# Metallo-dielectric core-shell nanospheres and their application as building blocks for isotropic 3d optical metamaterials

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### Abstract

Here we propose a fully 3D, isotropic metamaterial with strong electric and magnetic responses in the optical regime, based on metallo-dielectric core-shell nanospheres. The magnetic response stems from the lowest, magnetic-dipole resonance of the dielectric shell with high refractive index, and can be tuned to coincide with the plasmon resonance of the metal core, responsible for the electric response. Since the response does not originate from coupling between structures, no particular periodic arrangement needs to be imposed. Moreover, due to the geometry of the constituents, the metamaterial is intrinsically isotropic and polarization independent.

## 1. Introduction

Once the possibility of building a negative-index metamaterial (NIM) was proven in the microwave regime [1], extraordinary effort has been made to obtain analogous behaviours for increasingly higher frequencies up to the visible range of the electromagnetic spectrum. The major obstacle found when trying to translate ideas to higher frequencies was how to achieve a strong diamagnetic response in the systems designed. Many attempts to tackle the problem were simple miniaturizations of the canonical designs. Apart from the fundamental limitations inherited from those designs, e.g. anisotropy, and the increasingly complexity in the fabrication procedures, several drawbacks have been found in this process, some of them being consequence of the different behaviour of metals when excited with higher frequency waves. Moreover, in many cases, the behaviour of the proposed designs stems from coupling between the different constituents, thus making particular arrangements necessary. As a consequence, spatial dispersion effects often appear due to propagation of waves in the lattice.

Here we report a design that tackle many of the previously mentioned limitations. The use of metallo-dielectric core-shell nanospheres allows to obtain, with one single structure, both the strong diamagnetic and electric responses. Since the magnetic response stems from the magnetic Mie resonance of the dielectric shell, it is possible to avoid the high losses associated to ohmic currents. The electric response is due to the localized plasmon resonance of the metallic core. Calculations for core-shell structures built up with realistic materials (Ag@Si and Ag@Ge) demonstrate the possibility to obtain NIM operating at 1.2 $\mu$m-1.55$\mu$m [2].

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## 2. Optical properties of metallo-dielectric core-shell meta-atoms

The optical properties of a spherical core-shell under plane wave (wavelength $\lambda$) illumination can be determined without any approximation as an extension of Mie theory, as obtained first by Aden and Kerker [3]. For these particles, the extinction and scattering efficiencies can be written as:

$$Q_{sca} = \frac{2}{y^2}\sum_{l=1}^{\infty}(2l+1)(|a_l|^2+|b_l|^2), \quad Q_{ext} = \frac{2}{y^2}\sum_{l=1}^{\infty}(2l+1)\mathrm{Re}(a_l+b_l), \tag{1}$$

where $a_l$ and $b_l$ are the coefficients asssociated to the $l^{th}$ electric and magnetic multipoles in the field expansion and depend on the propoerties of both the shell and the core. Now, it is known that, for high permittivity small particles, the first Mie resonance is the magnetic dipolar [4]. High permittivity shells give additional degrees of freedom, associated to the possibility of varying the inner radius. The physical mechanism operating behind, is a strong circulation of the displacement field due to the abrupt jump of the index, as explained for a dielectric ring in [5]. Moreover, the use of shells open the possibility of adding a core with some desired properties. In our case, we can add a plasmonic core, in order to achieve a strong electric response.

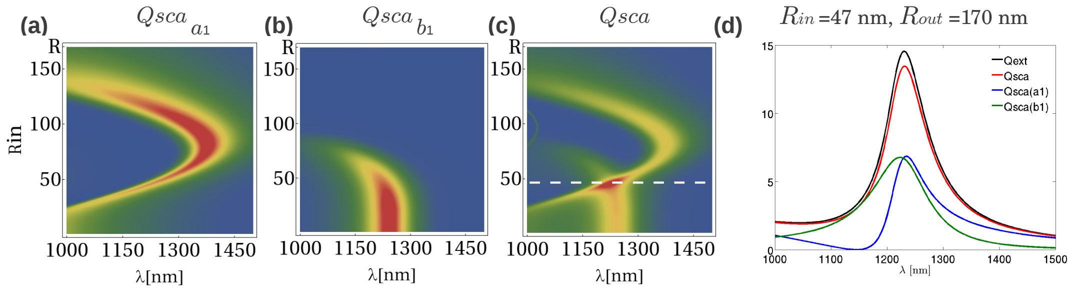

Fig. 1: Dipolar electric (a) and dipolar magnetic (b) contributions to the total scattering efficiency (c) of a Ag@Si core-shell nanosphere (radius $R = 170\ nm$), as a function of the inner radius ($R_{in}$). (d) The particular case with R =170nm and $R_{in} = \ 47nm$.

The question is, then, whether the magnetic resonance from the shell and the electic resonance from the core can co-exist. Let us take as specific materials Silicon as a high refractive index material, and Silver as the plasmonic one. Figure 1 (a)-(c) depicts both, the electric dipolar and the magnetic dipolar contributions to the scattering efficiency, together with the total scattering efficiency for a Ag@Si core-shell system of outer radius $R = 170$ nm as a function of $R_{in}$ and the incidence wavelength. It can be clearly seen an overlap between the electric and magnetic resonances within 1150 nm and 1300 nm and for inner radius between $R_{in} = 40 - 50$ nm. We have explicitly plotted the case $R_{in} = 47$ nm (Figure 1(d)). Near-field patterns at the combined magnetic (shell) and electric (core) resonance (not shown here) for the system, reveal that the distinctive behaviour of both contributions is preserved in the combined electromagnetic resonance.

## 3. Effective medium properties

For composites made of a cubic or random arrangement of dipolar particles, Lorentz-Lorenz theory is widely used, leading to the well known Clausius-Mossotti formulas relating the effective permittivity and permeability with the polarizabilities of the particles and the filling fraction. For the core-shell structures considered $R/\lambda \sim 1/7$. Therefore, we expect them to be well within the approximation considered in the theory, thus behaving essentially as electric and magnetic point dipoles. The electric and magnetic polarizabilities, $\alpha_E$ and $\alpha_M$, respectively, are directly proportional to the scattering coefficients $a_1$ and $b_1$ [factor $i(k^3/6\pi)^{-1}$]. Since both resonances are spectrally overlapping, there is a spectral region, where

$\alpha_E$ and $\alpha_M$ are simultaneously negative. Therefore, it is possible to obtain, for certain filling fractions, simulataneously negative effective permittivity and permeability.

This is shown in Figure 2, where we plot $\epsilon_{\mathtt{eff}}$, $\mu_{\mathtt{eff}}$ and $n_{\mathtt{eff}}$ for the Ag@Si core-shell system of Figure 1(d).

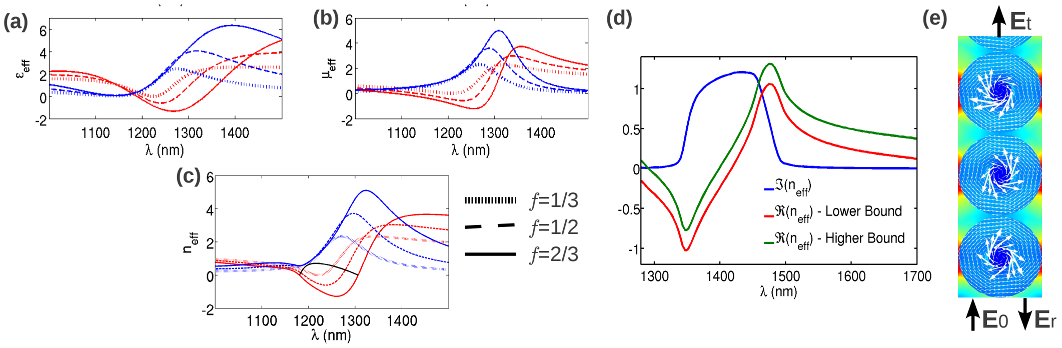

Fig. 2: Analytical effective permittivity (a), permeability (b) and refractive index (c) for different filling fractions of a metamaterial of randomly arranged a Ag@Si core-shell nanospheres with $R = 170\ nm$ and $R_{in} = 47\ nm$. (d) Effective refractive index of a simple cubic arrangement of Ag@Ge core-shells with $R = 190\ nm$ and $R_{in} = 55\ nm$, and lattice period 385 nm, extracted from full numerical simulations. (e) Detail of the norm of the electric field (colour) and displacement field (arrows) inside the periodic structure.

It is also possible to extract, using standard retrieval procedures [6], the effective medium properties of a periodic arrangement of these core-shells, while taking into account all possible coupling effects between particles. As an example, Figure 2 (d) represents the extracted parameters for a metamaterial composed by Ag@Ge core-shells with $R = 190\ nm$ and $R_{in} = 55\ nm$, arranged in a simple cubic lattice with period 385 nm.

## 4. Conclusion

Here we presented a new design based on core-shell nanospheres that operates in the near-infrared, which tackles the problems of anisotropy, polarization dependence and lack of three-dimensionality. In our system, the effective response of the metamaterial is due to every isolated “meta-atom”. Therefore, no particular arrangement of the constituents is needed. Specifically, we demonstrated with realistic materials that, for a random arrangement, the system achieves double-negative index of refraction for different filling fractions. We also tested the validity when a very simple periodic realization is assumed.

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