overviews of the historic basis of dielectric mixture to Van Beck and Landauer. Some of the earliest works on the electric properties of composite materials were performed by Clausius and Mossotti. They derived independently a mean-field theory for a disordered system of polarizable spheres. Since this pioneering work, the subject has been the focus of an intense research effort and, presently various forms of ''effective medium theory'' exist.[3–16] There are many other theoretical approaches for calculating the effective dielectric constant of two-component composite materials. These include the virial approach, variational principles, and analytic properties of the component parameters to obtain upper and lower bounds for this parameter.[2,17–19] In recent numerical studies, Felderhof *et al.* developed a virial approach by taking into account multipole corrections, e.g., cluster expansion, to obtain formal expressions for virial coefficients.[20]

Our work deals with a composite medium composed of monodisperse inhomogeneities of permittivity $\varepsilon_1$ randomly placed within the host material. The host permittivity is $\varepsilon_2$ within a volume $\Omega$. Materials are assumed to be nonmagnetic $(\mu_1=\mu_2=1)$. Moreover, the two permittivities are considered to be real. The volume fraction occupied by the inclusions is denoted $f$. Usually, when one considers the propagation of an electromagnetic wave in a random medium, two length scales are of importance. The first scale is the wavelength $\lambda$ of the electromagnetic wave probing the medium. The second one is the typical scale $\xi$ of the inhomogeneities. When the two conditions $k_1\xi \ll 1$ and $k_2\xi \ll 1$ are met, where we have set $k_i=(2\pi/\lambda)\sqrt{\varepsilon_i\mu_i}$, $i = 1,2$, so that the wave cannot discern the individual scatters immersed in the host medium. In this quasistatic limit for which scattering losses can be neglected, the system can be described by an effective (average) dielectric constant $\varepsilon$. Mixing formulas for discrete scatterers immersed in a host medium in terms of the material properties of the components as well as the volume fraction and spatial arrangement of inclusions have been proposed. A classical effective medium analysis neglects the correlations between inclusions that become significant as their concentration increases. Formulas for two-component mixtures with homogeneous ellipsoids, needles and discs have been presented in various forms in the literature.[5,8,10–13] First, several equations for mixtures of randomly oriented ellipsoidal inclusions will be identified. In that case, there is no preferred direction in the mixture and the effective permittivity can be written according to Sihvola and Kong[12] as

$$\varepsilon=\varepsilon_2+\frac{1}{3}(\varepsilon_1-\varepsilon_2)f\sum_{i=x,y,z}\frac{\varepsilon_a}{\varepsilon_a+L_i(\varepsilon_1-\varepsilon_a)}, \tag{1}$$

where $\varepsilon_a$ is the apparent permittivity $(\varepsilon_2 \leqslant \varepsilon_a \leqslant \varepsilon)$ and $L_i$ denote the depolarization factors of the ellipsoid in the three orthogonal directions. The parameter $L_x$ in Eq. (1) is obtained from a standard result of electrostatics and can be written as

$$L_x=\frac{abc}{2}\int_0^{+\infty}\frac{du}{(u+a^2)\sqrt{(u+a^2)(u+b^2)(u+c^2)}}, \tag{2}$$

where $a$, $b$, and $c$ denote the semiaxes of the ellipsoid in the $x$, $y$, and $z$ directions, respectively. To evaluate $L_y$ and $L_z$, interchange $b$ and $a$, and $c$ and $a$, respectively. Note that the depolarization factors verify $\Sigma_{i=x,y,z}L_i=1$. If $\varepsilon_a=\varepsilon$, Eq. (1) is known as the Polder–Van Santen mixing formula. The Polder–Van Santen formula has been extensively used to analyze the dielectric behavior of snow.[12,16] If $\varepsilon_a=\varepsilon_2$ Eq. (1) is termed the Fricke formula. Another well known mixture equation for randomly oriented ellipsoids has been reported by Bohren and Battan,

$$\varepsilon=\varepsilon_2+\frac{(\varepsilon_1-\varepsilon_2)fu}{1-(1-u)f} \tag{3}$$

where we have set

$$u=\frac{1}{3}\sum_{i=x,y,z}\frac{\varepsilon_2}{\varepsilon_2+L_i(\varepsilon_1-\varepsilon_2)}. \tag{4}$$

At this point it should be noticed that the effective permittivity for a mixture of randomly oriented discs can be written as[5]

$$\varepsilon=\varepsilon_2+\frac{(\varepsilon_a+2\varepsilon_1)(\varepsilon_1-\varepsilon_2)f}{3\varepsilon_1}, \tag{5}$$

where again $\varepsilon_2 \leqslant \varepsilon_a \leqslant \varepsilon$. If $\varepsilon_a$ is set equal to $\varepsilon$, Eq. (5) reduces to the mixture equation originally reported by Bruggeman

$$\varepsilon = \varepsilon_1 \frac{3\varepsilon_2 + 2(\varepsilon_1 - \varepsilon_2) f}{3\varepsilon_1 - f(\varepsilon_1 - \varepsilon_2)}, \tag{6}$$

which has been extensively used in the literature. When $\varepsilon_a$ is set equal to $\varepsilon_2$, the Van Beek result is found again.

Finally, in the case of lamellae, the effective permittivity can be expressed in the form

$$\varepsilon = \sqrt{\frac{f\varepsilon_1 + (1-f)\varepsilon_2}{\dfrac{f}{\varepsilon_1} + \dfrac{1-f}{\varepsilon_2}}}. \tag{7}$$

### B. Principle of the numerical approach for periodic composite structures

BIE and FE are numerical techniques which allow to compute the solution of Laplace's equation by determining the electric field and potential distributions from both the physical properties of the materials and the boundary conditions in the domain studied. Recent works have shown that the BIE method could be successfully applied to compute the effective permittivity of periodic composite materials.[1,21] The basic scheme of the BIE method is now briefly recalled.

Consider a spatial domain $\Omega$ with a density of charge equals to zero everywhere. Using Green's theorem, the local potential $V(M \in \Omega)$ can be written in terms of $V(P)$ and of the normal derivative $(\partial V / \partial n)(P)$, with $P$ being any point on the boundary $\Sigma$ (with no overhangs) of $\Omega$:

$$V(M) = -\frac{4\pi}{A} \int_{\Sigma} \left( V(P) \frac{\partial G}{\partial n} - G \frac{\partial V}{\partial n}(P) \right) ds, \tag{8}$$

where $A$ stands for the solid angle under which the point $M$ sees the oriented surface $\Sigma$, $n$ is the normal unit vector oriented outward from $\Sigma$, $ds$ is a surface element of $\Sigma$ and $G$ denotes the Green function.

Referring to the schematic representation of the configuration displayed in Fig. 1, a two-component periodic composite can be considered. It can be divided into elementary cells. The constituent of permittivity $\varepsilon_1$ occupying the volume $\Omega_1$ is embedded in the region $\Omega_2$ of permittivity $\varepsilon_2$. Absence of charge density will be tacitly assumed through our analysis. Given these assumptions, Eq. (8) reads as:

$$V = -\frac{4\pi}{A} \int_{\Sigma_1} \left( V \frac{\partial G}{\partial n} - G \left. \frac{\partial V}{\partial n} \right|_1 \right) ds \tag{9}$$

for domain 1, and

$$V = -\frac{4\pi}{A} \int_{\Sigma_2} \left( V \frac{\partial G}{\partial n} - G \left. \frac{\partial V}{\partial n} \right|_2 \right) ds \tag{10}$$

for domain 2. Moreover, the following relation is obtained

$$\varepsilon_1 \left. \frac{\partial V}{\partial n} \right|_1 = \varepsilon_2 \left. \frac{\partial V}{\partial n} \right|_2 \tag{11}$$

by virtue of the conservation of the normal component of the electric displacement at the interface. Consequently, the two integral Eqs. (9) and (10) have to be solved to evaluate numerically the electrostatic potential distribution. For that purpose, the implementation of the BIE method consists in dividing the boundaries into finite elements and for each finite element, the calculation is carried out by interpolation of $V$ and $\partial V / \partial n$ with the corresponding nodal values:

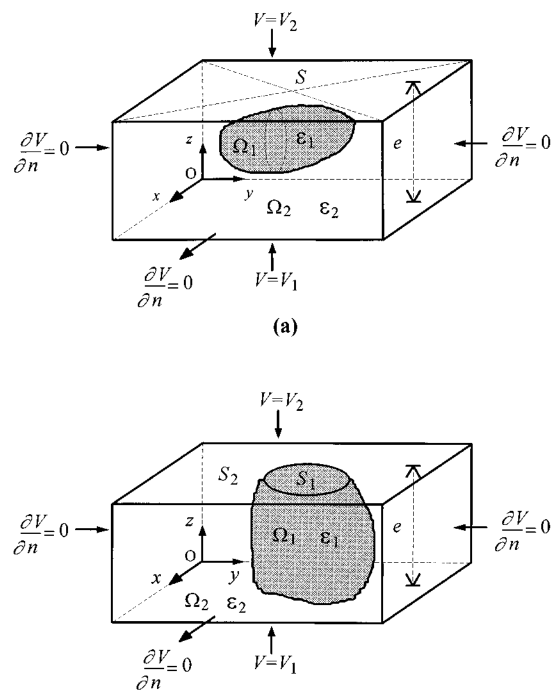


FIG. 1. Notation and boundary conditions related to a the three-dimensional composite: (a) isolated particle of permittivity $\varepsilon_1$, (b) fused particle of permittivity $\varepsilon_1$.

$$\begin{cases} V = \sum_j \lambda_j V_j \\ \dfrac{\partial V}{\partial n} = \sum_j \lambda_j \left( \dfrac{\partial V}{\partial n} \right)_j \end{cases} \tag{12}$$

where $\lambda_j$ denotes the interpolating functions. The generation of these functions, relevant to our computational requirements, and the detailed methodology used in this work are similar to those reported elsewhere.[22,23] In this way, integral equations are transformed into a matrix equation which is solved numerically using the boundary conditions on each side of the unit cell as displayed in Figs. 1(a) and 1(b). The permittivity is then obtained from the knowledge of the potential distribution and of its normal derivative.

Two types of configurations are distinguished for specification of the structure of the composite material. In Fig. 1(a), there is a single inclusion and thus the medium of permittivity $\varepsilon_1$ cannot intercept the sides of the parallelipipedic cell. Consequently, the effective permittivity, in the direction corresponding to the applied field, is calculated using the following relation:

$$\int_S \varepsilon_2 \left. \frac{\partial V}{\partial n} \right|_2 ds = \varepsilon_z \frac{V_2 - V_1}{e} S, \tag{13}$$

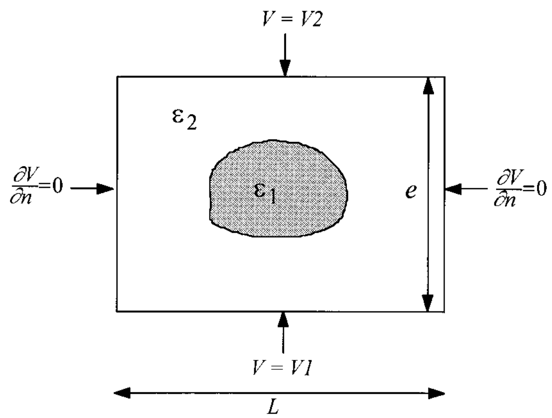


FIG. 2. Notation and boundary conditions related to a two-dimensional periodic composite.

where $V_2-V_1$ denotes the difference of potential imposed in the $z$-direction, $e$ stands for the composite thickness in the same direction, and $S$ denotes the surface of the unit cell perpendicular to the applied field. In Fig. 1(b) the inclusion is allowed to intercept the sides of the parallelipipedic cell. In that case we must take into account the electric displacement flux through the area $S_1$ associated to the medium of permittivity $\varepsilon_1$ to calculate the effective permittivity in the direction corresponding to the applied field. Then Eq. (13) is transformed into Eq. (14).

$$\int_{S_2}\varepsilon_2\frac{\partial V}{\partial n}\bigg|_2 ds+\int_{S_1}\varepsilon_1\frac{\partial V}{\partial n}\bigg|_1 ds=\varepsilon_z\frac{V_2-V_1}{e}(S_1+S_2), \tag{14}$$

where $S_1$ and $S_2$ are the surfaces resulting from the intersection of the volumic regions of permittivity $\varepsilon_1$ and $\varepsilon_2$, respectively, with the upper side of the unit cell, perpendicular to the applied field. At this point it should be emphasized that the BIE method gives an accurate description of the electric potential by including all order multipoles and by taking into account edge and proximity effects even at low and high concentration of inhomogeneities. Hence, this numerical technique does not suffer from the disadvantages of the traditional boundary-value approach.

In this paper we also study two-dimensional media, that we characterize by FE.[23] The system displayed in Fig. 2 is considered; an arbitrarily shaped homogeneous inclusion with permittivity $\varepsilon_1$ is embedded within a homogeneous matrix with permittivity $\varepsilon_2$. The implementation of the FE method consists in dividing the two-dimensional domain into triangular finite elements and interpolating the potential $V$ and its normal derivative $\partial V/\partial n$ on each finite element similarly to the BIE method [see Eq. (12)]. Following this analysis, the solution of Laplace's equation is obtained using the Galerkin method and by solving the resulting matrix equation from the boundary conditions thanks to a standard numerical technique, i.e., Gauss procedure. Having computed the potential and its normal derivative on each triangle of the computational mesh, the electrostatic energy can be expressed as

$$\delta W_e(k)=\frac{1}{2}\int_{S_k}\varepsilon_k\left[\left(\frac{\partial V}{\partial x}\right)^2+\left(\frac{\partial V}{\partial y}\right)^2\right] dx\ dy \tag{15}$$

for each triangular element, where $\varepsilon_k$ and $S_k$ denote the permittivity and the surface of the $k$th triangular element, respectively. Thus, the total energy in the entire composite can be written by summation over the $n_k$ elements such as

$$W_e=\sum_{k=1}^{n_k}\delta W_e(k). \tag{16}$$

In the problem at hand, we consider a portion of the composite material which fills a parallel capacitor. In this manner we obtain the effective permittivity in the corresponding direction of the applied electric field from the electrostatic energy stored in such a capacitor, i.e.,

$$W_e=\frac{1}{2}\varepsilon\frac{S}{e}(V_2-V_1)^2 \tag{17}$$

when a given potential slope is applied across the plates (see Fig. 2). In this equation $S=Ld$ stands for the surface of the plates with side of length $L$ (for the two-dimensional structures considered below, and $d$ is set equal to 1 unit of length).

### C. Extension of the numerical approach to random composite structures

The effective permittivity of random composites cannot be calculated so easily. The actual simulation of the random geometries requires description of a considerable number of cells. Moreover, for three-dimensional structures, the dimension of the matrix systems to be solved becomes very large. Then, the resulting CPU times to obtain solutions increase dramatically. In the case of three-dimensional random composites the effective permittivity is computed by considering the equivalent periodic material (with identical inclusions oriented in the same direction) and taking a statistical mean of the permittivity in the three directions $x$, $y$, and $z$:

$$\varepsilon=\frac{1}{3}(\varepsilon_x+\varepsilon_y+\varepsilon_z). \tag{18}$$

This procedure may be justified by the fact that these media are macroscopically homogeneous and isotropic as previously mentioned. The effective permittivity is a scalar parameter that can be derived from the effective permittivity tensor obtained for an anisotropic medium corresponding to periodic composites with oriented inclusions. In terminating this subsection it should be mentioned that while the BIE method is exact for periodic structures, its application to random media through Eq. (18) constitutes an approximation. It gets worse at high volume fractions of inclusions.

## III. RESULTS AND DISCUSSION

Three series of different numerical experiments were performed. The results of our simulations on the effective permittivity of dielectric mixtures are compared with simple analytical equations. The data obtained on the effects of inhomogeneities orientation are given first. Then, the influence of the scatterers geometry is assessed by computing the ef-

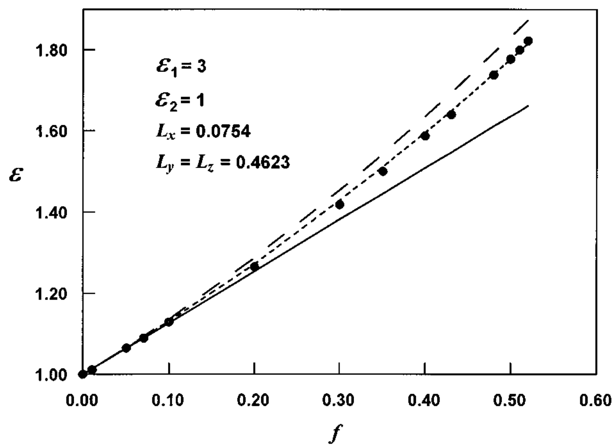


FIG. 3. The effective permittivity is plotted as a function of the volume fraction $f$ of inhomogeneities. The results are shown for randomly oriented ellipsoidal inclusions with permittivity $\varepsilon_1=3$ in the background matrix with permittivity $\varepsilon_2=1$. The depolarization factors of the ellipsoid are $L_x=0.0754$ and $L_y=L_z=0.4623$. Comparison between numerical and analytical evaluations: BIE results are shown as solid circles; predictions from the Polder–Van Santen formula, Bohren–Battan formula, and Fricke formula are displayed as dashed, dotted and solid lines, respectively.

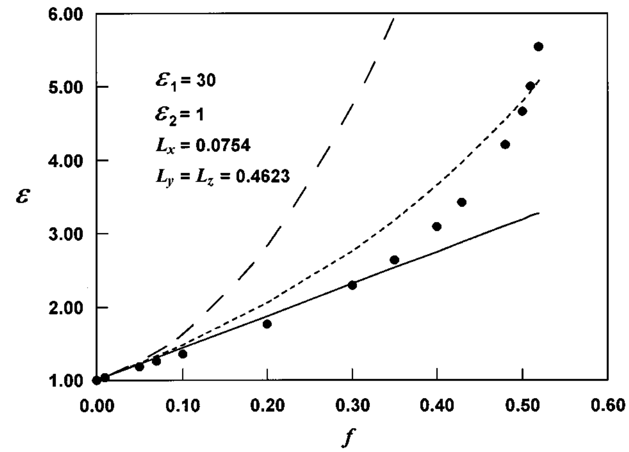


FIG. 4. The effective permittivity is plotted as a function of the volume fraction $f$ of inhomogeneities. The results are shown for randomly oriented ellipsoidal inclusions with permittivity $\varepsilon_1=30$ in the background matrix with permittivity $\varepsilon_2=1$. The depolarization factors of the ellipsoid are $L_x=0.0754$ and $L_y=L_z=0.4623$. Comparison between numerical and analytical evaluations: BIE results are shown as solid circles; predictions from the Polder–Van Santen formula ($\varepsilon_a=\varepsilon$), Bohren–Battan formula, and Fricke formula ($\varepsilon_a=\varepsilon_2$) are displayed as dashed, dotted, and solid lines respectively.

fective permittivity of the mixture. The effect of the components' random distribution in space is dealt with. It should be noted that in our experiments, the background relative permittivity is that of free space ($\varepsilon_2=1$). Impenetrable inclusions were considered for simplicity. The materials being non-lossy, their permittivities are real numbers.

### A. Dependence on the orientation of the inhomogeneities

It was necessary to investigate the influence of inhomogeneities orientation in the mixture for it can affect the establishment of the local electric fields. The effective permittivity of ellipsoidal inclusions randomly oriented in the mixture was first computed in accordance with the method described in the above section. To simplify the analysis, it was set that $b=c=a/4$. Comparing these numerical results with analytical ones resulting from Eqs. (1) and (3) is quite interesting. Two sets of permittivity components values were studied: $\varepsilon_1=3$, $\varepsilon_2=1$ and $\varepsilon_1=30$, $\varepsilon_2=1$. The plots of these data for the effective permittivity are, respectively, displayed in Figs. 3 and 4. Figure 3 corresponds to the case of a low permittivity contrast ratio between background and inclusions. Effective permittivity values provided by the BIE method are in good agreement with the values predicted from the Bohren–Battan equation [Eq. (3)]. This figure also indicates that the predictions from the different models are very close when the volume fraction of inclusions is less than 10% (dilute limit). The Polder–Van Santen equation gives $\epsilon$ values greater than the values obtained by the BIE method whereas values from the Fricke equation are smaller, over the range of volume fraction investigated. This behavior can be attributed to the small permittivity contrast between the inclusion and the host matrix. As this contrast increases, the numerical values of $\varepsilon$ are different from Eqs. (1) and (3) as one can see from Fig. 4. Therefore, the point of importance is that depending on the value of the contrast ratio and the volume fraction of inclusions, the results from the BIE method and those given by mixture formulas can diverge significantly, reflecting the differences in the very basic assumptions made. For a high contrast ratio the response of the system to a potential is found from coupled multipole equations which are not contained in simple dipole mixture rules.[20] The BIE results include all multipoles while Eqs. (1), (3), (5)–(7) are dipolar formulas. Attention was also focused on mixtures of discoidal inclusions with radius $r$ and thickness $h=r/5$, randomly oriented in the host matrix. Simulation results are shown in Figs. 5 and 6 and compared with analytical models [Eqs. (4), (5), (6) and (7)] for $\varepsilon_1=3$, $\varepsilon_2=1$ and $\varepsilon_1=30$, $\varepsilon_2=1$. Conclusions are similar to the case of randomly oriented ellipsoidal inclusions. These graphs show a good agreement between numerical and analytical data at a low value of the permittivity contrast ratio, but indicate a significant discrepancy when this parameter increases.

### B. Geometric shape of the components

To investigate the effect of mixture-components geometry on the effective permittivity, simulations with randomly oriented inclusions of different shapes, such as cube, sphere, ellipsoid, rod, and disc were carried out. The results obtained by the BIE method are presented in Figs. 7 and 8 for $\varepsilon_2=1$ and $\varepsilon_1=30$, $\varepsilon_2=1$, respectively. It shows first that the two curves plotted in Fig. 7 are rather similar: only very slight differences can be noticed. For a low permittivity contrast ratio between background and inclusions, the effective permittivity is not much affected by the inhomogeneities shape. At a high contrast ratio, Fig. 8, this behavior changes and the effective permittivity is higher for ellipsoid-like, or rod-like inclusions. Moreover, Fig. 8 exhibits that the dis-

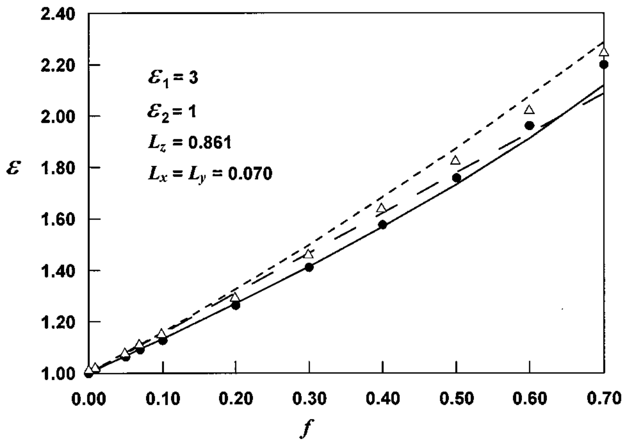


FIG. 5. The effective permittivity is plotted as a function of the volume fraction $f$ of inhomogeneities. The results are shown for randomly oriented discoidal inclusions with permittivity $\varepsilon_1 = 3$, thickness $h = r/5$, in the background matrix with permittivity $\varepsilon_2 = 1$. The solid circles are obtained by the BIE method. Dotted, dashed, and solid lines correspond to the Bruggeman ($\varepsilon_a = \varepsilon$), Van Beek ($\varepsilon_a = \varepsilon_2$), and lamellae formulae, respectively. The open triangles represent the solution calculated from the Bohren–Battan formula when discoidal inclusions are modeled as equivalent oblate ellipsoids with depolarization factors $L_x = L_y = 0.070$ and $L_z = 0.86$.

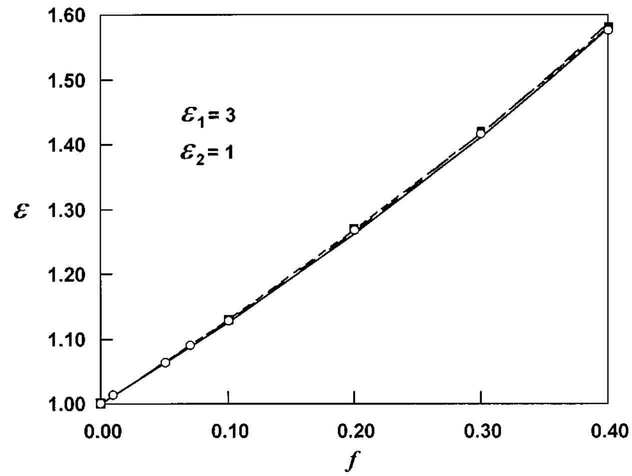


FIG. 7. Influence of the shape of inhomogeneities on the effective permittivity. Inclusions with permittivity $\varepsilon_1 = 3$ are randomly oriented in the background matrix with permittivity $\varepsilon_2 = 1$. The effective permittivity is computed by the BIE method for different shapes of inhomogeneities: open circles are obtained for discoidal inclusions of radius $r$ and thickness $h = r/5$, solid squares for cubic inclusions, dashed line describes ellipsoidal inclusions with semi-axes $b = c = a/4$, dotted line represents cylindrical inclusions (rods) with radius $r$ and height $h = 16r$, and solid line corresponds to spherical inclusions.

crepancy between the different models prediction is less than 20% in the range of volume fraction investigated.

### C. Type of random distribution

To investigate the influence of the component random arrangement in space, several simulations were carried out. The unit cell of the two-dimensional composite material is presented in Fig. 9. This unit cell consists of a square diamond of permittivity $\varepsilon_1$ inside a square of permittivity $\varepsilon_2$ and side $L = 2$.[24] The corresponding volume fraction of the inclusion phase is given by $f = \eta^2/4$. To take into account the effect of the inhomogeneities distribution on the effective permittivity, the domain of permittivity $\varepsilon_2$ is 100 times duplicated and the inclusions of permittivity $\varepsilon_1$ are arranged randomly with concentration equals to $C$ (see Fig. 10). We define this concentration by the ratio of the number of inclusions to the total number of unit cells of permittivity $\varepsilon_2$. Then, the resulting volume fraction of the random composite

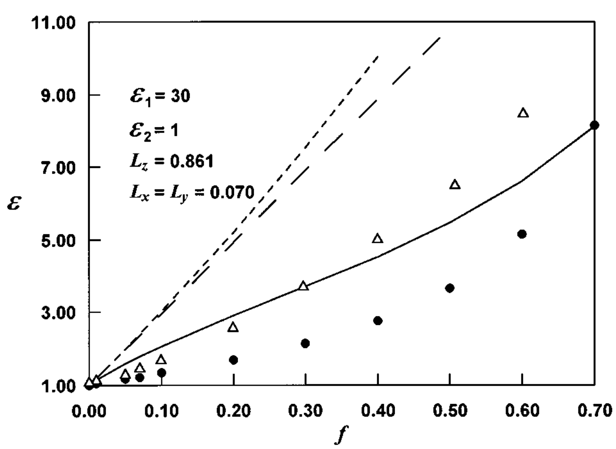


FIG. 6. The effective permittivity is plotted as a function of the volume fraction $f$ of inhomogeneities. The results are shown for randomly oriented discoidal inclusions with permittivity $\varepsilon_1 = 30$, thickness $h = r/5$, in the background matrix with permittivity $\varepsilon_2 = 1$. The solid circles are obtained by the BIE method. Dotted, dashed, and solid lines correspond to the Bruggeman ($\varepsilon_a = \varepsilon$), Van Beek ($\varepsilon_a = \varepsilon_2$), and lamellae formulae, respectively. The open triangles represent the solution calculated from the Bohren–Battan formula when discoidal inclusions are modeled as equivalent oblate ellipsoids with depolarization factors $L_x = L_y = 0.070$ and $L_z = 0.861$.

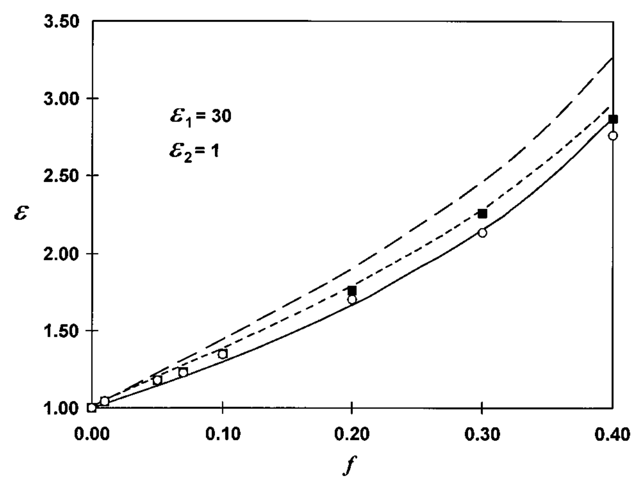


FIG. 8. Influence of the shape of inhomogeneities on the effective permittivity. Inclusions with permittivity $\varepsilon_1 = 30$ are randomly oriented in the background matrix with permittivity $\varepsilon_2 = 1$. The effective permittivity is computed by the BIE method for different shapes of inhomogeneities: open circles are for discoidal inclusions of radius $r$ and thickness $h = r/5$, solid square for cubic inclusions, dashed line describes ellipsoidal inclusions with semi-axes $b = c = a/4$, dotted line represents cylindrical inclusions (rods) with radius $r$ and height $h = 16r$, and solid line corresponds to spherical inclusions.

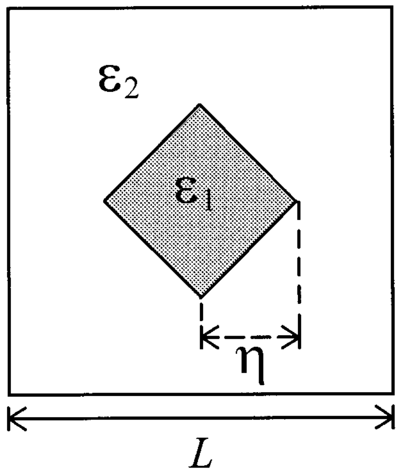


FIG. 9. Geometry of the unit cell of the two-dimensional composite material investigated. The unit cell consists of a square diamond of permittivity $\varepsilon_1$ and half-diagonal $\eta$ inside a square of permittivity $\varepsilon_2$ and side 2.

is expressed by $\tilde{f}=Cf$. Examples of different kinds of distributions are displayed in Fig. 11. The first pattern (a) concerns inclusions that are regularly distributed in the host medium (low disorder). In this case, the arrangement is quasi-periodic. The second pattern (b) deals with agglomerate distribution of inhomogeneities. Inclusions aggregate to form isolate clusters. The last pattern (c) describes a random arrangement of inhomogeneities (strong disorder). The effective permittivity is computed by the FE method for these types of distributions and for two values of the permittivity ratio $\varepsilon_1=2$, $\varepsilon_2=1$ and $\varepsilon_1=5$, $\varepsilon_2=1$. The resulting effective permittivities are compared with those obtained from periodically arranged inclusions (Figs. 12 and 13). The effect of the distribution on the dielectric properties of the composites is thus observed. For a low contrast ratio between the permittivities of the background and inclusions, the effective permittivity of the composite is not affected by the inhomogeneities distribution. Increasing this contrast ratio shows the influence of randomness. The deviation between the results obtained for the three distributions investigated and those computed in the case of a periodic arrangement is large for a strong disorder and when inclusions form isolated clusters.

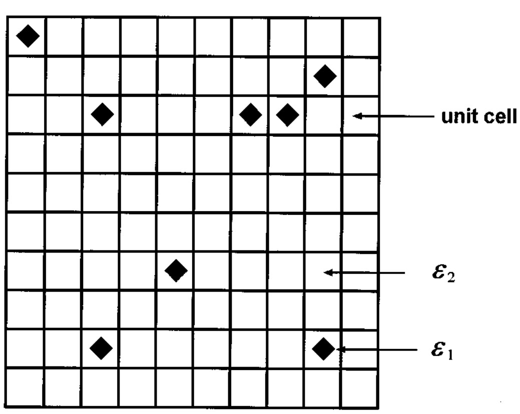


FIG. 10. Geometry of the two-dimensional composite with random distribution of inclusions.

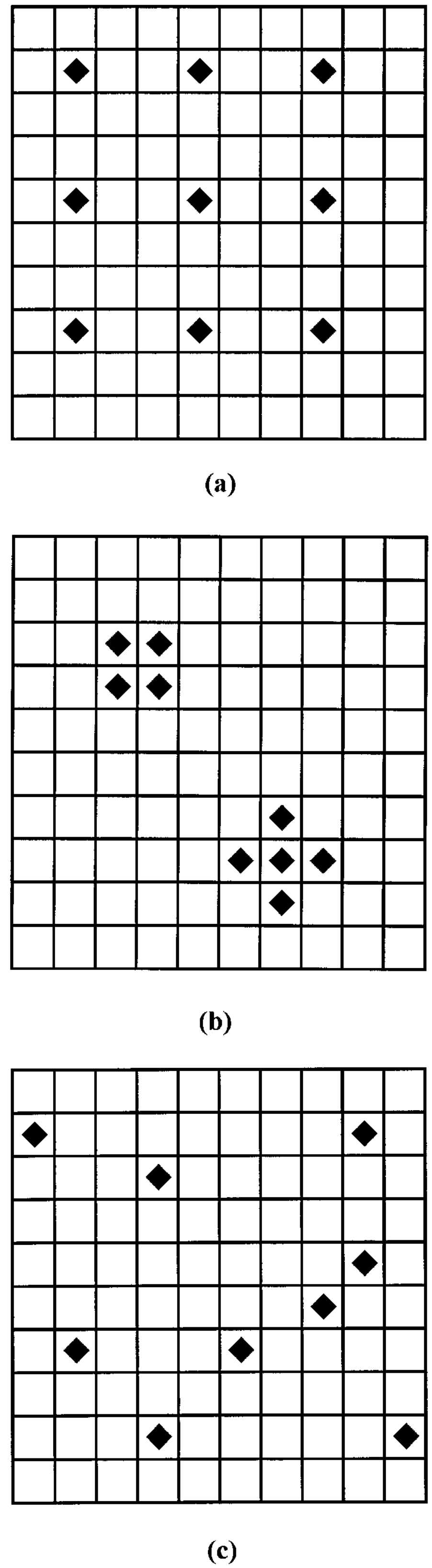


FIG. 11. Some of the patterns investigated by the FE method: (a) regular distribution, (b) agglomerate distribution, and (c) random distribution.

## IV. CONCLUSIONS

This paper describes an *ab initio* simulation approach to evaluate the effective dielectric constant of random arrays of

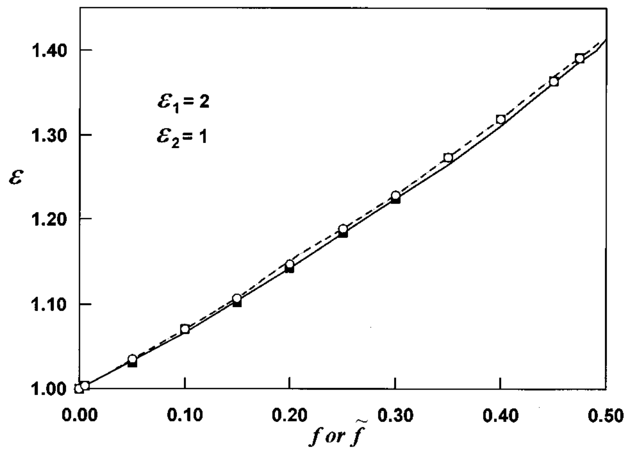


FIG. 12. Influence of the distribution of inhomogeneities on the effective permittivity. Inclusions with permittivity $\varepsilon_1 = 2$ are embedded in the background matrix with permittivity $\varepsilon_2 = 1$. The effective permittivity is computed by the FE method for the different types of distributions investigated. The open circles, solid squares, and the dashed line correspond to random, regular, and agglomerate distributions of inhomogeneities, respectively. The dashed curve describes the effective permittivity of the equivalent composite with a periodic arrangement of inclusions.

inclusions characterized by a permittivity $\varepsilon_1$ in a dielectric matrix of permittivity $\varepsilon_2$ under the quasistatic assumption, i.e., large scale regime. The computational results presented here, combined with others previously published by our team, show the potential interest of this method for interpretation of experimental data. Our results are in addition, compared to previously published analytical analysis. Our calculations confirm that FE and BIE methods can be applied to high concentrations of inclusions for which mean-field approaches generally do not hold good for they are unable to take into account interactions among inclusions. Therefore, our numerical method can fairly determine when these analyses are applicable. Another focus of our efforts was to investigate high contrast ratios between the permittivity of background and inclusions. The methodology presented here is useful to investigate the dielectric properties of dense composite materials which are of importance in many technological applications, e.g., medical applications of microwaves, characterization of geophysical media, and materials science. The effectiveness and flexibility of this simulation approach enables us to generalize it to multicomponent mixtures of arbitrary shapes. It can be extended for a wide variety of problems. For reasons of mathematical analogy, the results are also valid for the magnetic permeability and the diffusivity of such materials. We intend in the near future to present the results of numerical experiments beyond the quasistatic approximation or when the dielectric constant has a nonzero imaginary part. For instance, scattering effects become important as the frequency of the electromagnetic excitation is increased and the concept of effective permittivity loses its physical significance. Clearly, we are still a long way from developing a predictive theory to describe the dielectric properties of realistic composite materials but this analysis shows the need for well-controlled numerical approaches in the field of dielectric mixtures. While these computations are useful in highlighting physical situations which reproduce the qualitative features of real laboratory experiments, it is difficult to interpret the parameters appearing in these models in terms of measurable quantities in actual systems. In order to make progress in the understanding of the physical behavior of dielectric mixtures, new experiments are needed to quantify as carefully as possible the geometric microstructures of materials.

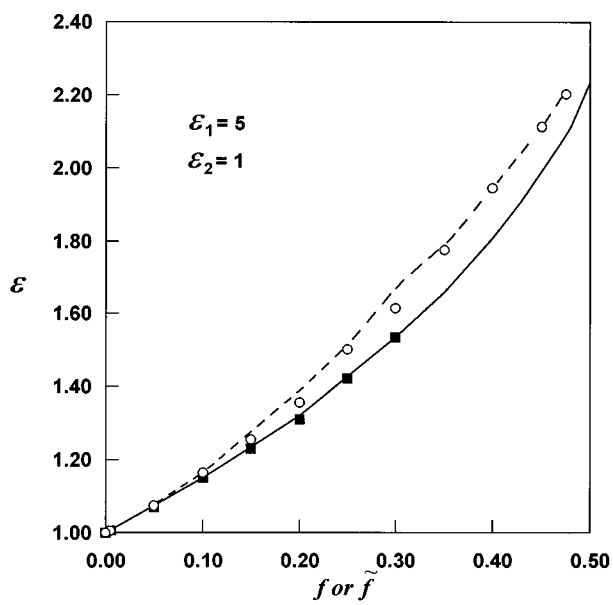


FIG. 13. Influence of the distribution of inhomogeneities on the effective permittivity. Inclusions with permittivity $\varepsilon_1 = 5$ are embedded in the background matrix with permittivity $\varepsilon_2 = 1$. The effective permittivity is computed by the FE method for the different types of distributions investigated. The open circles, solid squares, and the dashed line correspond to a random, regular, and agglomerate distribution of inhomogeneities, respectively. The dashed curve describes the effective perimittivity of the equivalent composite with a periodic arrangement of inclusions.


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