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Extracting Metamaterial Properties of Negative-Index and Plasmonic scatterers from the Mie Coefficients

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***Abstract*— An alternative theoretical approach is presented for improving the understanding of the scattering properties of simple negative-index and plasmonic spheres. The Mie scattering coefficients are broken up into two terms, the first representing the expected fields of a corresponding positive-index sphere, the second emerging from the intrinsic metamaterial nature of the scatterer. Some limiting cases are considered, e.g. dipole approximation), together with their circuital representation in terms of equivalent lumped elements. This formalism may be adopted, for example, in the evaluation of the particular internal and scattered electromagnetic field patterns observed for these particular classes of particles.**

***Index Terms*—** Metamaterials, Mie scattering, Negative index materials, Plasmonics.

I. INTRODUCTION

Metamaterials are one of today's most promising and growing research fields. From Engineering and Physics to Medicine and Biology, plasmonics and negative refractive index (NRI) materials have received great attention from scientific communities worldwide due to their intriguing and sometimes unexpected response to electromagnetic fields and waves. Applications such as perfect flat lenses, subwavelength waveguides and resonant cavities, nano-antennas and nano-resonators, photonic switches and gratings, nano-circuits with lumped elements, cancer treatment, imaging and sensing have turned these artificial classes of engineered-structured materials into one of the biggest scientific breakthroughs and achievements of our century [1-9].

The scattering properties of spherically symmetric metamaterial structures, regardless of the optical regime assumed, are exactly treated by the Lorenz-Mie theory and, for arbitrary shaped beams, by its generalized versions [10-13]. In this particular formalism – based on expansions of the electromagnetic fields in terms of spherical harmonic functions – the characteristics of the scattered and internal fields comes from the knowledge of the Mie coefficients (MCs) and, even though a direct physical significance may not be readily attributed to them, their determination is of utmost importance in the calculation of all the corresponding physical quantities of interest [12].

Let us consider, for a moment, the refraction/reflection properties of a single ray in geometrical optics when losses are absent. When a light ray reaches an interface between a positive refractive

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index (PRI) and a NRI medium, one observes (in comparison with a PRI-PRI interface) the inversion of Snell's law [1]. Therefore, if θ_r is the transmitted angle, there is a $2\theta_r$ offset between the transmitted rays on PRI-NRI and PRI-PRI interfaces possessing the same (absolute) electromagnetic parameters, and we can naturally ask ourselves what are the additional properties or constraints of a PRI-NRI interface responsible for this offset given that the refracted ray along a PRI-PRI is known. The same may be asked for scatterers in the Mie or Rayleigh optical regimes in terms of EM fields.

Since the electric (**E**) and magnetic (**H**) fields are the fundamental quantities associated with light scattering phenomena, one can answer the above questions by exploiting the additional fields ($\mathbf{E}_{add}$,$\mathbf{H}_{add}$) that emerge from the replacement PRI-PRI → PRI-NRI, i.e., when the relative refractive index M of the particle changes sign. This problem, in turn, should be equivalent to the expansion of MCs in such a way that these additional fields come to light in a natural fashion. Specifically, because of the linear relation between (**E**,**H**) and MCs, we should be able to rewrite such coefficients (without introducing any new poles or singularities) as a sum of two terms: the first being the expected one for a PRI particle, the second the responsible for the emergence of $\mathbf{E}_{add}$ and $\mathbf{H}_{add}$.

This work is therefore devoted to the task of providing an alternative theoretical formalism for the understanding of the scattering properties of NRI spheres in terms of such additional contributions which arises exclusively from the metamaterial nature of the scatterers. It may be of interest in the theoretical and numerical analysis of optical forces and torques, EM fields, scattering amplitudes and cross-sections and any other optical parameter or property of interest. It may be further exploited in future works by finding equivalent Debye series, thus helping elucidating some physical aspects of such additional MCs.

II. Theoretical Background

In the framework of the Lorenz-Mie theory (LMT), there is a linear relationship between EM fields (**E**,**H**) and MCs [12]. This simply means that splitting MCs into two additive terms is equivalent to doing the same with (**E**,**H**) and, therefore, we shall focus our analysis only on MCs. According to one of our previous works, they can be compactly written, for a PRI spherical scatterer, as [15]

$$\left\{\begin{matrix} a_n^{PRI} \\ b_n^{PRI} \end{matrix}\right\} = \frac{A\psi_n(\alpha)\psi_n'(\beta) - B\psi_n'(\alpha)\psi_n(\beta)}{A\xi_n(\alpha)\psi_n'(\beta) - B\xi_n'(\alpha)\psi_n(\beta)}, \qquad \left\{\begin{matrix} \text{TM} \\ \text{TE} \end{matrix}\right. \tag{1}$$

where n is a positive integer, $\alpha = ka$, $\beta = k_{sp}a$, k and k_{sp} being, respectively, the wave numbers of the host medium and the particle of radius a. Besides,

$$A = \begin{cases} 1, & \text{TM} \\ N, & \text{TE} \end{cases}; \qquad B = \begin{cases} N & \text{TM} \\ 1, & \text{TE} \end{cases} \tag{2}$$

with $N = \eta/\eta_{sp}$ being the ratio between the intrinsic impedances of the external medium (η) and of the particle (η_{sp}). The Ricatti-Bessel functions (RBFs) and their derivatives (with respect to their

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arguments) $\psi_n(.), \xi_n(.), \psi'_n(.), \xi'_n(.)$ are written using current terminology [10]. In order to proceed further, the following relations for the RBFs, valid for real *x*, are introduced [16]:

$$\begin{aligned} \psi_n(-x) &= -(-1)^n \psi_n(x) \\ \psi'_n(-x) &= +(-1)^n \psi'_n(x) \end{aligned} \quad \text{and} \quad \begin{aligned} \xi_n(-x) &= -(-1)^n \Gamma_n(x) \\ \xi'_n(-x) &= +(-1)^n \Gamma'_n(x) \end{aligned} \tag{3}$$

The wave number of a lossless NRI particle is negative, so $\beta = -|k_{sp}|a = -|\beta| = -\omega(|\mu||\varepsilon|)^{1/2}$ and the set of relations (3) may be substituted in (1) (had we assumed a PRI particle, then $\beta = +|\beta|$; α is taken as positive real number):

$$\begin{Bmatrix} a_n^{NRI} \\ b_n^{NRI} \end{Bmatrix} = \frac{A\psi_n(\alpha)\psi'_n(|\beta|) + B\psi'_n(\alpha)\psi_n(|\beta|)}{A\xi_n(\alpha)\psi'_n(|\beta|) + B\xi'_n(\alpha)\psi_n(|\beta|)}, \qquad \begin{cases} \text{TM} \\ \text{TE} \end{cases} \tag{4}$$

Just as for a single ray impinging on the surface of a NRI particle, we can ask ourselves what is the “pure” NRI contribution to the EM fields arising from the changes of sign in (3) which modify (1) in accordance with (4). Because EM fields are directly proportional to MCs, the answer to this question comes from rewriting (4) as follows:

$$\begin{Bmatrix} a_n^{NRI} \\ b_n^{NRI} \end{Bmatrix} = \begin{Bmatrix} a_n^{PRI} \\ b_n^{PRI} \end{Bmatrix} + \begin{Bmatrix} a_n^{add} \\ b_n^{add} \end{Bmatrix} \tag{5}$$

For instance, consider the scattered electric field components in spherical coordinates (r,θ,ϕ) for TM modes,

$$E^s_{r,TM} = -kE_0 \sum_{n=1}^{\infty}\sum_{m=-n}^{n} a_n c_n^{pw} g_{n,TM}^m \left[\xi''_n(kr) + \xi_n(kr)\right] P_n^{|m|}(\cos\theta) e^{im\varphi} \tag{6}$$

$$E^s_{\theta,TM} = -\frac{E_0}{r} \sum_{n=1}^{\infty}\sum_{m=-n}^{n} a_n c_n^{pw} g_{n,TM}^m \xi'_n(kr) \tau_n^{|m|}(\cos\theta) e^{im\varphi} \tag{7}$$

$$E^s_{\varphi,TM} = -i\frac{E_0}{r} \sum_{n=1}^{\infty}\sum_{m=-n}^{n} a_n m c_n^{pw} g_{n,TM}^m \xi'_n(kr) \pi_n^{|m|}(\cos\theta) e^{im\varphi} \tag{8}$$

where E_0 is the field strength, $g_{n,TM}^m$ are the beam-shape coefficients, c_n^{pw} is a constant for a given *n* and $P_n^{|m|}(.), \pi_n^{|m|}(.), \tau_n^{|m|}(.)$ are associated and generalized Legendre functions. Similar expressions hold for magnetic fields, and TE modes contain the transverse electric MCs b_n [12].

From (5)-(8) one clearly sees that the field $\mathbf{E}^s$ and $\mathbf{H}^s$ scattered by an NRI sphere are, in fact, a sum of two terms: one that comes from imposing (1) in (6)-(8) assuming a PRI scatterer with both the same geometry and absolute value of electromagnetic parameters, and a second one that emerges as an additional contribution due to the metamaterial nature of the particle.

In this way, let us explicitly evaluate a_n^{add} and b_n^{add} in (5). By subtracting (4) and (5) one finds, after some algebra,

$$\begin{Bmatrix} a_n^{add} \\ b_n^{add} \end{Bmatrix} = \frac{i2AB\psi_n(|\beta|)\psi'_n(|\beta|)}{A^2\xi_n^2(\alpha)\psi_n'^2(|\beta|) - B^2\xi_n'^2(\alpha)\psi_n^2(|\beta|)}. \tag{9}$$

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and the additional EM fields are found by replacing (a_n,b_n) by (a_n^{add}, b_n^{add}) in (6)-(8) and all other equivalent expressions for TM modes and **H** field. Notice that (5) and (9) do not explicitly involve negative refractive indices, even though we implicitly made use of such a condition in deriving (4). Due to a typo in a previous work (which nonetheless did not change the results and conclusions), the numerator in (9) is slightly different from the one found in equation (8) of [15].

The same procedure, once applied to internal fields, leads to the following expressions for the internal additional MCs:

$$\begin{Bmatrix} c_n^{add} \\ d_n^{add} \end{Bmatrix} = -i \begin{Bmatrix} |n_{sp}| \\ |\varepsilon_{rel}| \end{Bmatrix} \left\{ A\xi_n(\alpha)\psi_n'(|\beta|)\left[1+(-1)^n\right] + \frac{B\xi_n'(\alpha)\psi_n(|\beta|)\left[1-(-1)^n\right]\}}{A^2\xi_n^2(\alpha)\psi_n'^2(|\beta|)-B^2\xi_n'^2(\alpha)\psi_n^2(|\beta|)} \right\} \quad (10)$$

where n_{sp} is the refractive index of the scatterer.

These are quite interesting results by themselves. In fact, one can forget about negative refraction altogether and assert that a lossless NRI sphere is simply the one which replaces an equivalent PRI particle and modifies the EM fields according to (9) and (10). In other words, every time we are able to produce the EM fields imposed by (9) and (10) we should, in fact, create a theoretical NRI metamaterial. This is an alternative way of thinking about a NRI material in terms of scattering or internal fields.

Another possible interpretation of (9) can be appreciated as follows. Consider a given PRI sphere. If, by any means, we are capable of generating the electric and magnetic scattering fields associated with (9), such fields will superpose those scattered by the corresponding PRI particle and a far-field detector should conclude that they come from some sort of lossless NRI material with spherical symmetry.

For very small particles and far from resonances, the dipole approximation asserts that only the MCs a_1 and b_1 significantly contribute to $\mathbf{E}^s$ and $\mathbf{H}^s$. Accordingly, c_1 and d_1 are the only non-negligible internal MCs. Therefore, (9) and (10) reduce to the much simpler expressions

$$\begin{gathered} a_1^{add} = 4i\frac{|\varepsilon_{rel}|}{|\varepsilon_{rel}|^2-4}\alpha^3; \quad b_1^{add} = 4i\frac{|\mu_{rel}|}{|\mu_{rel}|^2-4}\alpha^3 \\ c_1^{add} = -\frac{6|\varepsilon_{rel}|}{|\varepsilon_{rel}|^2-4}; \quad d_1^{add} = -\frac{6|\mu_{rel}|}{|\mu_{rel}|^2-4} \end{gathered} \quad (11)$$

and we may now assume a_1^{add} and b_1^{add} in (11) to be identical to the dipole terms expected for some particular PRI scatterer:

$$\begin{Bmatrix} a_1' \\ b_1' \end{Bmatrix} = i\frac{2}{3}\frac{\gamma-1}{\gamma+2}\alpha^3 \quad (12)$$

γ being the relative permittivity (for a_1) or permeability (b_1). Equating (11) and (12) we find the conditions

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$$\gamma = \begin{cases} \dfrac{|\varepsilon_{rel}|^2 + 12|\varepsilon_{rel}| - 4}{|\varepsilon_{rel}|^2 - 6|\varepsilon_{rel}| - 4}, & \text{for } a_1' \\ \dfrac{|\mu_{rel}|^2 + 12|\mu_{rel}| - 4}{|\mu_{rel}|^2 - 6|\mu_{rel}| - 4}, & \text{for } b_1' \end{cases} \tag{13}$$

which means that, in order to have $\mathbf{E}^{s,add}$ and $\mathbf{H}^{s,add}$ generated by additional "naturally-available" PRI (magnetic and electric) dipoles, one must ensure $0 < |\varepsilon_{rel}| < 0.325$ *or* $|\varepsilon_{rel}| > 6.606$ (the same being valid for $|\mu_{rel}|$). If any of such conditions are matched then (13) ensures that the scattering field of a NRI nano-sphere can be theoretically represented by the sum of the scattering fields produced by its corresponding PRI equivalent plus those produced by a PRI dipole with dipole terms satisfying (12). Notice that references to a negative index have been suppressed and are no longer necessary. Incidentally, a NRI particle can then be viewed as a structure which simultaneously contains, in some limited region of space, two homogenous PRI structures with distinct EM parameters and without field interferences. They produce identical far-field patterns when compared to the original NRI scatterer. By the same token, repetition of the above procedure for internal fields gives, instead of (13),

$$\gamma = \begin{cases} \dfrac{-|\varepsilon_{rel}|^2 - 4|\varepsilon_{rel}| + 4}{2|\varepsilon_{rel}|}, & \text{for } c_1' \\ \dfrac{-|\mu_{rel}|^2 - 4|\mu_{rel}| + 4}{2|\mu_{rel}|}, & \text{for } d_1' \end{cases} \tag{14}$$

and so the pure metamaterial contribution to the internal fields may be obtained by placing, at the origin of the coordinate system, an hypothetical additional PRI sphere with electromagnetic parameters constrained between $0 < \mu_{rel} < 0.828$ and $0 < \varepsilon_{rel} < 0.828$. The above derivations may be extended to include lossy (active) NRI scatterers with the additional EM fields generated by such metamaterial particles theoretically originating from active (lossy) PRI structures. This can be verified by constructing the Ricatti-Bessel functions from the following identities [17]:

$$J_n(-z_R - iz_I) = J_n(-(z_R + iz_I)) \equiv J_n(-z) = (-1)^n J_n(z) \tag{15}$$

and similar for modified Bessel and Hankel functions.

In (15), $a > 0$, $b > 0$ and $z = a + ib$, i.e., z is associated with an active PRI structure. Finally, note that lossless plasmonic scatterers with arbitrary radius have analytical expressions involving Bessel functions of purely imaginary arguments. This poses additional mathematical difficulties [one can easily check, however, that for plasmonic dipoles (11)-(13) remain valid for the electric coefficient a_1]. Nonetheless, because real plasmonic materials always present absorption, this should not be a problem in modeling and studying practical situations.

In the particular case of dipole approximation, some situations may require equivalent descriptions in terms of RLC ladder networks. The MCs are then represented as combinations of lumped circuit

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elements. This has been done, for instance, for both PRI and NRI spheres [14,18]. Focusing on the additional MCs, it is easy to see that RLC networks associated with (11) are exactly those found for the PRI case [18], with the *proviso* that both permeability and permittivity satisfies (12) and (13).

Our formalism may help in deriving equivalent electrical circuits with lumped nanocapacitors, nanoinductors and nanoresistors for lossless plasmonic nanospheres [6,19]. Indeed, from Mie theory and imposing μ_{rel} = 1 in (11), the equivalent internal displacement current density can be rewritten as

$$\begin{aligned} i\omega\varepsilon E_0\left(c_1-1\right) &= i\omega\left|\varepsilon\right|E_0\left(c_1^{PRI}+c_1^{add}-1\right) \\ &= i\omega\left|\varepsilon\right|E_0\left(c_1^{PRI}-1\right)+i\omega\left|\varepsilon\right|E_0c_1^{add} \\ &= J_d^{PRI}+J_d^{add} \end{aligned} \tag{16}$$

and similarly for the average potential difference

$$\begin{aligned} \left\langle V_{sp}\right\rangle &= aE_0\left(c_1-1\right)=aE_0\left(c_1^{PRI}-1\right)+aE_0c_1^{add} \\ &= \left\langle V_{sp}\right\rangle_{PRI}+\left\langle V_{sp}\right\rangle_{add} \end{aligned} \tag{17}$$

so that, from (16) and (17), one finds the following nano-impedance:

$$Z_{sp}=Z_{sp}^{PRI}+Z_{sp}^{add}=\left(i\omega\pi a\left|\varepsilon\right|\right)^{-1}+i\left(2/\omega\pi a\left|\varepsilon\right|\right) \tag{18}$$

as expected so as to generate the inductive behavior observed for plasmonic nanoscatterers. According to (18), Z_{sp} for a dipolar plasmonic particle can be represented not only by $\left(i\omega\pi a\varepsilon\right)^{-1}$ but, instead, by a PRI nanocapacitor Z_{sp}^{PRI} plus an additional nanocapacitor Z_{sp}^{add} that comes into play because of the excitation of surface plasmons [6].

III. NUMERICAL EXAMPLES

The preceding section dealt with the theoretical background for the analysis of the scattering properties of metamaterial spheres giving emphasis to the additional fields which superpose those of PRI scatterers. As mentioned, MCs can be split into a sum of two terms, a very interesting trick considering the fact that some of the fundamental physical quantities are directly proportional to them. Even quantities like force, torque, scattering cross sections, etc, after some straightforward manipulation, may be analogously decomposed in such a form.

Let us consider the Cartesian version of (6)-(8) for a NRI sphere with $\alpha = ka = 0.01$ and $\mu_{rel} = -\mu_0$. For a +*z*-propagating (*x*-polarized) plane wave, Figs. 1 and 2 reveal the TM scattered E_x-field as composed by its additional and PRI contributions in $\theta = \pi/2$ plane (*x*-*y* plane) for a radial distance $r = 2a$ and as a function of ε_{rel} and azimuth angle ϕ. The TE field may be directly evaluated using expressions analogous to (6)-(8) [12]. In both plots, $|\varepsilon_{rel}| > 6.606$ in order to include only additional fields generated by "physical" PRI structures, as mentioned in section II.

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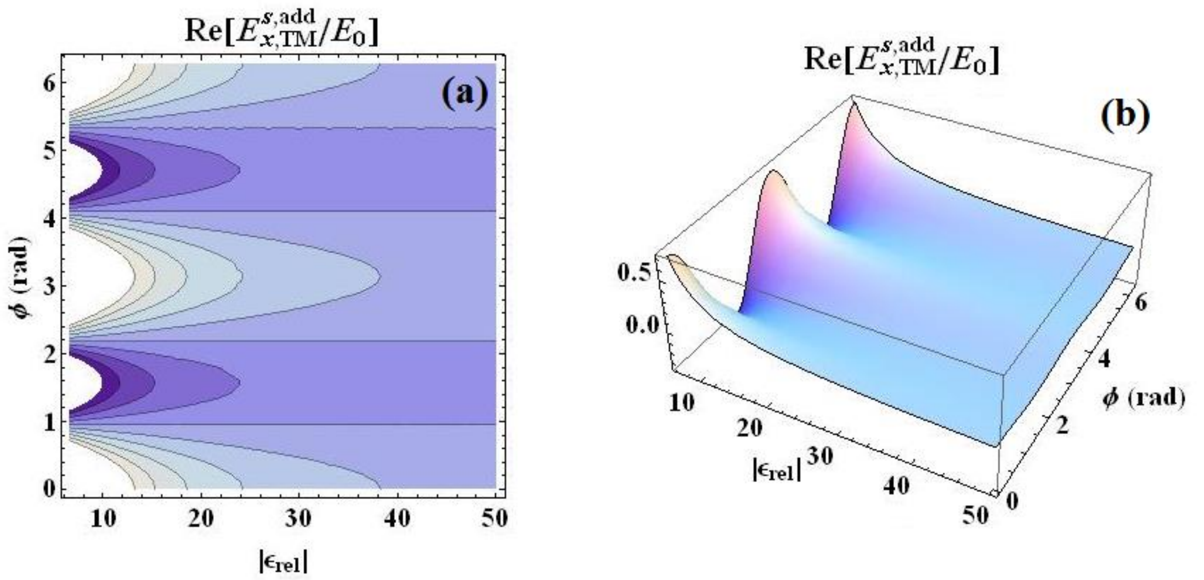

Fig. 1. Additional (pure metamaterial part of) TM E_x-field in the $\theta = \pi/2$ plane and at $r = 2a$ for a NRI sphere with $ka = 0.01$, $\mu_{rel} = -\mu_0$ under plane wave illumination. The wave propagates along $+z$ and is x-polarized.

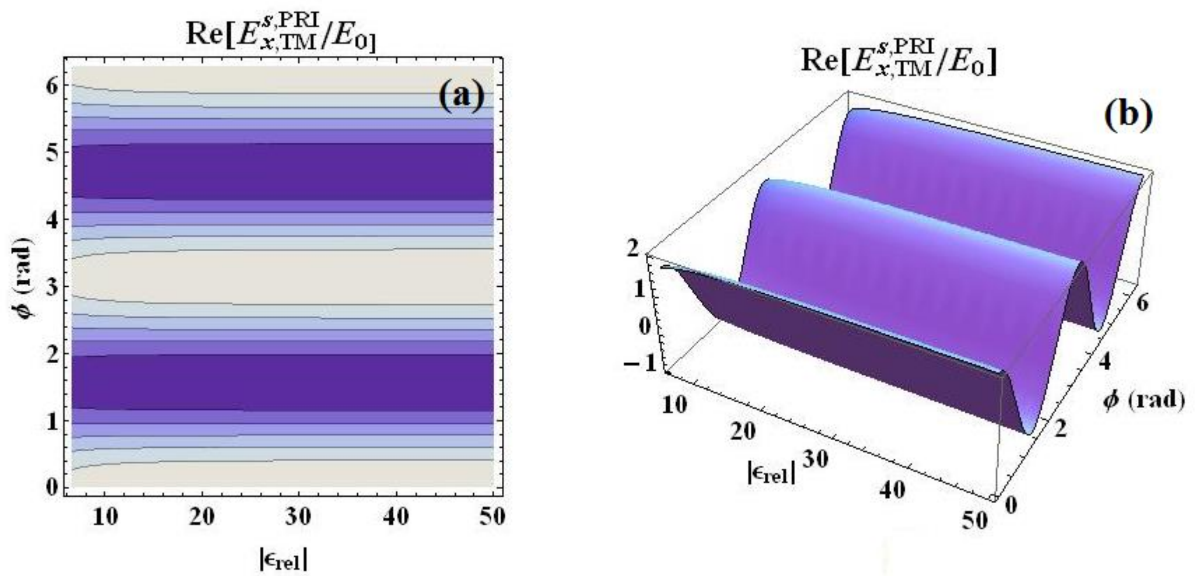

Fig. 2. PRI E_x-field counterpart for the NRI sphere of Fig. 1.

RLC ladder network circuits for the dipole coefficients also change radically for specific radius and particular relative permittivities. From (11)-(13), assuming an admittance model, the first four reactances for a_1' are

$$X_1'(\gamma) = \alpha^2 \frac{56(\gamma+1)}{3(\gamma^2+21\gamma+70)} \tag{19}$$

$$X_2'(\gamma) = 3\frac{9\gamma^2+49\gamma-210}{28(\gamma+1)} \tag{20}$$

$$X_3'(\gamma) = \frac{2(5\gamma^3+16\gamma^2+329\gamma-182)}{3(\gamma+2)(9\gamma^2+49\gamma-210)} \tag{21}$$

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$$X_4'(\gamma)=\alpha^2\,\frac{3(\gamma-1)\left(9\gamma^2+49\gamma-210\right)\left(5\gamma^3+16\gamma^2+329\gamma-182\right)}{280(\gamma+1)\left(5\gamma^3+16\gamma^2+329\gamma-182\right)} \quad (22)$$

In view of (13), (19)-(22) provide very distinct numerical outcomes as compared to those found in [18], even if their expressions look quite similar (here, in fact, the original ε_{rel} is simply replaced by γ). In fact, some of the capacitors in the original PRI RLC circuit may have to be replaced by inductors in the RLC circuit representation for a_1^{add} depending on the plus or minus signs carried by each of such passive circuit element due to the presence of additional reactances, with a direct consequence on the values of the NRI reactances as well. This is illustrated in Figs. 3 and 4 for X_1 and X_2 as functions of $|\varepsilon_{rel}|>6.606$ for α = 0.01. The corresponding electric dipole coefficients are shown in Fig. 5.

For particles whose dimensions are comparable or larger than the wavelength, (4), (5) and (9) can be used to evaluate NRI, PRI and additional Mie coefficients individually. Therefore, it is possible to study the individual properties of their corresponding scattered fields. For example, assuming λ = 1064 nm, MC a_1^{NRI} for a particle with $\mu=-\mu_0$ presents an oscillatory behavior as a function of both a and ε_{rel}, as revealed in Fig. 6. If it is broken up in its two terms as suggested by (5), then its PRI and additional dipole coefficients are as shown in Figs. 7 and 8, respectively. It becomes clear the new rearrangement of the ripples induced by virtue of a_1^{add}. Even if a direct physical interpretation cannot be readily extracted from such plots, expressing each MC as a Debye series could furnish interesting conclusions and insights for the equivalent series of reflection/refractions of a wave field at the surface of a NRI particle [15].

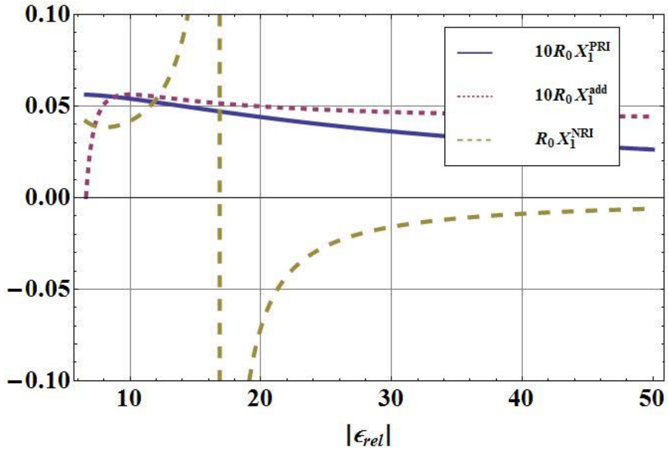

Fig. 3. First reactance X_1 (NRI, PRI and *add*) for $|\varepsilon_{rel}|$ > 6.606. It represents the first lumped element of an equivalent RLC (admittance) circuit model. For visualization purposes, multiplicative factors are included for X_1^{PRI} and X_1^{add}.

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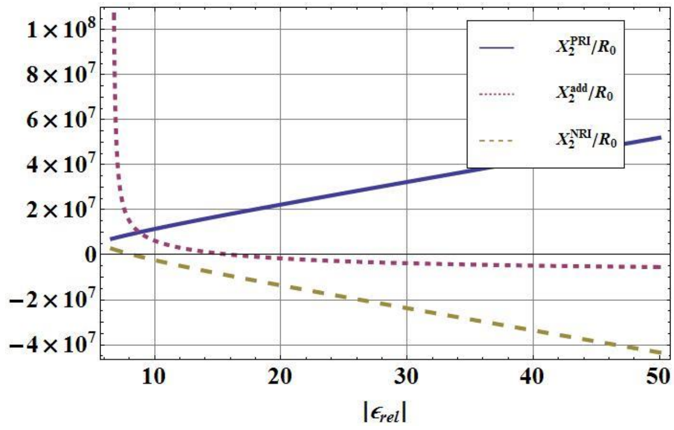

Fig. 4. Same as Fig. 3, for reactance X_2.

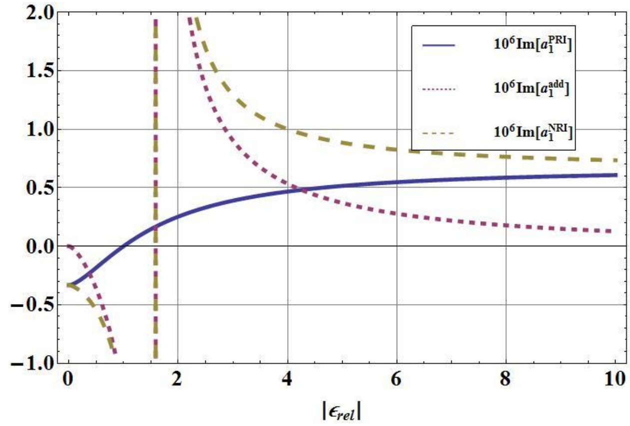

Fig. 5. Electric dipole coefficient a_1 as function of ε_{rel}.

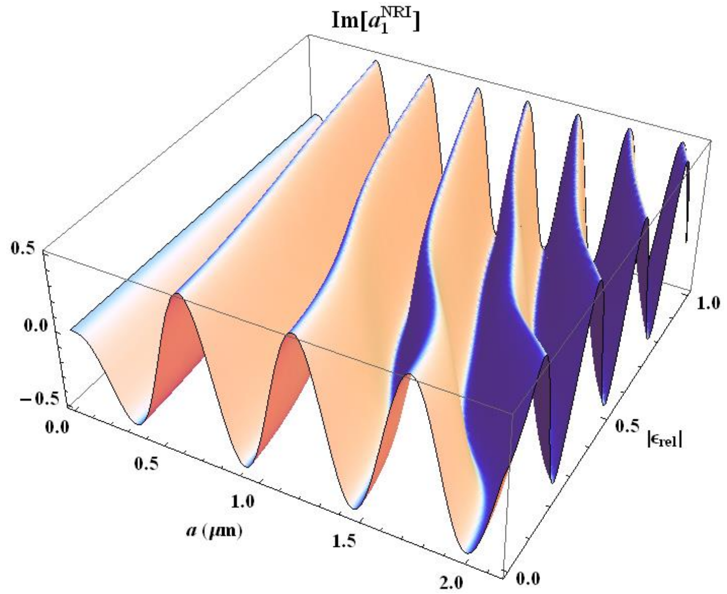

Fig. 6. a_1^{NRI} as function of ε_{rel} and a. λ = 1064 nm.

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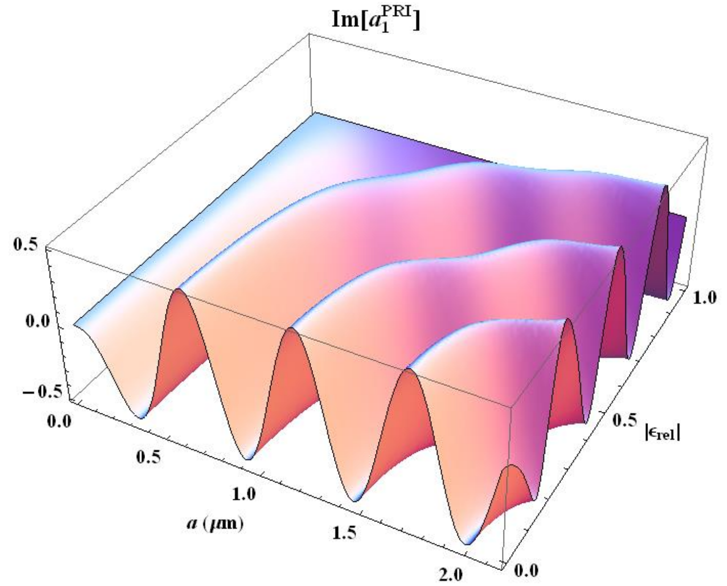

Fig. 7. PRI part (a_1^{PRI}) of a_1^{NRI} (shown in Fig. 6).

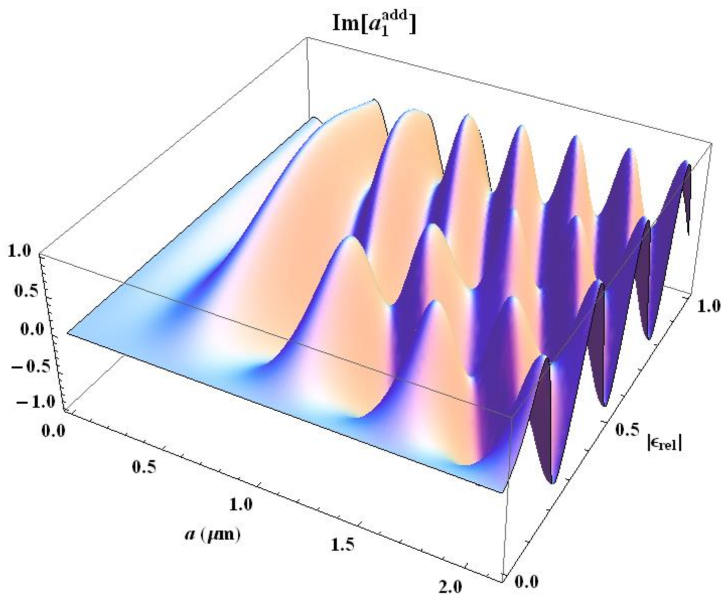

Fig. 8. Pure metamaterial or additional (a_1^{add}) part for a_1^{NRI} of Fig. 6.

IV. Conclusion

An analytical method is presented for expressing scattering properties of NRI particles as superpositions of two distinct terms: the first associated with a PRI particle carrying exactly the same absolute values of the electromagnetic parameters, the second reflecting the pure metamaterial properties of the original scatterer. Under certain conditions, the latter can also be represented as a function of (scattering) MCs representing dielectric scatterers of positive permittivity.

Within some specific range of values of permittivity and assuming Rayleigh scatterers far from any resonances, this formalism asserts that the expected NRI scattering properties may be equivalently interpreted as nothing but the result of field superpositions between two spatially coincident (or very close together) PRI particles. Additional EM fields are then the physical quantities which ultimately

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differentiate between PRI and NRI scatterers, being a sort of specific signature of a metamaterial structure.

We believe this approach could provide new ways for designing NRI and metamaterial composites from material science techniques and bottom-up approaches using effective medium theory.

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