Scattering cancellation of the magnetic dipole field from macroscopic spheres

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Abstract: Based on the scattering cancellation technique we suggest a cloak that allows to conceal macroscopic objects, i.e. objects with an optical size comparable to wavelengths in the visible and whose scattering response is dominated by a magnetic dipole contribution. The key idea in our approach is to use a shell of polaritonic spheres around the object to be cloaked. These spheres exhibit an artificial magnetism. In a systematic investigation, where we progressively increase the complexity of the considered structure, we devise the requirements imposed on the shell and outline how it can be implemented with natural available materials.

OCIS codes: (160.3918) Metamaterials; (230.3205) Invisibility cloaks; (160.4670) Optical materials.

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1. Introduction

Metamaterials are composite structures whose properties transcend those of natural materials. Their exploration for electromagnetic applications has been a notable trend in recent years. As far as optical applications are concerned, it is anticipated that the properties of light can be controlled to an unprecedented extend by an appropriate design of the meta-atoms. The periodic or aperiodic arrangements of these meta-atoms form the metamaterial. One challenging part for designers of optical metamaterials is to find the appropriate configurations with a strong artificial magnetism in the visible. For the microwaves regime, split-ring resonators were historically the first candidates investigated in 1999 by Pendry *et al.* [1] and were used later for realizing the first invisibility cloak in the GHz regime [2]. The mechanism that made the cloak working was a geometrical transformation that maps a point into a disc (or a sphere in 3D), creating thus a zone of space which does not interact at all with its surrounding [3]. Parallel to these seminal studies, other methods to cloak objects have been investigated ranging from anomalous resonance of the so-called perfect lens [4] to homogenization of *quasi*-photonic crystals to mimic anisotropy and space dispersion [5–7]. Nevertheless, the optical cloak remained a theoretical idea because of the high losses of metals due to interband transitions in this spectral window and the technological challenges in manufacturing anisotropic and inhomogeneous meta-atoms at the nanometer scale. A theoretical simplification of the mechanism was proposed in 2008 by Li and Pendry and allowed to relax the required parameters at the expense of a reduced functionality. The carpet cloak, as it was called, allowed to hide an object in a small space above a ground plate [8]. The experimental realization of this concept came soon after, for the IR regime first [9] and the optical regime recently [10–12] where woodpile photonic crystals have been tailored to render a bump invisible to incident unpolarized light for a finite range of frequencies in the visible.

A different technique to cloak objects proposed back in 2005 by Engheta and Alù relies on matching the impedance of the surrounding media by canceling the scattering response of the object to be cloaked [13]. This method has the advantage of requiring homogeneous and isotropic cloaking shells unlike the cloaks designed within the framework of transformation optics. This allows one to think of practical applications for such devices. Referential examples would be a cloaked sensor [14] or non-invasive imaging techniques by a cloaking Scanning Optical Microscope tip [15]. Experimental evidence of such invisibility devices was demonstrated in the microwaves regime for cylinders [16] and, more recently, for spheres [17].

Moreover, it has been shown how to use clusters of silver nanospheres as cloaking shells by arranging them in a random manner at the surface of a dielectric sphere [18]. In addition to the increased degrees of freedom, i.e. changing the filling fraction allows to adjust the frequency of operation, the composite cloak has been shown to be superior when compared to a continuous metal-film as the shell material. This results from the fact that parasitic absorption was reduced in the specific configuration. Moreover, the device was a prime example where an analytical analysis considering the shell as an effective medium could be compared to full-wave simulations that took into account all the individual silver nanospheres the shell was made of. Excellent agreement between both approaches provided evidence for the applicability of effective material parameters to describe the shell. In Ref. [19, 20], an epsilon-near-zero artificial material was suggested to cloak cylindrical objects or to realize optical angular filters. It is also worth to mention that not just nanospheres can be used as shell material, but also suitably patterned surfaces thin films (Frequency Selective Surfaces), that mimic almost surface-cloaks, can be used [21–23].

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However, all those cloaking devices that cancel the scattering response were based solely on plasmonic materials thus far. Such materials possess a negative permittivity; or at least a permittivity smaller than unity. This, unfortunately, only allows to cloak electrically small objects whose scattering response is dominated by that of an electric dipole, i.e. objects for which the electrostatic approximation is sufficient to describe their response. This obstacle needs to be lifted and it is naturally to ask, how structures could look like that allow to suppress the scattering response of the next higher order electromagnetic multipole, i.e. the magnetic dipole. Its contribution to the scattering response cannot be neglected anymore for objects of moderate optical sizes where (kr) tends to be comparable to unity. Here k is the wavenumber associated to the light in the sphere and r its radius.

In our contribution we suggest to extend the technique of scattering cancellation towards particles whose scattered field has a noticeable contribution from both electric and magnetic dipoles. While concentrating on how to suppress the magnetic dipolar contribution to the scattered field, we show that a suitably designed shell that possess a dispersive permeability is suitable for that purpose. Moreover, we go one step further and show that small nanospheres made from a medium with a large permittivity provide an effective medium that meets the requirement for the shell. The key is that such nanospheres sustain a strong magnetic dipole as the lowest order Mie resonance [24–27]. Once they are assembled sufficiently dense, the emerging effective material possesses a dispersive permeability. By carefully adjusting the free parameters describing the geometry, e.g. size, permittivity, and density of the nanospheres [28–31], allows to devise a shell that cancels the scattered field of the magnetic dipole of its core. Experimental evidence of the feasibility of that approach has been shown in Ref. [29, 32]. In a last step we even go further and analyze, exemplarily, a realistic design based on Copper Chloride (CuCl) nanospheres. Such material is known for its excitonic resonance at optical frequencies [33, 34] and for attaining a very high permittivity around this resonance. A shell made of CuCl nanospheres therefore perfectly serves the desired purpose.

Unique to our approach is to rely on rigorous simulations based on Mie scattering theory [35] for randomly arranged spheres. It is used to model the cloaking shell made of an disordered arrangement of nanospheres and takes into account precisely the object to be cloaked. It is shown that with such device a significant suppression of the scattering response over a finite range of frequencies is possible. The device is accessible in a possible experiment and it can be expected to be implemented in the near future thanks to the progress made recently in fabricating and characterizing nanospheres of various materials [36–40]. Moreover, although shown here only for CuCl nanospheres, other materials for the nanospheres are applicable as well that are known to support magnetic Mie resonance in certain frequency domains [29, 30].

2. Cloaking a magnetic dipole

To simplify the analysis, let us consider a dielectric sphere as the object to be cloaked. The sphere is centered at the origin of the coordinate system, without loss of generality. We want to investigate the scattering response of such object upon illumination by an electromagnetic plane wave. To fix the geometry, we consider a sphere of radius $r_{\mathrm{c}} = 66$ nm and relative permittivity $\varepsilon_{\mathrm{c}} = 8$. It could be shown [13] that the scattered electric and magnetic fields $\mathbf{E}_{\mathrm{s}}$ and $\mathbf{H}_{\mathrm{s}}$ can be derived from expressions for the Mie coefficients c_n^{TE} and c_n^{TM}, where TE and TM refer to the electric and magnetic spherical harmonics, respectively. These Mie coefficients relate the amplitudes of the spherical harmonics of the scattered to those of the incident field [41–43]. Figure 1(a) shows in addition to the total scattering cross section (SCS) the contributions to the SCS of the most dominant multipoles of the sphere under consideration, namely the electric and magnetic dipolar contribution. As could be noticed, only contributions up to a given order n are relevant since the scattering amplitudes scale as $|c_{\mathrm{n}}(\omega)| = o\left[(kr_c)^{2\mathrm{n}+1}\right]$ with $k = (\omega/c)\sqrt{\varepsilon\mu}$

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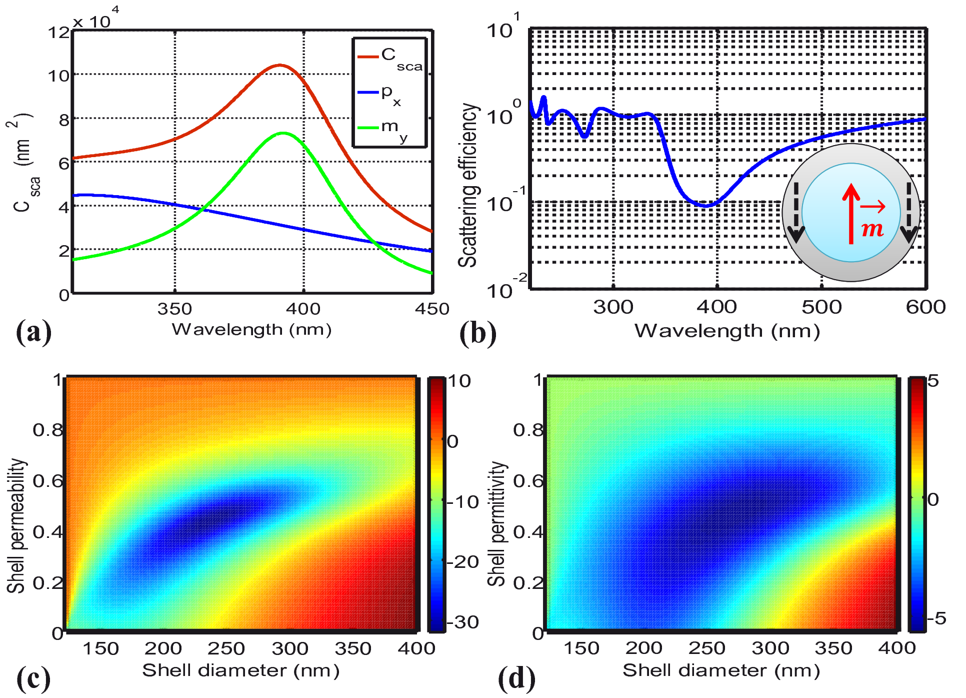

Fig. 1. (a) Decomposition of the scattered field of a dielectric sphere of radius $r_c = 66$ nm and permittivity $\varepsilon_c = 8$ into the contributions from electromagnetic multipoles. The overall SCS (scattering cross section) is dominated by the magnetic dipolar contribution. (b) Scattering efficiency of the same dielectric obstacle surrounded by a magnetic shell with effective permeability $\mu_s = 0.25$ and radius $r_s = 98$ nm. A drastic scattering reduction for a finite range of frequencies coinciding with the magnetic resonance of the bare obstacle can be seen. Scattering efficiency (in dB) through the variation of the shell diameter as well as the permeability (c) and the permittivity (d), where we neglected their imaginary parts. Please note that a suitable permeability alone can already significantly suppress the scattering response.

being the wave number associated with the material the object is made of [13]. In our case $n = 1$, and the scattering is dominated by the electric and magnetic dipoles. Moreover, in the wavelength window [350 nm, 450 nm], the object possesses a magnetic resonance at around 390 nm as reflected by the total SCS.

In the general case, for a core-shell system, the n^{th} coefficient is given by:

$$c_n^{\text{TE}}(\omega) = -\frac{U_n^{\text{TE}}(\omega)}{U_n^{\text{TE}}(\omega) + V_n^{\text{TE}}(\omega)}, \qquad c_n^{\text{TM}}(\omega) = -\frac{U_n^{\text{TM}}(\omega)}{U_n^{\text{TM}}(\omega) + V_n^{\text{TM}}(\omega)} \tag{1}$$

where $U_{\text{n}}^{\text{TE}}(\omega)$ could be expressed as

$$U_n^{\text{TM}} = \begin{vmatrix} j_n(k_c r_c) & j_n(k_s r_c) & y_n(k_s r_c) & 0 \\ [k_c r_c j_n(k_c r_c)]'/\varepsilon_c & [k_s r_c j_n(k_s r_c)]'/\varepsilon_s & [k_s r_c y_n(k_s r_c)]'/\varepsilon_s & 0 \\ 0 & j_n(k_s r_s) & y_n(k_s r_s) & j_n(k_0 r_s) \\ 0 & [k_s r_s j_n(k_s r_s)]'/\varepsilon_s & [k_s r_s y_n(k_s r_s)]'/\varepsilon_s & [k_0 r_s j_n(k_0 r_s)]'/\varepsilon_0 \end{vmatrix}, \tag{2}$$

where $j_n(.), y_n(.)$ are spherical Bessel functions, and $[.]'$ denotes derivation with respect to their argument, and k_c and k_s are the wavenumbers inside the core sphere (of radius r_c and permit-

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tivity ε_c) and the covering shell (of radius r_s and permittivity ε_s), respectively [$V_{\mathrm{n}}^{\mathrm{TE,TM}}(\omega)$ is obtained by changing $j_n(.)$ with $y_n(.)$ in the last column]. The TE coefficients are obtained from Eq. 2 by substituting ε with μ.

The scattering cross section per unit length of an spherical object is a quantitative measure of its overall visibility and is given by the expression [13,44]

$$C_{\mathrm{sca}} = \frac{2\pi}{|k_0|^2} \sum_{n=1}^{n=+\infty} (2n+1) \left[\left|c_n^{\mathrm{TE}}\right|^2 + \left|c_n^{\mathrm{TM}}\right|^2 \right] \ . \tag{3}$$

Generally, if an observer is in the far or near field, its possibility to detect the object is fully determined by the amplitude of σ_{SCS}. So that minimizing or totally canceling it would lead to the undetectability of the object.

We further investigate the scattering efficiency (defined as the ratio of the SCS of the core-shell system and the one of the bare sphere) when a magnetic [Fig. 1(c)] or a dielectric [Fig. 1(d)] shell surrounds the object. It is then evident, that when only a plasmonic material is used for the shell ($\varepsilon_{\mathrm{s}} < 1$), the maximum of scattering reduction that could be achieved is about 5dB which leaves the object almost completely detectable. It also shows that a permeability lower than one is required to achieve better scattering reduction since the magnetic dipole dominates the scattering response of the object. We can see from Fig. 1(c) that for some specific values of shell permeability μ_{s} and shell radius r_{s}, the visibility of the object can be suppressed by 30 dB (in the blue region).

Thus, by covering the object with a material that has a sufficient low permeability, it is possible to significantly reduce the SCS of the covered object at certain frequencies. In fact, by considering that the magnetic dipole ($n = 1$) to be given by an integral of the magnetization vector $\mathbf{M}_1(\mathbf{r},\omega) = \mu_0 [\mu_1(\mathbf{r},\omega) - 1] \mathbf{H}_1(\mathbf{r},\omega)$ across the scattering volume [44], with $\mu_1(\mathbf{r},\omega)$ being the local permeability and $\mathbf{H}_1(\mathbf{r},\omega)$ the local magnetic field, as can be schematized in the inset of Fig. 1(b), it can be shown that a metamaterial with small or negative permeability induces a local magnetization vector out of phase to the local magnetic field; thus permitting partial or even entire cancellation of the scattering signal caused by the object. Since in each case a negative permeability is sufficient for structures whose scattering response is dominated by a magnetic dipole, it is for further considerations sufficient to target a medium which possesses at least the sufficiently small permeability. One can also mention that in this unrealistic lowest-order approximation where we considered, non-dispersive materials, the cloaking bandwidth is very wide [see the region of invisibility that extends over almost 150 nm in Fig. 1(b)].

3. Achieving magnetic shells through polaritonic nanospheres

The more practical question to be solved consists in finding an artificial material that can be used for the shell. It is well established that SRRs would provide the desired magnetic response. But such materials suffer from the disadvantage of being highly anisotropic. Besides being mismatched in size, it is unfortunately not suitable for the present application that requires an isotropic magnetic material. The only option thus far to achieve such isotropic response is to use all-dielectric structures such as nanospheres that are randomly arranged. The metamaterials achievable with such meta-atoms have the advantage of being moderately lossy as well as isotropic. This is a strong motivation to build the cloaking shell out of nanospheres.

Let us consider a bulk medium made of randomly arranged dielectric nanospheres with a permittivity of ε_{d} and embedded in a host medium described by the permittivity ε_{h} and permeability $\mu_{\mathrm{h}} = 1$. We denote by f the volume filling fraction of the nanospheres in the surrounding medium and by x the relative size parameter of the nanospheres, i.e. $x(\lambda) = 2\pi r_{\mathrm{d}}/\lambda$, where r_{d} is the radius of the nanospheres and λ the wavelength in the host medium. For $x \ll 1$, the effective

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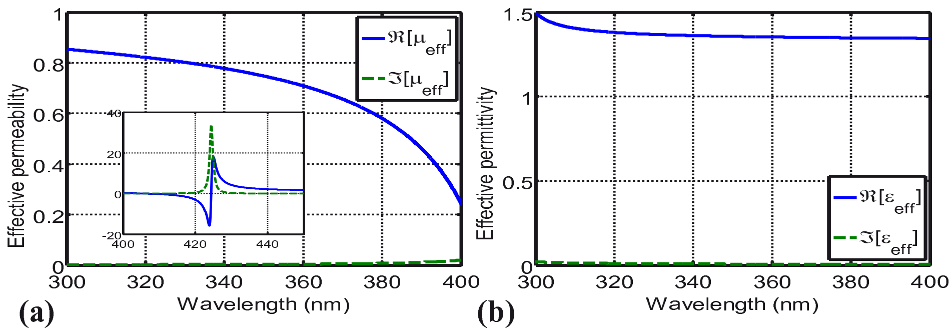

Fig. 2. Effective permeability (a) and permittivity (b) of a medium consisting of dielectric nanospheres of radius $r_\mathrm{d} = 16$ nm made from a material with a high permittivity of $\varepsilon_\mathrm{d} = 169$, and their filling fraction being $f = 0.1$. At wavelengths beyond the resonance of the permeability, it is clearly shown that $\mu_\mathrm{eff} < 1$ could be achieved coinciding with the range of Fig. 1(b). The inset shows the magnetic resonance which occurs at 425 nm.

permittivity of the homogenized material ε_eff can be calculated by the well-known Maxwell-Garnett mixing rules [45]. When the parameter $x_\mathrm{d}(\lambda_\mathrm{d}) = 2\pi r_\mathrm{d}/\lambda_\mathrm{d}$, where λ_d is the wavelength inside the sphere, does not satisfy $x_\mathrm{d} \ll 1$ this approximation is no longer valid. In that case, Mie theory is required to calculate the effective parameters of the medium as:

$$\varepsilon_\mathrm{eff}(\omega) = \varepsilon_\mathrm{h}\frac{x(\omega)^3 - 3ifT_1^\mathrm{E}(\omega)}{x(\omega)^3 + 3/2ifT_1^\mathrm{E}(\omega)} \quad , \quad \mu_\mathrm{eff}(\omega) = \mu_h\frac{x(\omega)^3 - 3ifT_1^\mathrm{H}(\omega)}{x(\omega)^3 + 3/2ifT_1^\mathrm{H}(\omega)} \tag{4}$$

where $T_1^\mathrm{E}(\omega)$ and $T_1^\mathrm{H}(\omega)$ are the electric and magnetic contributions to the scattered field of the single sphere, respectively (their expressions can be found for example in [13, 44]).

These considerations facilitate the characterization of the effective medium as it depends on the geometry and the properties of the nanospherical meta-atoms. In Fig. 2 the effective parameters of such medium are displayed when the radius of the dielectric nanospheres is $r_\mathrm{d} = 16$ nm, their permittivity $\varepsilon = 169$, and the filling fraction $f = 0.1$. It can be clearly seen that in the spectral domain of interest, the permeability is less than unity. Moreover, the imaginary part is relatively small, suggesting a high figure-of-merit. In the same frequency range, the effective permittivity is almost constant $\varepsilon_\mathrm{eff}(\omega) \approx 1.4$ since the magnetic and electric dipole of the nanosphere resonate at different frequencies.

Having a metamaterial for the shell at hand that provides the desired properties, we show in Fig. 3(a) the spectrally resolved scattering efficiency of the cloaked sphere. Two types of simulations have been performed. At first we have performed rigorous simulations taking into account all the physical interactions occurring in our device, i.e. the interactions between the dielectric nanospheres among each other and with the dielectric core to be cloaked. The structure as shown in the inset of Fig. 3(a) has been considered. The numerical analysis was performed using extensions to Mie theory to handle clusters of nanospheres [35] which is a rigorous solution of Maxwell's equations. At second an approximate theory was applied where the shell was made from an equivalent homogeneous isotropic material whose properties have been deduced for the respective system using Eq. 4. It can be seen that both approaches are in excellent agreement around the wavelength where the scattering signal was expected to be suppressed, hence justifying the application of an effective medium theory. It should be mentioned also that at higher wavelengths, a deviation between both approaches could be noticed which could be explained by the polaritonic resonances of the nanoparticles that occur at these wavelengths. Thus the a strong coupling between adjacent particles arise and dominates the optical response

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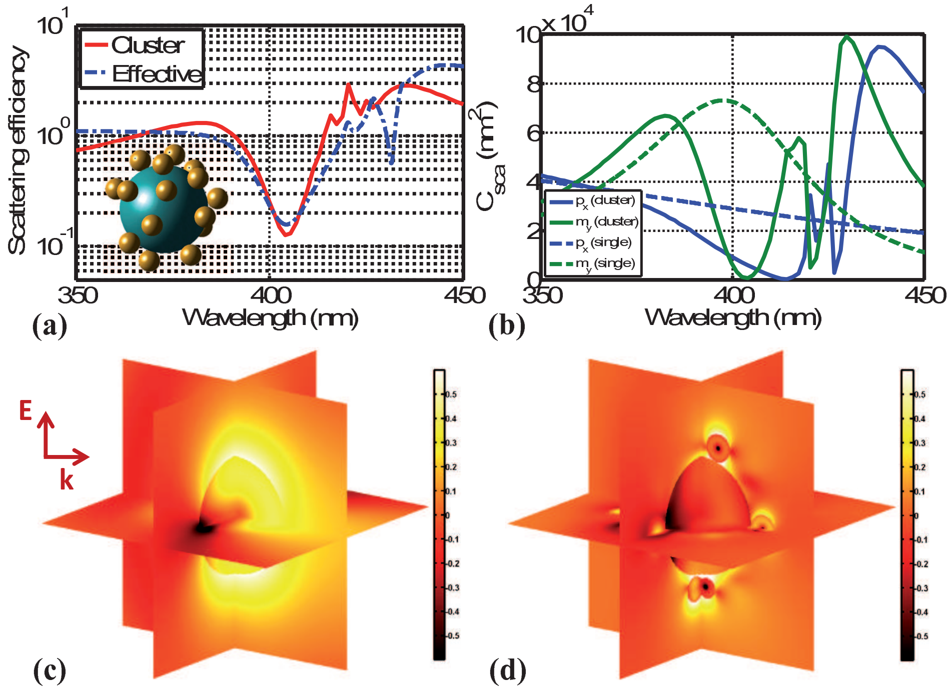

Fig. 3. (a) Numerical calculation of the scattering efficiency for the core shell system as a function of wavelength; red solid line - core-shell system rigorously calculated where the fine details of the structure are accounted for; blue dot-dashed line - core shell system calculated using the effective medium approach (Clausius-Mossotti equation) (Eq. 4). Note the scattering reduction in the wavelengths domain predicted by the theory. The inset shows a schematic of the dielectric sphere to be cloaked surrounded by 17 polaritonic nanospheres with filling fraction $f = 0.1$, radius $r_{\rm d} = 16$ nm and a permittivity $\varepsilon_{\rm d} = 169$, the axis unit is nm. (b) Different contributions to the scattering response by different electromagnetic multipoles of the structure showing a drastic reduction of the magnetic component around the spectral region of interest. (c) Time averaged amplitude of the electric field distributions in logarithmic scale of the bare dielectric sphere and (d) the cloaked one. The structures are illuminated with a unit amplitude plane wave (405 nm) where incident field propagates parallel to the horizontal plane and the electric field is polarized perpendicular to it (as sketched by the arrows).

of the structure and the effective medium models are no longer suitable for its description.

As can be seen from Fig. 3(b), showing the contributions to the SCS by the different multipoles, both the electric and the magnetic dipole contributions are drastically reduced for a finite range of wavelengths permitting thus a lower scattering efficiency. It has to be stressed that the scattering from the core-shell structure is ten times less than that from the bare dielectric sphere for a relatively wide range of wavelengths (380 nm to 415 nm). However, such broad response is only possible while considering the nanospheres to be made of a non-dispersive material with a large permittivity. Such materials do not exist and this assumption has to be lifted. To further check the functionality of our cloak, we show in Fig. 3 maps of the amplitude of the electric field at 405 nm scattered by the core sphere (c) and by the core-shell system (d). When the core sphere is surrounded by the cloak, the scattering is drastically reduced in contrast to the uncloaked case consistent with the scattering reduction shown in Fig. 3(a). As explained above, this is due to the choice of the permeability of the shell made of polaritonic nanospheres.

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4. The use of copper chloride nanospheres

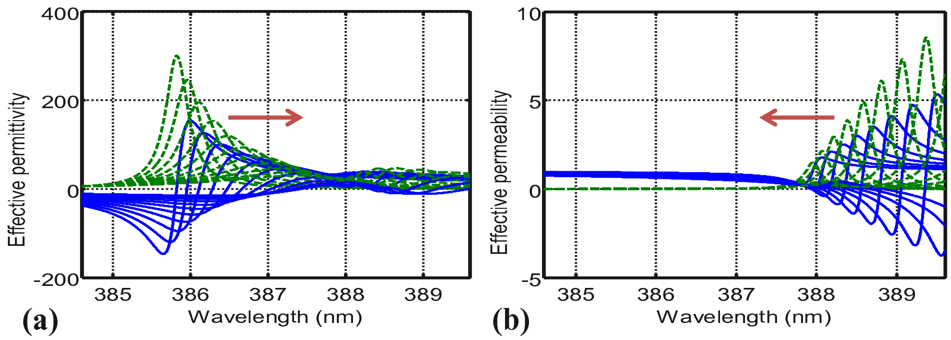

Fig. 4. Effective permittivity (a) and permeability (b) of a medium consisting of copper chloride nanospheres of different radii ranging from 16 nm to 32 nm and showing the possibility of tuning their resonance frequency. The (blue) solid lines are for the real parts while the (green) dashed ones are for the imaginary parts. The red arrows indicate the direction of increasing r_d, by steps of 4 nm.

In this last section, we will focus on realistic polaritonic materials to build meta-atoms for the cloaking shell. In fact, an amorphous arrangement of copper chloride (CuCl) nanospheres, described by its effective permeability and permittivity can approximate the features discussed in the previous section. These particles are known to exhibit a Z_3 exciton resonance around 387 nm [46] and can be fabricated by colloidal nano-crystallization in solutions involving $\mathrm{CuCl_2}$ in the presence of ascorbic acid and PVP (PolyVinylPyrrolidone) [47]. Around this particular frequency, the permittivity of this semiconductor is given by $\varepsilon_{\mathrm{CuCl}}(\omega) = \varepsilon_\infty + A\gamma/(\omega_0 - \omega - i\gamma)$, where $\varepsilon_\infty = 5.59$, $\omega_0 = 4862$ THz and $\gamma = 0.076$ THz. The factor $A = 632$ is proportional to the exciton oscillator strength for CuCl [33, 34, 46]. Due to its high permittivity, a medium of CuCl nanospheres exhibits a magnetic resonance where the effective permeability will show a strong dispersion, i.e. it features a resonance. This is shown in Fig. 4 for different radii of nanospheres. Tuning across a wide spectral range by varying r_d can be observed. This allows for a robust and tunable cloak. The dispersion in the permittivity is undesired for our purpose. But in the wavelengths range of interest, it can be safely assumed to be weakly dispersive, as can be seen from Fig. 5(a) whereas around 388 nm a permeability less than unity can be recognized in Fig. 5(b).

We finally discuss the scattering behavior of a cloak made from such CuCl nanospheres. In Fig. 5(c) the SCS is shown of a dielectric sphere coated with CuCl nanospheres of radius 16 nm each and filling fraction $f = 0.35$ sketched in Fig. 5(d). These nanospheres are not touching and do have indeed a minimum distance to their nearest neighbor to avoid undesired non-local effects. Again, simulations were made rigorously with Mie theory by taking into account all the spheres individually and their coupling among each other as well as their higher dumping compared to bulk medium. As can be noticed, a significant reduction of the visibility (scattering cross section) is achieved around the resonance wavelengths where the effective permeability shows a strong dispersion. The dashed curve in Fig. 5(c) gives the spectral response for the uncloaked sphere of radius 66 nm for comparison. It is evident that an excellent scattering reduction, reaching 72 percent, may be achieved close to the resonance at around a wavelength of 388 nm. We have verified herewith that the magnetic dipole contribution to the scattered field is significantly reduced due to the presence of the polaritonic nanospheres, i.e. the effective medium made from these provides a magnetization of same amplitude and opposite sign when

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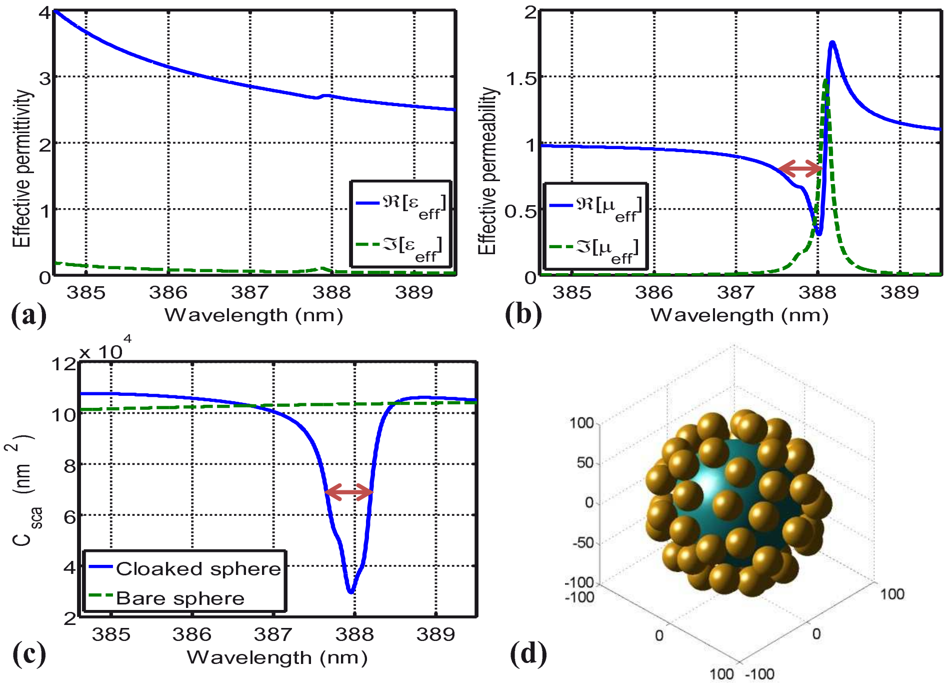

Fig. 5. Effective permittivity (a) and permeability (b) of the medium made up of CuCl nanospheres of radius $r_d = 16$ nm with filling fraction $f = 0.35$. This cluster consists of $N = 55$ polaritonic spheres randomly arranged on top of a dielectric obstacle of size 66 nm and permittivity of 8. Reduced scattering cross section (c) around the low permeability region, when the magnetic dipole moment of the total structure is nil. The region of low scattering efficiency is indicated by the red double arrows. (d) Schematic showing the core-shell cloaking structure.

compared to the the dielectric sphere. This allows thus the annihilation of the contribution to the scattering due to the magnetic dipole resonance.

5. Conclusion

In conclusion, we have studied analytically and numerically the extension of the cloaking mechanism described in Ref. [13, 18] to obstacles with an optical size such that the quasi-static limit is no longer applicable. First, we have shown numerically that a homogeneous layer with a permeability close to zero is required to drastically reduce the visibility of a dielectric sphere with a high permittivity and an optical size comparable to the wavelength. We have concentrated our attention to objects with a scattering response dominated by a magnetic dipolar contribution. We then exploited the magnetic resonances from polaritonic nanospheres to implement a shell that possesses a highly dispersive permeability that can strongly suppress the scattering response. A realistic design based on copper chloride nanospheres was considered and showed a robust cloaking efficiency. In addition the wavelength of operation can be tuned by varying the radius of the nanospheres. We believe that our results constitute one of the first steps to develop efficient cloaks that rely on the scattering cancelation techniques that can hide large objects. To cloak objects that are bigger compared to the incident wavelength requires more than just suppressing the electric dipolar contribution to the scattered field as was done for almost all the studies based on plasmonic cloaking. Our theoretical concept was backed by a feasible de-

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sign that can be investigated in experiments. Such cloaks may also represent a viable way for noninvasive sensing and probing with improved bandwidth, following the ideas of cloaking the sensor originally introduced in [14].

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