Localized magnetic plasmons in all-dielectric $\mu < 0$ metastructures

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Metamaterials have been shown to exhibit a variety of electromagnetic properties non-existing in Nature. In this Letter, we show that an all-dielectric (non-magnetic) system consisting of deep subwavelength, high permittivity resonant spheres posses effective negative magnetic permeability (dielectric permittivity being positive and small). Furthermore, we show that, due to the symmetry of the electromagnetic wave equations in classical electrodynamics, localized “magnetic” plasmon resonances can be excited in a metasphere made of such metamaterial. This is theoretically demonstrated by the coupled-dipole approximation and numerically for real spheres, in full agreement with the exact analytical solution for the scattering process by the same metasphere with effective material properties predicted by effective medium theory. The emergence of this phenomenon as a function of structural order within the metastructures is also studied. Our proposal paves the way towards (all-dielectric) magnetic plasmonics, with a wealth of fascinating applications.

The scattering of electromagnetic (EM) waves from macroscopic media is a classical problem of widespread interest throughout the entire EM spectrum, from radio and microwave waves, with increasing frequency through the THz, IR, and visible domains towards the high-energy UV and x-ray band [1, 2]. The variety of phenomena where EM wave scattering plays a leading role is overwhelming: radar, lidar, remote sensing, metamaterials, plasmonics, etc. Not to mention other classical waves (acoustic, seismic) or formally analogous problems (electron transport, neutron scattering, etc.).

Within classical electrodynamics, the scatterers’ response is described within macroscopic Maxwell equations in terms of a dielectric permittivity $\varepsilon$ and a magnetic permeability $\mu$ (cf. Refs.[3, 4]). In general, and particularly in the high frequency range, non-magnetic media have been considered, based on the fact that most materials found in Nature exhibit no magnetic permeability; except for a few theoretical works playing with artificial magnetic permeabilities [5, 6]. Nonetheless, the advent of the so-called metamaterials has made it possible to achieve a variety of electromagnetic responses; these are artificial materials structured at scales much shorter than the wavelength with exotic (effective) $\varepsilon$ and $\mu$. In this manner, not only large values of the magnetic permeability are possible, but also negative-$\mu$ metamaterials. Needless to say, such negative magnetic response is crucial in fabricating the so called “left-handed” media (LHM), a novel kind of materials predicted to exhibit peculiar electromagnetic properties in which both the permittivity and the permeability are negative [5, 7, 8].

Thus, it seems quite natural to address in the classical scattering of EM waves the impact of media with relative permittivities $\mu \neq 1$ [9, 10], either positive or negative. Although both cases are interesting enough, it can be argued that $\mu < 0$ media, behaving as magnetic metals, can be expected to present a richer associated phenomenology, as conventional metals do when compared to usual dielectrics. Among this phenomenology, the excitation of surface plasmons, i.e. collective oscillations of the conduction electrons, plays a major role due, mainly, to the fact that they concentrate the electromagnetic energy within subwavelength regions.

Interestingly, while the excitation of propagating surface plasmon polaritons in $\mu < 0$ and left-handed toy-model materials [6, 11–14], very few works deal with the localized version of these collective modes. In fact, most of the studies related to scattering from bodies with $\mu < 0$ consider simultaneously $\varepsilon < 0$, thus being left-handed [15–18]. Regarding those dealing with purely $\mu < 0$ structures, the studies of plane wave scattering from small homogeneous spheres [19, 20] are of particular interest. In any case, despite the amount of theoretical articles devoted to the subject, to the best of our knowledge, none of them has proposed any realistic, purely $\mu < 0$ mesoscale structures in which these effects could be observed.

In these letter, we propose as a purely $\mu < 0$ ($\varepsilon > 0$) system a collection of high refractive index (HRI) spheres. Moreover, we show that, when arranged as a finite metastructure, in particular a metasphere, this system may, in turn, support localized magnetic plasmon resonances (LMPRs). Note that similar magnetic localized plasmon resonances terminology has been used in the work by Huidobro et al. [21], but in a totally different context. While here, it is used to describe the excitation of plasmons in effective magnetic plasmas, it is introduced there in the context of localized spoof plasmons, in which the authors identify magnetic resonances in the extinction spectrum of Perfect Electric Conductor corrugated disks.

It is well known that the scattering of electromagnetic waves from a homogeneous sphere accepts an analytical solution which, in the case of illumination by a $x$-polarized plane wave travelling along the positive $z$-axis, gives the following form for the scattering and extinction
efficiencies:

\[ Q_{\text{sca}} = \frac{2}{\pi^2} \sum_n (2n+1)(|a_n|^2 + |b_n|^2), \]
\[ Q_{\text{ext}} = \frac{2}{\pi^2} \sum_n (2n+1) \text{Re}(a_n + b_n), \]

with

\[ a_n = \frac{\mu \psi_n(x) \psi_n'(mx) - m \psi_n(mx) \psi_n'(x)}{\mu \xi_n(x) \psi_n'(mx) - m \psi_n(mx) \xi_n'(x)}, \]
\[ b_n = \frac{m \psi_n(x) \psi_n'(mx) - \mu \psi_n(mx) \psi_n'(x)}{m \xi_n(x) \psi_n'(mx) - \mu \psi_n(mx) \xi_n'(x)}, \]

where \( x = 2\pi n_0 R/\lambda \) is the size parameter \((R = D/2\) being the sphere radius and \( \lambda = 2\pi c/\omega \) the wavelength), and \( n_0 = (\varepsilon /\varepsilon_0 \mu_0) \) is the refraction index (relative to the surrounding medium) and \( \mu \) the relative permeability.

From the equations that fully describe the EM scattering process, i.e., Maxwell’s equations and the continuity conditions:

\[
\begin{align*}
\mathbf{n} \cdot \mathbf{H} & = \varepsilon \mathbf{n} \times \mathbf{H}, \quad \mu_0 \mathbf{n} \times \mathbf{H} = \mu_1 \mathbf{n} \times \mathbf{H}, \quad (5a) \\
\mathbf{n} \cdot \mathbf{E} & = \varepsilon \mathbf{n} \times \mathbf{E}, \quad \varepsilon_0 \mathbf{n} \times \mathbf{E} = \varepsilon_1 \mathbf{n} \times \mathbf{E}, \quad (5b)
\end{align*}
\]

where \( \mathbf{n} \) is the unit vector normal to the scatterer surface, it is easy to demonstrate that, upon exchanging the value of the relative dielectric permittivity \( \varepsilon \) with that of the relative magnetic permeability \( \mu \), and viceversa, the resulting electric (magnetic) fields are identical to the initial magnetic (electric) fields (except for a sign change preserving the chirality of the EM waves). In particular, due to the rotational symmetry of this problem, one has:

\[
E, \mathbf{H} \iff \tilde{E} = -H, \tilde{\mathbf{H}} = \mathbf{E}
\]

\[
\varepsilon, \mu \iff \tilde{\varepsilon} = \mu, \tilde{\mu} = \varepsilon
\]

\[
a_n, b_n \iff \tilde{a}_n = b_n, \tilde{b}_n = a_n.
\]

This symmetry implies that the properties of a magnetic media with \( \mu \neq 1, \varepsilon = 1 \) can be inferred from those of a non-magnetic material with \( \mu = 1, \varepsilon = \mu \). Specifically, under the assumptions \( \lambda \gg R \) and \( \lambda \gg R/\sqrt{\mu} \), the scattering process can be accurately described retaining only the two first terms in Lorenz-Mie expansion \([2, 9]\), \( a_1, b_1 \) representing, respectively, the dipolar electric and magnetic contributions. In the limits previously indicated, they acquire the following form \([9]\):

\[
a_1 = \frac{2i \varepsilon - 1}{3 \varepsilon + 2x^3}, \quad b_1 = \frac{2i \mu - 1}{3 \mu + 2x^3}.
\]

Therefore, for small particles having \( \varepsilon = 1 \) and \( \mu \sim -2 \) it would be possible, in principle, to excite dipolar resonances with analogous properties to those excited in usual metallic particles (for which \( \mu = 1 \) and \( \varepsilon \sim -2 \)).

The question now arises if there is a medium that fulfills all necessary conditions. In order to address this question, let us consider first the optical properties of a HRI non-magnetic sphere \((\varepsilon \gg 1, \mu = 1)\). Particularly well known among the microwave community \([22, 23]\), but quickly extending to people working at optical frequencies \([24–26]\), it is the fact that in HRI particles the lowest-order resonance found in their extinction spectrum typically presents a definite dipolar magnetic character. Moreover, once a certain value of \( \varepsilon \) is reached, the position of this resonance scales as \( \lambda_{\text{res}}^{\mu} \sim 2\sqrt{\varepsilon} R \), which opens the possibility to use these HRI systems as building blocks of effective magnetic media. For large enough \( \varepsilon \) values, \( \lambda_{\text{res}}^\mu \gg R \), which makes the system homogenizable and, at the same time, such that \( \lambda_{\text{res}}^\mu \gg \lambda_{\text{res}}^\varepsilon \), for any other resonance. That makes the individual building blocks be accurately described as purely magnetic dipoles, and the effective medium so obtained to have \( \mu_{\text{eff}} \neq 1 \), but \( \varepsilon_{\text{eff}} \sim 1 \). Thus, consider for example a

![FIG. 1. Optical properties under plane wave illumination of a sphere with \( R = 1 \text{ mm} \), \( \varepsilon = 1000 \) and \( \mu = 1 \). (a) Scattering efficiency (black), together with the dipolar electric (red) and dipolar magnetic (green) contributions. (b) Real (black) and imaginary (red) parts of the electric polarizability. (c) Real (black) and imaginary (red) parts of the magnetic polarizability. The inset shows a detail for frequencies around the resonance.](image-url)
sphere with $\varepsilon = 1000$, which is a reasonable value for certain ceramic materials such as the BST operating in the gigahertz range [23], and let us fix the radius of a sphere of such material to $R = 1$ mm. The optical properties of this system under plane wave illumination are depicted in Fig.1. In the top panel, the scattering efficiency of the sphere is plotted, together with the electric and magnetic dipolar contributions. A resonance can be observed at a wavelength $\lambda_{\text{res}}^0/R \sim 2\sqrt{\varepsilon} \sim 63$. The multipole decomposition allows identification of its dipolar magnetic character. Moreover, the electric contribution is almost negligible, as can be also appreciated in the polarizabilities:

$$\alpha_e = 6\pi i a_1/k^3, \quad \alpha_m = 6\pi i b_1/k^3,$$  \hspace{1cm} (10)

shown in Figs.1(b) and (c), the non-resonant electric dipole polarizability being up to two orders of magnitude smaller than the (resonant) magnetic one. Since the sum of the remaining, higher-order contributions is about five orders of magnitude lower, we conclude that each of the HRI spheres can be accurately described retaining only the dipolar contributions, namely, by means of a pair of electric and magnetic dipoles.

Let us now consider an effective medium built with a collection of these HRI particles. Since the size of the particles is extremely subwavelength and the dipolar approximation holds, we expect the usual effective medium theories to give accurate results for the effective properties of the medium, at least, away from the resonance and for low volume filling fractions, $f$. Incidentally, it has been recently reported that the effective optical response of colloidal particles, in general, may exhibit a non-local magnetic permeability [27, 28]; despite that, as a first estimate, we make use of Clausius-Mossotti relations. The results so obtained for a medium with a filling fraction of $f \sim 0.263$ (the election of such number will be clarified afterwards) are shown in Fig.2. While the effective permittivity of the system remains approximately constant with low values ($\varepsilon_{\text{eff}} \sim 2$), the case of the magnetic permeability completely differs. It shows a resonant behaviour reaching negative values (extremely large in magnitude) for frequencies above the magnetic resonance completely differs. It shows a resonantly constant with low values ($\varepsilon_{\text{eff}} \sim 2$), the case of the magnetic permeability completely differs. It shows a resonant behaviour reaching negative values (extremely large in magnitude) for frequencies above the magnetic resonance. Moreover, the scattering cross section (SCS) of the individual constituents at that frequency is only about three times the geometrical one. Thus, one could expect that the effective medium theory can still hold, giving accurate results without further refinements [27–29], as we will show below.

One may wonder now, what would be the optical properties of a small particle made of such effective medium? In order to address this question, let us choose a sphere with these effective properties (those of Fig. 2) and radius $R_{\text{eff}}$. If $R_{\text{eff}}$ is small enough, recalling expressions (9) for the dipolar contributions in the small particle limit, we expect a resonance appearing at frequencies $\omega_0^{\text{res}}$ such that $\text{Re}(\mu_{\text{eff}}(\omega_0^{\text{res}})) \sim -2$. For this particular system, if we choose $R_{\text{eff}} = 6$ mm, the corresponding wavelength at resonance still fulfills $\lambda_0^{\text{res}} \gg \sqrt{\mu_{\text{eff}}\varepsilon_{\text{eff}}}R_{\text{eff}} > R_{\text{eff}}$. Therefore, we expect a dipolar magnetic resonance, together with an electrical contribution going as $Q_{\text{ext}}(a_1) \sim x^4/6$. Additionally, based in what is known for small metallic particles and considering the electric size of the sphere, one could also expect the emergence of a quadrupolar magnetic contribution for frequencies above the dipolar resonance, analogous to the electric quadrupole appearing in the spectra of relatively large (still deeply subwavelength) metallic spheres. In Fig.3 (a), the analytical (Mie) scattering and extinction efficiencies are plotted as a function of the wavelength of incidence. A relatively broad resonance can be observed at a wavelength such that $\mu_{\text{eff}} = -3.262 + 0.014i$, together with a narrow peak at smaller wavelengths. Multipolar decomposition allows a direct identification of the character of these resonances, as depicted, the broader at $\lambda = 64$ mm being the magnetic dipole, the sharper at $\lambda = 63$ mm corresponding to the magnetic quadrupole, as expected. For larger wavelengths a large number of resonances appear together, with higher order contributions playing an important role in the total extinction efficiency of the system. This is due to the fact that the effective permeability acquires extremely high positive values once it flips sign, hence making the effective index of refraction also large and allowing excitation of such collection of geometrical resonances, some of them, in turn, closely

![FIG. 2. Effective material properties of a medium made of the spherical particles of Fig.1 with a filling fraction $f = 0.263$. (a) Real (black) and imaginary (red) parts of the effective electric permittivity. (b) Real (black) and imaginary (red) parts of the effective magnetic permeability.](image-url)
All the previous would be nothing but a mere theoretical toy-model if there was no realistic system that could exhibit this sort of behaviour. Although a practical realization of HRI spheres (of, e.g., BST) is feasible for the range of geometrical parameters studied here [23], the question of whether or not a collection of them will behave as an effective magnetic plasma, as expected from the effective medium theory, is still open. We tackle this problem by a direct comparison of the analytical predictions for the extinction of Fig.3, calculated using Mie theory with the effective parameters of Fig.2, with simulations of the full scattering process, involving the whole set of HRI particles.

We do it numerically in two alternative ways. First, through the Coupled Electric and Magnetic Dipoles (CEMD) method [30], which describes each dielectric particle as a pair of coupled electric and magnetic dipoles with polarizabilities given by eqs. (10). The problem is self-consistently solved by expressing the electric and magnetic fields at the position of each dipole as a superposition of the incident field and the field generated by the rest of dipoles. Once the fields are known at each dipole position the far-field radiated is computed, together with the total SCS and ECS. Second, through full rigorous numerical calculations based on finite element method (FEM). Both approaches take fully into account inter-particle coupling. The results for the CEMD method in a single realization are shown as a blue curve (with circles) in Fig.3 (a). The particular distribution of HRI spheres inside the spherical volume of radius $R_{\text{eff}}$ for this realization is shown as an inset in the figure (efficiently occupying the volume of the equivalent sphere), and we assume the pair of dipoles to be located in the center of each HRI sphere. In this realization, we consider $N = 57$ particles, which makes a volume filling fraction $f = 0.263$.

Interestingly, even for a single realization, the result of CEMD closely reproduces that predicted for a negative-$\mu$ effective medium with the same radius, aside from the emergence of some kinks that depend on the particular realization. Investigation of the radiated far-field pattern at the maximum of the resonance $\lambda_{\text{res}} \sim 0.064$ (shown as an inset) reveals the magnetic dipole character of this collective resonance. Features related with the excitation of the magnetic quadrupole, as well as the set of resonances happening at wavelengths such that $\mu_{\text{eff}} > 1$, can also be observed in the CEMD simulation. Results of full numerical simulations carried out using COMSOL v4.3b (a FEM commercial software) are also shown as a black-dashed curve (with squares). These are in very good agreement as well, demonstrating the effect in realistic systems that could be readily measured in the lab. The inset, showing the magnetic field in the surface of the spheres (red corresponding to high intensity and blue to low), alongside with the field lines of the scattered magnetic field, further corroborates that the whole system collectively resonates with the characteristic pattern of a magnetic dipole, providing further support to our $\mu < 0$ effective medium approach for the metasphere.

Finally, we study the emergence of this resonance in connection with structural order in random arrangements of particles. To do so we apply the CEMD method and consider realization of $N = 57$ dipoles placed randomly with only two restrictions: we set a maximum distance from the origin to a dipole site equal to $R_{\text{max}} = R_{\text{eff}} - R$,
and an exclusion radius between dipoles equal to $R_{excl}$. Increasing $R_{excl}$ we are able to force the dipoles to efficiently occupy the volume of the effective sphere. Averaged results for $N_{excl} = 10^5$ realizations and different exclusion radii are shown in Fig.3 (b). A clear signature of the resonance is obtained even in the case of lowest ordering. Nevertheless, the resonance is better resolved (and the quadrupole peak starts to pop up) when a certain ordering is imposed. Since variations in the shape of structures supporting localized plasmons are known to strongly affect their spectral features, we attribute this effect to the fact that in the latter case the dipoles are located in such a way that the shape of the equivalent sphere is better preserved in the random realizations.

In conclusion, we have theoretically and numerically demonstrated the existence of the so-called Localized Magnetic Surface Plasmon Resonances in effective media characterized by a negative permeability (effective magnetic plasmas). We have shown that the excitation of these resonances occurs at frequencies such that $\mu_{\text{eff}} \sim -2$ when the overall size of the system is small compared with the incident wavelength, as expected from the analytical Mie expressions in conjunction with standard effective medium theories. To the best of our knowledge, this is the first time in which a realistic system is proposed that could exhibit this behaviour. It would be of interest to investigate the rich phenomenology analogous to standard $\varepsilon < 0$ plasmonics regarding, in particular, the zoology of magnetic localized plasmon resonances, including coupled metastructures (dimers, trimers, etc.), just to mention some. We, moreover, foresee that the same effective medium could be used to test the excitation of Magnetic Surface Plasmon Polaritons on flat surfaces. Our theoretical work thus paves the way towards a realistic $\mu < 0$ Plasmonics with the wealth of applications of "electric" plasmonics expected reciprocally in its "magnetic" counterpart.

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