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Effective electric and magnetic susceptibility of dilute systems of dielectric and metallic Mie particles ☆

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Abstract

Based on a recent effective medium theory, applicable to the case of spherical particles of radius comparable or larger than the wavelength, we compare the behavior of the effective optical coefficients of random systems of transparent-dielectric and metallic particles as a function of the particles radius. We show numerical calculations and discuss the appearance of negative imaginary parts in either the effective electric or magnetic susceptibility in a system of Mie particles.

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1. Introduction

The description of the interaction of light with systems composed by a collection of randomly located inclusions embedded in an otherwise homogeneous matrix is commonly done using effective-medium theories. In these theories, the system is replaced by an equivalent, or *effective*, homogeneous medium, with effective optical coefficients, that is, an effective electric permittivity and an effective magnetic permeability. Up to now, effective medium theories have been limited to particles small compared to the wavelength of the incident radiation. The best-known example of such a theory is the one of Maxwell Garnett [1] (MG). Some years ago, extended MG theories in which the wavelength inside the particle can be small, as compared to the dimensions of the particles, have been proposed [2–6]. Basically, in these theories, the static polarizability of the inclusions that appears in the MG theory is replaced by a dynamic one. However, they are still limited to particles that are small with respect to the wavelength of light within the matrix. We recall that when the size of the particles is comparable or larger that the wavelength of light

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within the matrix (large particles), light gets scattered by the randomly located inclusions, and the electromagnetic field within the system can then be split in two components: an average component, sometimes called the coherent component, and a diffuse component coming from the random scattering. When the particles are small with respect to the incident radiation, the power carried by the diffuse component is so much smaller that the one carried by the coherent component, that its contribution is usually neglected in the traditional treatment of continuum electrodynamics. However, in the case of large particles, the power carried by the diffuse component can be comparable or even larger than the one carried by the coherent component, and in this case the concept of an effective medium is related only to the behavior of the coherent component. In recent works [7,8], we have derived expressions for effective optical coefficients for a slab composed of a dilute, random system of spherical particles, which are valid when the radius of the particles is comparable or larger than the wavelength of the incident radiation. These expressions are given in terms of the scattering properties of an isolated sphere, limiting their validity to dilute systems. An interesting effect that appears is that an effective magnetic permeability different from one appears even if the constituents of the system are non-magnetic. Here we investigate how do the relative effective electric and magnetic susceptibility of systems composed by collections of metallic and dielectric particles compare to each other, as the particle radius increases.

2. Effective optical coefficients

We consider a dilute random distribution of spherical particles in vacuum (no matrix) contained in a slab region parallel to the XY plane, boundless in this plane, and constrained to $0<z<d$. The system is in the presence of an incident plane wave with an electric field given by $\mathbf{E}^{\mathrm{i}}(\mathbf{r},t) = E_0 \exp \mathrm{i}(\mathbf{k}^{\mathrm{i}} \cdot \mathbf{r} - \omega t)\,\hat{\mathbf{e}}_{\mathrm{i}}$, where $\mathbf{r}$ and t are the position vector and time, respectively, ω is the radial frequency, $\hat{\mathbf{e}}_{\mathrm{i}}$ is a unit vector in the direction of polarization, $\mathbf{k}^{\mathrm{i}} = k_y^{\mathrm{i}}\hat{\mathbf{a}}_y + k_z^{\mathrm{i}}\hat{\mathbf{a}}_z$ is the incident wave vector assumed to lie on the YZ plane, and $\hat{\mathbf{a}}_x$, $\hat{\mathbf{a}}_y$, and $\hat{\mathbf{a}}_z$ are unit vectors along the axes of Cartesian coordinates. The electric field satisfies $\hat{\mathbf{e}}_{\mathrm{i}} \cdot \mathbf{k}^{\mathrm{i}} = 0$, and $|\mathbf{k}^{\mathrm{i}}| = k$, 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. We will be using the SI system of units.

It has been shown that the behavior of the coherent component of the electromagnetic field can be described by an effective medium with a relative effective electrical permittivity $\tilde{\varepsilon}_{\mathrm{eff}} \equiv \varepsilon_{\mathrm{eff}}/\varepsilon_0$ and a relative effective magnetic permeability $\tilde{\mu}_{\mathrm{eff}} \equiv \mu_{\mathrm{eff}}/\mu_0$ that are polarization dependent, and are given by [7,8]

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

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

$$\tilde{\varepsilon}_{\mathrm{eff}}^{\mathrm{TM}}(\theta_{\mathrm{i}}) = 1 + \mathrm{i}\gamma \frac{S_-^{(2)}(\theta_{\mathrm{i}})}{\cos^2\theta_{\mathrm{i}}}, \tag{3}$$

$$\tilde{\mu}_{\mathrm{eff}}^{\mathrm{TM}}(\theta_{\mathrm{i}}) = 1 + \mathrm{i}\gamma[2S_+^{(2)}(\theta_{\mathrm{i}}) - S_-^{(2)}(\theta_{\mathrm{i}})\tan^2(\theta_{\mathrm{i}})], \tag{4}$$

where

$$S_+^{(m)}(\theta_{\mathrm{i}}) \equiv \tfrac{1}{2}[S(0) + S_m(\pi - 2\theta_{\mathrm{i}})], \tag{5}$$

$$S_-^{(m)}(\theta_{\mathrm{i}}) \equiv S(0) - S_m(\pi - 2\theta_{\mathrm{i}}) \tag{6}$$

with $m = 1$ or 2, and

$$\gamma \equiv 3f/2x^3, \tag{7}$$

where f is the volume filling fraction of the spheres, and $x = ka$, is the size parameter where a is the particles radius. Here $S(0)$ is the forward scattering amplitude and $S_m(\pi - 2\theta_{\mathrm{i}})$ are the diagonal components of the scattering matrix. The effective optical coefficients can also be written as diagonal tensors as shown in Ref. [8]. It is not difficult to show [7,8] that for small particles, when the angular distribution of the scattered field becomes isotropic, the above effective optical coefficients coincide with the low-density limit of the extended Maxwell–Garnet theories; and for normal incidence they also coincide with the expressions proposed by Bohren [9]. Note that the effective optical coefficients $\tilde{\varepsilon}_{\mathrm{eff}}$ and $\tilde{\mu}_{\mathrm{eff}}$ depend on the angle of incidence and on the polarization, and therefore, they are not unrestricted. They are restricted to the slab geometry. These expressions are linear in γ and they are valid only in dilute systems. The effective index of refraction is given by $n_{\text{eff}}(\theta_{\text{i}}) = \sqrt{\tilde{\varepsilon}^{(m)}_{\text{eff}}(\theta_{\text{i}})\tilde{\mu}^{(m)}_{\text{eff}}(\theta_{\text{i}})} \approx 1 + \text{i}\gamma S(0)$, which is isotropic and independent of polarization. The effective electric susceptibility and effective magnetic susceptibility for either polarization, are given by

$$\chi^{\text{E}}_{\text{eff}} = \tilde{\varepsilon}_{\text{eff}} - 1, \tag{8}$$

$$\chi^{\text{H}}_{\text{eff}} = \tilde{\mu}_{\text{eff}} - 1. \tag{9}$$

Substituting the above effective optical coefficients in Fresnel relations yield the coherent reflection amplitude of a half-space [7,8]. With these expressions, one can calculate the reflection and transmission amplitudes of slabs of different thickness composed by the inhomogeneous material.

3. Transparent dielectric versus metallic particles

We will consider random systems of spherical particles made of TiO_2 (rutile) and copper (Cu) in vacuum. We will take a value of the index of refraction corresponding to the red part of the spectrum in both types of particles, this is $n_{TiO_2} \simeq 2.8$ and $n_{\text{Cu}} \simeq 0.21 + \text{i}4.05$, and we will take these values as characterizing a typical transparent dielectric and a metal. In Fig. 1 we show the contribution from the particles to the real and imaginary parts of the effective index of refraction divided by the filling fraction of the spheres, as a function of the relative size of the particle, that is, as a function of a/λ, where a is the radius of the particle and λ is the wavelength of radiation in vacuum. We must recall that these results are valid only for $f \ll 1$. As it was already pointed out in the previous section, the effective optical coefficients are functions of the angle of incidence and of polarization. Here we show the dependence of the effective electric and magnetic susceptibility, $\chi^{\text{E}}_{\text{eff}}/f$ and $\chi^{\text{H}}_{\text{eff}}/f$, as a function of a/λ for two angles of incidence, $\theta_{\text{i}} = 45°$ and $\theta_{\text{i}} = 89°$. Let us recall that we have chosen the index of refraction of the constituent materials at a fixed λ in the red side of the spectrum, thus our plots should be read as functions of the radius. Plots of $\chi^{\text{E}}_{\text{eff}}/f$ and $\chi^{\text{H}}_{\text{eff}}/f$ as a function of the angle of incidence are shown in Ref. [7,10]. In Fig. 2 we plot the real and imaginary parts of $\chi^{\text{E}}_{\text{eff}}/f$ and $\chi^{\text{H}}_{\text{eff}}/f$ in TE polarization as a function of the relative size of the particles. In Fig. 3 we present the same plots but for TM polarization.

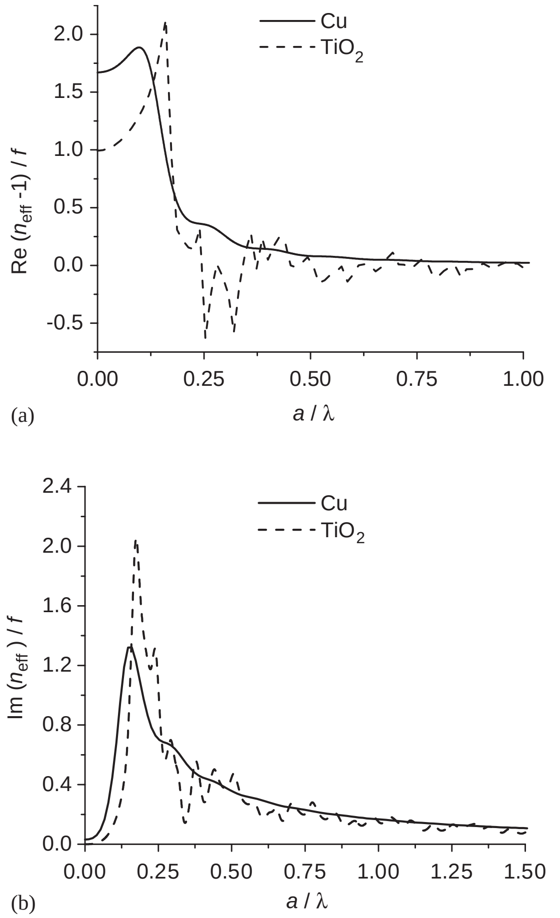

Fig. 1. Plot of the contribution of the particles to the (a) real and (b) imaginary parts of the effective refractive index normalized to the filling fraction of the spheres. The full curves are for copper particles and dashed curve for TiO_2 (rutile) particles. The indexes of refraction used are $n_{TiO_2} \simeq 2.8$ and $n_{\text{Cu}} \simeq 0.21 + \text{i}4.05$, which correspond to red light.

We make now some definite observations related to the difference in behavior between the systems of metallic and dielectric particles chosen here.

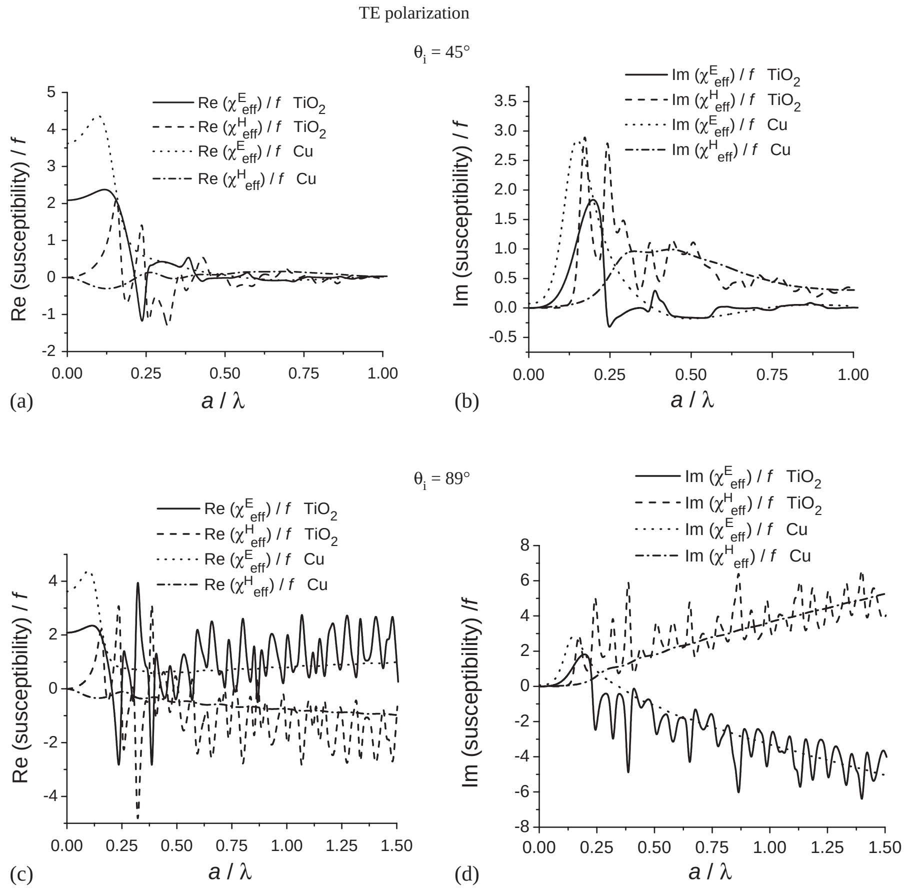

Fig. 2. Plots of the real and imaginary parts of the normalized electric and magnetic susceptibility for two different angles of incidence. Full lines are for $\mathrm{Re}(\chi_e)/f$ for the case of TiO_2 particles, dashed lines are for $\mathrm{Re}(\chi_h)/f$ for the case of TiO_2 particles, dotted lines are for $\mathrm{Re}(\chi_e)/f$ for the case of Cu particles, and dash-dot lines are for $\mathrm{Re}(\chi_h)/f$ for the case of Cu particles. Plots (a) and (b) are for $\theta_i = 45°$, and (c) and (d) are for $\theta_i = 89°$. All plots are for TE polarization.

1. The ripple, or resonance structure of the plots shown in the figures above, is smoothed out in the curves corresponding to metallic particles because the mode resonances are much wider than those of the transparent dielectric particles due to the large imaginary part of the refractive index.

2. As it is well known, even if the real part of the refractive index of a metal is less than one, the real part of the effective refractive index of this system of metallic particles is larger than one. This is due to the appearance of new resonances in the constrained spherical geometry.

3. The curves for the imaginary part behave similarly for both types of particles, except for the strong resonance structure in the curves for rutile. Because of the resonance peaks, the maxima in the curves for rutile particles are larger than for the metallic particles. In the curves for Cu, we can see that, except for the lack of ripple structure, the scattering of light plays a more important role than the absorption in relation to the attenuation of the coherent wave. Of course, for the diffuse light the scenario is very different.

4. We see that in the limit $a \to 0$, the imaginary part of the effective index of refraction remains finite for the metallic particles system, and this is due only to absorption.

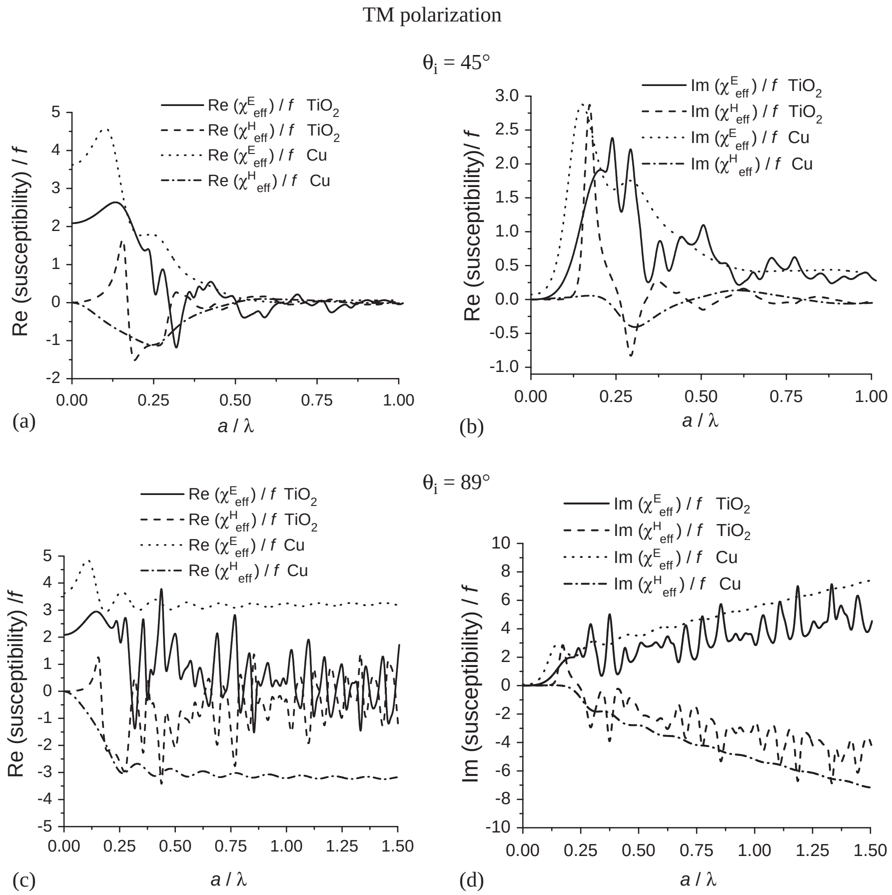

Fig. 3. Plots of the real and imaginary parts of the normalized electric and magnetic susceptibility for two different angles of incidence. Full lines are for $\mathrm{Re}(\chi_e)/f$ for the case of TiO_2 particles, dashed lines are for $\mathrm{Re}(\chi_h)/f$ for the case of TiO_2 particles, dotted lines are for $\mathrm{Re}(\chi_e)/f$ for the case of Cu particles, and dash-dot lines are for $\mathrm{Re}(\chi_h)/f$ for the case of Cu particles. Plots (a) and (b) are for $\theta_i = 45^\circ$, and (c) and (d) are for $\theta_i = 89^\circ$. All plots are for TM polarization.

5. In the case of small particles (for $a/\lambda \lesssim 0.1$), we can see that the real part of the effective magnetic susceptibility changes in opposite direction in one type of particles with respect to the other type as a/λ increases.

4. Energy balance

Perhaps, what is more striking in the previous plots, is that the imaginary part of either effective susceptibility takes negative values. In common materials it is argued that the imaginary part of both, the electric and magnetic susceptibility, must be positive [11]. The reason is that the energy lost is transformed into heat, and the second law of thermodynamics require both imaginary parts to be positive at all frequencies. However, this argument does not apply in the present case, because, part, or even all, of the energy lost from the coherent wave is due to scattering, and thus, it is transformed into diffuse radiation. Now, this process does not involve heat generation, and therefore, there is no inconsistency in having negative imaginary parts of either $\chi_{\mathrm{eff}}^{\mathrm{E}}$ or $\chi_{\mathrm{eff}}^{\mathrm{H}}$. However, what now must be satisfied, is that the sum of the imaginary parts of $\chi_{\mathrm{eff}}^{\mathrm{E}}$ and $\chi_{\mathrm{eff}}^{\mathrm{H}}$ must be positive at all frequencies. This assures the attenuation of the coherent wave as it travels through the random medium.

From Eqs. (1)–(4), we can see that,

$$\mathrm{Im}[\tilde{\varepsilon}_{\mathrm{eff}}^{\mathrm{TE}}(\theta_{\mathrm{i}})+\tilde{\mu}_{\mathrm{eff}}^{\mathrm{TE}}(\theta_{\mathrm{i}})] = \mathrm{Im}[\tilde{\varepsilon}_{\mathrm{eff}}^{\mathrm{TM}}(\theta_{\mathrm{i}})+\tilde{\mu}_{\mathrm{eff}}^{\mathrm{TM}}(\theta_{\mathrm{i}})] = 2\gamma\,\mathrm{Re}[S(0)]. \quad (10)$$

Other way to understand this equation, is by showing that in the dilute limit, $n_{\mathrm{eff}} = 1+\frac{1}{2}(\chi_{\mathrm{eff}}^{\mathrm{E}}+\chi_{\mathrm{eff}}^{\mathrm{H}})$. Thus, $\mathrm{Im}(\tilde{\varepsilon}_{\mathrm{eff}}+\tilde{\mu}_{\mathrm{eff}}) = 2\,\mathrm{Im}(n_{\mathrm{eff}})$. Then, if we were to plot $\mathrm{Im}[\chi_{\mathrm{eff}}^{\mathrm{E}}+\chi_{\mathrm{eff}}^{\mathrm{H}}]$ versus a/λ from the data in Figs. 2 and 3, we would obtain the curves in Fig. 1a but multiplied by a factor of 2. Now, the extinction cross section of a single particle is given by $C_{\mathrm{ext}} = (4\pi/k^2)\,\mathrm{Re}[S(0)]$ [12], and, as it is well known, C_{ext} determines the losses due to scattering and absorption. Therefore, $\mathrm{Re}[S(0)]$, and thus $\mathrm{Im}[\chi_{\mathrm{eff}}^{\mathrm{E}}+\chi_{\mathrm{eff}}^{\mathrm{H}}]$, is always positive.

5. Discussion and conclusion

The observations made above with respect to the behavior of the real part of $\chi_{\mathrm{eff}}^{\mathrm{E}}$ and $\chi_{\mathrm{eff}}^{\mathrm{H}}$ for small particles (for $a/\lambda \lesssim 0.1$), can be understood on physical grounds as follows. The physical origin of the effective electric and magnetic response can be explained as due to effective open and closed currents in the effective medium [7]. Now, these effective open and closed currents must arise as an average over the currents within the particles. If the particles are non-magnetic, the current lines inside the particles consists of induced polarization or displacement currents, which are proportional to the electric field lines through the electric susceptibility of the particles, $\mathbf{J}_{\chi} = -\mathrm{i}\omega\varepsilon_0(\chi_{\mathrm{e}}'+\mathrm{i}\chi_{\mathrm{e}}'')\mathbf{E}$. For transparent materials we have that $\chi_{\mathrm{e}}''=0$. In metallic particles, we can have, in addition, large conduction current lines which are proportional to the electric field lines through the complex conductivity of the particles, $\mathbf{J}_{\sigma} = (\sigma'+\mathrm{i}\sigma'')\mathbf{E}$. Now, in the Drude model of the conductivity [12] we have $\sigma''>0$. Then, comparing $\mathbf{J}_{\chi}$ and $\mathbf{J}_{\sigma}$, we observe the following equivalences, $\omega\varepsilon_0\chi_{\mathrm{e}}'' \rightleftharpoons \sigma'$, and $\omega\varepsilon_0\chi_{\mathrm{e}}' \rightleftharpoons -\sigma''$. Thus, if the medium is conducting, the real part of the conductivity causes currents equivalent to those due to the imaginary part of the electric susceptibility in a lossy dielectric, whereas the imaginary part of the conductivity causes currents opposite to those due to the real part of the electric susceptibility of a dielectric. In the case of particles not too large ($a/\lambda \lesssim 0.1$), we can see from Figs. 2 and 3 that the real part of the effective electric susceptibility in both systems of TiO_2 and Cu particles is positive. Therefore we must have that the microscopic electric field lines (the electric field lines inside the particles) leading to the effective open currents, are on the average in opposite directions in metallic and dielectric particles (they are out of phase by π). This is because of the strong screening of the external field by the surface charges on the particle. However, the real parts of the magnetic susceptibility for both types of particles are of opposite sign in the case of small particles. This means that the microscopic electric fields, leading to the effective closed currents, are on the average, in phase in both types of particles. These fields are not strongly affected by the screening charges.

In summary, we have found that in general terms the effective magnetic and electric susceptibilities of dielectric and metallic Mie particles show similar qualitative features with numerical values in the same order of magnitude. The main difference is the smoothing of the resonance, or ripple structure in the curves for the metallic particles. This means that in relation to the behavior of the coherent wave, there is not a strong difference between a system of metallic or dielectric spherical particles. This does not mean, however, that the "appearance" of the metallic and dielectric systems of spheres are similar. The appearance is more directly linked with the diffuse light rather than to the coherent light. For diffuse light, one would find strong qualitative difference due to absorption. For non-spherical particles and special 'designs' of particles, the behavior of the effective optical coefficients may be very different (see for example Ref. [13]). In addition, we argued that the effective optical coefficients, remain consistent with effective-medium theories, even if the electric or magnetic susceptibilities take negative values in some range of parameters. We showed, that the sum of the imaginary parts is always positive, leading to the attenuation of the coherent field.

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