Nonlinear Properties of Left-Handed Metamaterials

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We analyze the properties of microstructured materials with negative refraction, the so-called *left-handed metamaterials*. We consider a two-dimensional periodic structure created by arrays of wires and split-ring resonators embedded into a nonlinear dielectric, and calculate the effective nonlinear electric permittivity and magnetic permeability. We demonstrate that the hysteresis-type dependence of the magnetic permeability on the field intensity allows changing the material properties from left- to right-handed and back. These effects can be treated as *the second-order phase transitions* in the transmission properties induced by the variation of an external field.

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Recent theoretical studies [1–3] and experimental results [4–6] have shown the possibility of creating novel types of microstructured materials that demonstrate the property of negative refraction. In particular, the composite materials created by arrays of wires and split-ring resonators (SRR) were shown to possess a negative real part of the magnetic permeability and dielectric permittivity for microwaves. These materials are often referred to as *left-handed (LH) materials* or *materials with negative refraction*. The properties of such materials were analyzed theoretically by Veselago many years ago [7], but they were demonstrated experimentally only recently. As was shown by Veselago [7], left-handed materials possess a number of peculiar properties, including negative refraction for interface scattering, inverse light pressure, and reverse Doppler and Vavilov-Cherenkov effects.

Thus far, all properties of left-handed materials were studied in the linear regime of wave propagation when both the magnetic permeability and the dielectric permittivity of the material were assumed to be independent of the intensity of the electromagnetic field. However, any future effort in creating *tunable structures* where the field intensity changes the transmission properties of the composite structure would require the knowledge of nonlinear properties of such metamaterials, which may be quite unusual. In this Letter we analyze, for the first time to our knowledge, *nonlinear properties of left-handed metamaterials* for the example of a lattice of the split-ring resonators and wires with a nonlinear dielectric. We show that the effective magnetic permeability depends on the intensity of the macroscopic magnetic field in a nontrivial way, allowing *switching between the left- and right-handed (RH) materials by varying the field intensity.* We believe that our findings may stimulate future experiments in this field, as well as studies of nonlinear effects in photonic crystals, where the phenomenon of negative refraction is analyzed now very intensely [8,9].

We consider a two-dimensional composite structure consisting of a square lattice of the periodic arrays of conducting wires and split-ring resonators shown schematically in Fig. 1. We assume that the unit cell size d of the structure is much smaller then the wavelength of the propagating electromagnetic field and, for simplicity, we choose the single-ring geometry of a lattice of cylindrical SRRs. The results obtained for this case are qualitatively similar to those obtained in the more involved cases of double SRRs. This type of microstructured material has recently been suggested and built in order to create left-handed metamaterials with negative refraction in the microwave region [4].

The negative real part of the effective dielectric permittivity of such a composite structure appears due to the metallic wires, whereas a negative sign of the magnetic permeability becomes possible due to the SRR lattice. As a result, these materials demonstrate the properties of negative refraction in the finite frequency band, $\omega_0 < \omega < \min(\omega_p, \omega_{\|m})$, where ω_0 is the eigenfrequency of the SRRs, $\omega_{\|m}$ is the frequency of the longitudinal magnetic plasmon, ω_p is an effective plasma frequency,

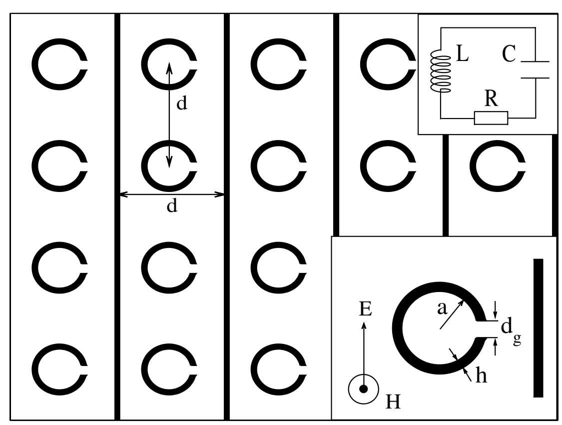

FIG. 1. Schematic of the composite metamaterial structure. The lower inset shows a unit cell of the periodic structure while the upper inset shows the SRR equivalent oscillator with the parameters used in the derivation.

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and ω is the angular frequency of the propagating electromagnetic wave, $(\mathcal{E}, \mathcal{H}) \sim (\mathbf{E}, \mathbf{H}) \exp(i\omega t)$. The split-ring resonator can be described as an effective LC oscillator (see Ref. [10]) with the capacitance of the SRR gap, as well as an effective inductance and resistance (see the upper inset in Fig. 1).

The *nonlinear response* of such a composite structure can be characterized by two different contributions. The first one is an intensity-dependent part of the effective dielectric permittivity of the infilling dielectric. For simplicity, we assume that the metallic structure is embedded into a nonlinear dielectric with a permittivity that depends on the intensity of the electric field in a general form, $\epsilon_D = \epsilon_D(|\mathbf{E}|^2)$. For detailed calculations presented below, we take the linear dependence that corresponds to the Kerr nonlinearity.

The second contribution into the nonlinear properties of the composite material comes from the lattice of resonators, since the SRR capacitance (and, therefore, the SRR eigenfrequency) depends on the strength of the local electric field in a narrow slot. Additionally, we can expect a nonlinear eigenfrequency detuning due to a resonant growth of the charge density at the edges of the SRR gap. The intensity of the local electric field in the SRR gap depends on the electromotive force in the resonator loop, which is induced by the magnetic field. Therefore, the effective magnetic permeability $\mu_{\rm eff}$ should depend on the macroscopic (average) magnetic field $\mathbf{H}$.

For the polarization shown in Fig. 1 (lower inset), the main contribution to the dielectric function is given by the array of wires. When the wire length is large enough, so that the frequency of the fundamental (dipole) mode of an individual wire becomes much smaller than ω, only the resistance and inductance of the wires give a contribution to the lattice impedance. Hence, the Ohm law for the current can be written in the form

$$\mathbf{j}_\omega \approx \frac{\sigma}{1 + i\omega\sigma S L_w}\mathbf{E}', \qquad (1)$$

where $\mathbf{j}_\omega$ is the electric current density in the wire, $\mathbf{E}'$ is the local electric field, σ is the conductivity of the wire metal, $L_w \approx 2c^{-2}\ln(d/r)$ $(d \gg r)$ is the inductance of the wire per unit length, c is the speed of light, r is the wire radius, S is the effective area of the wire cross section, $S \approx \pi r^2$, for $\delta > r$, and $S \approx \pi\delta(2r - \delta)$, for $\delta < r$, where $\delta = c/\sqrt{2\pi\sigma\omega}$ is the thickness of the skin layer. The average current density in the unit cell can be written in the form

$$\langle \mathbf{j}_\omega \rangle = \frac{S}{d^2}\mathbf{j}_\omega. \qquad (2)$$

For waves polarized along the wires the average macroscopic electric field $\mathbf{E}$ is approximately equal to the local field $\mathbf{E}'$. Taking into account the general relation between the electric field $\mathbf{E}$ and the electric induction $\mathbf{D}$,

$$\mathbf{D} = \epsilon_D(|\mathbf{E}|^2)\mathbf{E} + \frac{4\pi}{i\omega}\langle \mathbf{j}_\omega \rangle, \qquad (3)$$

we can obtain the expression for the effective nonlinear dielectric permittivity,

$$\epsilon_{\rm eff}(|E|^2) = \epsilon_D(|E|^2) - \frac{\omega_p^2}{\omega(\omega - i\gamma_\epsilon)}, \qquad (4)$$

where $\omega_p \approx (c/d)[2\pi/\ln(d/r)]^{1/2}$ is the effective plasma frequency and $\gamma_\epsilon = c^2/2\sigma S \ln(d/r)$. The second term on the right-hand side of Eq. (4) is in complete agreement with the earlier result obtained by Pendry and coauthors [1]. One should note that the low losses case, i.e., $\gamma_\epsilon \ll \omega$, corresponds to the condition $\delta \ll r$.

The analysis becomes more involved for calculating *the nonlinear magnetic response* of the composite structure, which is determined by the intrinsic properties of the interacting nonlinear oscillators in the presence of an external periodic force. For the structure under consideration, the current induced in each resonator can be found as

$$I = -i\pi a^2\left(\frac{\omega}{c}\right)|\mathbf{H}_0|Z^{-1}, \qquad (5)$$

where $\mathbf{H}_0$ is the amplitude of the external field applied to a SRR, $Z = i\omega L + R + (i\omega C)^{-1}$ is the SRR impedance, and the other parameters are marked in Fig. 1: a is the radius, L, R, and C are the inductance, resistance, and capacitance, respectively. The amplitude of the electric field in the gap of a SRR can be found as follows:

$$|\mathbf{E}_g| \approx \frac{I}{i\omega C d_g}, \qquad (6)$$

where d_g is the size of the SRR gap. Nonlinear effects in Eqs. (5) and (6), appear due to the capacitance C which is proportional to $\epsilon_D(|E_g|^2)$. Therefore, Eq. (6) gives an implicit relation between the amplitude of the local electric field in the gap and the amplitude of the (external to the SRR) magnetic field.

The relation between magnetic inductance $\mathbf{B}$ and macroscopic magnetic field $\mathbf{H_0}$ is given by the formula

$$\mathbf{B} = \mathbf{H_0} + F\mathbf{H}', \qquad (7)$$

where $\mathbf{H}'$ is an additional magnetic field induced by the alternating external magnetic field $\mathbf{H}_0$ in a cylindrical SRR, which determines the magnetization of the composite, and $F = \pi a^2/d^2$.

Taking into account that $\mathbf{H}' = 0$ outside the SRR, from the boundary conditions we obtain the relation

$$|\mathbf{H}'| = \frac{4\pi}{c}|\mathbf{j}_s|, \qquad (8)$$

where $\mathbf{j}_s$ is an equivalent surface current in the SRR, which is equal to the current per unit length. From Eqs. (5), (7), and (8) we obtain an explicit expression for the effective magnetic permeability of the composite structure (for $F \ll 1$):

$$\mu_{\rm eff}(\mathbf{H}) = 1 + \frac{F\,\omega^2}{\omega_{0\rm NL}^2(\mathbf{H}) - \omega^2 + i\Gamma\omega}, \qquad (9)$$

where

$$\omega_{0\mathrm{NL}}^2(\mathbf{H}) = \left(\frac{c}{a}\right)^2 \frac{d_g}{\pi h \epsilon_D(|\mathbf{E}_g(\mathbf{H})|^2)}$$

is the eigenfrequency of oscillations in the presence of the external field of a finite amplitude, h is the width of the ring, $\Gamma = c^2/2\pi\sigma a h$, for $h < \delta$, and $\Gamma = c^2/2\pi\sigma a \delta$, for $h > \delta$. It is important to note that Eq. (9) has a simple physical interpretation: The resonant frequency of the artificial magnetic structure depends on the amplitude of the external magnetic field and, in turn, this leads to the intensity-dependent function μ_{eff}.

To be more specific, we consider the Kerr nonlinearity of the dielectric in the composite material, i.e.,

$$\epsilon_D(|E|^2) = \epsilon_{D0} + \alpha |E|^2/E_c^2, \tag{10}$$

where E_c is a characteristic electric field, and $\alpha = \pm 1$ stands for a focusing or defocusing nonlinearity, respectively. Then, the relation between the macroscopic magnetic field and the dimensionless nonlinear resonant frequency can be obtained from Eqs. (5)–(8) and (10) as

$$|\mathbf{H}|^2 = \alpha A^2 E_c^2 \frac{(1 - X^2)[(X^2 - \Omega^2)^2 + \Omega^2 \gamma^2]}{X^6}, \tag{11}$$

where $A^2 = 16\epsilon_{D0}^3 \omega_0^2 h^2/c^2$, $\Omega = \omega/\omega_0$, $\omega_0 = (c/a) \times [d_g/\pi h \epsilon_{D0}]^{1/2}$ is the eigenfrequency of the system of SRRs in the linear limit, $X = \omega_{0\mathrm{NL}}/\omega_0$, and $\gamma = \Gamma/\omega_0$. Therefore, we find that the dimensionless eigenfrequency of the SRR lattice $X(|H|^2)$ is a multivalued function of the magnetic field. This result reveals a general property of nonlinear oscillators with a high quality factor [11].

The parametric dependence of the effective magnetic permeability on the magnetic field is determined completely by Eqs. (9) and (11). Figures 2 and 3 summarize different types of nonlinear magnetic properties of the composite, which are defined by the dimensionless frequency of the external field Ω, for both a *focusing* [Figs. 2(a) and 2(b) and 3(a) and 3(b)] and a *defocusing* [Fig. 2(c) and 2(d) and 3(c) and 3(d)] nonlinearity of the dielectric.

In the case of a *focusing nonlinearity* (i.e., when $\alpha = 1$), the SRR eigenfrequency decreases with increasing intensity of the electromagnetic field because of the growth of the SRR capacitance. Then, for $\Omega > 1$, the effective magnetic permeability of the composite structure grows with the field intensity, as shown in Fig. 2(a). If in the linear limit the composite material is left-handed, i.e., $\mathrm{Re}(\mu_{\mathrm{eff}}) < 0$, it will become right-handed for higher intensities of the magnetic field [in this reasoning we assume $\mathrm{Re}(\epsilon_{\mathrm{eff}}) < 0$].

A more complicated behavior of the magnetic permeability is observed for $\Omega < 1$; this is shown in Fig. 2(b). Here, in the linear limit the real part of the magnetic permeability is always positive, but the eigenfrequency of the SRR decreases with the growth of the magnetic field, thus driving the system into resonance. Since a nonlinear oscillator has a hysteresis structure of its response with a change of an external force (see, e.g., Ref. [11]), this leads to multivalued dependencies. In our problem, the nonlinear eigenfrequency is a three-valued function of the external magnetic field, and this results in jumps of the magnetic permeability with the growth of the magnetic field. As follows from Fig. 2(b), the magnetic field intensity displays a jump of the magnetic permeability from positive to negative values at some H_{c1}. Thus, the initially opaque medium with positive refraction becomes a negative refraction transparent medium with the growth of the field intensity. This effect can be treated as a second-order phase transition induced by the external electromagnetic field. The reverse transition takes place when the magnetic field intensity decreases to the value $H_{c2} < H_{c1}$.

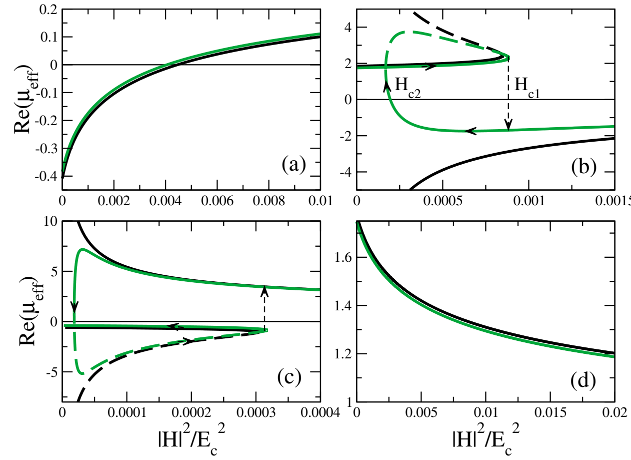

FIG. 2 (color online). The real part of the effective magnetic permeability versus intensity of the magnetic field: (a) $\Omega > 1$, $\alpha = 1$; (b) $\Omega < 1$, $\alpha = 1$; (c) $\Omega > 1$, $\alpha = -1$; (d) $\Omega < 1$, $\alpha = -1$. Black: the lossless case ($\gamma = 0$); grey: the lossy case ($\gamma = 0.05$). The dashed curves show unstable branches.

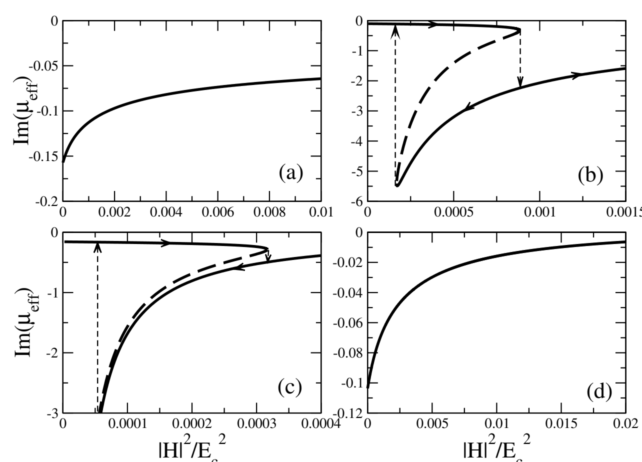

FIG. 3. The imaginary part of the effective magnetic permeability versus intensity of the magnetic field for $\gamma = 0.05$: (a) $\Omega > 1$, $\alpha = 1$; (b) $\Omega < 1$, $\alpha = 1$; (c) $\Omega > 1$, $\alpha = -1$; (d) $\Omega < 1$, $\alpha = -1$. The dashed curves show unstable branches.

In the case of a *defocusing nonlinearity* (i.e., when $\alpha = -1$), the SRR eigenfrequency increases with the amplitude of the external field. That is why the resonance effects take place for $\Omega > 1$, as shown in Figs. 2(c) and 2(d) and 3(c) and 3(d). Here, we observe the opposite behavior when the transition from the case $\mathrm{Re}(\mu_{\mathrm{eff}}) < 0$ to the case $\mathrm{Re}(\mu_{\mathrm{eff}}) > 0$ takes place at high values of the external field, and the reverse transition occurs at lower field intensities. In the latter case, $\mathrm{Re}(\mu_{\mathrm{eff}})$ is always positive for $\Omega < 1$; see Fig. 2(d).

Our results show that the imaginary part of the effective magnetic permeability, which determines the structure losses, can be controlled rather effectively by a proper choice of the intensity of the external high-frequency magnetic field; see Fig. 3. We believe that this feature may be important for the future applications of left-handed materials.

Because of the high values of the electric field in the slot of SRR as well as resonant interaction of the electromagnetic field with the SRR lattice, the characteristic magnetic nonlinearity in such structures is much stronger then the corresponding electric nonlinearity. Therefore, *magnetic nonlinearity should dominate* in the composite materials that display the phenomenon of negative refraction. More importantly, the nonlinear medium can be created by inserting nonlinear elements into the slots of SRRs, allowing an easy tuning by an external field.

The critical fields for switching between LH and RH states, shown in Figs. 2 and 3 can be reduced to a desirable value by choosing the frequency close to the resonant frequency of SRRs. Even for a relatively large difference between the SRR eigenfrequency and the external frequency, as we have in Fig. 2(b) where $\Omega = 0.8$ (i.e., $\omega = 0.8\omega_0$), the switching amplitude of the magnetic field is $\sim 0.03E_c$. The characteristic values of the focusing nonlinearity can be estimated for some materials such as n-InSb for which $E_c = 200$ V/cm [12]. As a result, the strength of the critical magnetic field is found as $H_{c1} \approx 1.6$ A/m. Strong defocusing properties for microwave frequencies are found in $\mathrm{Ba}_x\mathrm{Sr}_{1-x}\mathrm{TiO}_3$ (see Ref. [13] and references therein). The critical nonlinear field of a thin film of this material is $E_c = 4 \times 10^4$ V/cm, and the corresponding field of the transition from the LH to RH state [see Fig. 2(c)] can be found as $H_c \approx 55.4$ A/m.

The possibility of strongly enhanced nonlinearities in left-handed metamaterials revealed here may lead to an essential revision of the concepts based on the linear theory, since the electromagnetic waves propagating in such materials always have a finite amplitude. At the same time, the engineering of nonlinear composite materials will open a number of their novel applications such as frequency multipliers, beam spatial spectrum transformers, switchers, limiters, etc.

We notice that the hysteresis behavior with jumps in the dependencies of the effective material parameters has been described above for stationary processes only. Such transitions will display a characteristic scale in time or space for initial or boundary problems, respectively. Such spatial or temporal scales are determined by the relaxation microprocesses in the SRR lattice.

In conclusion, we have presented, for the first time to our knowledge, a systematic analysis of nonlinear properties of microstructured materials which display negative refraction, the left-handed metamaterials. We have shown that the composite metamaterials composed of a lattice of wires and split-ring resonators possess an effective magnetic permeability that depends on the intensity of the macroscopic magnetic field in a nontrivial way. The magnetic nonlinearity is found to be much stronger than the nonlinearity in the dielectric properties due to the field enhancement in the split-ring resonators. The dependence of the effective magnetic permeability on the field intensity allows switching between its positive and negative values, i.e., a change of the material properties from left- to right-handed and back. Such processes can be treated as the second-order phase transitions induced by varying the external electromagnetic field.

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