Magnetic Resonance in Metal Nanoantennas

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ABSTRACT

It is predicted that metal nanostructures can have a magnetic plasmon resonance (MPR) in the optical spectral range. Similarly to the known electrical surface plasmon resonance (SPR), the MPR depends only on metal properties and geometry of the system, rather than on the wavelength, and it can occur in structures much smaller in size than the optical wavelength. The MPR can have as large optical cross-section as the SPR so that nanostructures supporting the two resonances can provide a strong coupling to both field components of light, electrical and magnetic. Above the resonance, the magnetic plasmon polarizability may acquire negative values making possible to use this phenomenon for developing left-handed materials in the optical range.

1 Introduction

The optical properties of nanostructured metamaterials have been intensively studied during the last decade. It has been proposed by Pendry, who further developed earlier studies on negative refraction,[2,1] that a metamaterial with negative dielectric permittivity ε and negative magnetic permeability μ could be used for developing a super-lens providing a sub-wavelength resolution. According to Pendry, when the scattered light passes through a

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material with a negative refractive index (specifically, when $n = \sqrt{\varepsilon\mu} = -1$ and the two impedances are matched), the evanescent components of the scattered field grow exponentially, allowing the restoration of the scattered image with a subwavelength resolution. Smith, Padilla, Vier, and Shultz[3] have demonstrated negative-refraction materials in the microwave range. These materials are also referred to as double-negative or left-handed materials (LHMs), because the electric field and magnetic field along with the wavevector form a left-handed system in this case. In addition to super resolution, the unusual and sometimes counter-intuitive properties of LHMs make them very promising for applications in resonators, waveguides and other microwave and optical elements (see[4] and[5–7]).

In spite of large efforts LHMs have not been demonstrated yet in the optical range. To obtain a negative refraction in the optical range, one needs to have a metamaterial with optical magnetism, which is a challenging problem because magnetism is typically weak in the high-frequency range. Relaxation times of paramagnetic and ferromagnetic processes are long in comparison with the optical period and collective magnetic responses become small at high frequencies. With no collective effects, the magnetic susceptibility is very small since it is proportional to $\sim v^2/c^2 \sim \beta^2 \sim 10^{-4}$, where v is the velocity of electron in atom, c is the speed of light, and $\beta = e^2/\hbar c \cong 1/137$ is the electromagnetic constant. [This is because the ratio v/c appears first with the magnetic field $\mathbf{H}$ in the interaction Hamiltonian and again in the magnetic moment $\mathbf{M}$ of atoms.] We note that for microwave LHMs, artificial magnetic elements, such as split-ring resonators and Swiss Roll structures, have been suggested and successfully implemented (see, for example,[8,3] and[4] p.709).

In this paper we predict a magnetic plasmon resonance (MPR) for the optical spectral range. The existence of such a magnetic resonance makes possible to develop structures with optical magnetism and implement them for LHMs in the optical range. In our previous papers we proposed optical LHMs based on half-wavelength-long metal rods so that a magnetic resonance in this case was directly related to the wavelength.[9] Here we show that a magnetic resonance can occur in structures much smaller than the wavelength. Moreover, in close analogy with the electrical surface plasmon resonance (SPR), the MPR depends only on material properties and geometry of the system rather than on the wavelength. Similar to the electrical SPR where the optical cross section of a nanostructure with size $a << \lambda$ can be as large as λ^2, the MPR can also be characterized by a large

optical cross-section. When nanostructures support simultaneously SPR and MPR, a strong coupling to both field components of light, electrical and magnetic, becomes possible. Therefore, such plasmonic nanostructures can act as efficient couplers across the micro-nano interface for the optical and infrared part of the spectrum, where the light wavelength is on the micron scale so that interactions with nanostructures are typically weak, without the resonant enhancement provided by SPR and MPR.

2 Magnetic Plasmon Resonance

The electrical SPR occurs in the optical and infrared part of the spectrum and results from a collective electron oscillations in metal structures. Consider, for example, an elliptical metal particle that has the electrical dipole polarizability $\alpha_E \sim [1+\gamma(\varepsilon_m - 1)]^{-1}$, where ε_m is the metal permittivity and $\gamma < 1$ is the depolarization factor, which depends on the aspect ratio. For "good" optical metals (Ag, Au, Al, etc.), the real part of ε_m is negative and large while its imaginary part is relatively small in the optical range. The plasmon resonance corresponds to the condition $\mathrm{Re}\,\varepsilon_m(\omega) = 1 - 1/\gamma$ and it critically depends on metal properties and the shape of a metal nanoparticle. For particles much smaller than the wavelength, the SPR is size- and wavelength-independent. Many important plasmon-enhanced optical phenomena and applications of metal nanocomposites are based on the electrical SPR (see, for example,[10]).

Bellow we show that along with the electrical SPR, specially arranged metal nanoparticles can support a magnetic plasmon resonance (MPR), with the resonance frequency ω_r independent of the size and λ. Such structures act as optical nanoantennas by concentrating large electric and magnetic energies on the nanoscale at the optical frequencies. The magnetic response is characterized by the magnetic polarizability α_M with a resonant behavior similar to α_E: its real part part changes the sign near the resonance and becomes negative for $\omega > \omega_r$, as required for LHMs.

We consider first a pair of parallel metal rods. The external magnetic field excites the electric current in the pair of the rods as shown in Fig. 1. The magnetic moment associated with the circular current flowing in the

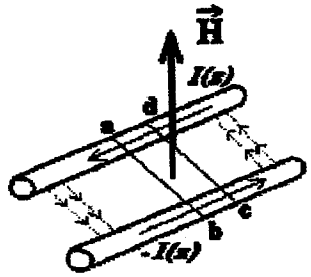

Figure 1: Current in the two-rod circuit excited by external magnetic field **H**. The displacement currents, "closing" the circuit, are shown by dashed lines.

rods results in a magnetic response of the system. Suppose that an external magnetic field $H = H_0 \exp(-i\omega t)$ is applied perpendicular to the plane of the pair. The circular current $I(z)$ excited by the magnetic field flows in opposite directions in the nanowire pair, as shown in Fig. 1. The displacement currents flowing between the nanowires close the circuit. We introduce the electric potential $U(z) = \int_a^b \mathbf{E}d\mathbf{l}$ between the pair where the integration is along the line $\{a(z), b(z)\}$. To find the current $I(z)$, we integrate the Maxwell equation $\mathrm{curl}\,\mathbf{E} = ik(\mathbf{H_0}+\mathbf{H_{in}})$ over the contour $\{a, b, c, d\}$ in Fig. 1, where $k = \omega/c$ and $\mathbf{H_{in}} = \mathrm{curl}\,\mathbf{A}$ is the magnetic field induced by the current. We take the nanowire length $2a$ to be much larger than the distance d between the nanowires and the radius of a nanowire $b \ll d$. We also assume that $kd \ll 1$. Under these assumptions, the vector potential **A** is directed along the nanowires (z direction) and the integration of the Maxwell equation gives

$$(IR - ikA_z + dU/dz)\Delta z = ikH_0 d\Delta z, \tag{1}$$

where the pair impedance $R \simeq 2/(\sigma\pi b^2) = 8i/(\varepsilon b^2\omega)$ [$\varepsilon = i4\pi\sigma/\omega$ is the metal permittivity] and $\pm IR/2$ are the electric fields on the surface of the nanowires. The electric potential $U(z)$ between the pair is given by the solution to Maxwell's equations that can be written for $a \gg d \gg b$ as

$$U(z) \cong 2q(z)\int_{-a}^{a}\left(\frac{1}{r_1}-\frac{1}{r_2}\right)dz' + \int_{-a}^{a}\left[\frac{q(z')\exp(ikr_1)-q(z)}{r_1} - \frac{q(z')\exp(ikr_2)-q(z)}{r_2}\right]dz' \tag{2}$$

where $q(z)$ is the electric charge per unit length, $r = \sqrt{(z-z')^2+b^2}$, $r_2 = \sqrt{(z-z')^2+h^2}$ and the terms $\sim (b/a)^2$ are neglected. We explicitly separate in Eq. (2) the first term, which has a singularity when $b \to$

0; it can be estimated as $4q(z)\ln(d/b)$. The second term in Eq. (2) is regular for $b \to 0$ and we can expand it over $d/a \ll 1$. Thus we obtain a local relation between U and q: $U(z) = C\,q(z)$, where $C = \left[4\log(d/b) - 3(d/a)^2 + (dk)^2\,(2\log(2a/d) - 1)/2\right]^{-1}$. The vector potential A_z can be found following the same procedure which results in $A_z(z) = (L/c)\,I(z)$, where the inductance is given by $L = 4\ln(d/b) - (d/a)^2 + (dk)^2\,[3 + 4iak + 6\log(2a/d)]/6$. We substitute $U(z)$ and $A_z(z)$ into Eq. (1), taking into account the charge conservation law $dI/dz = i\omega q(z)$, and obtain the second-order differential equation for the current,

$$\frac{d^2 I(z)}{dz^2} = -g^2 I(z) + \frac{Cd\omega^2}{c} H_0, \tag{3}$$

where $-a < z < a$, $I(-a) = I(a) = 0$, and parameter g is given as $g^2 = k^2\left[LC - 8C\left[(kb)^2\varepsilon_m\right]^{-1}\right]$. The product LC can be estimated as $LC \sim 1$. We consider here the "quasistatic" case $\left|8C\left[(kb)^2\varepsilon_m\right]^{-1}\right| \gg 1$ when parameter g does not depend on the wavelength of light:

$$g^2 \simeq -2\ln(d/b)\,/\,(b^2\varepsilon_m). \tag{4}$$

The case of strong skin effect ($\left|(kb)^2\varepsilon_m\right|^{-1} \ll 1$, $g \simeq k$), when the pair of metal wires has a so-called antenna resonance at $ka = \pi/2$, was numerically simulated in our previous papers[11,9] and in papers by Panina et al..[12]

We solve Eq. (3) for the current $I(z)$ and calculate the magnetic moment $\mathbf{m} = (2c)^{-1}\int[\mathbf{r}\times\mathbf{j}(\mathbf{r})]\,d\mathbf{r}$, where $\mathbf{j}(\mathbf{r})$ is the density of the current and the integration is over the two nanowires as well as over the space between them where the displacement currents are flowing. Thus we obtain

$$m = \frac{1}{2} H_0 a^3 \ln(d/b)\,(kd)^2\,\frac{\tan(ga) - ga}{(ga)^3}. \tag{5}$$

The metal permittivity ε_m has a large negative value in the optical range while its imaginary part is small; therefore, the magnetic moment m has a resonance at $ga \approx (a/b)\sqrt{2\ln(d/b)\,|\varepsilon_m(\omega)|} = \pi/2$ when m attains large values. The magnetic resonance frequency $\omega = \omega_r$ depends on geometry of the system and material properties. In analogy with the the electric surface plasmon resonance (SPR) in metal nanoparticles, we see that for the magnetic plasmon resonance (MPR), the size of the sticks can also be arbitrary small in comparison with the wavelength of the incident light. This is in a striking difference with the previously considered magnetic resonance at $ka = \pi/2$.[9] For a perfect metal, ($\mathrm{Im}\,\varepsilon_m = 0$) the magnetic polarizability $4\pi(m/H_0)$ goes to $-\infty$ at the resonance. Thus, the MPR opens the possibility for engineering efficient LHM in the optical range.

For a typical metal, the permittivity $\varepsilon_m(\omega)$ can be well approximated by the Drude formula for the red and infrared parts of the spectrum: $\varepsilon_m(\omega) \cong -(\omega/\omega_p)^2 / (1 - i\omega_\tau/\omega)$, where ω_p is the plasma frequency and the relaxation parameter is small, $\omega_\tau/\omega \ll 1$. Then the polarizability α_M normalized to the volume $V = 4abd$ of the pair has the following form near the magnetic resonance:

$$\alpha_M = \frac{4\pi m}{H_0 V} = \frac{16\, a\, d\, \omega_p}{\lambda^2\, \omega_r \sqrt{2\log(d/b)}} \left[1 - \omega/\omega_r - i\omega_\tau/(2\omega_r)\right]^{-1}, \tag{6}$$

where the resonance frequency $\omega_r = b\pi\omega_p\sqrt{2\log(d/b)}/(4a)$. The plasma frequency ω_p is typically in the ultraviolet part of the spectrum so that $\omega_p \gg \omega_r$. Therefore, the pre-factor in Eq. (6) can be on the order of one, even for a nanowire length $2a$ much smaller than the wavelength λ of the incident light, so that a strong MPR can be observed. We can also estimate the optical cross-section for the MPR, $\sigma_M \sim \alpha_M V/\lambda$, assuming that the logarithm factor is ~ 1 and that radiation losses dominate (so that $\omega_\tau \sim \omega V/\lambda^3$); this gives $\sigma_M \sim d^2 a/b$. Thus, the magnetic cross-section σ_M can be very large and, in particular, comparable to λ^2 (as in the case of the electric SPR in spheroids, where $\sigma_E \sim \lambda^2$), despite the fact that all sizes involved are much smaller than the wavelength. Thus, by employing both resonances, SPR and MPR, one can accomplish a strong coupling of nanostructures to both components of light, electrical and magnetic. For a material composed of plasmonic nanostructures that support SPR and MPR, the dielectric permittivity and magnetic permeability can be both negative above the resonance, resulting in a negative refraction (for simplicity, we assume here that the two resonances do not interact).

We now consider a metal nanoantenna that has a $\sqsubset$ shape, which is obtained from a pair of nanowires by shorting it at one of the ends (see Fig. 2). When the quasistatic condition $\left|8C\left[(kb)^2\varepsilon_m\right]^{-1}\right| \gg 1$ holds, the electric current $I(z)$ in a $\sqsubset$ nanoantenna can be obtained from Eq. (3), where the boundary condition changes to $I_{z=a} = (dI/dz)_{z=0} = 0$ and, as above, $a \gg d \gg b$. It is easy to check that the magnetic polarizability α_M is still given by Eq. (6), where a is now equal to the total length of the $\sqsubset$ nanoantenna. Therefore, the $\sqsubset$ nanoantenna provides the same magnetic polarizability α_M at twice shorter length of the parallel rods than in the case above.

Consider now a magnetic permeability μ for a metamaterial where the stick pairs (or $\sqsubset$ nanoantennas) are oriented in one direction ("z" direction in Fig. 1) and are organized in the periodic square lattice. The tensor

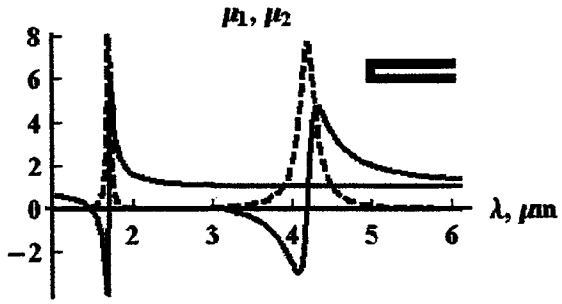

Figure 2: Optical magnetic permeability $\mu = \mu_1 + \mu_2$ (μ_1– continous line, μ_2–dashed line) of the composite containing $\sqsubset$ shaped silver nanoantennas; volume concentration $p = 0.3$; left curves: $a = 200\,nm$, $d = 50\,nm$, $b = 13\,nm$; right curves $a = 600\,nm$, $d = 90\,nm$, $b = 13\,nm$.

μ component, which is in the direction perpendicular to the plane of the sticks (**H** direction in Fig. 1), can be estimated from the Lorenz-Lorentz formula[14] $(\mu_e - 1)/(\mu_e + 1) = p\alpha_M/3$, where p is the volume concentration of the pairs.

Results of our calculating $\mu_e = \mu_1 + i\mu_2$ for silver $\sqsubset$ nanoantennas are shown in Fig. 2; the optical parameters for silver were taken from.[10,15] As one can see in the figure, the negative magnetism can be observed, for example, in the near-infrared part of the spectrum, including the telecommunication wavelength of 1.5 μm. By varying nanoantenna parameters, one can tune the position of the MPR for any frequency in the visible and infrared parts of the spectrum.

For practical applications of the optical magnetism losses may play an important role. We estimate losses (given by μ_2) at the wavelength corresponding to the condition $\mu_1 = -1$ as $\mu_{2r} = \lambda_r^2\,\omega_\tau\,\sqrt{2\log(d/b)}/(8\,\omega_p a\,d\,p)$, where λ_r is the resonance wavelength. For metals at room temperatures losses are significant (for silver, $\mu_{2r} \sim 0.3$, see Fig. 2) but still they are relatively small. These losses can be much smaller at low temperatures and atomic quality of metal crystals. We also note that the radiative losses (which are given by the small imaginary part of the inductance L) are of no importance for nanoantennas arranged in a periodic array (i.e., in a plasmonic crystal); the radiation corrections, in this case, result in a change in the spatial dispersion rather than in an

increase of μ_2.

3 Conclusions

We considered here two types of nanoantennas that support the magnetic plasmon resonance (MPR) in the optical range. Other possible designs could include, for example, nanosized metal spheres sectored into eight equal parts by thin dielectric slits and split-ring resonators (SRRs). The SRRs were successfully used earlier for the microwave LHMs.[3] A subwavelength SRR can provide a large magnetic polarizability at the resonance, when the radius is as follows $R \simeq d\,|\varepsilon_m|/(2\pi)$, with d being the thickness of the dielectric slit in the ring. However, it seems hard, if not impossible, to have the concentration of SRRs large enough to provide a reasonable negative magnetic permeability in the optical range. When the distance between the rings becomes less than their radii, the rings strongly interact. Then, any particular ring is subjected to the external magnetic field and the fields of the neighbor rings. For a negative magnetic response ($\mathrm{Re}\,\alpha_M < 0$), the magnetic field induced by the rings is opposite to the external field so that the SRRs effectively screen the external field and a collective magnetic response decreases with a further increase of the concentration. Our estimates show that for the optimal concentration, a negative magnetic response of a SRR metamaterial is significantly smaller than for the $\sqsubset$ nanoantenna metamaterial considered above. Yet we would like to stress out that SRR metamaterials can have a large paramagnetic response in the optical range (with large and positive μ) with many interesting applications.

In conclusion, we show that a specially designed metal nanoantenna, which is much smaller than the light wavelength, can have a magnetic plasmon resonance (MPR) with the resonant frequency depending on the shape and material properties of the nanoantenna rather than on the wavelength. In this sense, the MPR is similar to the electric surface plasmon resonance (SPR) in a metal nanoparticle. We show that composites comprising such non-magnetic nanoantennas may have a large magnetic response in the optical spectral range. Metamaterials based on plasmonic nanoantennas supporting both SPR and MPR can have a dielectric permittivity and magnetic permeability, which are simultaneously negative, and thus act as left-handed materials in the optical and infrared spectral ranges.

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