Artificial Magnetism at Optical Frequencies in Composite Materials Made of Particles with Pairs of Tightly Coupled Metallic Nanospheres

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Abstract

We show how artificial magnetism at optical or infrared frequencies is produced by particles made of two tightly coupled subwavelength metallic spheres. This coupled particles system exhibits two main plasmon resonances, one of which corresponds to an antisymmetric mode with the induced electrical dipoles oscillating out-of-phase and creating an effective current loop. This phenomenon, which can be rationalized in terms of an equivalent magnetic dipole moment, enhances the magnetic field in the region between spheres. Due to this strong magnetic response, such particle pairs assembled in a periodic array form a material with high-frequency permeability.

1. Introduction

The design of material media exhibiting negative refraction, as a result of both negative dielectric permittivity and negative magnetic permeability, represents one of the most prominent application in the field of engineered optical materials or so-called metamaterials. In this context, a crucial role can be played by arrays of metallic nanoparticles, whose optical response is dominated by collective oscillations of the valence electrons known as particle plasmons. As a matter of fact, plasmonic structures inherently show negative permittivity at resonance. Furthermore, negative refraction has been shown to be closely connected to the resonant magnetic response of the plasmonic nanostructures [1]. Thus, a great deal of efforts has been focused on developing structures with a strong magnetic response, which is a challenging task especially for the visible spectral range.

High-frequency magnetism has been achieved for frequencies in the microwave to terahertz range using split-ring resonators (SRR) [2, 3], but the direct scaling of the demonstrated microwave media to visible optics is impracticable because of both technological issues and different electromagnetic responses of materials to visible light and microwaves. However, recently, two novel types of nanostructure assemblies have been demonstrated to exhibit resonant magnetic properties also at visible frequencies. In [1], pairs of gold nanopillars arranged in a planar array produce a strong magnetic field enhancement in the region between the nanopillars as a consequence of locally induced electric currents associated with an antisymmetric plasmon resonance. A similar plasmonic excitation is supported by an array of gold nanowire pairs on a glass substrate as demonstrated in [4]. Some further concepts for media exhibiting a magnetic response in the visible range have been reported in [5, 6] using ensembles of metallic nanospheres.

In this paper we consider a very simple medium made of pairs of tightly coupled silver nanospheres. We show that this structure can support an antisymmetric electromagnetic oscillation, which is responsible for artificial magnetism. This oscillation is revealed for a couple of silver nanospheres by using an approximated dipolar model and for periodical structures of pairs of coupled nanospheres through finite difference simulations [7].

2. Electromagnetic modes of two coupled metallic nanospheres

At first, we consider the two nanospheres problem in free space. The two nanospheres are made of silver, a noble metal that at optical frequencies exhibits a negative real-part of dielectric permittivity. We aim at finding the eigenvalues of the system of two subwavelength spheres by solving the coupled equations

$$\begin{aligned} \mathbf{E}(\mathbf{r}_2) &= \underline{\mathbf{G}}(\mathbf{r}_2,\mathbf{r}_1)\cdot\mathbf{p}_1, \\ \mathbf{E}(\mathbf{r}_1) &= \underline{\mathbf{G}}(\mathbf{r}_1,\mathbf{r}_2)\cdot\mathbf{p}_2, \end{aligned} \tag{1}$$

with the information that $\mathbf{p}_2 = \alpha\mathbf{E}(\mathbf{r}_2)$ and $\mathbf{p}_1 = \alpha\mathbf{E}(\mathbf{r}_1)$. Here $\underline{\mathbf{G}}(\mathbf{r}_2,\mathbf{r}_1) = \underline{\mathbf{G}}(\mathbf{r}_1,\mathbf{r}_2)$ is the dyadic Green's function; $\mathbf{r}_1$ and $\mathbf{r}_2$ denote the positions of nanospheres, which are treated as electric dipoles with dipole moments $\mathbf{p}_1$ and $\mathbf{p}_2$, respectively; α is the sphere polarizability [8] expressed in closed form as a function of silver permittivity [9]. Combining the equations (1) one obtains

$$\begin{aligned} \mathbf{p}_2 &= \alpha\,\underline{\mathbf{G}}(\mathbf{r}_2,\mathbf{r}_1)\cdot\mathbf{p}_1, \\ \mathbf{p}_1 &= \alpha\,\underline{\mathbf{G}}(\mathbf{r}_1,\mathbf{r}_2)\cdot\mathbf{p}_2. \end{aligned} \tag{2}$$

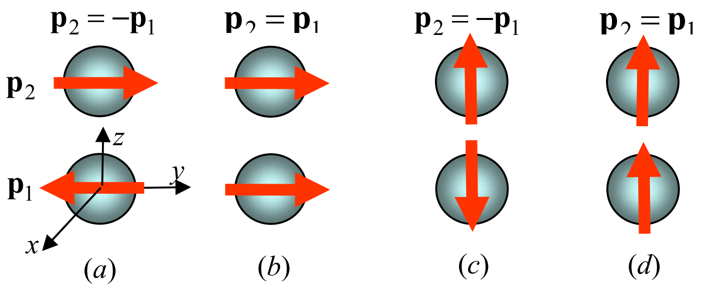

Fig. 1. Nanosphere pairs and resonance configurations: there are two resonances along the transverse direction (*x-y* plane) and two other resonances along *z*. The vectors on each particle indicate the electric dipole moments.

It can be easily shown that the system admits independent antisymmetric ($\mathbf{p}_2 = -\mathbf{p}_1$) and symmetric solutions ($\mathbf{p}_2 = \mathbf{p}_1$), which leads to two sets of solutions of the equation $\left[\underline{\mathbf{G}}(\mathbf{r}_1,\mathbf{r}_2) \pm \underline{\mathbf{I}}/\alpha\right]\cdot\mathbf{p}_1 = 0$, where the + sign holds for antisymmetric solutions, whereas the – sign holds for symmetric solutions. The eigenvectors associated to the four possible resonance configurations are shown in Fig. 1. Below we plot the two eigen-frequencies $f_{t,1}$ and $f_{t,2}$ associated to the two transverse resonances in Figs. 1(a) and 1(b), respectively. We also plot the two eigen-frequencies $f_{,1}$ and $f_{,2}$ relevant to the two longitudinal resonances in Figs. 1(c) and 1(d), respectively. The real and imaginary parts of the four resonance frequencies are plotted against spheres radius r_0 (Fig. 2), keeping fixed the spheres distance at $H = 75\,\mathrm{nm}$.

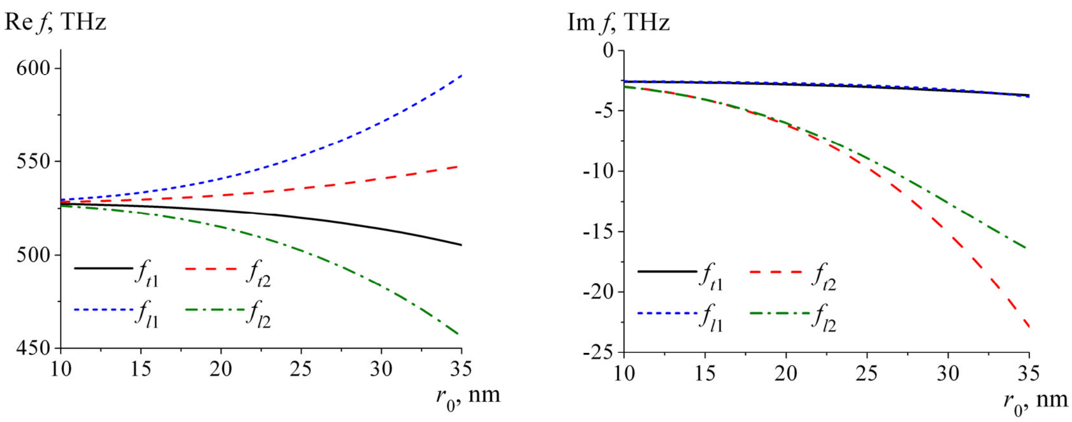

Fig. 2. Real and imaginary parts of the four resonance frequencies versus spheres radius r_0 for a fixed sphere distance $H = 75\,\mathrm{nm}$.

It appears that $f_{t,2}$ and $f_{,2}$, the resonances of the symmetric modes in Figs. 1(b) and 1(d), have a larger imaginary part than $f_{t,1}$ and $f_{,1}$, the resonances of the antisymmetric modes in Figs. 1(a) and 1(c). This is because of larger radiation losses for symmetric modes where the pair of spheres radiate like dipoles. Instead, radiations associated to the two antisymmetric dipoles tend to cancel each other and the antisymmetric dipole pair tends to radiate like a quadrupole. Equivalently, the antisymmetric mode is associated to a current loop and it is a classic result of antenna theory that a small loop radiates less than a small dipole. It is noted that the antisymmetric resonance $f_{t,1}$ is smaller than the symmetric one $f_{t,2}$, for all the considered cases; generally, the resonances are ordered as $f_{,2} < f_{t,1} < f_{t,2} < f_{,1}$. Moreover, for large ratios H/r_0 the real parts of the four resonance frequencies tend to be similar due to the reduced coupling between spheres.

3. Array of pairs of tightly coupled spheres

The structure under investigation is shown in Fig. 3. It consists of an infinite, two-dimensional periodic distribution, in the *x*-*y* plane, of pairs of tightly coupled spheres. In other words, each array element is made of the couple of nearby metallic spheres shown in Fig. 1. The lattice constants of the array are a and b along *x* and *y*, respectively. The coordinate origin is placed at the position of a sphere. The positions of the lower spheres are $\rho_{mn} = ma\hat{\mathbf{x}} + nb\hat{\mathbf{y}}$, $m,n = 0,\pm1,\pm2,\ldots$

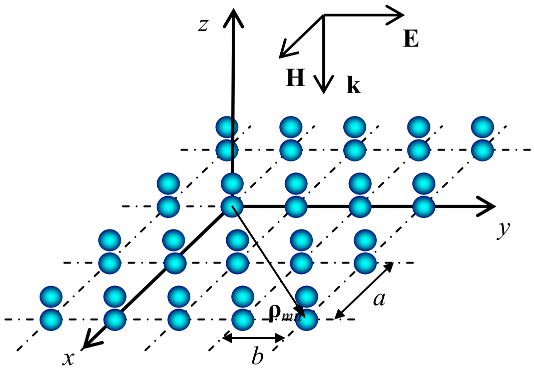

Fig. 3. Array of pairs of tightly coupled spheres.

We have investigated the transmission and reflection properties of an array of pairs of tightly coupled spheres, assuming a plane wave coming from the +*z* direction, as shown in Fig. 3. The magnitude of reflection and transmission coefficients shown in Fig. 4, for H = 75 nm, r_0 = 35 nm and a = b = 200 nm, are obtained by finite difference simulations [7]. For symmetry reasons, only the transverse resonance modes of Fig. 1(a) and 1(b) may be excited. Indeed, the sphere pair array shows two distinct resonances around f = 460 THz and f = 525 THz corresponding to the antisymmetric and symmetric modes, respectively,. To further strengthen this identification of the resonances, we performed theoretical calculations by an approximated dipolar model for periodic structures, deduced similarly to Eq. (2), and we numerically analyzed the electric and magnetic field behavior at resonances.

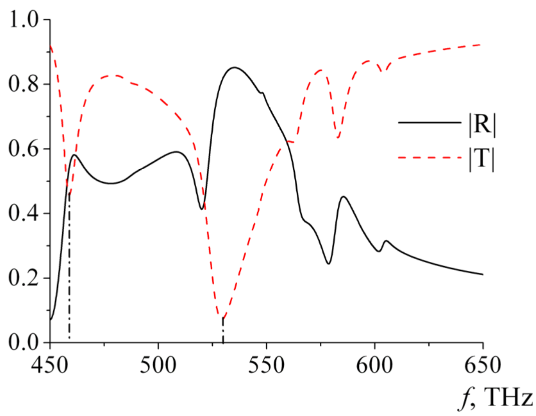

Fig. 4. Reflection by and transmission through a periodic array of sphere pairs with $r_0 = 35\,\text{nm}$ and $H = 75\,\text{nm}$.

These latter calculations, illustrated in Fig. 5, have confirmed that a normally incident plane wave at f = 458 THz excites anti-phase currents flowing mostly along the y axis (Fig. 5(a)), which is a characteristic of the antisymmetric mode. A current loop is effectively created in the y-z plane, which generates strong magnetic fields in the region between spheres and produces a substantial magnetic moment contributing to permeability. On the other hand, an incident plane wave at f = 525 THz excites mostly in-phase currents flowing along the y axis (Fig. 5(b)), which do not produce a significant enhancement of the magnetic field.

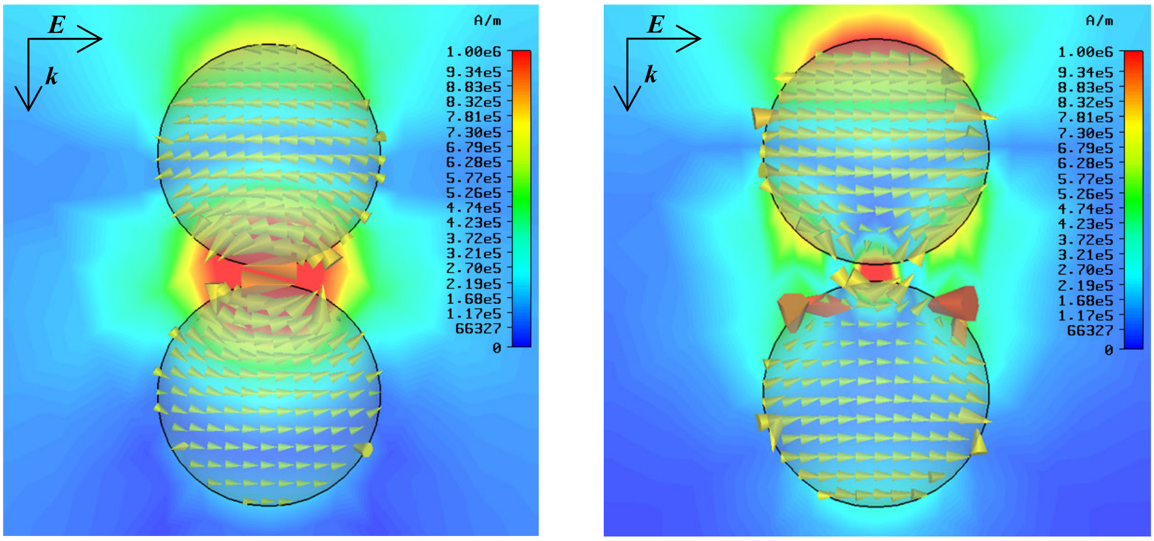

Fig. 5. Current and magnetic field amplitude distributions for an array of sphere pairs with r_0 = 35 nm, H = 75 nm, a = b = 200 nm: (a) antisymmetric resonance mode at f = 458 THz; (b) symmetric resonance mode at f = 525 THz.

4. References

1. A. N. Grigorenko, A. K. Geim, H. F. Gleeson, Y. Zhang, A. A. Firsov, I. Y., Khrushchev, J. Petrovic, “Nanofabricated media with negative permeability at visible frequency,” *Nature*, vol. 438, November 2005, pp. 17–20.

2. J. B. Pendry, S. O’Brien, “Magnetic activity at infrared frequencies in structured photonic crystals. *J. Phys. Condens. Matter*, vol. 14, 2002, pp. 6383–6394.

3. T. J. Yen *et al*., “Terahertz magnetic response from artificial materials,” *Science*, vol. 303, 2004, pp. 1494–1496.

4. V. M. Shalaev, W. Cai, U. K. Chettiar, H. Yuan, A. K. Sarychev, V. P. Drachev, and A. V. Kildishev, “Negative index of refraction in optical metamaterials,” *Optics Letters*, vol. 30, December 2005, pp. 3356–3358.

5. E.V. Ponizovskaya and A.M. Bratkovsky "Ensembles of plasmonic nanospheres at optical frequencies and a problem of negative index behavior," *Appl. Phys. A*, vol. 87, 2007, pp. 175–179.

6. A. Alù and N. Engheta "Three-dimensional nanotransmission lines at optical frequencies: A recipe for broadband negative-refraction optical metamaterials," *Physical Review B*, vol. 75, January 2007, pp. 024304-1–024304-20.

7. *CST Microwave Studio*, User Manual Ver. 5, CST GmbH, Darmstadt, Germany, February 2004.

8. C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles*, Wiley, New York, 1983.

9. I. El-Kady, M. M. Sigalas, R. Biswas, K. M. Ho, and C. M. Soukoulis, “Metallic photonic crystals at optical wavelengths,” *Phys. Rev. B*, vol. 62, 2000, pp. 15299–15302.