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Electromagnetic response of a random half-space of Mie scatterers within the effective-field approximation and the determination of the effective optical coefficients

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Abstract

We calculate the coherent reflectance of light from a half-space with randomly located spherical particles using the effective field approximation to solve the integral equation obeyed by the scattered fields. We obtain the effective refractive index and the coherent reflection coefficient to first order in the density of particles. Then we use basic principles of continuum electrodynamics to derive expressions for the effective permittivity and permeability tensors of the half-space. The results are not restricted to particles of radius small compared to the wavelength of the incident radiation. It is shown that an effective magnetic permeability arises, even in the case when the particles are non-magnetic. The present work provides a standard framework to pursue higher order approximations for denser media.

Keywords: Effective medium theory; Coherent reflection; Random media; Particle scattering

1. Introduction

The interaction of light with a random system of particles is of interest in many areas of science and engineering. Although many efforts have been devoted to the development of this subject many open questions still remain, due in part to the rather involved mathematical procedures that are usually required, and also to the difficulties in establishing the validity of the different types of approximations. Here we are interested in the dynamical description of the average electromagnetic

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field in the presence of a random system of polarizable particles and its relationship to the physical foundations of effective-medium theories. When the particles are small compared to the wavelength of the incident radiation, the optical properties of the system can be described in terms of the effective optical coefficients of an artificial, homogeneous medium, commonly known as: effective medium. Well known and rather popular examples of effective media are the ones related to the mixing formulas of Maxwell Garnett [1] and Bruggeman [2], which yield the effective dielectric function of a two-component composite material in terms of the filling fractions and dielectric functions of its components. It is now accepted that the electromagnetic properties of the effective medium depend on the specific microstructure of the composite. For example, the formulas of Maxwell Garnett yield a reasonable description of materials with a granular topology while the ones of Bruggeman are more adequate to composite materials with intermixed components, also called aggregate topology. In relation to the extent of applicability of effective medium theories, Ruppin, in a recently appeared critical review on the subject [3], calls *unrestricted* theories to the ones in which the proposed *effective* dielectric or magnetic responses can be used in continuum electrodynamics, exactly in the same way as one uses the dielectric function or the magnetic susceptibility of a common homogeneous material, and he reserves the name *restricted* to theories in which this is not the case.

In the last decades, the extension of the effective-medium approach to systems with inclusions of a larger size has attracted the interest and attention of the material- and atmospheric-science communities, and it has also posed some very fundamental questions. For example, in 1978 Stroud and Pan [4] and later on Wachniewski and McClung [5], extended Bruggeman's ideas to the dynamic response of a two-component composite, and they obtain expressions for the effective dielectric response of the composite material in terms of the scattering properties of isolated inclusions. With this approach they were able to include the contribution of the induced magnetic dipoles in the calculation of the electric dipolar polarizability and to determine the additional absorption coming from the induced eddy currents. In 1983 Chýlek and Srivastava [6] and later on Doyle [7] proposed a size-dependent generalization of Maxwell–Garnett theory, where again, the dependence on the grain size arises from the contribution of the induced magnetic dipoles to the electric dipolar polarizability. Grimes and Grimes [8] also showed that in a grain topology the effective dielectric and magnetic responses were not independent of each other, but that they were actually interrelated. For example, in a system consisting of a collection of non-magnetic, polarizable spherical inclusions, one obtains an actual effective magnetic susceptibility slightly different from the one of vacuum. More recently, there has been some efforts to extend the effective-medium theories to the case of composite particles [9,10].

One has to realize, however, that all these efforts to extend the effective-medium approach to larger size inclusions rely on an adequate determination and interpretation of the dipolar electric and magnetic responses of isolated scatterers, yielding interesting effects in the effective electromagnetic response of the composite system, due to the finite size of the inclusions. Nevertheless, the validity of the dipolar approximation in the treatment of the scattering process restricts the application of all these extended effective medium theories to the case in which the size of the inclusions is still very small in comparison to the wavelength of the incident radiation.

In 1986 Bohren [11] calculated the average of the fields reflected and transmitted by a plane slab with large spherical, non-magnetic, polarizable inclusions driven by an incident plane wave at normal incidence. By large we mean here a size comparable to the wavelength of the incident radiation. He found that in order to interpret the reflection and transmission amplitudes of the slab in terms of an effective medium and continuum electrodynamics, two different effective indices of refraction were required: one for reflection and another one for transmission. Instead of accepting this awkward situation he noted that continuum electrodynamics could still be used if one accepts that besides an effective dielectric response, the effective medium possesses also an effective magnetic response even if neither the particles nor the surrounding medium were magnetic. He provided expressions for the effective dielectric and magnetic responses in terms of the scattering properties of isolated spheres, but the physical origin of the magnetic response was never clarified and it was later explicitly disputed as, for example, in Ref. [10]. Bohren concluded in his paper [11] that the concept of an effective medium was not strictly adequate for systems with large inclusions, pointing out that it was not even clear if this concept was plausible for cases of either off-normal incidence or a non-plane geometry.

First of all, it is important to recall that in a system with large spherical inclusions the incident plane wave gets scattered in all directions by each of the inclusions. The total scattered field can be decomposed in two components: an average plus a fluctuating component, the average component is also known as the coherent field while the fluctuating one as the diffuse field. It is pertinent to remark that the concept of an effective medium is related only to the behavior of the coherent field, leaving out the contribution of the diffuse field, and is thus a partial view of the whole physical problem. Thus, for example, one cannot calculate the total power absorption in the system without looking to both, the coherent and diffuse components. In the case of systems with large inclusions the power carried by the diffuse field can be as large as the one carried by the coherent field, while for systems with small inclusions the power carried by the diffuse field is so small it can be neglected, as it is usually done in the standard treatments of continuum electrodynamics.

In this paper we deal with the behavior of an electromagnetic wave in the presence of a half-space of a dilute random system of identical, polarizable, spherical particles in vacuum. We use a standard integral-equation formalism to determine the scattered field, and then a configurational average is performed. In the integral-equation formalism the field scattered by an incident wave from a system of N randomly located particles is given in terms of an integral equation, where the unknowns are the exciting fields within each of the N particles. But the average of the exciting fields at each particle are coupled to each other through a hierarchy of equations involving higher and higher orders of statistical correlations among the particles. Truncation at the first stage in this hierarchy of equations is known as the effective-field approximation (EFA) (see e.g., [12,13]). Basically, the EFA assumes that the exciting field within each particle can be approximated by the average of the total field (coherent field) itself, and the integral equation yields a self-consistent condition for its amplitude and phase. The phase can be interpreted in terms of an effective propagation wavevector and the amplitude can be related to the reflection amplitude of the half space. This approximation is valid for systems with a dilute concentration of inclusions (dilute systems).

Here we apply the EFA to the half-space of a random collection of spheres, and we determine the effective propagation wavevector of the coherent field that travels within, as well as its reflection amplitude from the interface. These results are valuable by themselves and have no direct relationship to the concept of an effective medium. Furthermore, our results go beyond the extended effective medium theories mentioned above, in the sense that they are applicable to systems with actually large inclusions. They also go beyond the calculations of the reflection and transmission amplitudes performed by Bohren, because here we also consider oblique angles of incidence. They are, however, restricted to dilute systems and to a planar interface. Nevertheless, the present approach is the first step in a systematic procedure to construct better approximations, as it has been already delineated in the book of Tsang and Kong [14].

Now, if one would like to interpret our results in terms of the electromagnetic response of an effective medium, the simplest thing would be to associate an effective index of refraction n_{eff} to the effective propagation wavevector and to regard this index of refraction n_{eff} as the one corresponding to the equivalent effective medium. Nevertheless, internal consistency with continuum electrodynamics would require that the reflection amplitude calculated within the EFA should be consistent with the one given by Fresnel's relations together with n_{eff}. But Fresnel's relations require not only on an effective index of refraction, they also require, depending on the incident polarization, either an effective dielectric function ε_{eff} or an effective magnetic susceptibility μ_{eff}. If one now takes, in Fresnel's relations, the effective magnetic susceptibility equal to μ_0 the one in vacuum (non-magnetic), the reflection amplitude so obtained is not consistent with the one calculated using the EFA. One possible explanation of this inconsistency would be, following Bohren, that the effective medium should have a magnetic susceptibility different from the one in vacuum (magnetic). But in order to calculate either the effective dielectric or magnetic responses of the composite system, one should find the effective currents induced in the system and then find their relationship with the average of the total field. We have already done this [15] and have found expressions for both, the effective dielectric and magnetic responses in terms of the scattering properties of the spherical inclusions. Our expressions, besides being related to a planar interface, depend on both, the angle of incidence and the polarization of the incident beam, and they reduce to the ones given by Bohren in the case of normal incidence. In this sense, they do not represent *unrestricted* effective optical constants of the composite system, nevertheless, they presumedly describe correctly, in the context of continuum electrodynamics, the reflection and transmission amplitudes of the coherent beam in a system with large polarizable inclusions. Furthermore, we have also shown [15] that the magnetic response in this system is a true magnetic response due to closed currents induced within the inclusions by the time variations of the magnetic field. As a final remark, let us comment that although both the dielectric and the magnetic responses depend on the angle of incidence, their product, and consequently the effective index of refraction $n_{eff} = \sqrt{\varepsilon_{eff}\,\mu_{eff}}$, does not, at least to lowest order in the density of the inclusions.

In this paper we use a less intuitive and a rather more formal approach, based on the EFA, to obtain the same expressions, as in Ref. [15], for the effective dielectric and magnetic responses of the half-space of randomly located polarizable spheres. As mentioned above, the EFA can provide the effective index of refraction n_{eff} and the reflection amplitude, but it does not provide the effective dielectric or magnetic responses, separately, thus in order to obtain them one has to rely in some other idea. What we do here is to use the boundary conditions for the coherent field at a planar interface. Since the boundary conditions relate the dielectric and magnetic responses at each side of the interface, we then use them and we are then able to obtain explicit expressions for ε_{eff} and μ_{eff}. Finally, we propose some specific reflectance experiments that can provide evidence about the magnetic response of the system at optical frequencies.

2. Effective-field approximation

Consider an ensemble of spherical particles located at random in the half-space $z > 0$ surrounded by vacuum, as shown in Fig. 1. The position of each particle is specified by the coordinates of its

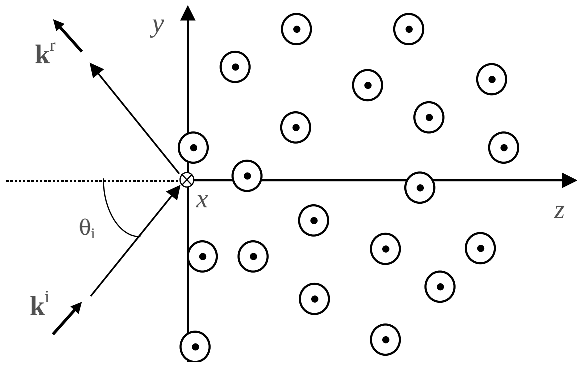

Fig. 1. Geometry of the problem. The plane of incidence is the yz-plane and the particles are located at random in the half-space $z > 0$. This means that the coordinates of the center of all particles have positive values of z.

center. Let us assume a plane wave is incident on the half-space at an angle $\theta_{\rm i}$, being the plane of incidence the y–z plane. The electric field is,

$$\mathbf{E}^{\rm i} = E_0 \exp(\mathrm{i}\mathbf{k}^{\rm i}\cdot\mathbf{r})\hat{\mathbf{e}}_{\rm i}, \tag{1}$$

where $\mathbf{k}^{\rm i} = k_y^{\rm i}\hat{\mathbf{a}}_y + k_z^{\rm i}\hat{\mathbf{a}}_z$ and $\hat{\mathbf{e}}_{\rm i} = \hat{\mathbf{a}}_x$ or $\hat{\mathbf{e}}_{\rm i} = \cos\theta_{\rm i}\hat{\mathbf{a}}_y - \sin\theta_{\rm i}\hat{\mathbf{a}}_z$ for TE or TM polarization, respectively. The electric field satisfies $\hat{\mathbf{e}}_{\rm i}\cdot\mathbf{k}^{\rm i} = 0$, and $|\mathbf{k}^{\rm i}| = k$, where $k = \omega/c = 2\pi/\lambda$ is the wavenumber in vacuum, λ is the corresponding wavelength and c is the speed of light. We will be using 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. The total scattered field is given by the sum of the fields scattered by each of the particles. Therefore, the scattered field $\mathbf{E}^{\rm S}$ due to a collection of N spherical particles with their centers located at $\{\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_{\rm p}, \ldots, \mathbf{r}_N\}$ can be written as [16],

$$\mathbf{E}^{\rm S}(\mathbf{r}) = \sum_{p=1}^{N} \int \mathrm{d}^3 r' \int \mathrm{d}^3 r'' \overleftrightarrow{G}_0(\mathbf{r},\mathbf{r}') \cdot \overleftrightarrow{T}(\mathbf{r}' - \mathbf{r}_{\rm p}, \mathbf{r}'' - \mathbf{r}_{\rm p}) \cdot \mathbf{E}_{\rm p}^{\rm E}(\mathbf{r}''), \tag{2}$$

where $\overleftrightarrow{G}_0(\mathbf{r},\mathbf{r}')$ is the dyadic Green's function in free space, $\overleftrightarrow{T}(\mathbf{r}',\mathbf{r}'')$ is the transition operator for a sphere, and $\mathbf{E}_{\rm p}^{\rm E}$ 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 $\mathbf{E}_{\rm p}^{\rm E}$ depends parametrically on the location of the rest $N-1$ particles. To deal with a half-space geometry it is convenient to work in a plane-wave representation, thus we substitute the plane-wave expansion of the dyadic Green's function

$$\overleftrightarrow{G}_0(\mathbf{r},\mathbf{r}') = \frac{\mathrm{i}}{8\pi^2} \int\int \mathrm{d}k_x^{\rm s}\,\mathrm{d}k_y^{\rm s} \frac{1}{k_z^{\rm s}} (\overleftrightarrow{\mathbf{1}} - \hat{\mathbf{k}}_{\pm}^{\rm s}\hat{\mathbf{k}}_{\pm}^{\rm s}) \exp[\mathrm{i}\,\mathbf{k}_{\pm}^{\rm s}\cdot(\mathbf{r}-\mathbf{r}')], \tag{3}$$

valid in the region outside the particle ($r > r'$), the momentum representation of the transition operator

$$\overleftrightarrow{T}(\mathbf{r}' - \mathbf{r}_\mathrm{p}, \mathbf{r}'' - \mathbf{r}_\mathrm{p}) = \frac{1}{(2\pi)^6} \int \mathrm{d}^3 p' \int \mathrm{d}^3 p'' \exp[\mathrm{i}\mathbf{p}' \cdot (\mathbf{r}' - \mathbf{r}_\mathrm{p})] \overleftrightarrow{T}(\mathbf{p}', \mathbf{p}'') \times \exp[-\mathrm{i}\mathbf{p}'' \cdot (\mathbf{r}'' - \mathbf{r}_\mathrm{p})] \tag{4}$$

and the plane-wave expansion of the exciting field

$$\mathbf{E}_\mathrm{p}^\mathrm{E}(\mathbf{r}'') = \frac{1}{(2\pi)^3} \int \mathrm{d}^3 k^\mathrm{E} \exp(\mathrm{i}\, \mathbf{k}^\mathrm{E} \cdot \mathbf{r}'') \mathbf{E}_\mathrm{p}^\mathrm{E}(\mathbf{k}^\mathrm{E}) \tag{5}$$

into Eq. (2), to get

$$\mathbf{E}^\mathrm{S}(\mathbf{r}) = \frac{\mathrm{i}}{8\pi^2} \frac{1}{(2\pi)^3} \sum_{p=1}^{N} \int \mathrm{d}^3 k^\mathrm{E} \int\int \mathrm{d}k_x^\mathrm{s}\, \mathrm{d}k_y^\mathrm{s} \frac{(\overleftrightarrow{\mathbf{1}} - \hat{\mathbf{k}}_\pm^\mathrm{s} \hat{\mathbf{k}}_\pm^\mathrm{s})}{k_z^\mathrm{s}} \cdot \overleftrightarrow{T}(\mathbf{k}_\pm^\mathrm{s}, \mathbf{k}^\mathrm{E}) \cdot \mathbf{E}_\mathrm{p}^\mathrm{E}(\mathbf{k}^\mathrm{E}) \exp[-\mathrm{i}(\mathbf{k}_\pm^\mathrm{s} - \mathbf{k}^\mathrm{E}) \cdot \mathbf{r}_\mathrm{p}] \exp(\mathrm{i}\mathbf{k}_\pm^\mathrm{s} \cdot \mathbf{r}). \tag{6}$$

Here $\mathbf{k}_\pm^\mathrm{s} = k_x^\mathrm{s} \hat{\mathbf{a}}_x + k_y^\mathrm{s} \hat{\mathbf{a}}_y \pm k_z^\mathrm{s} \hat{\mathbf{a}}_z$, $k_z^\mathrm{s} = \sqrt{k^2 - (k_x^\mathrm{s})^2 - (k_y^\mathrm{s})^2}$, $\overleftrightarrow{T}(\mathbf{p}', \mathbf{p}'')$ is the momentum representation of the transition operator $\overleftrightarrow{T}(\mathbf{r}', \mathbf{r}'')$ of an isolated sphere, and $\mathbf{E}_\mathrm{p}^\mathrm{E}(\mathbf{k}^\mathrm{E})$ is the Fourier component of $\mathbf{E}_\mathrm{p}^\mathrm{E}(\mathbf{r}'')$ with wavevector $\mathbf{k}^\mathrm{E}$. This is the plane-wave expansion of the scattered field, this means that the scattered field is expressed as a sum of plane waves propagating along the $\mathbf{k}_\pm^\mathrm{s}$ directions, the signs $\pm$ refer to the field propagating to the right ($+$) and to the left ($-$) of each particle. The factor $\exp[-\mathrm{i}(\mathbf{k}_\pm^\mathrm{s} - \mathbf{k}^\mathrm{E}) \cdot \mathbf{r}_\mathrm{p}]$ keeps track of the phase difference of the field scattered by different particles. Let us recall that both the amplitude $\mathbf{E}_\mathrm{p}^\mathrm{E}(\mathbf{k}^\mathrm{E})$ and the phase $\mathbf{k}^\mathrm{E}$ are functions of the location of the particles $\{\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N\}$.

The total field within the system is the sum of the incident field, the scattered fields. Its configurational average can be written as

$$\langle \mathbf{E}^\mathrm{T}(\mathbf{r}) \rangle = \mathbf{E}^\mathrm{i}(\mathbf{r}) + \langle \mathbf{E}^\mathrm{S}(\mathbf{r}) \rangle. \tag{7}$$

We now calculate $\langle \mathbf{E}^\mathrm{S}(\mathbf{r}) \rangle$, that is

$$\langle \mathbf{E}^\mathrm{S}(\mathbf{r}) \rangle = \frac{\mathrm{i}}{8\pi^2} \frac{1}{(2\pi)^3} \int\int \mathrm{d}k_x^\mathrm{s}\, \mathrm{d}k_y^\mathrm{s} \frac{(\overleftrightarrow{\mathbf{1}} - \hat{\mathbf{k}}_\pm^\mathrm{s} \hat{\mathbf{k}}_\pm^\mathrm{s})}{k_z^\mathrm{s}} \cdot \left\langle \sum_{p=1}^{N} \int \mathrm{d}^3 k^\mathrm{E} \overleftrightarrow{T}(\mathbf{k}_\pm^\mathrm{s}, \mathbf{k}^\mathrm{E}) \cdot \mathbf{E}_\mathrm{p}^\mathrm{E}(\mathbf{k}^\mathrm{E}) \exp[-\mathrm{i}(\mathbf{k}_\pm^\mathrm{s} - \mathbf{k}^\mathrm{E}) \cdot \mathbf{r}_\mathrm{p}] \right\rangle \exp(\mathrm{i}\mathbf{k}_\pm^\mathrm{s} \cdot \mathbf{r}). \tag{8}$$

The main problem here is the dependence of $\mathbf{E}_\mathrm{p}^\mathrm{E}(\mathbf{k}^\mathrm{E})$ and $\mathbf{k}^\mathrm{E}$ on the location of the particles $\{\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N\}$. To handle this problem we now assume that the exciting field can be approximated by the average field $\langle \mathbf{E}^\mathrm{T}(\mathbf{r}) \rangle$, something like a mean-field approximation, and furthermore, that it is given by

$$\mathbf{E}^{\mathrm{E}}_{\mathrm{p}}(\mathbf{k}^{\mathrm{E}}) = (2\pi)^3 \hat{\mathbf{e}}_{eff}\, tE_0 \delta(\mathbf{k}^{\mathrm{E}} - \mathbf{k}^{eff}) \tag{9}$$

where $t, \hat{\mathbf{e}}_{eff}$ and $\mathbf{k}^{eff}$ are the transmission coefficient, the polarization vector, and the wavevector of the effective field, respectively (the average or coherent field). These parameters are unknown and must be determined using self-consistency requirements. By substituting this expression into Eq. (8) one gets

$$\langle \mathbf{E}^{\mathrm{S}}(\mathbf{r})\rangle = \frac{\mathrm{i}}{8\pi^2} tE_0 \int\int \mathrm{d}k_x^{\mathrm{s}}\, \mathrm{d}k_y^{\mathrm{s}} \left\langle \frac{(\overleftrightarrow{\mathbf{1}} - \hat{\mathbf{k}}^{\mathrm{s}}_{\pm}\hat{\mathbf{k}}^{\mathrm{s}}_{\pm})}{k_z^{\mathrm{s}}} \cdot \overleftrightarrow{T}(\mathbf{k}^{\mathrm{s}}_{\pm}, \mathbf{k}^{eff}) \cdot \hat{\mathbf{e}}_{eff} \right.$$
$$\left. \times \sum_{p=1}^{N} \exp[-\mathrm{i}(\mathbf{k}^{\mathrm{s}}_{\pm} - \mathbf{k}^{eff}) \cdot \mathbf{r}_{\mathrm{p}}] \exp(\mathrm{i}\mathbf{k}^{\mathrm{s}}_{\pm} \cdot \mathbf{r}) \right\rangle . \tag{10}$$

We now perform the configurational average $\langle \cdots \rangle$. In the averaging procedure we will further assume that the positions of the particles are independent of each other, i.e., we ignore the exclusion volume, and that the probability to find a particle with its center inside the volume $\mathrm{d}^3\mathbf{r}$ is uniform and given by $\mathrm{d}^3\mathbf{r}/V$, where V is the volume of the slab. Therefore, the configurational average is calculated by integrating the location of each particle $\mathbf{r}_{\mathrm{p}}$ over the volume of the system keeping $N/V \equiv \rho$ constant. The integrals over $\mathrm{d}x_{\mathrm{p}}$ and $\mathrm{d}y_{\mathrm{p}}$ yield delta functions $2\pi\delta(k_x^{\mathrm{s}} - k_x^{eff})$ and $2\pi\delta(k_y^{\mathrm{s}} - k_y^{eff})$ thus $\langle \mathbf{E}^{\mathrm{S}}(\mathbf{r})\rangle$ can be written as

$$\langle \mathbf{E}^{\mathrm{S}}(\mathbf{r})\rangle = \frac{\mathrm{i}}{2} tE_0 \frac{N}{V} \frac{(\overleftrightarrow{\mathbf{1}} - \hat{\mathbf{k}}_{\pm}\hat{\mathbf{k}}_{\pm})}{k_z^{\mathrm{s}}} \cdot \overleftrightarrow{T}(\mathbf{k}_{\pm}, \mathbf{k}^{eff}) \cdot \hat{\mathbf{e}}_{eff}$$
$$\int \exp[-\mathrm{i}(\pm k_z^{\mathrm{s}} - k_z^{eff}) z_{\mathrm{p}}]\, \mathrm{d}z_{\mathrm{p}} \exp(\mathrm{i}\mathbf{k}_{\pm} \cdot \mathbf{r}), \tag{11}$$

where $\mathbf{k}_{\pm} = (k_x^{eff}, k_y^{eff}, \pm k_z^{\mathrm{s}})$, $k_z^{\mathrm{s}} = \sqrt{k^2 - (k_x^{eff})^2 - (k_y^{eff})^2}$ and $k_z^{eff} = \sqrt{(k^{eff})^2 - (k_x^{eff})^2 - (k_y^{eff})^2}$, where the $\pm$ sign refer to the field propagating to the right $(+)$ or to the left $(-)$ of each particle.

Let us now assume the observation point is at $z > 0$, that is, inside the system. The integral in Eq. (11) must be divided in two parts, from $z_{\mathrm{p}} = 0$ to $z_{\mathrm{p}} = z$, and from $z_{\mathrm{p}} = z$ to $z_{\mathrm{p}} = \infty$. In the first integral we use the $+$ sign while in the second we use the $-$ sign. The total field is given by the incident field plus the scattered field. Carrying out the integrals in Eq. (11), while assuming that $\mathrm{Im}\, k_z^{eff} \geqslant 0$ (so that the last integral vanishes at ∞), simplifying, and adding the incident field yields,

$$tE_0 \exp(\mathrm{i}\mathbf{k}^{eff} \cdot \mathbf{r})\hat{\mathbf{e}}_{eff} = E_0 \exp(\mathrm{i}\mathbf{k}^{\mathrm{i}} \cdot \mathbf{r})\hat{\mathbf{e}}_{\mathrm{i}} + \frac{\mathrm{i}}{2} tE_0 \rho$$
$$\times \left[\frac{(\overleftrightarrow{\mathbf{1}} - \hat{\mathbf{k}}_{+}\hat{\mathbf{k}}_{+})}{k_z^{\mathrm{s}}} \cdot \overleftrightarrow{T}(\mathbf{k}_{+}, \mathbf{k}^{eff}) \cdot \hat{\mathbf{e}}_{eff} \frac{\exp(\mathrm{i}\mathbf{k}_{+} \cdot \mathbf{r}) - \exp(\mathrm{i}\mathbf{k}^{eff} \cdot \mathbf{r})}{\mathrm{i}(k_z^{\mathrm{s}} - k_z^{eff})} \right.$$
$$\left. - \frac{(\overleftrightarrow{\mathbf{1}} - \hat{\mathbf{k}}_{-}\hat{\mathbf{k}}_{-})}{k_z^{\mathrm{s}}} \cdot \overleftrightarrow{T}(\mathbf{k}_{-}, \mathbf{k}^{eff}) \cdot \hat{\mathbf{e}}_{eff} \frac{\exp[\mathrm{i}\mathbf{k}^{eff} \cdot \mathbf{r}]}{\mathrm{i}(k_z^{\mathrm{s}} + k_z^{eff})} \right] . \tag{12}$$

In order for this equation to be fulfilled, it is required that,

$$\exp(\mathrm{i}\mathbf{k}^{\mathrm{i}}\cdot\mathbf{r})\hat{\mathbf{e}}_{\mathrm{i}}+\frac{\mathrm{i}}{2}t\rho\,\frac{(\overleftrightarrow{\mathbf{1}}-\hat{\mathbf{k}}_{+}\hat{\mathbf{k}}_{+})}{\mathrm{i}(k_z^{\mathrm{s}}-k_z^{eff})k_z^{\mathrm{s}}}\cdot\overleftrightarrow{T}(\mathbf{k}_{+},\mathbf{k}^{eff})\cdot\hat{\mathbf{e}}_{eff}\exp(\mathrm{i}\mathbf{k}_{+}\cdot\mathbf{r})=0 \tag{13}$$

and

$$\hat{\mathbf{e}}_{eff}=-\frac{\mathrm{i}}{2}\rho\left[\frac{(\overleftrightarrow{\mathbf{1}}-\hat{\mathbf{k}}_{+}\hat{\mathbf{k}}_{+})}{\mathrm{i}(k_z^{\mathrm{s}}-k_z^{eff})k_z^{\mathrm{s}}}\cdot\overleftrightarrow{T}(\mathbf{k}_{+},\mathbf{k}^{eff})\cdot\hat{\mathbf{e}}_{eff}\right.$$
$$\left.+\frac{(\overleftrightarrow{\mathbf{1}}-\hat{\mathbf{k}}_{-}\hat{\mathbf{k}}_{-})}{\mathrm{i}(k_z^{\mathrm{s}}+k_z^{eff})k_z^{\mathrm{s}}}\cdot\overleftrightarrow{T}(\mathbf{k}_{-},\mathbf{k}^{eff})\cdot\hat{\mathbf{e}}_{eff}\right]. \tag{14}$$

These equations can be recognized as the ones corresponding to the Ewald–Oseen theorem.

For Eq. (13) to be satisfied for all $\mathbf{r}$ $(z>0)$, one needs that,

$$\mathbf{k}^{\mathrm{i}}=\mathbf{k}_{+} \tag{15}$$

and $\hat{\mathbf{e}}_{eff}=\hat{\mathbf{e}}_{\mathrm{i}}$. This implies, $k_x^{\mathrm{i}}=k_x^{eff}$ and $k_y^{\mathrm{i}}=k_y^{eff}$. This is Snell's law; thus this simply means that the coherent field satisfies Snell's law. It also implies $\mathbf{k}_{-}=\mathbf{k}^{\mathrm{r}}\equiv(k_x^{\mathrm{i}},k_y^{\mathrm{i}},-k_z^{\mathrm{i}})$. A further approximation will be to consider only independent scattering, this means that field scattered by a sphere and impinging on another sphere, is already in the far-zone. This approximation, together with Snell's law, is expressed by the replacement of $\overleftrightarrow{T}(\mathbf{k}_{+},\mathbf{k}^{eff})$ by $\overleftrightarrow{T}(\mathbf{k}^{\mathrm{i}},\mathbf{k}^{\mathrm{i}})$ and of $\overleftrightarrow{T}(\mathbf{k}_{-},\mathbf{k}^{eff})$ by $\overleftrightarrow{T}(\mathbf{k}^{\mathrm{r}},\mathbf{k}^{\mathrm{i}})$, and it will be valid in the dilute regime.

By comparing the fields scattered by a sphere, when illuminated by a plane wave, in terms of the T-matrix, with the expression for these same fields in the far-zone, in terms of the amplitude scattering matrix, it can be shown that,

$$\mathrm{i}\,\tfrac{1}{2}\,\rho(\overleftrightarrow{\mathbf{1}}-\hat{\mathbf{k}}^{\mathrm{i}}\hat{\mathbf{k}}^{\mathrm{i}})\cdot\overleftrightarrow{T}(\mathbf{k}^{\mathrm{i}},\mathbf{k}^{\mathrm{i}})\cdot\hat{\mathbf{e}}_{\mathrm{i}}=-\gamma k^2S(0)\hat{\mathbf{e}}_{\mathrm{i}}, \tag{16}$$

$$\mathrm{i}\,\tfrac{1}{2}\,\rho(\overleftrightarrow{\mathbf{1}}-\hat{\mathbf{k}}^{\mathrm{r}}\hat{\mathbf{k}}^{\mathrm{r}})\cdot\overleftrightarrow{T}(\mathbf{k}^{\mathrm{r}},\mathbf{k}^{\mathrm{i}})\cdot\hat{\mathbf{e}}_{\mathrm{i}}=-\gamma k^2S_m(\pi-2\theta_{\mathrm{i}})\hat{\mathbf{e}}_{\mathrm{r}}, \tag{17}$$

where $m=1$ or 2 and $\hat{\mathbf{e}}_{\mathrm{r}}=\hat{\mathbf{a}}_x$ or $\hat{\mathbf{e}}_{\mathrm{r}}=-(\cos\theta_{\mathrm{i}}\hat{\mathbf{a}}_y+\sin\theta_{\mathrm{i}}\hat{\mathbf{a}}_z)$ for TE or TM polarization, respectively, and S_1 and S_2 are elements of the amplitude scattering matrix (as defined by Bohren and Huffman in their book [17]). The scattered field in the far-zone is transverse and is given by the product of the amplitude scattering matrix times the incident plane wave. Being the fields transverse, in an adequate reference frame only two components of polarization are needed, so the dimension of the scattering matrix is 2×2 and it has, in general, four elements; but for a spherical particle S_3 and S_4 are zero. S_1 and S_2 are functions of only the scattering angle θ, and they can be calculated using Mie theory. Here, $S(0)\equiv S_1(\theta=0)=S_2(\theta=0)$ is called the forward scattering amplitude, and

$$\gamma=3f/2x^3, \tag{18}$$

where $x \equiv ka$ is the size parameter, $f = \rho 4\pi a^3/3$ is the filling fraction of spheres, and a is their radius. Since γ is proportional to ρ, to keep results up to order ρ^n will be equivalent to keep them to order γ^n.

Using $k_z^{\mathrm{s}} = k_z^{\mathrm{i}}$ and Eqs. (16) and (17), we can rewrite Eqs. (13) and (14) as,

$$\hat{\mathbf{e}}_{\mathrm{i}} - t\,\frac{\gamma k^2 S(0)}{\mathrm{i}(k_z^{\mathrm{i}} - k_z^{eff})k_z^{\mathrm{i}}}\,\hat{\mathbf{e}}_{\mathrm{i}} = 0 \tag{19}$$

and

$$\hat{\mathbf{e}}_{eff} = \left[\frac{\gamma k^2 S(0)}{\mathrm{i}(k_z^{\mathrm{i}} - k_z^{eff})k_z^{\mathrm{i}}}\,\hat{\mathbf{e}}_{\mathrm{i}} + \frac{\gamma k^2 S_m(\pi - 2\theta_{\mathrm{i}})}{\mathrm{i}(k_z^{\mathrm{i}} + k_z^{eff})k_z^{\mathrm{i}}}\,\hat{\mathbf{e}}_{\mathrm{r}}\right]. \tag{20}$$

Now these two equations must be solved for the two unknowns: k_z^{eff} and t. From Eq. (19) one gets

$$t = \frac{\mathrm{i}(k_z^{\mathrm{i}} - k_z^{eff})k_z^{\mathrm{i}}}{\gamma k^2 S(0)}. \tag{21}$$

Notice that, as $\theta_{\mathrm{i}} \to \pi/2$ (grazing incidence), $k_z^{\mathrm{i}} \to 0$ thus $t \to 0$, which is correct. However, the expression for t is not complete to first order in γ. This can be seen from Eq. (21) in which γ appears in the denominator. Therefore, the calculation of k^{eff} in the numerator should be correct to order γ^2 in order to get t complete to first order in γ.

Now, we dot multiply the second equation (20) by $\hat{\mathbf{a}}_x$ when considering TE polarization, or by $\hat{\mathbf{a}}_z$ when considering TM polarization. Notice that the difference between $\hat{\mathbf{e}}_{eff} \cdot \hat{\mathbf{a}}_z$ and $\hat{\mathbf{e}}_{\mathrm{i}} \cdot \hat{\mathbf{a}}_z$ is of first order in γ (this is for TM polarization). We now simplify Eq. (20) for either polarization, to first order in γ, as

$$1 = -\left[\frac{\mathrm{i}\gamma k^2 S(0)}{(k_z^{\mathrm{i}} - k_z^{eff})k_z^{\mathrm{i}}} + \frac{\mathrm{i}\gamma k^2 S_m(\pi - 2\theta_{\mathrm{i}})}{(k_z^{\mathrm{i}} + k_z^{eff})k_z^{\mathrm{i}}}\right]. \tag{22}$$

Solving for k_z^{eff} and keeping terms to first order in γ, yields

$$(k_z^{eff})^2 = (k_z^{\mathrm{i}})^2 + 2\mathrm{i}\gamma k^2 S(0). \tag{23}$$

We take the square root on both sides and write $k_z^{eff} = \sqrt{(k_z^{\mathrm{i}})^2 + 2\mathrm{i}\gamma k^2 S(0)}$. The effective index of refraction may be obtained from the relation $k_{eff} = n_{eff}\,k$, and gives

$$n_{eff} = \sqrt{1 + 2\mathrm{i}\gamma k^2 S(0)} \simeq 1 + 2\mathrm{i}\gamma k^2 S(0). \tag{24}$$

This result has been already obtained by several authors [12,13,18–20]. As it is well known, the imaginary part of n_{eff} gives the attenuation (extinction) of the coherent light due to both, scattering and absorption, and by taking $\mathrm{Im}\,n_{eff}$ in Eq. (24) one gets Beer–Lambert's law. The real part of n_{eff} gives the phase delay of the coherent light. This can be measured directly by interferometry [21,22], or indirectly by the critical-angle effect [23–25].

Now using Eqs. (21) and (23) we obtain the following expression for t,

$$t=\frac{2k_z^{\mathrm{i}}}{(k_z^{\mathrm{i}}+k_z^{eff})}. \tag{25}$$

Notice that this expression is independent of the polarization and this should not be so for oblique angles of incidence (except at grazing incidence). This is a consequence of the fact that this expression is not complete to order γ^1. Furthermore, the right-hand side of Eq. (12) provides another expression for $\langle \mathbf{E}^{\mathrm{T}}(\mathbf{r})\rangle = \mathbf{E}_{coh}$ from which we could try to get t; however, neither the amplitude nor phase of the scattered field, calculated in this way, are complete to first order in γ. The reason is that a term given by the inverse of t [see Eq. (21)] appears in this expression. If we were to include a higher order approximation to the exciting field, second-order terms should appear in the numerator of this term, resulting in additional terms of order γ^1, after dividing by $(k_z^{\mathrm{i}}-k_z^{eff})k_z^{\mathrm{i}}$. (This would be so even if we do not ignore the exclusion volumes of the particles when performing the ensemble average.) Then we conclude that EFA cannot yield directly and expression, correct to order γ^1, for the transmission coefficient into the composite half-space.

2.1. The reflected field

Let us assume that the observation point is just outside the system, i.e., for $z<0$, and let us go back to Eq. (11). In this case all particles are now to the right of the observation point. Thus, we may write

$$\langle \mathbf{E}^{\mathrm{S}}(\mathbf{r})\rangle = \frac{\mathrm{i}}{2} tE_0 \frac{N}{V}\frac{(\overleftrightarrow{\mathbf{1}}-\hat{\mathbf{k}}_-\hat{\mathbf{k}}_-)}{k_z^{\mathrm{i}}}\cdot \overleftrightarrow{T}(\mathbf{k}_-,\mathbf{k}^{eff})\cdot \hat{\mathbf{e}}_{eff}$$
$$\times\int_0^{\infty}\exp[-\mathrm{i}(-k_z^{\mathrm{i}}-k_z^{eff})z_{\mathrm{p}}]\,\mathrm{d}z_{\mathrm{p}}\exp(\mathrm{i}\mathbf{k}_-\cdot\mathbf{r}). \tag{26}$$

We now substitute $\mathbf{k}^{eff}\to\mathbf{k}^{\mathrm{i}}$ in the arguments of $\overleftrightarrow{T}$, replace $\hat{\mathbf{e}}_{eff}\to\hat{\mathbf{e}}_{\mathrm{i}}$ and evaluate the integral. Then use Eq. (17), and write $\langle \mathbf{E}^{\mathrm{S}}(\mathbf{r})\rangle|_{z<0}=\mathbf{E}^{\mathrm{r}}=rE_0\exp(\mathrm{i}\mathbf{k}_-\cdot\mathbf{r})$, where r is the reflection coefficient. One gets

$$r=t\,\frac{\gamma k^2 S_m(\pi-2\theta_{\mathrm{i}})}{\mathrm{i}(k_z^{\mathrm{i}}+k_z^{eff})k_z^{\mathrm{i}}} \tag{27}$$

By using $t=2k_z^{\mathrm{i}}/(k_z^{\mathrm{i}}+k_z^{eff})$ and Eq. (23), one can write,

$$r=\frac{\gamma k^2 S_m(\pi-2\theta_{\mathrm{i}})}{\mathrm{i}(k_z^{\mathrm{i}}+k_z^{eff})k_z^{\mathrm{i}}-\gamma k^2 S(0)}. \tag{28}$$

This result is correct to order γ^1. One can get convinced of this by noting that if we go to a higher order approximation in the exciting field, and add to it terms of order γ^1, the reflected field would change to order γ^2. Note also that $r \to -1$ at grazing incidence, since $S_m(\pi - 2\theta_i) \to S(0)$ as $\theta_i \to \pi/2$, and this is correct. Also, since $S_1(\pi) = -S_2(\pi)$ we have that $r^{TE} = -r^{TM}$ at normal incidence ($\theta_i = 0$) which must be the case in the present problem. Thus, Eq. (28) gives an expression consistent to all orders in the density of particles at grazing and normal incidence (although it is complete only to order γ^1).

3. Effective optical coefficients

An effective medium corresponds to an equivalent, artificial, homogeneous medium that describes the propagation of the average field by using continuum electrodynamics. We now obtain the effective optical coefficients by averaging Maxwell's equations. By averaging Faraday's law, one gets

$$\nabla \times \mathbf{E}_{coh} = -i\omega \mathbf{B}_{coh}, \tag{29}$$

where $\mathbf{B}_{coh} = \langle \mathbf{B}^T(\mathbf{r}) \rangle$. By averaging Ampere–Maxwell's equation, one obtains,

$$\nabla \times \mathbf{B}_{coh} = \mu_0 \langle \mathbf{J} \rangle + i\omega\varepsilon_0\mu_0 \mathbf{E}_{coh}. \tag{30}$$

In any given material, the current $\mathbf{J}$, usually arises by the induction of polarization, conduction, or magnetization currents in the system. In conventional continuum electrodynamics one defines the vector fields, $\mathbf{H}_{coh}$ and $\mathbf{D}_{coh}$, such that they take account of the average of the induced currents, obtaining

$$\nabla \times \mathbf{H}_{coh} = i\omega \mathbf{D}_{coh}. \tag{31}$$

Then the effective optical coefficients are defined as the tensors relating $\mathbf{H}_{coh}$ with $\mathbf{B}_{coh}$ and $\mathbf{D}_{coh}$ with $\mathbf{E}_{coh}$. Specifically, one writes $\mathbf{B}_{coh} = \mu_0 \overleftrightarrow{\tilde{\mu}}^{eff} \cdot \mathbf{H}_{coh}$ where we introduced the effective relative magnetic permeability tensor, $\overleftrightarrow{\tilde{\mu}}^{eff} = \overleftrightarrow{1} + \overleftrightarrow{\chi}_h$, where $\overleftrightarrow{\chi}_h$ is an effective magnetic susceptibility. The tilde on $\overleftrightarrow{\tilde{\mu}}^{eff}$ means that its components are measured in units of μ_0. Similarly, we write $\mathbf{D}_{coh} = \varepsilon_0 \overleftrightarrow{\tilde{\varepsilon}}_{eff} \cdot \mathbf{E}_{coh}$, where $\overleftrightarrow{\tilde{\varepsilon}}_{eff}$ is the effective relative electric permittivity tensor measured in units of ε_0. One can also write $\overleftrightarrow{\tilde{\varepsilon}}_{eff} = \overleftrightarrow{1} + \overleftrightarrow{\chi}_e$, where $\overleftrightarrow{\chi}_e$ is an effective electric susceptibility. Unfortunately, a direct calculation of the coherent fields $\mathbf{E}_{coh}$, $\mathbf{B}_{coh}$, $\mathbf{D}_{coh}$, and $\mathbf{H}_{coh}$ within the composite half-space, requires more elaborated approximations than the EFA. Nevertheless, we can still use the EFA results to obtain first order expressions to the effective optical coefficients as it will be shown below.

Our strategy here is very simple. The effective-medium equations (29) and (31) imply the continuity of the tangential components of the $\mathbf{E}_{coh}$ and $\mathbf{H}_{coh}$ at the interface,

$$\mathbf{E}^T \times \hat{\mathbf{a}}_z|_{z=0^-} = \mathbf{E}^T \times \hat{\mathbf{a}}_z|_{z=0^+}, \tag{32}$$

$$\mathbf{H}^T \times \hat{\mathbf{a}}_z|_{z=0^-} = \mathbf{H}^T \times \hat{\mathbf{a}}_z|_{z=0^+}. \tag{33}$$

These equations relate the tangential components of the fields outside ($z=0^-$) and inside ($z=0^+$) the half-space, and from these relations and symmetry requirements one can obtain the relationship between the effective permeability $\overset{\leftrightarrow}{\tilde{\varepsilon}}^{eff}$ and permittivity $\overset{\leftrightarrow}{\tilde{\mu}}^{eff}$ tensors inside with the corresponding ones outside (μ_0 and ε_0). Symmetry requirements here demand that in our reference frame, the effective optical tensors, $\overset{\leftrightarrow}{\tilde{\varepsilon}}^{eff}$ and $\overset{\leftrightarrow}{\tilde{\mu}}^{eff}$, should be diagonal. Below we perform the calculation of the effective optical tensors and show that this procedure requires to know only the coherent reflection coefficient, which we do to order γ^1 (Eq. (28) above). To avoid possible confusions in what follows, we will add a subscript e or h to the transmission coefficient, to denote that it refers to the electric field or magnetic coherent wave. We do not need to do that for the reflection coefficient, since it is the same for the electric and magnetic wave. We will also add a superscript to indicate the polarization.

3.1. TE polarization

In the present geometry, and for TE polarization, Eqs. (32) and (33) can be written as,

$$E_x^{\mathrm{i}}(z=0^-)+E_x^{\mathrm{r}}(z=0^-)=E_x^{coh}(z=0^+), \tag{34}$$

$$H_y^{\mathrm{i}}(z=0^-)+H_y^{\mathrm{r}}(z=0^-)=H_y^{coh}(z=0^+), \tag{35}$$

where $E_x^{\mathrm{i}}=E_0\exp(\mathrm{i}\mathbf{k}^{\mathrm{i}}\cdot\mathbf{r})$, $E_x^{\mathrm{r}}=r^{\mathrm{TE}}E_0\exp(\mathrm{i}\mathbf{k}^{\mathrm{r}}\cdot\mathbf{r})$, and $E_x^{coh}=t_{\mathrm{e}}^{\mathrm{TE}}E_0\exp(\mathrm{i}\mathbf{k}^{eff}\cdot\mathbf{r})$, and $\mathbf{k}^{\mathrm{i}}=k_y^{\mathrm{i}}\hat{\mathbf{a}}_y+k_z^{\mathrm{i}}\hat{\mathbf{a}}_z$, $\mathbf{k}^{\mathrm{r}}=k_y^{\mathrm{i}}\hat{\mathbf{a}}_y-k_z^{\mathrm{i}}\hat{\mathbf{a}}_z$, and $\mathbf{k}^{eff}=k_y^{\mathrm{i}}\hat{\mathbf{a}}_y+k_z^{eff}\hat{\mathbf{a}}_z$. Outside the system of particles, i.e., for $z<0$, one has that Faraday's law reads, $\mathbf{H}^{\mathrm{i}}=(1/\mathrm{i}\omega\mu_0)\nabla\times\mathbf{E}^{\mathrm{i}}$ and $\mathbf{H}^{\mathrm{r}}=(1/\mathrm{i}\omega\mu_0)\nabla\times\mathbf{E}^{\mathrm{r}}$, while inside the system, i.e., for $z>0$, one has $\nabla\times\mathbf{E}_{coh}=\mathrm{i}\omega\mu_0\,\overset{\leftrightarrow}{\tilde{\mu}}^{eff}\cdot\mathbf{H}_{coh}$. Since the tensor $\overset{\leftrightarrow}{\tilde{\mu}}^{eff}$ must be diagonal, the y-component of the latter equation yields,

$$\tilde{\mu}_{yy}^{eff}H_y^{coh}=\frac{k_z^{eff}}{\omega\mu_0}E_x^{coh}=\frac{k_z^{eff}}{\omega\mu_0}t_{\mathrm{e}}^{\mathrm{TE}}E_0\exp(\mathrm{i}\mathbf{k}^{eff}\cdot\mathbf{r}). \tag{36}$$

Thus, we write Eqs. (34) and (35) as

$$1+r^{\mathrm{TE}}=t_{\mathrm{e}}^{\mathrm{TE}}, \tag{37}$$

$$\frac{k_z^{\mathrm{i}}}{\omega\mu_0}(1-r^{\mathrm{TE}})=\frac{1}{\omega\mu_0}\frac{k_z^{eff}}{\tilde{\mu}_{yy}^{eff}}t_{\mathrm{e}}^{\mathrm{TE}}, \tag{38}$$

where we have made use of Snell's law. Since from the EFA we have calculated r^{TE} correct to first order in γ, from Eq. (37) we get $t_{\mathrm{e}}^{\mathrm{TE}}$ correct to first order in γ. By substituting Eq. (28) with $m=1$ into Eq. (37), one gets,

$$t_{\mathrm{e}}^{\mathrm{TE}}=\frac{\mathrm{i}(k_z^{\mathrm{i}}+k_z^{eff})k_z^{\mathrm{i}}-\gamma k^2[S(0)-S_1(\pi-2\theta_{\mathrm{i}})]}{\mathrm{i}(k_z^{\mathrm{i}}+k_z^{eff})k_z^{\mathrm{i}}-\gamma k^2 S(0)}. \tag{39}$$

This expression for the transmission coefficient is correct to order γ^1. One can see that this expression is different from the one in (21). Therefore, we can write,

$$\mathbf{E}_{coh} = t_{\mathrm{e}}^{\mathrm{TE}} E_0 \exp(\mathrm{i}\mathbf{k}^{eff} \cdot \mathbf{r})\hat{\mathbf{a}}_x, \tag{40}$$

and from Faraday's law, $\nabla \times \mathbf{E}_{coh} = \mathrm{i}\omega \mathbf{B}_{coh}$, we get

$$\mathbf{B}_{coh} = t_{\mathrm{e}}^{\mathrm{TE}} E_0/\omega (k_z^{eff}\hat{\mathbf{a}}_y - k_y^{\mathrm{i}}\hat{\mathbf{a}}_z)\exp(\mathrm{i}\mathbf{k}^{eff} \cdot \mathbf{r}). \tag{41}$$

Both of these expressions are now correct to first order in γ. However, we cannot write an expression for $\mathbf{H}_{coh}$ since it involves an effective magnetic susceptibility which we must find first.

By using $t_{\mathrm{e}}^{\mathrm{TE}} = 1 + r^{\mathrm{TE}}$ in (38) and solving for $\tilde{\mu}_{yy}^{eff}$, one gets,

$$\tilde{\mu}_{yy}^{eff} = \frac{k_z^{eff}}{k_z^{\mathrm{i}}} \frac{(1 + r^{\mathrm{TE}})}{(1 - r^{\mathrm{TE}})}. \tag{42}$$

We now use Eq. (23), expand $k_z^{eff}/k_z^{\mathrm{i}}$ in powers of γ and keep in the resulting expression only terms up to first order in γ. One gets

$$\tilde{\mu}_{yy}^{eff} \simeq 1 + \mathrm{i}\gamma \frac{S_-^1(\theta_{\mathrm{i}})}{\cos^2\theta_{\mathrm{i}}}, \tag{43}$$

where $S_-^1(\theta_{\mathrm{i}}) = S(0) - S_1(\pi - 2\theta_{\mathrm{i}})$. Also, from the symmetry of the system one must have,

$$\tilde{\mu}_{yy}^{eff} = \tilde{\mu}_{zz}^{eff}. \tag{44}$$

Now, we write Ampere–Maxwell equation, Eq. (31), as $\mathrm{i}\mathbf{k}^{eff} \times \mathbf{H}_{coh} = -\mathrm{i}\omega\varepsilon_0 \overset{\leftrightarrow}{\tilde{\varepsilon}}{}^{eff} \cdot \mathbf{E}_{coh}$. Since the tensor must be is diagonal, the x-component of the latter equation is

$$k_y^{\mathrm{i}} H_z^{coh} - k_y^{eff} H_y^{coh} = -\omega\varepsilon_0 \tilde{\varepsilon}_{xx}^{eff} E_x^{coh}. \tag{45}$$

But we have that $H_z^{coh} = B_z^{coh}/\mu_0\tilde{\mu}_{zz}^{eff}$ and $H_y^{coh} = B_y^{coh}/\mu_0\tilde{\mu}_{yy}^{eff}$. Since $\tilde{\mu}_{zz}^{eff} = \tilde{\mu}_{yy}^{eff}$ we can solve for $\tilde{\varepsilon}_{xx}^{eff}$,

$$\tilde{\varepsilon}_{xx}^{eff} = -\frac{k_y^{\mathrm{i}} B_z^{coh} - k_y^{eff} B_y^{coh}}{\omega\varepsilon_0\mu_0 E_x^{coh}\tilde{\mu}_{yy}^{eff}}. \tag{46}$$

By using Eqs. (43), and (41), $(k_y^{eff})^2 - (k_y^{\mathrm{i}})^2 = 2\mathrm{i}\gamma k^2 S(0)$, and $1/\omega^2\varepsilon_0\mu_0 = c^2/\omega^2 = 1/k^2$, one can simplify this equation and arrive to

$$\tilde{\varepsilon}_{xx}^{eff} = 1 + 2\mathrm{i}\gamma S_+^1(\theta_{\mathrm{i}}) - \mathrm{i}\gamma S_-^1(\theta_{\mathrm{i}})\tan^2\theta_{\mathrm{i}} \tag{47}$$

where $S_+^1(\theta_{\mathrm{i}}) = \frac{1}{2}[S(0) + S_1(\pi - 2\theta_{\mathrm{i}})]$.

3.2. TM polarization

In this case we have $\mathbf{E}^{\mathrm{i}} = E_0 \exp(\mathrm{i}\mathbf{k}^{\mathrm{i}} \cdot \mathbf{r})\hat{\mathbf{e}}_{\mathrm{i}}$ and $\mathbf{E}^{\mathrm{r}} = r^{\mathrm{TM}} E_0 \exp(\mathrm{i}\mathbf{k}^{\mathrm{r}} \cdot \mathbf{r})\hat{\mathbf{e}}_{\mathrm{r}}$, where $\mathbf{k}^{\mathrm{i}} = k_y^{\mathrm{i}}\hat{\mathbf{a}}_y + k_z^{\mathrm{i}}\hat{\mathbf{a}}_z$, $\hat{\mathbf{e}}_{\mathrm{i}} = \cos\theta_{\mathrm{i}}\hat{\mathbf{a}}_y - \sin\theta_{\mathrm{i}}\hat{\mathbf{a}}_z$ and $\mathbf{k}^{\mathrm{r}} = k_y^{\mathrm{i}}\hat{\mathbf{a}}_y - k_z^{\mathrm{i}}\hat{\mathbf{a}}_z$, $\hat{\mathbf{e}}_{\mathrm{r}} = -\cos\theta_{\mathrm{i}}\hat{\mathbf{a}}_y - \sin\theta_{\mathrm{i}}\hat{\mathbf{a}}_z$, and r^{TM} is given by Eq. (28) with $m = 2$. The electric field outside the system (i.e., for $z < 0$) is given by $\mathbf{E}^{\mathrm{i}} + \mathbf{E}^{\mathrm{r}}$. From this, we can calculate the magnetic field, $\mathbf{H}^{\mathrm{i}} + \mathbf{H}^{\mathrm{r}}$ by using Faraday's law: $\nabla \times \mathbf{E}^{\mathrm{i}} = \mathrm{i}\omega\mu_0\mathbf{H}^{\mathrm{i}}$, and $\nabla \times \mathbf{E}^{\mathrm{r}} = \mathrm{i}\omega\mu_0\mathbf{H}^{\mathrm{r}}$. These equations yield,

$$\mathbf{H}^{\mathrm{i}} + \mathbf{H}^{\mathrm{r}} = \frac{-kE_0}{\omega\mu_0}[\exp(\mathrm{i}\mathbf{k}^{\mathrm{i}} \cdot \mathbf{r}) + r^{\mathrm{TM}} \exp(\mathrm{i}\mathbf{k}^{\mathrm{r}} \cdot \mathbf{r})]\hat{\mathbf{a}}_x \tag{48}$$

From the continuity of the tangential components of the magnetic field at the interface, i.e., at $z = 0$, and writing $\mathbf{H}^{coh}$ as $\mathbf{H}^{coh} = t_h^{\mathrm{TM}} H_0 \exp(\mathrm{i}\mathbf{k}^{eff} \cdot \mathbf{r})\hat{\mathbf{a}}_x$, where $H_0 = -kE_0/(\omega\mu_0)$, one gets,

$$t_h^{\mathrm{TM}} = 1 + r^{\mathrm{TM}}. \tag{49}$$

By substituting Eq. (28) with $m = 2$ in this equation, one gets an expression for t_h^{TM} similar to Eq. (39) but with S_1 replaced by S_2. Now, from Ampere–Maxwell's equation we have,

$$\nabla \times \mathbf{H}_{coh} = -\mathrm{i}\omega\varepsilon_0 \overleftrightarrow{\tilde{\varepsilon}}_{eff} \cdot \mathbf{E}_{coh} \tag{50}$$

Now, since the tangential component of the electric field is continuous, i.e., E_y is continuous, and $\overleftrightarrow{\tilde{\varepsilon}}_{eff}$ is diagonal, the y-component of this equation yields,

$$k_z^{eff} H_{coh} = -\omega\varepsilon_0 \tilde{\varepsilon}_{yy}^{eff} E_y^{coh}. \tag{51}$$

At $z = 0^+$ we have $E_y^{coh} = E_y^{\mathrm{i}} + E_y^{\mathrm{r}} = E_0 \cos\theta_{\mathrm{i}}(1 - r^{\mathrm{TM}})\exp(\mathrm{i}k_x^{\mathrm{i}}x + \mathrm{i}k_y^{\mathrm{i}}y)$. Then solving for $\tilde{\varepsilon}_{yy}^{eff}$, one can write

$$\tilde{\varepsilon}_{yy}^{eff} = \frac{k_z^{eff}(1 + r^{\mathrm{TM}})}{k_z^{\mathrm{i}}(1 - r^{\mathrm{TM}})}. \tag{52}$$

In this equation, we now substitute r^{TM} from Eq. (28) with $m = 2$, expand $k_z^{eff}/k_z^{\mathrm{i}}$ in powers of γ, and keep in the resulting expression only terms up to first order in γ. One obtains

$$\tilde{\varepsilon}_{yy}^{eff} = 1 + \mathrm{i}\gamma \frac{S_-^{(2)}(\theta_{\mathrm{i}})}{\cos^2\theta_{\mathrm{i}}}. \tag{53}$$

Now, due to symmetry requirements, we assume $\tilde{\varepsilon}_{yy}^{eff} = \tilde{\varepsilon}_{zz}^{eff}$. Then, from Faraday's law, $\nabla \times \mathbf{E}_{coh} = \mathrm{i}\omega\mu_0 \overleftrightarrow{\tilde{\mu}}^{eff} \cdot \mathbf{H}_{coh}$ we get

$$(k_y^{\mathrm{i}} E_z^{coh} - k_z^{eff} E_y^{coh}) = \omega\mu_0 \tilde{\mu}_{xx}^{eff} H_{coh}, \tag{54}$$

and from Ampere–Maxwell's equation, Eq. (31), one obtains

$$(k_z^{eff}\hat{\mathbf{a}}_y - k_y^{\mathrm{i}}\hat{\mathbf{a}}_z)H_{coh} = -\omega\varepsilon_0(\tilde{\varepsilon}_{yy}^{eff} E_y^{coh}\hat{\mathbf{a}}_y + \tilde{\varepsilon}_{zz}^{eff} E_z^{coh}\hat{\mathbf{a}}_z)$$

and since $\tilde{\varepsilon}^{eff}_{yy} = \tilde{\varepsilon}^{eff}_{zz}$, we get from Eq. (54),

$$\frac{(k^{\mathrm{i}}_y)^2 + (k^{eff}_z)^2}{\omega\varepsilon_0\tilde{\varepsilon}^{eff}_{yy}} = \omega\mu_0\tilde{\mu}^{eff}_{xx}.$$

We now use $(k^{\mathrm{i}}_y)^2 = k^2 - (k^{\mathrm{i}}_z)^2$ and $\tilde{\varepsilon}^{eff}_{yy}$ from Eq. (53) and by keeping terms up to first order in γ, we finally arrive to,

$$\tilde{\mu}^{eff}_{xx} = 1 + 2\mathrm{i}\gamma S^{(2)}_{+}(\theta_{\mathrm{i}}) - \mathrm{i}\gamma\tan^2\theta_{\mathrm{i}} S^{(2)}_{-}(\theta_{\mathrm{i}}). \tag{55}$$

4. Results

Summarizing, we have that the effective magnetic permeability and permittivity tensors are,

$$\overleftrightarrow{\mu}_{eff} = \begin{pmatrix} \mu^{\mathrm{TM}}_{eff} & 0 & 0 \\ 0 & \mu^{\mathrm{TE}}_{eff} & 0 \\ 0 & 0 & \mu^{\mathrm{TE}}_{eff} \end{pmatrix} \quad \text{and} \quad \overleftrightarrow{\varepsilon}_{eff} = \begin{pmatrix} \varepsilon^{\mathrm{TE}}_{eff} & 0 & 0 \\ 0 & \varepsilon^{\mathrm{TM}}_{eff} & 0 \\ 0 & 0 & \varepsilon^{\mathrm{TM}}_{eff} \end{pmatrix}, \tag{56}$$

where

$$\tilde{\mu}^{\mathrm{TM}}_{eff} = 1 + 2\mathrm{i}\gamma S^{(2)}_{+}(\theta_{\mathrm{i}}) - \mathrm{i}\gamma\tan^2\theta_{\mathrm{i}} S^{(2)}_{-}(\theta_{\mathrm{i}}), \tag{57}$$

$$\tilde{\mu}^{\mathrm{TE}}_{eff} = 1 + \mathrm{i}\gamma\frac{S^{(1)}_{-}(\theta_{\mathrm{i}})}{\cos^2\theta_{\mathrm{i}}}, \tag{58}$$

$$\tilde{\varepsilon}^{\mathrm{TE}}_{eff} = 1 + 2\mathrm{i}\gamma S^{(1)}_{+}(\theta_{\mathrm{i}}) - \mathrm{i}\gamma\tan^2\theta_{\mathrm{i}} S^{(1)}_{-}(\theta_{\mathrm{i}}), \tag{59}$$

$$\tilde{\varepsilon}^{\mathrm{TM}}_{eff} = 1 + \mathrm{i}\gamma\frac{S^{(2)}_{-}(\theta_{\mathrm{i}})}{\cos^2\theta_{\mathrm{i}}}, \tag{60}$$

and $S^{(m)}_{-}(\theta_{\mathrm{i}}) = S(0) - S_m(\pi - 2\theta_{\mathrm{i}})$, and $S^{(m)}_{+}(\theta_{\mathrm{i}}) = \frac{1}{2}[S(0) + S_m(\pi - 2\theta_{\mathrm{i}})]$. Also, to first order in γ, we have that

$$n_{eff} = \sqrt{\tilde{\mu}^{\mathrm{TE}}_{eff}\tilde{\varepsilon}^{\mathrm{TE}}_{eff}} = \sqrt{\tilde{\mu}^{\mathrm{TM}}_{eff}\tilde{\varepsilon}^{\mathrm{TM}}_{eff}} \approx 1 + \mathrm{i}\gamma k^2 S(0), \tag{61}$$

which is the result obtained originally by Van de Hulst [20].

For particles comparable to the wavelength and larger, these optical coefficients are in general oscillatory functions of the angle of incidence. Let us consider some limiting cases. Noting that $S_m(\pi - 2\theta_{\mathrm{i}}) \to S(0)$ as $\theta_{\mathrm{i}} \to \pi/2$, it is not difficult to show that Eqs. (57)–(60) are well behaved at grazing incidence. Using $S_1(\pi) = -S_2(\pi)$ one can also see that for normal incidence these expressions reduce to the formulas given by Bohren [11]. For dielectric particles of size small compared to the wavelength ($x \ll 1$), we have that $S_1(\theta) \simeq -\mathrm{i}x^3\beta$ and $S_2(\theta) \simeq -\mathrm{i}x^3\beta\cos\theta$, where $\beta = (\tilde{\varepsilon}_{\mathrm{p}} - 1)/(\tilde{\varepsilon}_{\mathrm{p}} + 2)$ and $\tilde{\varepsilon}_{\mathrm{p}}$ is the relative dielectric permittivity of the particles. Substituting these expressions in the effective optical coefficients above give, $\tilde{\mu}^{\mathrm{TM}}_{eff} = \tilde{\mu}^{\mathrm{TE}}_{eff} = 1$, and $\tilde{\varepsilon}^{\mathrm{TE}}_{eff} = \tilde{\varepsilon}^{\mathrm{TM}}_{eff} = 1 + 3\beta f$. Thus, the optical coefficients reduce to scalar quantities and coincide with the low density limit of Maxwell–Garnett's formula. However, for particles which are not small compared to the wavelength, the scattering pattern of a single particle is anisotropic and, $S(0) \neq S_m(\theta)$, for $\theta \neq 0$. Thus, a magnetic effective permeability appears even if the particles are non-magnetic. The present results are valid for particles with a complex refractive index, such as metallic particles, and also for particles that have a magnetic permeability different from one. Since we considered only spherical particles only S_1 and S_2 are involved and one can use Mie theory to evaluate them.

With the above effective optical coefficients one can show that the reflection coefficients are given by the Fresnel relations,

$$r^{\mathrm{TE}} = \frac{\tilde{\mu}_{eff}^{\mathrm{TE}} k_z^{\mathrm{i}} + k_z^{eff}}{\tilde{\mu}_{eff}^{\mathrm{TE}} k_z^{\mathrm{i}} - k_z^{eff}}, \tag{62}$$

$$r^{\mathrm{TM}} = \frac{\tilde{\varepsilon}_{eff}^{\mathrm{TM}} k_z^{\mathrm{i}} + k_z^{eff}}{\tilde{\varepsilon}_{eff}^{\mathrm{TM}} k_z^{\mathrm{i}} - k_z^{eff}}. \tag{63}$$

If one were to ignore the effective magnetic permeability, one would use $\tilde{\mu}_{eff} = 1$ and $\tilde{\varepsilon}_{eff} = \sqrt{n_{eff}}$, where $n_{eff} = 1 + \mathrm{i}\gamma k^2 S(0)$ (being $\tilde{\varepsilon}_{eff}$ and $\tilde{\mu}_{eff}$ scalar quantities) in the Fresnel formulas above, and obtain different reflection coefficients. We will refer to these reflection coefficients as the non-magnetic approximations. The non-magnetic approximation is suggested in Refs. [4,5].

We could provide an alternative derivation of the same effective optical coefficients by postulating the Fresnel reflection relations, Eqs. (62) and (63), and equating them to the coherent reflection coefficients, Eq. (28) with $m = 1$ or 2. By doing so one immediately gets Eqs. (42) and (52) and from them, the effective optical coefficients Eqs. (58) and (60). The other two coefficients would be obtained by requiring $n_{eff} = \sqrt{\tilde{\mu}_{eff}^{\mathrm{TE}} \tilde{\varepsilon}_{eff}^{\mathrm{TE}}} = \sqrt{\tilde{\mu}_{eff}^{\mathrm{TM}} \tilde{\varepsilon}_{eff}^{\mathrm{TM}}}$. Doing this, is in fact equivalent to the derivation we have presented above, nevertheless we believe this is less clear, thus we have preferred to set explicitly the boundary value problem, for the clarity of the ideas and the approximations involved.

In an experiment involving optical measurements, the particles would most probably be immersed in a matrix material, and one would have to take into account the reflection at the matrix interface. For a dilute system of particles, the coherent reflectivity due to the particles will in general be small compared to the reflectivity from the matrix interface, except near grazing incidence. In order to detect the presence of the particles one might be obliged to perform differential measurements. For example, to measure the difference between the reflectivity in each polarization. Another possibility is to take advantage of the Brewester angle of the matrix interface or the critical angle effect. The use of the present formulas in problems involving a matrix interface will be treated in detail in a future publication. For the moment being let us only consider the particles in vacuum.

In order to illustrate the coherent reflectivity from a system of particles systems, we show the results of a few numerical calculations. In Figs. 2a–d we plot the reflectivity of linearly polarized light vs. the angle of incidence for a few values of the particle radius and for two different types of particles: glass and silver particles. The refractive index of the particles was taken to be $n_{\mathrm{p}} = 1.5$ and $0.12 + 3.45\mathrm{i}$ (at $\lambda = 0.59$ μm) respectively, and we used a filling fraction of $f = 0.1$. Although the filling fraction used is relatively large and the accuracy of the presents results may be questioned, the plots serve to illustrate the order of magnitude of the coherent reflectivity and its dependence

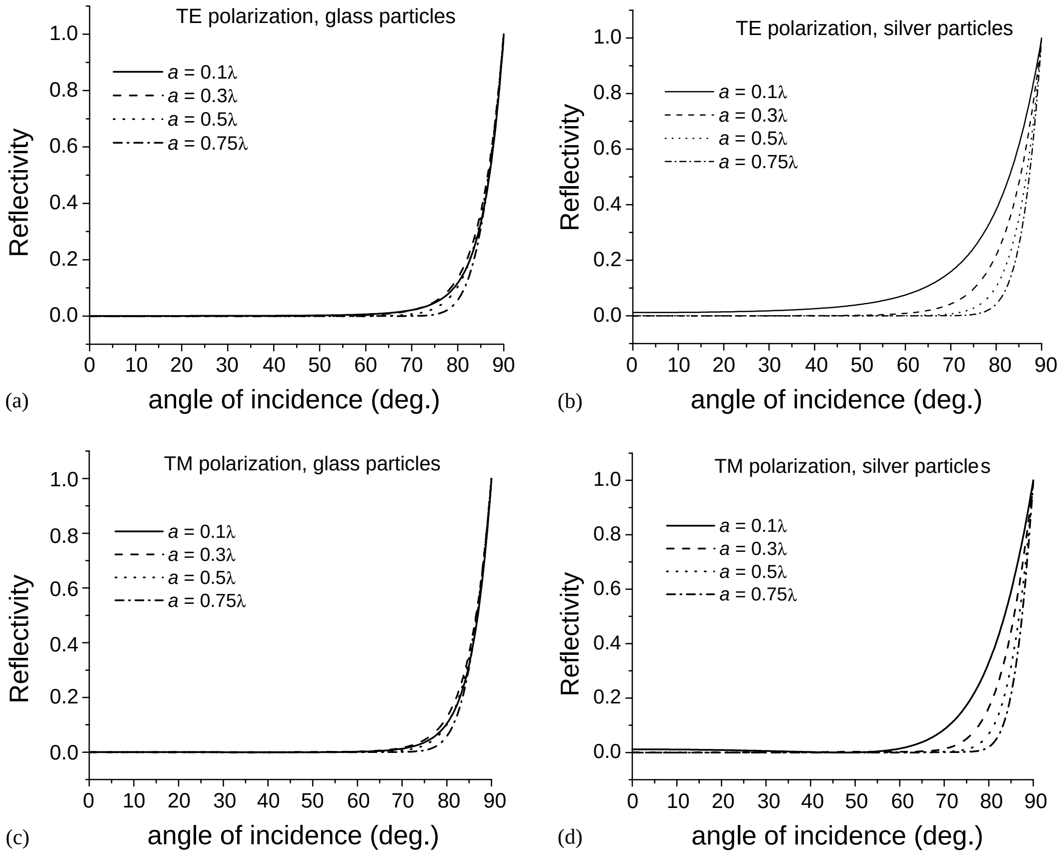

Fig. 2. Coherent reflectivity versus angle of incidence as calculated by the modulus squared of (62) and (63) for: glass and silver particles. Curves for a few values of the particle radius in terms of the wavelength are shown. The refractive index of the particles was taken to be $n_p = 1.5$ and $n_p = 0.12 + 3.45i$ (at $\lambda = 0.59$ μm) respectively, and we used a filling fraction of $f = 0.1$.

on the angle of incidence and particle radius. Clearly the coherent reflectivity of the silver particles start being noticeable before than for the glass particles of the same size. In Fig. 3a and b we show amplified a portion of the reflectivity plots for TM polarization. Curves for the two types of particles and for $a/\lambda = 0.1$ and 0.5 are shown. Note that in Fig. 3a the vertical axis is multiplied by 10^{-3} whereas in Fig. 3b the factor is 10^{-2}. We can see that the Brewster angle effect is still clear for particles of radius $a/\lambda = 0.1$, however, for particles of radius $a/\lambda = 0.5$ the effect is not noticeable anymore. At an even larger amplification factor one can see oscillations of the reflectivity which are due to the oscillations of the scattering amplitude S_2.as a function of the angle of incidence. In addition, we show the corresponding results using the non-magnetic approximation. The difference between using Eqs. (62) and (63), and the corresponding non-magnetic approximations is noticeable and can be large in relative terms. Generally, we find that the reflectivity is less than that predicted by the non-magnetic approximation. As the particle radius increases, the relative difference between both approximations increases.

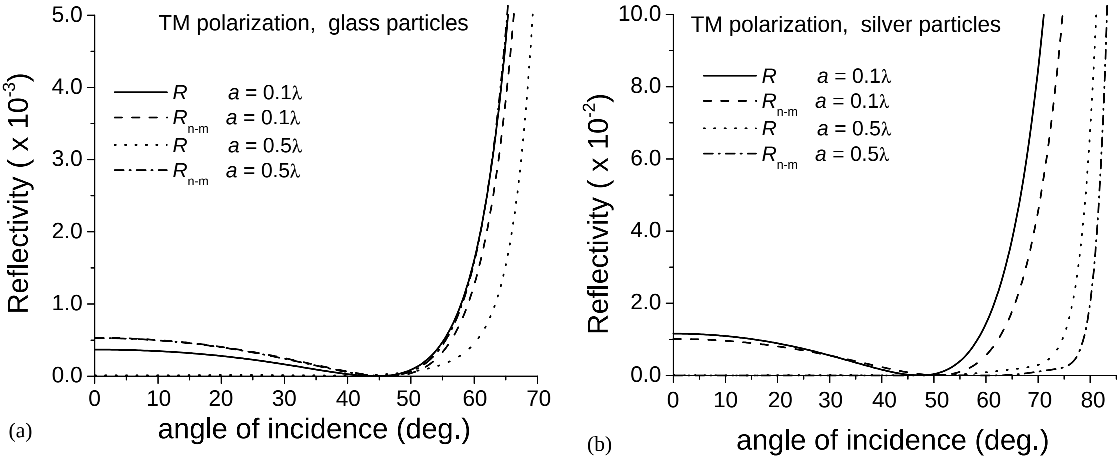

Fig. 3. Reflectivity versus angle of incidence for TM polarization for: (a) glass particles, and (b) silver particles, for two different values of the particles radius. The vertical scale in these plots is amplified and we added the corresponding curves calculated by the non-magnetic approximation ($R_{\text{n-m}}$).

Regarding the possible need for differential measurements in actual optical experiments, in Figs. 4a–d, we plot the difference between the reflectivity for TE and TM polarization divided by their sum for glass particles, $\Delta R/R = (R^{\text{TE}} - R^{\text{TM}})/(R^{\text{TE}} + R^{\text{TM}})$, as calculated from Eqs. (62) and (63). We also show the curves corresponding to the non-magnetic approximations, $(\Delta R/R)_{\text{n-m}} = (R^{\text{TE}}_{\text{n-m}} - R^{\text{TM}}_{\text{n-m}})/(R^{\text{TE}}_{\text{n-m}} + R^{\text{TM}}_{\text{n-m}})$. The filling fraction of the spheres was taken to be $f = 0.1$. It can be appreciated that for particle radius comparable to the wavelength the difference between the curves predicted by the magnetic response compared to the non-magnetic approximation are large. The magnetic response causes the curves to oscillate taking positive and negative values, whereas the non-magnetic approximation predicts only positive values and one single maximum. As the particle radius decreases, both curves approach each other, showing that the magnetic effects become less important. For particles with a larger refractive index, the curves oscillations are stronger reaching higher positive values. These figures suggest that differential measurements of polarized reflectance may be useful as an analytical tool in studying particle suspensions by optical reflection. Here, the errors from ignoring the magnetic response can be very large, as seen from the figures, once the particles have radius larger than about 0.1λ.

5. Discussion and conclusions

We used the effective-field approximation (EFA) to calculate the coherent reflection coefficient from a half-space of a dilute random system of spherical, polarizable particles in vacuum. The EFA leads to an integral equation from which one obtains a well-known approximation to the effective propagation wavevector, and from it, the effective refractive index. However, we found that the EFA could not provide expressions for the coherent fields inside the composite half-space correct to first order in the density of spheres. Nevertheless, the EFA provides an approximation to the coherent reflection coefficient, r, which is correct to first order in the density of particles, ρ. The

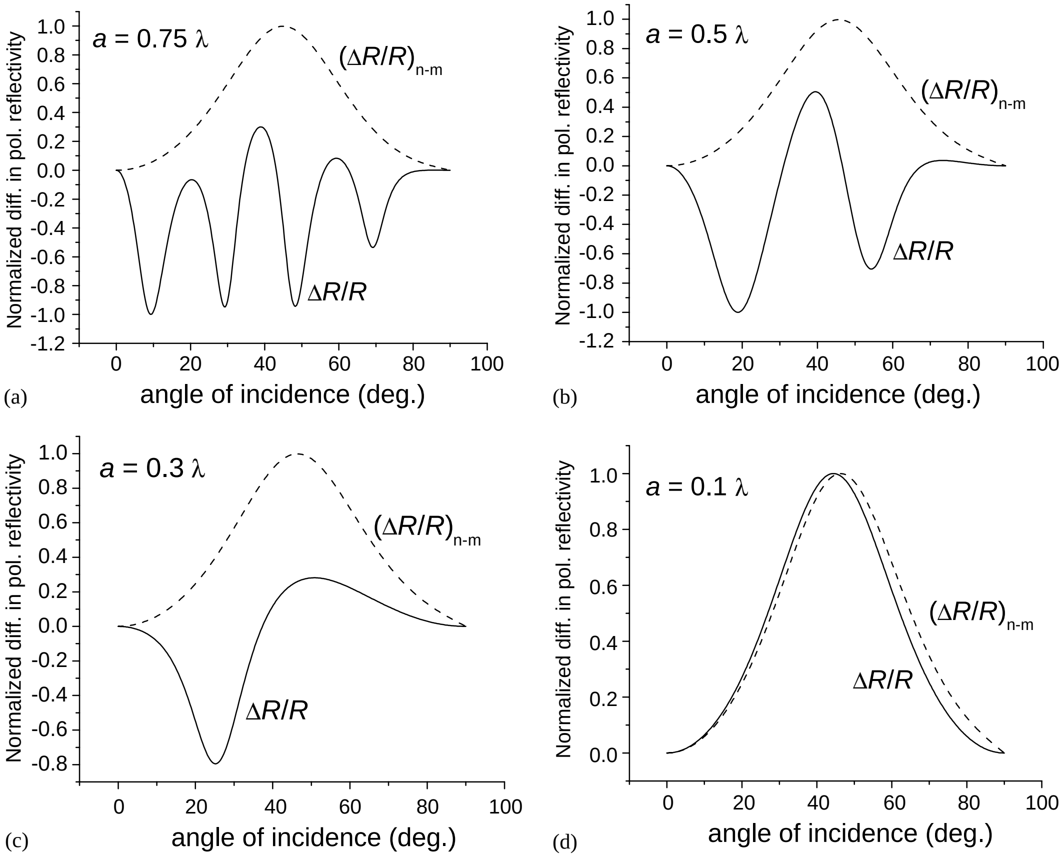

Fig. 4. Normalized difference in coherent reflectivity for linearly polarized light, $\Delta R/R = (R^{\mathrm{TE}} - R^{\mathrm{TM}})/(R^{\mathrm{TE}} + R^{\mathrm{TM}})$, versus the angle of incidence (full curves) for a few values of the radius of the particles, a. The curves predicted by the non-magnetic approximation, $(\Delta R/R)_{\text{n-m}} = (R_{\text{n-m}}^{\mathrm{TE}} - R_{\text{n-m}}^{\mathrm{TM}})/(R_{\text{n-m}}^{\mathrm{TE}} + R_{\text{n-m}}^{\mathrm{TM}})$, are also shown (dashed curves).

continuity of the tangential components of the coherent fields at the interface together with r was used to calculate the effective magnetic permeability and permittivity tensors. All quantities are given in terms of the elements of the amplitude scattering matrix (S_1 and S_2), and then, the coherent fields inside the half-space can be established correct to order ρ^1. These effective optical coefficients are functions of the angle of incidence because they are linked to the shape of the half-space. The present work show the need of an effective magnetic permeability, even when the particles are non-magnetic. The effective magnetic permeability becomes important, even if the particles are non-magnetic, as the radius of the particles increases. The effective optical coefficients of this work coincide, at normal incidence, with those given by Bohren in 1986 [11] and they may be regarded as an extension of Bohren's expressions. As a matter of fact, we have shown that in spite of Bohren's assertions about the questionable use of the effective-medium concept for large inclusions and off-normal incidence, it is possible to find expressions for effective optical coefficients that can be used in continuum electrodynamics, at least in a *restricted* manner. Extensions to particle size distributions as well as to inclusions with other shapes, is straightforward. Apparently, the applicability of effective medium theory to model the scattering from composite objects is limited by the fact that the optical coefficients will in general depend on the shape of the scattering object, as is indicated here by the angle dependence of the scattering coefficients. This has already been noted by Wachniewski and McClung [5] in 1986. In this sense the effective responses here derived should be called *restricted*, although we believe that further insight into this question is actually needed.

We have also derived [15] the same results presented here with a more intuitive approach, and it is possible to show that the physical origin of the appearance of the effective magnetic permeability may be explained as due to the existence of effective closed currents induced within the particles. Although the present approach does not provide a direct insight into the physical origin of the magnetic effect, it does provide a standard framework to pursue higher order approximations for denser media. The main contribution of the present work is to provide a formal and relatively simple derivation of the effective optical coefficients for a dilute suspension of particles, even if their size is not small compared to the wavelength of the incident radiation.

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