

## Double-nonlinear metamaterials

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We study a double-nonlinear metamaterial composed of a mixture of both nonlinear electric and nonlinear magnetic resonators. We predict multistable behavior in such metamaterial with the possibility to control the effective index of refraction by the electromagnetic field intensity. In contrast to the structure with just one type of nonlinear inclusions, our composite material may switch properties between the transparent negative index and the transparent positive index regimes. © 2010 American Institute of Physics. [doi:10.1063/1.3525172]

Left-handed materials were first theoretically studied by Pafomov<sup>1</sup> and Veselago,<sup>2</sup> and later they were experimentally demonstrated in the form of composite metamaterials by Smith *et al.*<sup>3</sup> First, metamaterials with negative refraction were composed of periodic arrays of split-ring resonators (SRRs) and thin conducting wires.<sup>4</sup> By now, a number of various designs of metamaterials have been suggested for realizing the negative refractive index.<sup>5–9</sup>

Recent studies have shown that *nonlinear metamaterials* exhibit a range of intensity-dependent phenomena, such as bistability,<sup>10,11</sup> harmonic generation,<sup>12</sup> and parametric amplification.<sup>13,14</sup> The nonlinear metamaterials can be made by embedding the SRRs in the nonlinear dielectric host medium or by inserting a varactor diode within the gap of SRRs.<sup>10,15</sup> It was shown that the magnetic or electric resonances in nonlinear magnetic or nonlinear electric metamaterials can be dynamically tuned by varying external fields.<sup>16–18</sup> Moreover, it was recently demonstrated that the same physical mechanism leads to the enhancement of nonlinearity in optical metamaterials.<sup>19</sup>

All nonlinear metamaterials studied so far are composed of either nonlinear electric or nonlinear magnetic resonators. In this letter, we consider a three-dimensional composite structure with both nonlinear electric and nonlinear magnetic inclusions, *double-nonlinear metamaterial*. For the magnetic components, we choose the nonlinear SRRs studied earlier,<sup>20</sup> while for electric inclusions, we develop a model based on nonlinear electric resonators (ERs), which were recently studied experimentally.<sup>18</sup> We predict that such nonlinear metamaterial can be engineered to exhibit tunable nonlinear response with a control over both electric and magnetic nonlinearities. Our results suggest that it is possible to realize metamaterial with the index of refraction changing from positive to negative values, while metamaterial remains mostly transparent.

The schematic of the unit cell of nonlinear ERs is shown in Fig. 1(a), where the nonlinear response is generated by the nonlinear element inserted into the central slit of the ER. We assume that the unit cell size  $d_e$  of the structure is much smaller than the wavelength of the propagating electromagnetic wave, and we use an equivalent resistance-inductance-

capacitance model for the ER, see Fig. 1(b). We assume that the nonlinear capacitance  $C_n$  has the form<sup>20</sup>

$$C_n(U_n) = \frac{r_e^2 \epsilon_{De}}{4d'_{ge}} \left( 1 + \alpha_e \frac{|U_n|^2}{U_c^2} \right), \quad (1)$$

where  $U_n$  and  $U_c$  are, respectively, voltage across the capacitor, and characteristic voltage of the nonlinear capacitor,  $d'_{ge}$  is the central slit size,  $\alpha_e = \pm 1$  describes focusing or defocusing types of nonlinear response,  $\epsilon_{De}$  is the linear dielectric permittivity of the dielectric infilling the capacitor,  $r_e$  is the radius of the capacitor plates. The current  $I$  in the effective circuit is governed by the equation

$$L \frac{dI}{dt} + U_n + U + RI = -E' d_{ge}, \quad (2)$$

where  $U$  is the voltage across the linear capacitor and  $d_{ge}$  is the size of the side slits of the ER.  $E'$  is the local microscopic electric field creating the electromotive force in the resonator, which is related to the external macroscopic electric field  $E$  in cubic lattice through the Lorenz-Lorentz relation  $E' = E + 4\pi P/3$ , where  $P = 2n_e d_{ge}^2 E' / i\omega Z$  is the electric polarization of the unit volume,  $Z = i\omega L + (i\omega C)^{-1} + (i\omega C_n)^{-1} + R$ ,  $n_e$  is the density of the ERs. Then, the expression for the effective electric permittivity of the composite can be obtained in the form

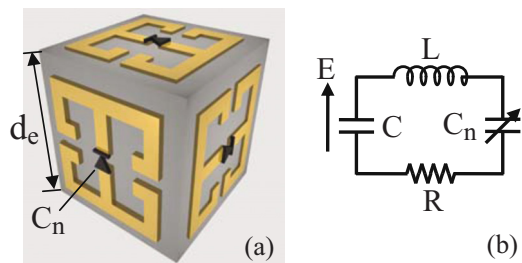


FIG. 1. (Color online) (a) Schematic of a unit cell of the nonlinear electric metamaterial and (b) the equivalent circuit with the parameters used in the derivations.

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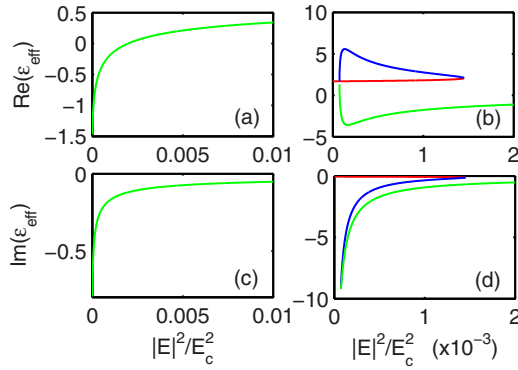


FIG. 2. (Color online)  $\text{Re}(\varepsilon_{\text{eff}})$  [(a) and (b)] and  $\text{Im}(\varepsilon_{\text{eff}})$  [(c) and (d)] of the electric metamaterial for a focusing nonlinearity  $\alpha_e=1$ . [(a) and (c)]  $\Omega_e=1.03$ ,  $\gamma_e=0.01$ ; [(b) and (d)]  $\Omega_e=0.95$ ,  $\gamma_e=0.01$ .

$$\varepsilon_{\text{eff}}(|E|^2) = 1 + \frac{F_e}{\omega_{0NL}^2(|E|^2) - \omega^2 + i\omega\Gamma_e + F_e/3}, \quad (3)$$

where  $\omega_{0NL}^2(|E|^2) = L^{-1}[1/C + 1/C_n(|E|^2)]$  is the eigenfrequency of nonlinear oscillations,  $\Gamma_e = (2a_e + 3h)/2L\sigma S_r$  is the damping coefficient,  $a_e$  is the length of the side slits of the resonator,  $\sigma$  is the conductivity of the metal wires,  $S_r$  is the effective area of the wire cross-section; and assuming circular cross-section,  $S_r \approx \pi r_e^2$ , for  $\delta_e > r_e$ , and  $S_r \approx \pi \delta_e(2r_e - \delta_e)$ , for  $\delta_e < r_e$ , where  $\delta_e = c/\sqrt{2\pi\sigma\omega}$  is the skin-layer thickness. Here  $F_e = 8\pi d_{ge}^2/Lh^3$ ,  $C = \varepsilon_{De}a_e\delta_e/2\pi d_{ge}$  and  $L = [\ln(h/r_e) + 2\ln(2h/r_e) + Y]h/c^2$  with  $Y = -\ln(1 + \sqrt{5})/2 - 2\ln(2 + \sqrt{5}) + 2\sqrt{5} - 21/4$ , where  $h$  is the height of the ER. Equation (3) shows that the effective electric permittivity depends on the intensity of the external electric field due to the intensity-dependent resonant frequency. The physical origin of the nonlinear behavior of the dielectric permittivity is identical to that of magnetic permeability.<sup>10,20</sup> The relation between the resonant frequency and the electric field can be implicitly obtained from Eqs. (1)–(3) as follows:

$$|E|^2 = \alpha_e |E_c|^2 \frac{1}{(1 - C_r)} \frac{(1 - X_e^2)[(X_e^2 - \Omega_e^2 + M)^2 + \Omega_e^2 \gamma_e^2]}{(X_e^2 - C_r)^2}, \quad (4)$$

where  $E_c = U_c/d_{ge}$  is the characteristic electric field of nonlinear dielectric,  $C_r = C_0/(C_0 + C)$  and  $M = 8\pi d_{ge}^2/3h^3 L \omega_{0e}^2$ , where  $C_0 = r_e^2 \varepsilon_{De}/4d_{ge}'$  is the linear part of  $C_n$ , and  $\omega_{0e}^2(|E|^2) = L^{-1}[1/C + 1/C_0]$  is the linear eigenfrequency of the resonators,  $\Omega_e = \omega/\omega_{0e}$ ,  $X_e = \omega_{0NL}/\omega_{0e}$  and  $\gamma_e = \Gamma_e/\omega_{0e}$  are, respectively, normalized angular frequency, normalized nonlinear frequency and normalized damping coefficient. Equation (3) implies that the nonlinear electric resonant frequency  $X_e$  can be a multivalued function of  $|E|^2$ . This, in turn, leads to the multivalued structure of dielectric permittivity as a function of  $|E|^2$ . Figures 2 and 3 demonstrate different types of nonlinear electric response of the electric microstructure, for both a *focusing* and a *defocusing* nonlinearity of the dielectric, respectively. Here, we assume the following parameters of the composite structure:  $d_e = 1.1$  cm,  $h = 0.9$  cm,  $a_e = 0.25$ ,  $\delta_e = 0.25$  cm,  $d_{ne}' = 0.04$  cm,  $d_{ge} = 0.15$  cm,  $r_e = 0.08$  cm,  $\varepsilon_{De} = 8.2$ , and  $\omega_{0e} = 32.18 \times 10^9$  rad/s.

For a *focusing nonlinearity* (i.e.,  $\alpha_e = 1$ ), the nonlinear eigenfrequency  $X_e$  decreases with the increasing intensity of the electric field due to the increase of the nonlinear capaci-

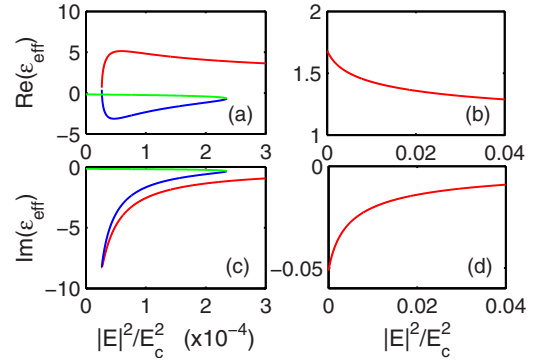


FIG. 3. (Color online) Same as in Fig. 2, but for a defocusing nonlinearity  $\alpha_e=-1$ . [(a) and (c)]  $\Omega_e=1.05$ ,  $\gamma_e=0.01$ ; [(b) and (d)]  $\Omega_e=0.95$ ,  $\gamma_e=0.01$ .

tance. Then, for  $\Omega_e > 1$ , the real and imaginary parts of effective electric permittivity of the composite structure increase with  $|E|^2$ , as shown in Figs. 2(a) and 2(c). From Fig. 2(a), we see that  $\text{Re}(\varepsilon_{\text{eff}})$  may be positive or negative depending on the intensity of electric field. For  $\Omega_e < 1$ ,  $X_e$  is a three-valued function of  $|E|^2$  in Eq. (4), leading to a three-valued dependencies of effective electric permittivity on  $|E|^2$  [see Figs. 2(b) and 2(d)]. This nonlinear behavior is generic to many nonlinear resonant systems, including the split-ring resonators for nonlinear magnetic metamaterials.<sup>10</sup>

For a *defocusing nonlinearity* (i.e.,  $\alpha_e = -1$ ), the nonlinear electric eigenfrequency  $X_e$  increases with the increasing intensity of the electric field, leading to the nonlinear permittivity behavior shown in Fig. 3. For  $\Omega_e > 1$ ,  $\text{Re}(\varepsilon_{\text{eff}})$  can switch from negative to positive values with the change of  $|E|^2$  [see Fig. 3(a)]. However, for  $\Omega_e < 1$ ,  $\text{Re}(\varepsilon_{\text{eff}})$  is always positive [see Fig. 3(b)], which is easy to understand from the moving resonance picture: the frequency of the wave is to the left of the resonance, and the resonance keeps moving to the right with increase of intensity, ensuring a positive sign of the dielectric permittivity.

It is natural to extend the concept to a metamaterial with simultaneously nonlinear electric and nonlinear magnetic responses, which are expected to lead to nontrivial nonlinear phenomena and tunability of metamaterials. Here, we combine the metamaterials of nonlinear electric structure studied above and nonlinear magnetic structure considered in the Ref. 20. The actual realization of such metamaterial may include the magnetic SRRs on the opposite sides of the boards that already contain electric resonators. In this work, we neglect the interaction between electric and magnetic resonators, which may lead to the nonlinear bianisotropy; this is a subject of future studies. As a result, our nonlinear magnetic substructure has an effective magnetic permeability depending on magnetic field,<sup>20</sup>  $\mu_{\text{eff}}(|H|^2)$ . We choose the parameters of the SRRs so that the magnetic linear eigenfrequency  $\omega_{0m}$  is close to the linear eigenfrequency  $\omega_{0e}$  of ERs, so that the regions of simultaneously negative epsilon and mu can overlap.

We assume that the behavior of the double-nonlinear metamaterial is described by the effective dielectric permittivity (3) and the effective magnetic permeability.<sup>20</sup> Here, we focus on two cases that lead to negative refractive index for a range of wave intensities. Figure 4 presents the  $\varepsilon_{\text{eff}}$ ,  $\mu_{\text{eff}}$ , and refractive index  $n_{\text{eff}}$  of the composite structure for the combination of the ERs with defocusing nonlinearity  $\alpha_e = -1$ , and SRRs with focusing nonlinearity  $\alpha_m = 1$  for

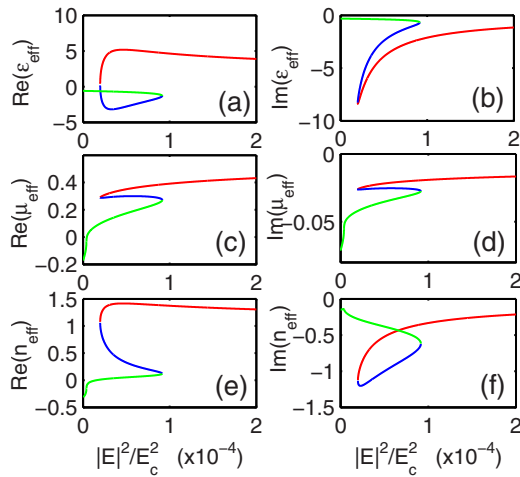


FIG. 4. (Color online)  $\epsilon_{\text{eff}}$ ,  $\mu_{\text{eff}}$ , and  $n_{\text{eff}}$  as functions of the intensity of the external field for  $\alpha_e = -1$ ,  $\alpha_m = 1$ ,  $\Omega_e = 1.04$ ,  $\Omega_m = 1.05$ , and  $\gamma_e = \gamma_m = 0.01$ .

$\Omega_e > 1$  and  $\Omega_m > 1$ . In this case, the ERs resonant frequency  $X_e$  increases with the increase of  $|E|^2$  and then  $\epsilon_{\text{eff}}$  is a three-valued function of  $|E|^2$ , as shown in Figs. 4(a) and 4(b). In a double-nonlinear metamaterial, the electric and magnetic fields of the *nonlinear plane wave* are related through the impedance,

$$Z = \frac{E}{H} = \sqrt{\frac{\mu_{\text{eff}}(|H|^2)}{\epsilon_{\text{eff}}(|E|^2)}}. \quad (5)$$

We note that for other types of nonlinear waves, such as solitons, the relation between electric and magnetic fields can be different. From Eq. (5), for any given electric field intensity  $E$ , and known functions  $\epsilon_{\text{eff}}(|E|^2)$  and  $\mu_{\text{eff}}(|H|^2)$ , we can numerically find the magnetic field  $H$ . Then we can express the effective magnetic permeability as a function of the electric field intensity, see Figs. 4(c) and 4(d). As we see from the Figs. 4(a) and 4(c), the real parts of permittivity and permeability are simultaneously negative for low intensity of electric field, indicating the negative refraction regime. The calculated refractive index of the composite is shown in Figs. 4(e) and 4(f). We see that low external field intensity can switch the effective index from negative values to positive, while keeping its imaginary part relatively small. Thus, in this case, the initially transparent negative refracting medium can become transparent positive refracting medium with the growth of the field intensity. This switching regime is dramatically different from what can be achieved in nonlinear metamaterials with just one of the parameters being nonlinear, where the material properties typically switch between transparent to opaque states.<sup>20</sup>

The switching between positive and negative-index states can also sharply occur, when the metamaterial contains electric resonators with  $\alpha_e = 1$  and magnetic resonators with  $\alpha_m = -1$ , and  $\Omega_e > 1$ ,  $\Omega_m > 1$ . We also studied other possible combinations of the nonlinear inclusions in resonators and observed that the nonlinear responses of such composite medium can be very diverse.

In conclusion, we have studied a double-negative metamaterial made of a mixture of nonlinear ERs and nonlinear SRRs and investigated its properties for different combinations of focusing and defocusing nonlinearities of dielectric inclusions. We have shown that in some regimes such nonlinear metamaterial can switch its refractive index between negative and positive values with the change of external field intensity. This switching regime is dramatically different from what can be achieved in nonlinear metamaterials with just one type of nonlinear inclusions. The diversity of possible nonlinear responses in the double-nonlinear composites suggests that they can be invaluable for engineering future nonlinear materials.

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