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Near-infrared photonic band gaps and nonlinear effects in negative magnetic metamaterials

S. O'Brien,[1] D. McPeake,[1] S. A. Ramakrishna,[2] and J. B. Pendry[3]
[1]*NMRC, University College, Lee Maltings, Prospect Row, Cork, Ireland*
[2]*Department of Physics, Indian Institute of Technology, Kanpur - 208 016, India*
[3]*Condensed Matter Theory Group, The Blackett Laboratory, Imperial College, London, SW7 2BZ United Kingdom*

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We describe and characterize a nanostructured metallic photonic crystal metamaterial which is magnetically active in the near-infrared region of the spectrum. The periodic array of modified split-ring resonator structures is numerically demonstrated to have a negative effective permeability at telecommunications wavelengths. Local electric fields in the structure can be many orders of magnitude larger than in free space thus allowing for enhanced nonlinear effects. We have derived an expression for the change in the resonance frequency of the structure due to the Kerr nonlinear index change and estimate a characteristic magnetic field strength for the observation of bistable behavior.

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Subwavelength structured metallic composites are currently the object of intense experimental and theoretical interest. Of particular interest is the split-ring resonator (SRR) structure which behaves as an artificial magnetic atom through a resonant overscreening response to a time varying applied magnetic field. Driven by the back electromotive force, an antiphased response above the resonance frequency makes a negative effective permeability possible in arrays of these structures.[1]

Artificial plasmas composed of arrays of thin conducting wires can have a negative effective permittivity.[2] When combined with arrays of SRRs a negative refractive index or left-handed medium can result where the permeability (μ) and permittivity (ϵ) are simultaneously negative.[3,4] Perhaps the most striking property of a negative refractive index material is the possibility of subwavelength imaging by a plane slab in vacuum at the single frequency where $\epsilon=\mu=-1$.[5]

An important simplification of the "perfect lens" condition earlier results when we consider the case when all length scales are much smaller than the wavelength of light (the quasistatic limit). Then, in the case of P-polarized fields, it can be shown that only the value of the permittivity is important. Subwavelength resolution is then possible with, for example, thin films of silver where the permittivity becomes equal to -1 in the visible region of the spectrum and with small losses.[6,7] Due to the inherent symmetry of the Maxwell equations, low-loss artificial magnetic materials with a negative permeability can also be expected to exhibit the remarkable properties of "negative epsilon" materials such as silver but of course with the roles of the magnetic and electric fields exchanged. Using a physical model for the quasistatic response of a SRR array and the results of transfer matrix simulations, we show here that SRR arrays can have a negative permeability at near-infrared frequencies.

Because the origin of the stop band is a single scatterer resonance and a negative permeability rather than a macroscopic Bragg resonance, significant attenuation is achievable with a small number of layers of SRR arrays. Thus, SRRs have much potential to form compact subwavelength photonic band gap media.

Local electric field enhancements in the SRR array can be intense.[1,8] This property thus allows for enhanced nonlinear effects. To illustrate this potential for nonlinear functionality, we have derived an expression for the change in the resonance frequency of the structure as a function of the incident magnetic field intensity when the structure is embedded in a Kerr nonlinear dielectric. We estimate a characteristic magnetic field strength which leads to bistable behavior and switching of the structure from negative to positive effective permeability.

The zeroth order transverse electric Mie resonance in dielectric particles can in principle lead to a nonzero magnetic susceptibility.[9] When the refractive index of the particles is large, spatial averaging of the highly inhomogeneous magnetic fields excited near resonance can lead to a bulk magnetization of the system.

At optical frequencies the dielectric response of noble metals is dominated by the plasma like behavior of the electron gas. It can be described by a complex dielectric function of the form

$$\tilde{\epsilon}(\omega) = (\epsilon_1, \epsilon_2) = \epsilon_\infty - \frac{\omega_p^2}{\omega(\omega + i\gamma)}. \tag{1}$$

Here ω_p is the plasma frequency, and γ is the relaxation rate which gives rise to the resistive dissipation.

For metallic scatterers [where $\tilde{\epsilon}(\omega)<0$], the transverse electric Mie modes become the surface plasmons of the particle. These modes are responsible for the very rich photonic band structure found in certain metallic systems at optical frequencies.[10,11] However, we cannot expect these resonant modes to be magnetically active as they begin with the dipolar surface plasmon varying as $\exp(im\phi)$, where $m\geqslant 1$. In any case, significant penetration of the fields into the particle interior is required in order that spatial averaging of fields give an effective magnetization. Analogous to the Mie resonance in dielectric particles, by structuring the metallic scatterer to form a SRR we can create a resonant mode with the required magnetic dipolar symmetry $(m=0)$. In each case

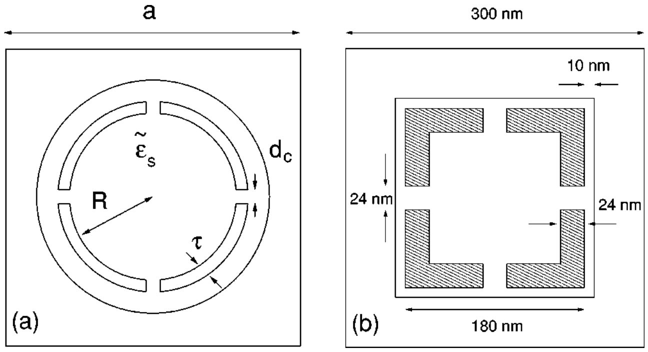

FIG. 1. Two-dimensional unit cells that we consider. (a) A circular resonator comprising a negative epsilon material with internal radius R and film thickness τ. (b) A square resonator with side length $b=180$ nm. The shaded areas are films of thickness $D=24$ nm and composed of silver. Each structure is embedded in a dielectric matrix of relative permittivity $\tilde{\epsilon}_s$ with a vacuum region separating neighboring cells. A total of four splits determine the capacitance of each structure.

then, to the external observer, the inclusions seem to oscillate as a plasma of fictitious magnetic poles.

By considering the quasistatic magnetic response of a two-dimensional SRR structure composed of a material with a negative permittivity, one can derive an important relation which leads to the optimal structure for a resonant magnetic response. The SRR consists of a thin circular film of negative epsilon material in a dielectric matrix of relative permittivity $\tilde{\epsilon}_s$. We assume that the thickness of the films, τ, is small in comparison to the skin depth, $\delta \simeq c_0/\omega_p$, and that the separations d_c are small in comparison to the internal radius R. Results presented here will be for SRRs composed of silver where $\delta \simeq 22$ nm. The structure incorporates n_c divisions around the circumference as shown in Fig. 1(a). By placing the divisions symmetrically, we ensure that the quasistatic distribution of induced charge has zero net electric dipole moment. This screening is an important factor limiting the dielectric response of the structure at resonance.

For a high frequency magnetic field applied perpendicular to the plane of the unit cell and assuming a small filling fraction, f, the quasistatic effective permeability of a two-dimensional array is given by [8]

$$\tilde{\mu}_{\text{eff}} = 1 - \frac{f' \omega^2}{\omega^2 - \omega_0^2 + i\Gamma\omega}, \tag{2}$$

where $f' = L_g f \cdot (L_g + L_i)^{-1}$, $\Gamma = L_i \gamma \cdot (L_g + L_i)^{-1}$, and $\omega_0^2 = (L_g + L_i)^{-1} C^{-1}$. In the earlier expressions $L_g = \mu_0 \pi R^2$ is the geometrical inductance per unit length of the structure and $C = \epsilon_0 \tilde{\epsilon}_s \tau / n_c d_c$ is the capacitance per unit length of the structure for series connection.

An additional inductive impedance in the structure, the inertial inductance, $L_i = 2\pi R/\epsilon_0 \omega_p^2 \tau = 2\mu_0 \pi R \delta^2/\tau$, determines the effective filling fraction and damping of the resonance through the ratio of the two contributions to the total inductance. This contribution to the inductance arises from the finite electron mass and implies that simply decreasing the size of the resonators indefinitely will not result in our being able to realize a strong magnetic response at near-infrared or optical frequencies.

Strong damping of the resonance will be avoided if the quantity $R\tau/2\delta^2$ is large. In fact, with δ equal to the London penetration depth, this ratio also determines the screening efficiency of low frequency magnetic fields by a thin layer of superconductor.[12] At optical frequencies, once $\tau \sim \delta$, the finite incident wave vector will give rise to multiple scattering within the metal film leading to increased absorption. Further, our use of the bulk dielectric function for silver will only be valid if the film thickness is sufficiently large. Measurements suggest that this minimum is greater than 20 nm.[13] Thus, key to the high frequency operation are the requirements $R \gg \delta \sim \tau$.

At submicron scales and in two dimensions, a nanostructured metallodielectric SRR such as that depicted in Fig. 1(a) represents a very ambitious goal for fabrication technologies. As a planar element, however, the structure could be argued to be within the capability of present day techniques. In Ref. 14 for example, monodisperse gold nanorings were prepared by colloidal lithography. By introducing the capacitance and stacking arrays of the structures, we can then approximate the response of the two-dimensional array.

Planar SRRs with a cross section where the capacitance in the structure might be easier to introduce are also of interest. As an example, we consider a unit cell comprising a structured square silver column embedded in a dielectric matrix as shown in Fig. 1(b). The two-dimensional unit cell has side length a which we take to be 300 nm. The metallic sheets are of uniform thickness $D=24$ nm while separations $d_c=24$ nm define the capacitance in the structure as shown. For the dielectric function of silver as defined in Eq. (1) we use the empirical values $\omega_p=9.013$ eV and $\gamma=0.018$ eV.[13]

The complex reflection and transmission coefficients of a slab of such a material can be calculated using the transfer matrix method. Assuming a homogeneous medium, these quantities can be inverted to find the refractive index, n, and the wave impedance, z. From these quantities we can calculate the effective permittivity, $\epsilon_{\text{eff}}=n/z$ and the effective permeability, $\mu_{\text{eff}}=nz$.[15] Here we assume these response functions are local quantities. With $\tilde{\epsilon}_s=4$, we have plotted the reflectance, transmittance, and absorbance, for four and for eight layers of the structure in Fig. 2. In the case of four layers, the transmittance we calculate is as small as 10^{-7} above the resonance. The reflectance in these regions is in excess of 90%. The SRR array therefore has much potential as a subwavelength photonic band gap medium.

The stop band region includes the technologically important telecommunications band near 1.5 μm or 200 THz. Thus, our free space wavelength at the resonance frequency is approximately five times the unit cell dimension. This degree of separation is probably not sufficient to qualify our resonant structure as a true effective medium. Nevertheless, for the purposes of illustration, we will assume that the effective permittivity and permeability we calculate are representative of the physical properties of the discrete crystal structure.

The refractive index and impedance and the effective permittivity and permeability we derived for the structure are

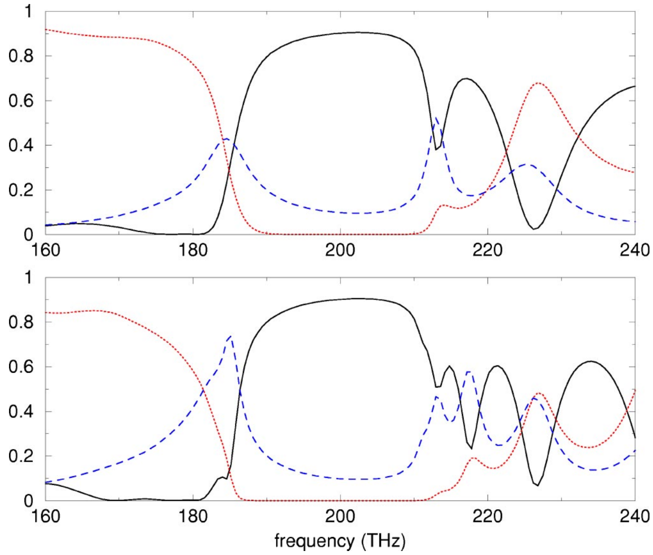

FIG. 2. (Color online) Reflectance, transmittance, and absorbance for four layers of the magnetic structure (top) and eight layers (bottom). The reflectance is the solid line, the transmittance is the dotted line, and the dashed line is the absorbance in each case. The dimensions defining the magnetic structure are as for Fig. 1(b).

given in Fig. 3. The effective permeability is indeed negative in the region near 200 THz where the crystal is highly reflecting. This result suggests that arrays of subwavelength SRR structures can form low loss negative magnetic metamaterials over an unprecedented range from microwave to optical frequencies.

The characteristic forms of the effective response functions shown in Fig. 3 were also found in previous numerical studies at microwave frequencies.[16,17] In Ref. 17 it was argued that the anomalous negative sign of the imaginary part of the effective permittivity is due to the finite spatial periodicity. Further theoretical work will be necessary to elucidate the roles of spatial periodicity, bianisotropy, and inadequate screening of the induced dipolar fields in determining the results of numerical simulations such as these.

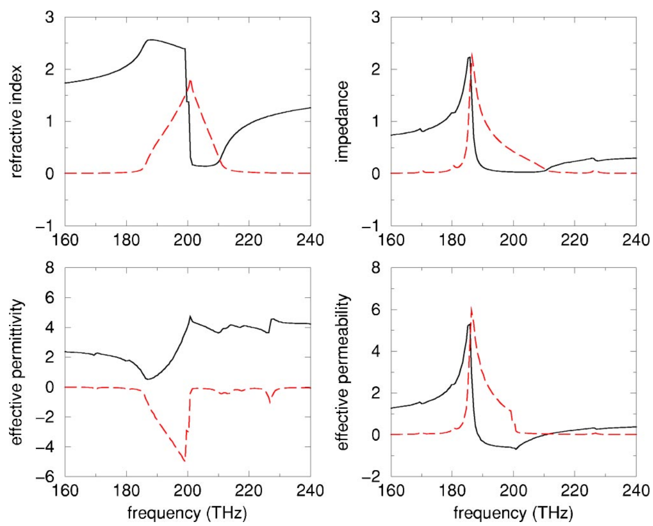

FIG. 3. (Color online) The calculated effective refractive index (top left) and effective impedance (top right) and the calculated effective permittivity (bottom left) and permeability (bottom right) of the magnetic structure of the magnetic structure. The solid lines are the real parts and the dashed lines are the imaginary parts in each case. The dimensions defining the magnetic structure are as for Fig. 1(b).

The steady-state nonlinear properties of a negative index medium composed of arrays of SRRs and metallic wires in a nonlinear dielectric was discussed in Ref.18. Here we perform a similar analysis for the SRR array alone and with parameters appropriate to optical frequencies. We consider the nonlinearity due to the intensity dependence of the refractive index where $n_s=\sqrt{\tilde{\epsilon}_s}=n_0+n_2I$. Here n_0 is the refractive index of the dielectric in the linear limit, I is the electric field intensity, and n_2 determines the intensity dependence of the refractive index. Of particular interest is the case of a defocusing nonlinearity, $(n_2<0)$, where a beam which is located in the band gap region where $\mu_{\text{eff}}<0$ could, as the intensity is increased, switch to the region below the resonance with reflectance close to zero and low transmission losses.

To illustrate the potential for bistable switching of the SRR array, we neglect the dispersion in the dielectric response of the composite and describe the magnetic response of the structure of Fig. 1(b) in an idealized picture through the quasistatic response. With geometric parameters describing an equivalent circular structure,as derived as in Ref.8,the resonance frequency we calculate is approximately 15% larger than the simulation result of 188 THz. For agreement, we increase the capacitance and attribute this discrepancy to the presence of additional parasitic capacitance in the square geometry.

Given that the assumption of uniform fields which leads to Eq. (2) is not valid near resonance, this simplified approach can only approximately describe the nonlinear properties of the composite. As the inset of Fig. 4 demonstrates, we can, however, obtain qualitative agreement with the simulation result for the reflectance and transmittance of an equivalent thickness of homogeneous material. Here we used the Maxwell-Garnett result for the effective permittivity of the composite. Taking $|\epsilon(\omega)|\gg 1$ and neglecting the dielectric layer enclosing the resonator, we have $\epsilon_{\text{eff}}^{\text{MG}}\simeq(1+f)/(1-f)=2.12$ which is in good agreement with the simulation result at frequencies below the resonance region.

We now define the dimensionless frequencies $X=\omega_{0\text{NL}}/\omega_0, \Omega=\omega/\omega_0$, and $\Gamma'=\Gamma/\omega_0$. Here $\omega_{0\text{NL}}$ is the intensity dependent resonance frequency of the SRR, ω_0 is the resonance frequency of the SRR in the linear limit and ω is the field frequency. Then the relation between the incident magnetic field strength and the nonlinear resonance frequency of the structure is given by

$$|H_{\text{ext}}|^2=\frac{Z_s n_c^2 d_c^2}{4n_2L_g^2\omega_0^2}\frac{(1-X^2)}{\Omega^2X^6}\{[X^2-\Omega^2]^2+\Omega^2\Gamma'^2\},$$

where Z_s is the impedance of the dielectric matrix.

In Fig. 4 we have plotted the dimensionless nonlinear resonance frequency of the SRR against the incident magnetic field strength. Here, $\omega=200$ THz, and the value of n_2 is

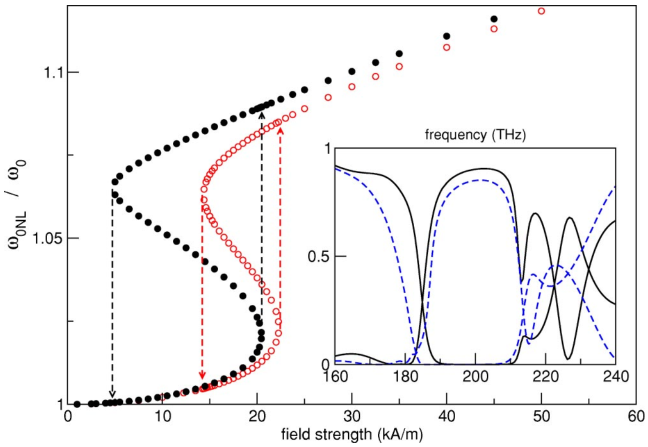

FIG. 4. (Color online) Dimensionless nonlinear resonance frequency vs field strength for two values of the resonance quality factor. Curves obtained using dissipation rates (γ) (filled circles) and (3γ) (open circles). Inset: Reflectance and transmittance of a slab of four layers of the composite calculated using the transfer matrix (solid lines, cf. Fig. 2) and of a homogeneous slab of equivalent thickness and with effective response functions determined by Eq. (2) and the Maxwell-Garnett effective permittivity (dashed lines).

-2.5×10^{-12} cm^2/W. Ultrafast nonresonant Kerr nonlinearities of this magnitude have recently been observed in the near-infrared spectral region.[19] Curves plotted in Fig. 4 are for two values of the resonance quality factor obtained using dissipation rates γ (filled circles) and 3γ (open circles). Dashed vertical arrows indicate the size of the shift in the nonlinear resonant frequency that we can expect at the onset of bistability in each case. The increased dissipation rate results in better agreement between the numerical and quasistatic results for the effective permeability. In the idealized picture, the corresponding data therefore determine a lower bound for the switching amplitude and indicates that the desired switching behavior with a change in sign of the effective permeability can be observed. The switching incident field strength of $\sim$20 kA/m corresponds to a minimum peak power requirement of $\sim$100 mW in free space. We conclude that the manufacture of compact SRR based optical switches using current materials and fabrication techniques is a possibility.

In conclusion, numerical calculations of the effective permittivity and permeability have been presented for a photonic crystal metamaterial composed of split-ring resonators structured on a nanometric scale. We have shown that metallic resonators can provide a means to realize a negative effective permeability up to telecommunications wavelengths. As passive photonic band gap media these structures are of interest as compact mirrors and in subwavelength optical waveguiding. In addition, they could offer enhanced nonlinear effects and bistable switching through the intensity dependence of the resonance frequency. These structures therefore have much potential for creating functional all optical elements.

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