AMPERIAN MAGNETISM IN THE DYNAMIC RESPONSE OF GRANULAR MATERIALS

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Abstract We study the coherent reflectance of electromagnetic waves from a random system of identical spheres with radius comparable to the wavelength of the incident radiation. An effective–medium theory for this system is developed and it is found that the effective–medium must posses an effective magnetic permeability, even if the spheres are non–magnetic, in order to be consistent with continuum electrodynamics. The physical origin of this magnetic effect is discussed and we conclude that it is due to the induction of closed currents in the spheres, being then analogous to the mechanism proposed by Ampère when he tried to explain the origin of magnetism. It turns out that the effective magnetic permeability as well as the effective electric permittivity depend on the angle of incidence and the polarization of the incident wave. We derive formulas for the coherent reflectance from a half–space and display numerical results.

Keywords: granular matter, electrodynamics, optical properties, random system, effective-medium theory

1. INTRODUCTION

The concept of an effective medium has been extremely useful in the description of the electromagnetic response of granular matter [1]. By granular matter we will understand a very general class of materials

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composed by granular inclusions of one type of material embedded in an otherwise homogeneous matrix of another type of material. By electromagnetic response we mean the polarization and magnetization processes induced in the material by an externally applied electromagnetic field. In electrodynamics of continuous media one introduces the concept of polarization and magnetization fields. They are called material fields because they are defined only in the regions occupied by the materials, they are attached to their presence, and outside these regions these fields vanish. The optical properties of the material are determined by the manner in which these material fields respond to the applied one, and this response is usually given in terms of response functions like the dielectric function and the magnetic susceptibility, which will be generally referred as the optical coefficients of the material.

Furthermore, one might think that behind all these concepts there is the assumption of a continuous material and that the values acquired by the material fields correspond to a quantitative measure of the induced polarization and magnetization phenomena. Nevertheless, one also knows that in essence all matter is granular, after all, one can think that any piece of matter is made of a large collection of small grains, these grains being the atoms and molecules. Therefore, due to this granular structure the induced electromagnetic field within any material is a highly varying function of space and time. This field is usually called the *microscopic* field. But when this microscopic field is decomposed as the sum of an average component plus a fluctuation component, one finds that the length scales of their spatial variations are very different, while the average component varies on a length scale of the order of the wavelength of the incident field, the fluctuation component varies on a length scale of atomic dimensions. Now, the fields that appear in Maxwell's equations of continuous media correspond only to the average component, the fluctuation component is neglected. The average component is also called the macroscopic field and one refers to the equations that govern its behavior as the macroscopic Maxwell's equations. As a consequence, all the laws derived in electrodynamics of continuous media, like Snell's law, Fresnel's relations and Poynting's theorem, neglect the contribution due to the field fluctuations, providing only relations between the average (macroscopic) values of the electromagnetic and material fields. Thus the concept of continuity in the macroscopic equations is somewhat artificial and is the result of an averaging procedure that smooths out the space–time variations of the fields by neglecting the field fluctuations. But in ordinary materials the power carried along by the average component is much greater than the one carried along by the fluctuations, and this fact is what justifies the successful application of continuous electrodynamics to ordinary materials. As a consequence, one can conclude that the concept of continuity in macroscopic electrodynamics is a matter of scale based in the actual possibility of neglecting the contribution of the small fluctuations of the fields caused by the molecular "granularity" of matter. Nevertheless, this does not mean that the field fluctuations are undetectable, because if this were true we would not be able to see, for example, a blue sky.

Now we move to the problem of the optical properties of granular materials where the characteristic size of the individual grains is not longer of atomic dimensions but is rather of macroscopic dimensions, and their optical properties are described by macroscopic electrodynamics. First we will assume that although the characteristic size of the grains is macroscopic, it is still much smaller than the wavelength of the incident radiation. In this case the power carried along by the average component of the electromagnetic field is still large as compared to the one carried along by the fluctuations; although not as large as in the case of ordinary materials with "molecular" granularity. If we now concentrate our attention in the physical description of *only* the average component of the fields, we can ask ourselves if it is now possible to extend the concept of the continuity of matter and to define a continuous medium in which the average component of the field behaves exactly in the same manner as in the actual granular material. This artificial continuous medium is commonly known as *effective* medium, in which effective material fields can be defined and whose response to an applied external field yield, for example, an effective dielectric function and an effective magnetic susceptibility. From this perspective one could regard the material fields of ordinary materials in macroscopic electrodynamics, also as properties of an artificial effective continuous medium which describes correctly the behavior that the average field has in the actual material with "molecular granularity". The main advantage of an effective–medium approach is that one can immediately use all the results of continuous electrodynamics by simply setting, in the relevant expressions, instead of the macroscopic response functions the effective response functions, and to certain extent one forgets about the granularity of the material. One has only to be careful about certain aspects of the physical interpretation of the results. For example, in macroscopic electrodynamics one interprets the imaginary part of the response functions as a quantity that is proportional to the absorption of energy by the system, in the case of an effective medium of a granular material, the imaginary part of the effective response functions is proportional not only to the energy that is absorbed but also to energy that is scattered. In this way, the energy balance forces one to look at the energy flux carried by the fluctuations, as an energy flux that is "taken away" from the flux carried by the average component of the field.

The problem is now to find a relationship between the effective response functions and the actual geometric and optical parameters of the grains and the matrix, as well as the statistical parameters that describe the way in which grains are mixed into the matrix. This is the problem that has attracted the attention of many researchers for more than a century, and has required the efforts of theoretical and experimental physicists as well as applied mathematicians and engineers [2]. Besides its interest as a problem in basic physics, its solution can be used in a wide variety of applications. First, because its range of application extends beyond the field of optical properties and comprises all physical properties involving a linear response to an external field, like in the elastic, electric, thermal, or hydrodynamic properties of either granular composites, rocks, emulsions, suspensions or colloids. The interest lies then in the calculation, in this type of systems, of properties like: the effective stress-strain tensor, the effective electrical conductivity, the effective thermal conductivity, the effective chirality or the effective viscosity. Second, because the knowledge of the relationship between the effective response properties and the parameters that characterize a granular system, opens the possibility for the construction and design of novel type of materials fulfilling requirements and specifications that cannot be found in ordinary materials. Although there has been a significant progress in the solution of this problem and new type of materials with unexpected properties have been produced and designed, we are still far from claiming that the problem has been finally solved. There is a wide collection of expressions, called "mixing rules", that propose explicit expressions that relate the parameters characterizing a granular system and its effective response, nevertheless their range and conditions of validity as well as the microstructure that is assumed for their derivations, are issues that very often are not clear. Also, the experimental characterization of the microstructure of a granular system is not an easy task, and one usually ends up with only a few from the total set of relevant parameters.

Finally, we address essentially the same problem of extending the idea of a continuous effective medium for the description of the optical properties of a granular system, but now when the characteristic size of the grains is of the same order of magnitude as the wavelength of the incident radiation. In this case the power carried along by the fluctuations of the field might be as large as the one carried along by the average component, and the average field is now called the coherent field, while the field fluctuations are called the diffuse field. Nevertheless, one can still ask oneself if in this case the behavior of the average component of the field can still be described by a continuous effective medium. There are several contributions towards the solution of this more complicated problem in which the scattering processes play a more important role. One finds in the literature explicit expressions that relate, for example, the effective index of refraction of a dilute system of randomly located identical spheres with the forward scattering amplitude of an individual sphere. Probably, the most popular derivation of this relation is the one given by van de Hulst in his book [3]. There have been also efforts to generalize this relation to systems with a larger concentration of spheres [4] or to different geometries, like a spherical matrix with spherical inclusions [5].

However, there are critical remarks about the use of the effective index of refraction derived by van de Hulst in expressions like the Fresnel's relations that yield the reflection and transmission amplitudes from a slab in terms of the index of refraction and the optical coefficients of the material. For example, C. Bohren has considered the simple case of a plane wave at normal incidence into a slab containing a dilute concentration of randomly located identical polarizable spherical inclusions, and he has calculated the coherent component of the reflected and transmitted fields [6]. He finds that in order to calculate the amplitudes of these fields by replacing the slab by a continuous medium with an effective index of refraction and Fresnel's relations, it would be necessary to define two different index of refraction: one for reflection and one for transmission. But instead of doing that he proposes to choose another two different optical coefficients: an effective electric permittivity and an effective magnetic permeability. This last one is proportional to the difference between the forward and backward scattering amplitude of an individual sphere. The problem is to justify how come a composite made of two nonmagnetic components turns out to be magnetic. The main objective of this paper is to clarify this issue as well as to extend the calculation to non–normal incidence. We do this by using scattering–wave and Mie theories to calculate the reflected and transmitted fields from a thin slab with containing a dilute concentration of randomly located identical polarizable spheres, and then we identify the current distributions that may act as sources of these fields. We find that these sources should be given by a superposition of open and closed currents induced in the spheres. The closed currents are induced by the time variations of the magnetic field, and in this sense they represent a true *bona fide* magnetic response of the system. We find that both the effective electrical permittivity and the effective magnetic permeability depend on the angle of incidence and the polarization of the incident field. Therefore they cannot be regarded as intrinsic properties of the granular system, nevertheless they provide the basis for the calculation of the reflected and transmitted fields. Also, their product being proportional to the square of the effective index of refraction turns out to be independent of the angle of incidence and the polarization of the incident beam, and its expression in terms of the scattering properties of the individual spheres coincides with the one derived by van de Hulst.

One might call this type of magnetic response in a granular system: *Amperian magnetism*, because in the beginning of electrodynamics, A. M. Ampère had envisioned the physical origin of magnetism as the result of closed currents induced in the molecules by the time variations of the magnetic field. Later on it was found that magnetism was a more complicated phenomenon related more to the spin of the electrons than to the induction of closed currents in the molecules. But in a system like a granular composite one has small macroscopic spheres instead of molecules and the induction of closed currents is more favorable in large spheres than in small spheres. In a more technical language, the contribution of the spheres to the effective magnetic permeability turns out to be proportional to the asymmetry in the scattering amplitude between the forward and the specular direction in the individual spheres, and from Mie theory one sees that this asymmetry increases with the size of the spheres. In the limit of very small spheres the scattering becomes quite isotropic thus the contribution of the spheres to the effective permeability vanishes, and one recovers the non-magnetic character of the effective medium.

A more transparent example of Amperian magnetism is perhaps the recently developed microstructured materials that operate in the microwave region. A particular type of these materials consists of an insulating matrix in which a collection of millimeter–size copper rings are embedded within forming a 3D periodic structure [7]. A time–varying magnetic field induces currents in the rings giving rise to closed currents that are responsible for the magnetic character of the response. This is a beautiful example of Amperian magnetism and shows how a composite of two non–magnetic materials becomes magnetic. By opening the rings with a small gap, there is also an induced capacitance, which together with the inductance due to the induced currents in the rings, gives rise to a resonance phenomenon, and this yields frequency regions in which the effective magnetic permeability becomes negative. Another interesting microstructured material is an insulating matrix in which a collection of very thin long wires are embedded within the matrix forming a well–defined cubic structure. It can be shown that this type of microstructured materials posses an effective dielectric response that is negative for certain frequencies. By combining the wire and the ring structures it has been shown that there are frequency regions in which both the effective electric permittivity and the magnetic permeability are negative yielding a negative effective index of refraction. It has been also argued that in the frequency regions in which the index of refraction is negative, one could use this uncommon property for the construction of a perfect lens.

Finally, we believe that the expressions derived in this paper, although limited to dilute systems, are not a pure and simple curiosity, but on the contrary they may be useful in several applications. For example, there is now interest to follow, in real time, different processes that take place in turbid media, through the changes in their effective index of refraction. Nevertheless, although measurements of the attenuation of light through turbid systems are done routinely in many laboratories, there are few transmission experiments which measure both, the real and imaginary part of their effective index of refraction [8], [9]. A simple and potentially very useful way of measuring the effective index of refraction in turbid media is by critical–angle refractometers [10], [11], [12]. In this method the real and imaginary parts of the effective index of refraction are obtained by inverting the relationship between the reflection amplitude and the effective index of refraction. The naive use of Fresnel expressions to perform this inversion would lead to errors in both, accuracy and interpretation. In this respect, the expressions for the reflection amplitude derived here could be used, together with data of critical–angle refractometers, to obtain not only more accurate results of the optical constants of turbid media, but also to start doing reliable modelling of the correlation between their changes and some of the specific processes that take place within the system.

2. BASIC CONCEPTS

First we review some basic concepts of linear response in ordinary materials. If the response is linear the polarization **P** and the magnetization **M** are proportional to the incident field. The relation between **P** and **M** and the incident electromagnetic field is given, in general, in terms of integral operators with kernels that are non-local in space and time. However, for the case homogeneous and isotropic ordinary materials one can write $\mathbf{P} = \epsilon_0 \chi^E \mathbf{E}$ and $\mathbf{M} = \chi^H \mathbf{H}$, where χ^E and χ^H are scalar algebraic functions. The functions χ^E and χ^H are called the electric and magnetic susceptibilities, respectively, and they are intrinsic properties of the material. The non–locality in space is avoided by relating **P** and **M** not to the incident fields but rather to the total fields **E** and **H**, which are given by the sum of the incident plus the (average) induced field. The non–locality in time is accounted for by expressing the susceptibilities in the Fourier space of frequencies. Thus for an isotropic and homogeneous material in the presence of an applied field oscillating with frequency ω, the susceptibilities χ^E and χ^H are only functions of ω. The propagation wavevector k of the field within the material is given by $k = (\omega/c)n$, where c is the speed of light and $n = \sqrt{\tilde{\epsilon}\tilde{\mu}}$ is the index of refraction. Here $\tilde{\epsilon} = 1+\chi^E$ and $\tilde{\mu} = 1+\chi^H$. The reflection amplitude r is defined as $r = E_r/E_i$ where is the amplitude of the reflected electric field while E_i is the amplitude of the incident electric field. For the case of reflection from a half space the Fresnel's relations are

$$r_{hs}^{TE} = \frac{\tilde{\mu}k_z^i - k_z}{\tilde{\mu}k_z^i + k_z} \tag{1}$$

and

$$r_{hs}^{TM} = \frac{\tilde{\epsilon}k_z^i - k_z}{\tilde{\epsilon}k_z^i + k_z}, \tag{2}$$

where $k_z = k\sqrt{n^2 - \sin^2\theta_i}$, n is the index of refraction of the material, and the superscripts TE and TM denotes transverse–electric and transverse–magnetic polarization, respectively, referring to the cases where the electric or the magnetic field are perpendicular to the plane of incidence.

As mentioned above, in electrodynamics of continuous media the set of Maxwell's equations describe the behavior of only the average component of the electromagnetic field. Moreover, when one tries to extend the idea of continuity to the case of granular composites through the concept of effective medium, one has to properly define the average of the fields. The average procedure smooths out the field variations to a given specified scale. From the experimental point of view this process can be thought as performed by the measuring apparatus when trying to detect highly varying fields. From a mathematical point of view the averaging procedure can be represented by a projection operator acting on the highly varying microscopic field. There are many ways of taking the average of the microscopic field, there is, for example, a spatial average in which a spatial integration of the field times a weight function is performed around any given point in space, there is truncation in Fourier space in which the spatial Fourier transform is truncated up to a maximum cut off wavevector and then is transformed back into real space. In this work we are dealing with a system with randomly located inclusions and we will consider a configurational average, that is, the field at a certain point in space is averaged by "moving around" the location of the spheres. This "moving around" in a system of randomly located spheres generates a finite set of different configurations characterized by a different location of the spheres. The average value is obtained by adding up the values of the field at any given point in space generated by each configuration and then dividing it by the total number of configurations. One should take a sufficiently large number of configurations in order to obtain a stable value for the average. A critical analysis on the dependence of the results on the type of average that is taken is out of the scope of this work, but interested readers can take a look at Ref. [13].

3. FORMALISM

Our approach to the effective medium theory consists of comparing the average scattered fields from a thin slab of the random system of spheres to the radiated fields by an equivalent homogeneous slab when a plane wave is incident on them. By matching the scattered and radiated fields the optical coefficients of the effective medium are obtained. In what follows we briefly describe the main steps in the calculation of the effective optical coefficients. More details can be found in Ref. [14].

First, we consider a dilute random distribution of spherical particles in vacuum (no matrix) contained in a boundless slab region parallel to the XY plane and $-d/2 < z < d/2$. The system is in the presence of an incident plane wave with an electric field given by $\mathbf{E}^i(\mathbf{r},t) = E_0 \exp i(\mathbf{k}^i \cdot \mathbf{r} - \omega t)\,\widehat{\mathbf{e}}_i$, where $\mathbf{r}$ and t are the position vector and time, respectively, ω is the radial frequency, $\widehat{\mathbf{e}}_i$ is a unit vector in the direction of polarization, $\mathbf{k}^i = k^i_y \widehat{\mathbf{a}}_y + k^i_z \widehat{\mathbf{a}}_z$ is the incident wave vector assumed to lie on the YZ plane, and $\widehat{\mathbf{a}}_x$, $\widehat{\mathbf{a}}_y$, and $\widehat{\mathbf{a}}_z$ are unit vectors along the Cartesian axes of coordinates (see Fig. 1). The electric field satisfies $\widehat{\mathbf{e}}_i \cdot \mathbf{k}^i = 0$, and $|\mathbf{k}^i| = k$, where $k = \omega/c = 2\pi/\lambda$ is the wave number in vacuum, λ is the corresponding wavelength and c is the speed of light. The time dependence $\exp(-i\omega t)$ will be assumed implicit and we use the SI system of units.

The incident field is scattered by the particles, and we assume that their number density is low enough so the independent–scattering approximation is valid. Within this approximation the total scattered field is given by the sum of the fields scattered by each of the particles in the slab region. Therefore, the scattered field $\mathbf{E}^S$ due to a collection of N spherical particles with their centers located at $\{\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_p, \ldots, \mathbf{r}_N\}$ can be written as [4],

$$\mathbf{E}^S(\mathbf{r}) = \sum_{p=1}^{N} \int d^3r' \int d^3r'' \overline{\overline{G}}_0(\mathbf{r},\mathbf{r}') \cdot \overline{\overline{T}}(\mathbf{r}' - \mathbf{r}_p, \mathbf{r}'' - \mathbf{r}_p) \cdot \mathbf{E}^E_p(\mathbf{r}''), \quad (3)$$

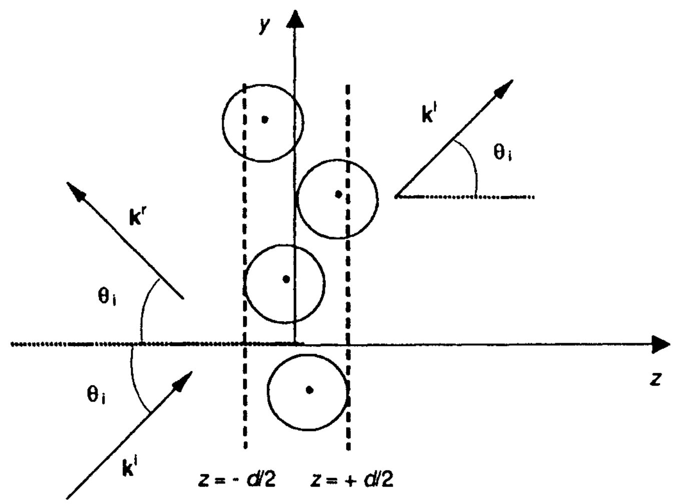

Figure 1. A slab of a dilute random–system of spheres. The centers of the particles are within the planes $z = -d/2$ and $z = d/2$.

where $\overline{\overline{G}}_0(\mathbf{r}, \mathbf{r}')$ is the dyadic Green's function in free space, $\overline{\overline{T}}(\mathbf{r}', \mathbf{r}'')$ is the transition operator for a sphere [4], and $\mathbf{E}_p^E$ denotes the exciting field. This is defined as the field that drives the scattering process in particle p, that is, the incident field plus the field scattered by the rest of the particles in a region within and around particle p. Thus $\mathbf{E}_p^E$ depends parametrically on the location of the rest $N-1$ particles.

Since we are assuming a dilute system of particles and thin slab, the exciting field may be approximated as the incident field to the slab, $\mathbf{E}_p^E \simeq \mathbf{E}^i$. By using the plane–wave expansion of the dyadic Green's function and the momentum representation of the transition operator $\overline{\overline{T}}(\mathbf{r}', \mathbf{r}'')$ of an isolated sphere: $\overline{\overline{T}}(\mathbf{p}', \mathbf{p}'')$ and finally averaging the scattered fields with respect to the position of the particles, one arrives to,

$$\langle \mathbf{E}^S(\mathbf{r}) \rangle_{slab} = \begin{cases} \mathbf{E}_+^S \exp(i\mathbf{k}^i \cdot \mathbf{r}) & \text{for } z > d/2 \\ \mathbf{E}_-^S \exp(i\mathbf{k}^r \cdot \mathbf{r}) & \text{for } z < d/2 \end{cases}, \tag{4}$$

where

$$\mathbf{E}_+^S = i\frac{E_0}{2}\rho \frac{(\overline{\overline{1}} - \widehat{\mathbf{k}}^i\widehat{\mathbf{k}}^i)}{k_z^i} \cdot \overline{\overline{T}}(\mathbf{k}^i, \mathbf{k}^i) \cdot \widehat{\mathbf{e}}_i \, d \tag{5}$$

$$\mathbf{E}_-^S = i\frac{E_0}{2}\rho \frac{(\overline{\overline{1}} - \widehat{\mathbf{k}}^r\widehat{\mathbf{k}}^r)}{k_z^i} \cdot \overline{\overline{T}}(\mathbf{k}^r, \mathbf{k}^i) \cdot \widehat{\mathbf{e}}_i \, \frac{\sin k_z^i d}{k_z^i}, \tag{6}$$

where $\mathbf{k}^r = k_x^i\widehat{\mathbf{a}}_x + k_y^i\widehat{\mathbf{a}}_y - k_z^i\widehat{\mathbf{a}}_z$ is the wave vector in the specular direction, $k_z^i = \sqrt{k^2 - (k_x^i)^2 - (k_y^i)^2}$, and ρ is the density of particles. In the averaging procedure we assumed that the positions of the particles are independent of each other (i.e., we ignored the exclusion volume) and that the probability to find a particle with its center inside the volume $d^3\mathbf{r}$ is uniform and given by $d^3\mathbf{r}/V$, where V is the volume of the slab. Eq. (4) means that the scattered field interferes constructively along two directions: $\mathbf{k}^i$ and $\mathbf{k}^r$, independently of the location of the scatterers, for this reason these are the only components of the field that survive after a configurational average.

Equations (5) and (6) can be put in terms of the scattering matrix elements commonly used in describing light scattering from small particles. (The scattering matrix is clearly defined in the book by Bohren and Huffman [15]) First, one recognizes that $(\overline{\overline{\mathbf{I}}} - \widehat{\mathbf{k}}^a\widehat{\mathbf{k}}^a)\cdot\overline{\overline{T}}(\mathbf{k}^a, \mathbf{k}^b) = 4\pi\overline{\overline{F}}(\widehat{\mathbf{k}}^a, \widehat{\mathbf{k}}^b)$, where $\overline{\overline{F}}$ is the far–field scattering dyad, and then express $\overline{\overline{F}}$ in terms of the scattering matrix elements S. When the particles are spherical, there are only two non–zero matrix elements, S_1 and S_2 and the following expression is obtained

$$\mathbf{E}_+^S = -E_0\gamma\frac{kd}{\cos\theta_i}S(0)\widehat{\mathbf{e}}_i \tag{7}$$

$$\mathbf{E}_-^S = -E_0\gamma\frac{k}{\cos\theta_i}\frac{\sin k_z^i d}{k_z^i}[-(\cos\theta_i\widehat{\mathbf{a}}_y + \sin\theta_i\widehat{\mathbf{a}}_z)(\cos\theta_i\widehat{\mathbf{a}}_y - \sin\theta_i\widehat{\mathbf{a}}_z) \times S_2(\pi - 2\theta_i) + \widehat{\mathbf{a}}_x\widehat{\mathbf{a}}_x S_1(\pi - 2\theta_i)]\cdot\widehat{\mathbf{e}}_i\,, \tag{8}$$

where $S(0) \equiv S_1(\theta = 0) = S_2(\theta = 0)$ is called the forward scattering amplitude, $\gamma \equiv 3f/2x^3$, $x \equiv ka$ is the size parameter, $f = N4\pi a^3/3V$ is the filling fraction of spheres, $\pi - 2\theta_i$ is the specular direction, and we recall that $k_z^i = k\cos\theta_i$. Notice that while $\mathbf{E}_+^S$ is directly proportional to d, $\mathbf{E}_-^S$ is proportional to $\sin k_z^i d/k_z^i$. Here d is the thickness of the averaging region where the centers of the spheres are randomly located. Since we are considering that the slab is thin enough for the independent scattering approximation to be valid, one can take d small enough and approximate $\sin k_z^i d\,/k_z^i \approx d$.

Notice also, that in general $|\mathbf{E}_+^S| \neq |\mathbf{E}_-^S|$, and this is a direct consequence of the forward–backward anisotropy of Mie scattering, that is, $S(0) \neq S_m(\pi - 2\theta_i)$ for $m = 1, 2$. We also recall that this anisotropy is more acute the larger the sphere. For spheres whose radii are very small with respect to the incident wavelength, this anisotropy almost disappears and one has $|\mathbf{E}_+^S| \approx |\mathbf{E}_-^S|$.

Now the idea is to find the effective current distribution that act as a source of these fields, and identify this effective currents with the average current distribution induced in an effective medium. To model this effective currents within the thin slab, we imagine the simplest possible geometry: a 2D homogeneous and isotropic sheet with no internal structure. We locate the sheet at the $z = 0$ plane and consider an incident plane wave with TE polarization: $\mathbf{E}^i(\mathbf{r},t) = E_0 \exp\left[i(k_y^i y + k_z^i z)\right] \widehat{\mathbf{a}}_x$. The fields radiated by this 2D-sheet of homogeneous material can be found by assuming some 2D–currents (i.e., surface currents) driven by the incident field and applying Maxwell's equation in the region about $z = 0$.

The radiated fields by the 2D–sheet are found in the form

$$\mathbf{E}^J = \begin{cases} E_+^J \exp(i\mathbf{k}^i \cdot \mathbf{r})\widehat{\mathbf{a}}_x & \text{for } z > 0 \\ E_-^J \exp(i\mathbf{k}^r \cdot \mathbf{r})\widehat{\mathbf{a}}_x & \text{for } z < 0, \end{cases} , \tag{9}$$

where $\mathbf{k}^i$ and $\mathbf{k}^r$ have the same meaning as before. The coefficients E_+^J and E_-^J are found in terms of the effective currents. Then these currents are assumed proportional to the incident field through some effective optical coefficients.

If we assume only open currents along the direction of the electric field, $\mathbf{J} = j_{0x}\delta(z)\exp(ik_y^i y)\widehat{\mathbf{a}}_x$, we get $E_{\pm x}^J = -\frac{1}{2}\mu_0 j_{0x}\omega/k_z^i$ where μ_0 is the magnetic permeability of vacuum. The resulting radiated field are similar to the ones radiated by the slab with spherical inclusions. However, in this case $E_{+x}^J = E_{-x}^J$, while in the case of the slab one has a right–left anisotropy, that is $E_{+x}^S \neq E_{-x}^S$, which comes from the anisotropy of Mie scattering. Furthermore, the result $E_{+x}^J = E_{-x}^J$ is a direct consequence of Faraday's law $\nabla \times \mathbf{E} = -i\omega\mathbf{B}$, that demands the continuity of E_x^J whenever B_y^J is finite. Here $\mathbf{B}$ is the magnetic field of the incident plane wave. Therefore, if one wants to find a distribution of induced currents that properly simulate the sources of the fields radiated by the slab, one is forced to conclude that this is not possible with an open current distribution. The fulfillment of Faraday's law requires a singular value of B_y at $z = 0$, as the only way to obtain a right-left anisotropy in the wave amplitudes of the radiated electric field. But the only way to get a singular value of B_y at $z = 0$ would be to have a distribution of closed currents that generate a magnetization $\mathbf{M}$ in the sheet along the y–direction. Only in this manner $B_y/\mu_0 = H_y + M_y$ can have a singular contribution. An average of closed currents running along the x–direction can be written as two surface current densities running in opposite directions These closed currents should be induced by an electric field generated by the time variations of the magnetic field along the y–direction. In a slab with spherical inclusions the closed currents can be induced at the inclusions. Let us now define the magnetization field $\mathbf{M}$ as $\mathbf{J} = \nabla \times \mathbf{M}$ where $\mathbf{J}$ is, in general, the average of the closed currents induced in the material. The magnetization in the y–direction can be written as $\mathbf{M} = m_{0y}\delta(z)\exp(ik_y^i y)\hat{\mathbf{e}}_y$, where m_{0y} is the surface magnetization. Now, one can show that the electric field radiated by this induced magnetization is also given by plane waves, as the ones in Eq. (9), but with amplitudes $E^J_{\pm x} = \pm\frac{i}{2}\omega\mu_0\, m_{0y}$, which are discontinuous at $z = 0$. This discontinuity obviously arises from the discontinuity of the closed–current distribution. However, if H_y can induce closed currents in the sheet, the same should happen with the time variations of H_z. In this case closed currents should be induced in the XY plane with a corresponding magnetization in the z–direction. Therefore in order to be consistent we should also consider the field radiated by a source like $\mathbf{M} = -m_{0z}\delta(z)\exp(ik_y^i y)\hat{\mathbf{e}}_z$. Adding up the contributions to the amplitude of the radiated field of the three sources, one gets

$$E^J_{\pm x} = \frac{1}{2}\mu_0\omega\left[-\frac{j_{0x}}{k_z^i} \pm im_{0y} + im_{0z}\frac{k_y^i}{k_z^i}\right]. \tag{10}$$

We now assume that the averages of the induced currents are proportional to the incident field through some effective response functions, and then try to find out the values for which one recovers the fields radiated by the thin slab with spherical inclusions. First we define the polarization field $\mathbf{P}$ as $\mathbf{J} = \partial\mathbf{P}/\partial\mathrm{t} \rightarrow -i\omega\mathbf{P}$,where $\mathbf{J}$ is the average of the induced current in the material. Then we define the electric susceptibility tensor $\overline{\overline{\chi}}^E$ as $\mathbf{P} =\epsilon_0\overline{\overline{\chi}}^E\cdot\mathbf{E}$, where is $\mathbf{E}$ the average electric field. In the same manner the magnetic susceptibility tensor $\overline{\overline{\chi}}^H$ is defined as $\mathbf{M} = \overline{\overline{\chi}}^H\cdot\mathbf{H}$,where $\mathbf{H}$ is the average H–field. For an object like the 2D sheet we can write $\overline{\overline{\chi}}_S^E = (\chi^E_{S\|},\chi^E_{S\|},\chi^E_{S\perp})$ and $\overline{\overline{\chi}}_S^H = (\chi^H_{S\|},\chi^H_{S\|},\chi^H_{S\perp})$, where the subindexes $\|$ and $\perp$ denote parallel and perpendicular to the sheet. The response of the 2D sheet is clearly anisotropic in the $\|$ and $\perp$ directions, but we are regarding the XY plane isotropic. Now we assume that the system is so dilute that the average induced current and magnetization distributions are proportional to the *incident* field, thus may write

$$j_{ox} = -i\omega\epsilon_0\chi^E_{S\|}E_0 \tag{11}$$

$$m_{0y} = \chi^H_{S\|}H_0\cos\theta_i = \chi^H_{S\|}\frac{k}{\omega\mu_0}E_0\cos\theta_i \tag{12}$$

$$m_{0z} = \chi^B_{S\perp}B_0\sin\theta_i = \chi^B_{S\perp}\frac{k}{\omega}E_0\sin\theta_i\,, \tag{13}$$

where we have used the relations between $\mathbf{E}$, $\mathbf{H}$ and $\mathbf{B}$ given by Maxwell's equations and we have introduced in Eq. (13) the surface response $\chi^B_{S\perp}$

to the **B** field instead of the response $\chi^H_{S\perp}$ to the **H** field. We do this because in case the magnetization is along the z–direction, the field $H_z = B_z/\mu_0 - m_{0z}\delta(z)\exp(ik^i_y y)$ is singular at $z = 0$, and it is not adequate to define a response to a singular field. On the contrary, the field B_z is continuous and can be regarded as the driving field of the induced magnetization.

Now we compare the amplitudes of the waves radiated by this sheet, characterized by three effective surface response functions, with the amplitudes of the waves radiated by the slab with spherical inclusions. In order to do this we first imagine that the effective response of the sheet is actually describing the response of a slab of a finite width d. One can regard the sheet as the ending shape of a limiting process which starts with a slab of a finite width. For example, one can define the surface susceptibility $\chi^E_{S\parallel}$ as $\chi^E_{S\parallel} = \lim_{d\to 0}\chi^E d$, where χ^E is the bulk susceptibility of a homogeneous and isotropic slab. Therefore, we have to perform, in Eqs. (11)-(13), the following replacements: $\chi^E_{S\parallel} \to \chi^E d$ and $\chi^H_{S\parallel} \to \chi^H d$, where χ^H is the bulk magnetic susceptibility of a homogeneous and isotropic slab, and $\chi^B_{S\perp} \to \chi^H d/\mu \approx \chi^H d/\mu_0$. In this last replacement we are taking into account that in the $\perp$ direction there is a surface magnetization at the two parallel faces of the slab that produces a difference between the average **B** and **H** field. This does not happen along the $\parallel$ direction because along this direction the system is boundless. Nevertheless, since we are considering here only the dilute limit, in which the driving field for the induced currents comes solely from the incident beam, we can take $\mathbf{B} \approx \mu_0\mathbf{H}$ and replace $\chi^B_{S\perp} \to \chi^H d/\mu_0$. We now substitute the replacements in Eqs. (11)–(13) into Eq. (10) and compare it with Eqs. (7) and (8) to yield

$$\chi^E + \chi^H\cos^2\theta_i + \chi^H\sin^2\theta_i = 2i\gamma S(0) \tag{14}$$

$$\chi^E - \chi^H\cos^2\theta_i + \chi^H\sin^2\theta_i = 2i\gamma S_1(\pi - 2\theta_i)\frac{\sin k^i_z d}{k^i_z d}, \tag{15}$$

If we assume $k^i_z d \ll 1$, we can approximate $\sin k^i_z d/k^i_z d \approx 1$. We now solve Eqs. (14) and (15) for χ^E and χ^H and use the definitions of the electrical permittivity $\tilde{\epsilon} \equiv \epsilon/\epsilon_0 = 1 + \chi^E$ and the magnetic permeability $\tilde{\mu} \equiv \mu/\mu_0 = 1 + \chi^H$ to get

$$\tilde{\mu}^{TE}_{eff}(\theta_i) = 1 + i\gamma\frac{S^{(1)}_-(\theta_i)}{\cos^2\theta_i} \tag{16}$$

$$\tilde{\epsilon}^{TE}_{eff}(\theta_i) = 1 + i\gamma\left[2S^{(1)}_+(\theta_i) - S^{(1)}_-(\theta_i)\tan^2(\theta_i)\right], \tag{17}$$

where $S_+^{(m)}(\theta_i) \equiv \frac{1}{2}[S(0) + S_m(\pi - 2\theta_i)]$ and $S_-^{(m)}(\theta_i) \equiv S(0) - S_m(\pi - 2\theta_i)$, and we have added, to $\tilde{\epsilon}$ and $\tilde{\mu}$, the superindex TE to denote the polarization and the subindex eff to emphasize the fact that they describe an effective response. In the case of TM polarization one performs an analogous procedure as the one developed above for TE polarization, and one can show that the corresponding optical coefficients are given by

$$\tilde{\epsilon}_{eff}^{TM}(\theta_i) = 1 + i\gamma \frac{S_-^{(2)}(\theta_i)}{\cos^2\theta_i} \tag{18}$$

$$\tilde{\mu}_{eff}^{TM}(\theta_i) = 1 + i\gamma \left[2S_+^{(2)}(\theta_i) - S_-^{(2)}(\theta_i)\tan^2(\theta_i)\right]. \tag{19}$$

These results can also be readily obtained from the symmetry relations in Maxwell's equations.

Note that the effective optical coefficients $\tilde{\epsilon}_{eff}$ and $\tilde{\mu}_{eff}$ depend on the angle of incidence and on the polarization, just like in an anisotropic medium. Also, the expressions for the effective optical coefficients in Eqs. (16)-(19) are linear in $\gamma \equiv 3f/2x^3$ and they are valid only to linear order in γ. This is consistent with the dilute–limit approximation adopted above, and therefore the validity of all of our results will be limited by this restriction.

According to continuum electrodynamics the effective index of refraction n_{eff} should be given by

$$n_{eff}^{(m)}(\theta_i) = \sqrt{\tilde{\epsilon}_{eff}^{(m)}(\theta_i)\tilde{\mu}_{eff}^{(m)}(\theta_i)} \simeq \sqrt{1 + 2i\gamma S(0)}\,, \tag{20}$$

where we dropped terms of second order in γ since our approximations are valid up to first order in γ only. This result is the same as the one derived by Foldy [16] long time ago We may expand the square root and to lowest order in γ we get $n_{eff} \approx 1 + i\gamma S(0)$ which is isotropic and independent of polarization, and is actually the same result as the one proposed by van de Hulst [3]. So we can see that although the optical coefficients $\tilde{\epsilon}_{eff}$ and $\tilde{\mu}_{eff}$ are highly anisotropic and polarization dependent, their dependence in the angle of incidence is such that the square root of their product is not.

Let us now look at some limiting cases. First we notice that for small particles ($x \ll 1$) the Mie forward–backward anisotropy in the angular distribution of scattered radiation becomes $S_1(\theta_i) \approx -ix^3\beta$ and $S_2(\theta_i) \approx -ix^3\beta\cos\theta_i$, where $\beta = (\tilde{\epsilon}_S - 1)/(\tilde{\epsilon}_S + 2)$ and $\tilde{\epsilon}_S = \epsilon_S/\epsilon_0$ is the electrical permittivity of the spheres. Then $S_+^{(1)} \approx -ix^3\beta$, $S_-^{(1)} \approx 0$, $S_+^{(2)} \approx -ix^3\beta\sin^2\theta_i$, and $S_-^{(2)} \approx -2ix^3\beta\cos^2\theta_i$. Substituting these values into Eqs. (16)-(19) we get

$$\tilde{\mu}^{TE}_{eff}(\theta_i) = \tilde{\mu}^{TM}_{eff}(\theta_i) \equiv \tilde{\mu}_{eff} = 1 \quad (21)$$

$$\tilde{\epsilon}^{TE}_{eff}(\theta_i) = \tilde{\epsilon}^{TM}_{eff}(\theta_i) \equiv \tilde{\epsilon}_{eff} = 1 + 3\beta f\,, \quad (22)$$

and these are the well-known results for the case of small particles, or for the case of an ordinary material, when one regards the material as a composite made of molecular inclusions in vacuum. Eq. (21) tells us that the system is non magnetic and Eq. (22) is the low–density limit of the effective dielectric response in Maxwell–Garnett theory or in the Clausius–Mossotti relation, when one interprets β as proportional to the molecular polarizability. One can also see that the magnetic character of the system appears only when the spheres are big enough and is related to the large forward–backward anisotropy in the Mie scattering of large particles ($x \sim 1$).

For normal incidence ($\theta_i = 0$) one gets

$$\tilde{\mu}^{TE}_{eff}(0) = \tilde{\mu}^{TM}_{eff}(0) \equiv \tilde{\mu}_{eff}(0) = 1 + i\gamma\,[S(0) - S_1(\pi)] \quad (23)$$

$$\tilde{\epsilon}^{TE}_{eff}(0) = \tilde{\epsilon}^{TM}_{eff}(0) \equiv \tilde{\epsilon}_{eff}(0) = 1 + i\gamma\,[S(0) + S_1(\pi)]\,, \quad (24)$$

where we have used $S_1(\pi) = -S_2(\pi)$. And these are the results proposed by C. Bohren [6] when he introduced the idea of a magnetic response in the optical properties of granular materials made with non–magnetic components.

At grazing incidence, $\theta_i \to \pi/2$, we have that $S_m(\pi - 2\theta_i) \to S(0)$, thus $S^{(m)}_{+}(\theta_i) \to S(0)$ and $S^{(m)}_{-}(\theta_i) \to 0$ and $S^{(m)}_{-}(\theta_i)/\cos^2\theta_i$ remains finite. We can see this by expanding $S^{(m)}_{-}(\theta_i)$ around $\theta_i = \pi/2$ and showing that $\lim_{\theta_i \to \pi/2} S^{(m)}_{-}(\theta_i)/\cos^2\theta_i = 2S''_m(0)$, where the primes indicate derivative with respect to the argument.

If we now accept the description of the optical properties of a granular material in terms of the effective optical coefficients given by Eqs. (16)-(19), the reflection amplitudes of a half space r_{hs} will be given by the Fresnel's relations of continuum electrodynamics, that is,

$$r^{TE}_{hs} = \frac{\tilde{\mu}^{TE}_{eff}(\theta_i)k^i_z - k^{eff}_z}{\tilde{\mu}^{TE}_{eff}(\theta_i)k^i_z + k^{eff}_z} \quad \text{and} \quad r^{TM}_{hs} = \frac{\tilde{\epsilon}^{TM}_{eff}(\theta_i)k^i_z - k^{eff}_z}{\tilde{\epsilon}^{TM}_{eff}(\theta_i)k^i_z + k^{eff}_z}\,, \quad (25)$$

where $k^{eff}_z = k\sqrt{(n^{eff})^2 - \sin^2\theta_i}$, and $n^{eff} = 1 + i\gamma S(0)$. One can see that these reflection amplitudes look very different from the ones we would have used by assuming a non-magnetic effective medium with $\tilde{\epsilon}_{eff} = n^2_{eff} = [1 + i\gamma S(0)]^2$ and $\tilde{\mu}_{eff} = 1$. We will denote the reflection coefficients calculated by r^{TE}_{n-m} and r^{TM}_{n-m} where the subscript n-m stands for *non–magnetic.*

Now, it is also possible to derive the coherent reflectance from the scattering approach by considering a semi–infinite pile of slabs with spherical particles and solving the multiple scattering of waves between slabs. The result from this approach can be shown to be consistent with Eq. (25) [14]. Extending the above results to a composite matrix consisting of spherical inclusions embedded in a homogeneous matrix is not difficult and is also discussed in Ref. [14].

4. NUMERICAL RESULTS

Now, we illustrate the behavior of the optical coefficients in a few examples by numerical calculations. We plot the normalized change in the optical coefficients (real and imaginary parts). By *normalized* we mean divided by the fractional volume occupied by the spheres f and by *change* in the optical coefficient we mean the difference with respect to the optical coefficient without the particles (vacuum, in this case). In all cases we assume that the particles are in vacuum (no matrix) and we should recall that the expressions used are valid for dilute systems only ($f \ll 1$). The scattering matrix elements S, involved in the formulas for the optical coefficients were calculated following the recipe given in the book by Bohren and Hoffman [15].

First, in Fig. 2, we show the effective index of refraction for a system of non–magnetic lossless glass spheres ($n_p = 1.50$) as a function of the particle radius divided by the wavelength, and similar plots for lossy spheres with increasing imaginary component of the refractive index. As it can be appreciated, even if the spheres are lossless the effective index of refraction has an imaginary component. This is entirely due to scattering losses and has a maximum near $a/\lambda \simeq 0.5$. For curves corresponding to lossy particles ($\Im(n_p) \neq 0$) the loss is due to both, absorption in the particles, and scattering from the particles. Also, note that the real part of the effective index of refraction can be less than one for some particle radius. As it may be expected, the imaginary part of the effective index of refraction reach the highest value for most absorbing particles. However, also the real part of the effective refractive index is higher than for the other curves. The reason is that the scattering efficiency is stronger for these particles, since the contrast of the particles with respect to vacuum is highest.

In Fig. 3 we plot the real and imaginary part of the normalized change in the effective optical coefficients, $\epsilon^{TE}_{eff}, \epsilon^{TM}_{eff}, \mu^{TE}_{eff}$, and μ^{TM}_{eff} as a function of the particle radius divided by the wavelength for a system of

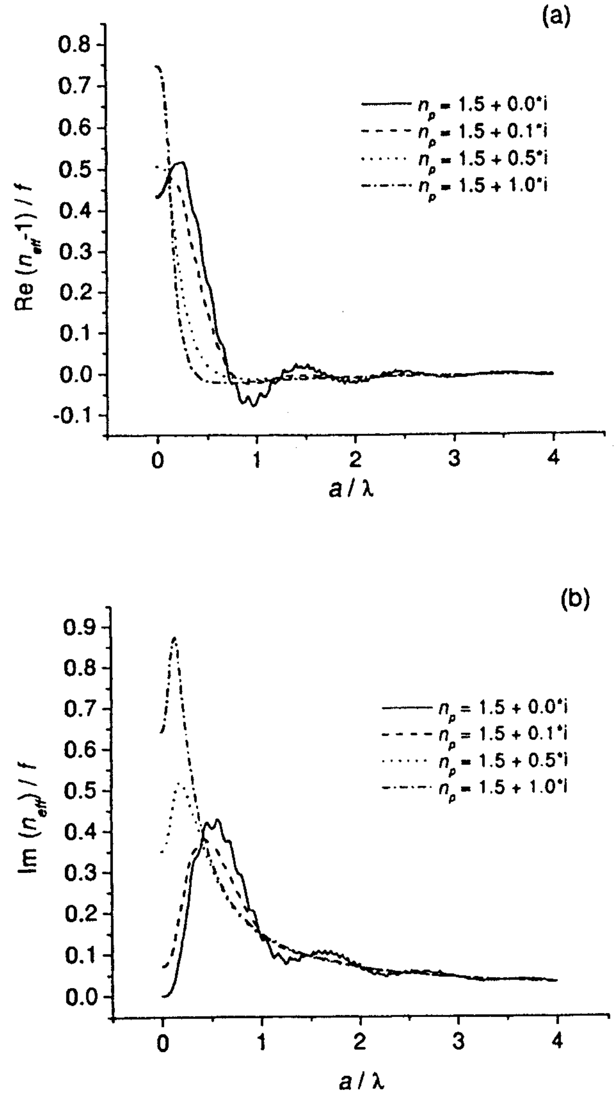

Figure 2. Plots of the normalized change in the real and imaginary part of the effective index of refraction for a system of non-magnetic glass spheres ($n_p = 1.50$) in vacuum, and similar plots for particles with different values of the imaginary part of their index of refraction.

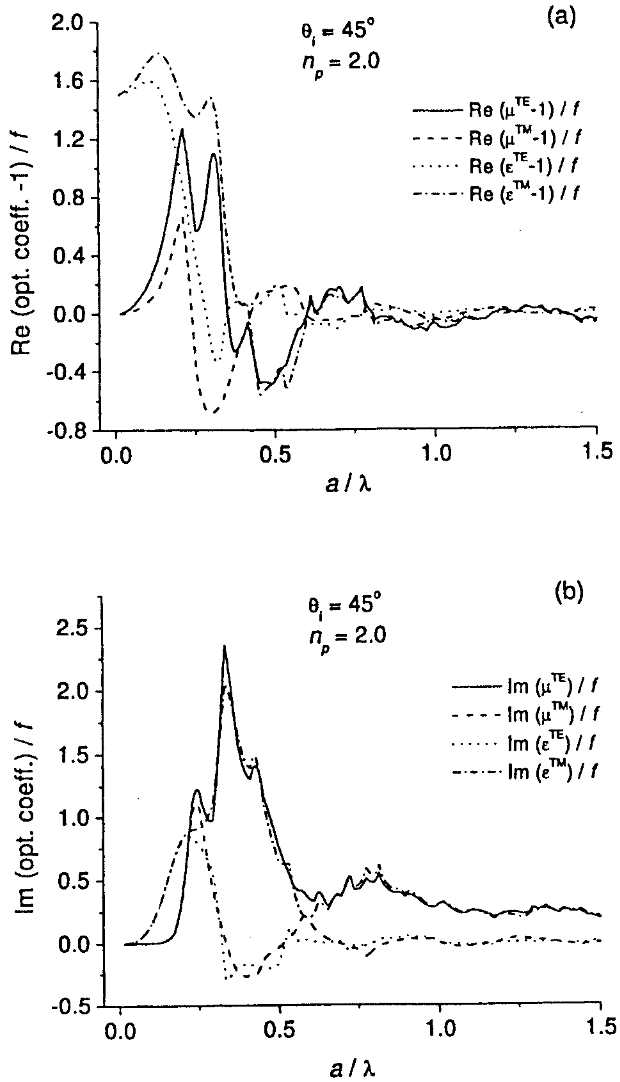

Figure 3. Plots of the normalized change in the real and imaginary part of the optical coefficients as a function of the particle radius a divided by the wavelength λ, for an angle of incidence of 45°. The plots are for a system of non-magnetic dielectric spheres ($n_p = 2.00$). The subindex in the optical coefficients *eff* was removed here for clarity; dot–dot lines are for ϵ^{TE}, dash–dot lines for ϵ^{TM}, solid lines for μ^{TE}, and dash–dash lines for μ^{TM}.

dielectric spheres with refractive index of $n_p = 2.0$. and for an angle of incidence of $\theta_i = 45°$. As it can be appreciated, these are irregular oscillatory function of the particle radius. Also, the effective magnetic permeability reaches values comparable to the effective electric permittivity for particles of radius $a/\lambda \sim 0.25$ and larger. In Fig. 3b it can be seen that the imaginary parts of ϵ_{eff}^{TE} and μ_{eff}^{TM} are negative within some range of particle radius. This, however, is not an inconsistency since the sums $\Im\epsilon_{eff}^{TE} + \Im\mu_{eff}^{TE}$, and $\Im\epsilon_{eff}^{TM} + \Im\mu_{eff}^{TM}$, remain always positive. In similar plots, but for fixed particle radius and as a function of the angle of incidence (not shown here) one finds that the change in the optical coefficients generally increases towards grazing incidence.

Since the appearance of the effective magnetic susceptibility for systems of non–magnetic particles is apparently due to the induced closed currents within the particles, it is interesting to compare the magnetic response of systems of dielectric particles with that of metallic particles. It turns out that the effective magnetic response for systems of particles of the same radius is in general similar in magnitude for dielectric and metallic particles; except when the particle radius is small compared to the wavelength. For small particles (say, $a \lesssim 0.1\lambda$) the imaginary component of the effective magnetic permeability is orders of magnitude larger for metallic particles than for dielectric ones. Although its value is small in absolute terms. Also, the change in the real part of μ_{eff} is negative for metallic particles and positive for dielectric particles. In Fig. 4 we plot the real and imaginary parts of the normalized change in μ_{eff} for both polarizations as a function of the angle of incidence for metallic particles (copper, $n_p = 0.21 + 4.05i$ at $\lambda = 0.69\,\mu$m) and dielectric (glass, $n_p = 1.5$) ones, and for particles radius of $a/\lambda = 0.1$.

Finally, with respect to the coherent reflectance, we have found that this is generally smaller than what would be predicted by a simpler model which ignores the effective magnetic response. In Fig. 5 we show the reflectance for TE and TM polarizations as a function of the angle of incidence for particles with $n_p = 2.50$ and two different radius: $a/\lambda = 0.1$ and $a/\lambda = 0.5$. The fractional volume of the particles is taken to be $f = 0.1$. In Fig. 5b the location of the Brewster angle can be appreciated and it can be seen that the location of the Brewster angle predicted by the non–magnetic model differs from our result. The curves for TM polarization and for particles $a/\lambda = 0.1$ are an exception and the reflectance predicted by the non–magnetic formula is lower than the coherent reflectance R. In plots of the TE reflectance for larger particles (not shown here) one finds zeros in the coherent reflectance, and these can be interpreted as a Brewster angle, which only exist when the medium has a magnetic permeability different from that of vacuum.

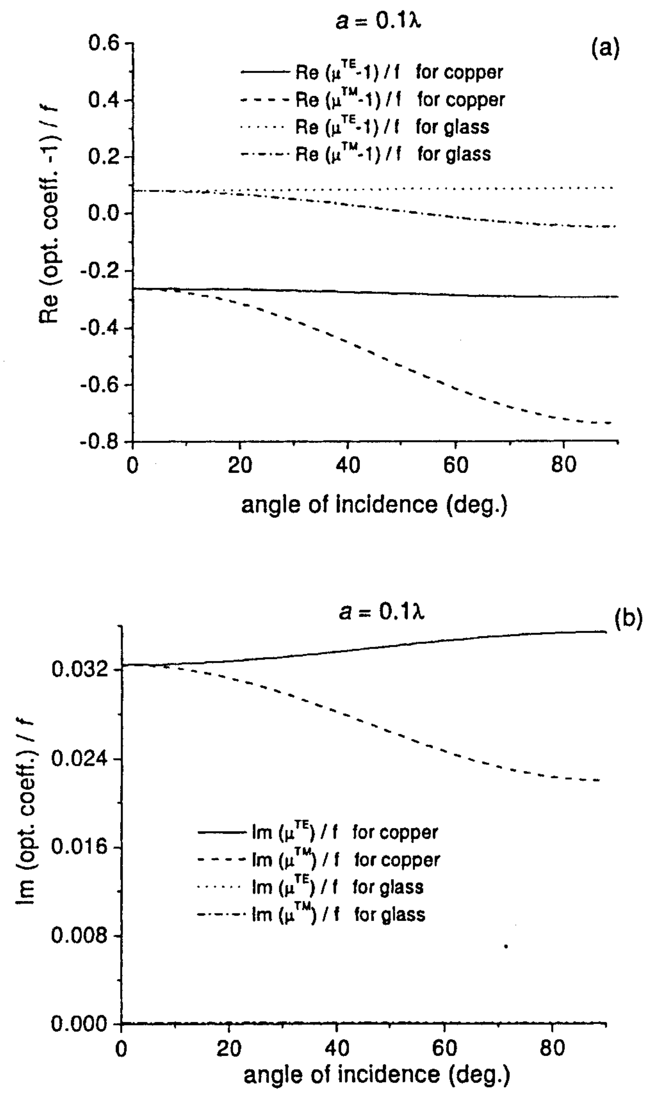

Figure 4. Plots of the normalized change in the (a) real and (b) imaginary part of the effective magnetic permeability as a function of the angle of incidence for metallic (copper) and glass particles of radius $a = 0.1\lambda$. Plots for both polarizations are shown. The subindex in the optical coefficients *eff* was removed here for clarity; dot–dot lines are for μ^{TE} and dash–dot lines for μ^{TM}, both for copper particles. Solid lines for μ^{TE} and dash–dash lines are for μ^{TM}, both for glass particles.

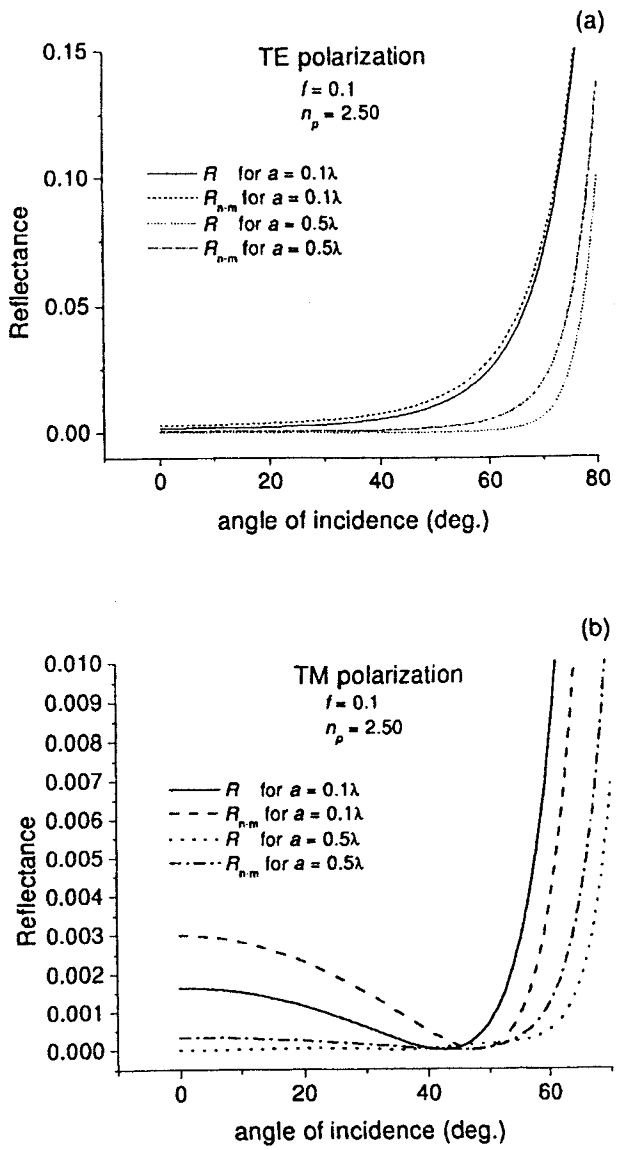

Figure 5. Plot of the coherent reflectance R of polarized light for a system of non-magnetic dielectric spheres of index of refraction $n_p = 2.50$ and a filling fraction of $f = 0.1$; (a) TE polarization and (B) TM polarization. For comparison we also plot the reflectance ignoring the effective magnetic susceptibility ($R_{\text{n-m}}$).

5. CONCLUSIONS

We have constructed an effective medium theory to describe the coherent reflection of electromagnetic waves from a random system of spheres. Our results can be regarded as an extension of ideas put forth previously by C. Bohren. We found that the effective medium must posses an effective magnetic permeability, even if the particles are non–magnetic, in order to have a theory consistent with continuum electrodynamics. The effective magnetic susceptibility becomes comparable to the effective electric permittivity as the particle radius increases and they attain their maximum value when the radius is comparable to the wavelength of the incident radiation. The origin of this magnetic effect appears in our theory from the identification of induced closed currents as sources of the fields radiated by the random system of spheres. These closed currents must be physically present within each sphere and when averaged they must act as the source of an effective magnetization. The coherent reflectance calculated including the effective magnetic response differs appreciably from the one calculated without it. The formulas put forth in this work are valid for a dilute system of spheres and they can be readily used in applications satisfying this criteria. Extensions of the present results to a polydispersed system of spheres is straightforward.

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