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# Four-wave mixing, quantum control, and compensating losses in doped negative-index photonic metamaterials

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The possibility of compensating absorption in negative-index metamaterials (NIMs) doped by resonant nonlinear-optical centers is shown. The role of quantum interference and the extraordinary properties of four-wave parametric amplification of counterpropagating electromagnetic waves in NIMs are discussed.

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Negative refractive index metamaterials (NIMs) present a novel class of materials that promise a revolutionary breakthrough in electromagnetics (for review, see, e.g., [1]). Nonlinear optics in such materials remains so far a less-developed branch of optics. The possibility of nonlinear electromagnetic responses in such materials attributed to the asymmetry of the voltage-current characteristics of their building blocks was predicted in [2,3]. Recent experimental demonstrations of the exciting opportunities to craft nonlinear optical materials with characteristics exceeding those in natural crystals are reported in [4]. Unique nonlinear-optical (NLO) propagation effects associated with three-wave ( $\chi^{(2)}$ ) coupling in NIMs, as compared with their well-known counterparts in natural materials, were revealed in [5–8]. The striking changes in the optical bistability in a layered structure including a NIM layer were shown in [9]. A review of the corresponding theoretical approaches is given in [10]. The most detrimental obstacle toward applications of NIMs is strong absorption that is inherent to this class of materials. The possibility to overcome such obstacles based on *three-wave* optical parametric amplification (OPA) in NIMs was shown in [7,8]. A great deal of technical problems must be solved, however, to match the frequency domains of negative index (NI), strong NLO response, and phase matching to realize such feasibility. Here, we propose and explore an alternative approach associated with *resonant four-wave mixing* (FWM) nonlinearities  $\chi^{(3)}$  embedded in NIMs and *tailored through quantum control*. The possibility of compensating losses and manipulating transparency, refractive index, and nonlinear response of the NIM sample with control laser(s) is shown.

The basic idea of the proposed approach is that a slab of NIM is doped by four-level nonlinear centers [Fig. 1(a)] so that the frequency  $\omega_4$  falls in the NI do-

main, whereas all the other frequencies are in the positive index domain. Below we show the feasibility of producing the transparency and amplification for the signal wave at  $\omega_4$  controlled by two lasers at  $\omega_1$  and  $\omega_3$ . These three fields generate an idler at  $\omega_2 = \omega_3 + \omega_1 - \omega_4$ , which experiences amplification due to population-inversion or Raman gain; either provided by the driving field at  $\omega_1$  and controlled by another driving field at  $\omega_3$ . The amplified idler contributes back to  $\omega_4 = \omega_3 + \omega_1 - \omega_2$  through FWM, which leads to strongly enhanced OPA. We assume that the wave vectors of all waves,  $\mathbf{k}_j$ , are codirected, which is required for phase matching. Since only  $\omega_4$  experiences negative refraction and all other frequencies are in the positive index domain, energy flow at  $\omega_4$  is counterdirected against other waves. Hence, this signal wave must enter the slab from its opposite side at  $z=L$ , which is the exit facet for all other waves entering the slab at  $z=0$  [Fig. 1(b)]. Correspondingly, the signal exits the slab at  $z=0$ . Such *backwardness* of the signal wave leads to counterintuitive, *distributed feedbacklike* behavior of the OPA process in NIMs described in [7,8].

With the photon fluxes given by  $S_{2,4}/\hbar\omega_{2,4} = (c/8\pi)|a_{2,4}|^2$  and the amplitudes of the control

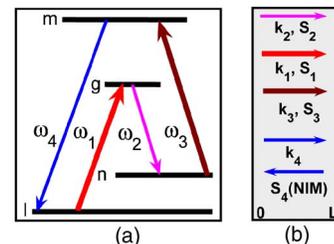


Fig. 1. (Color online) Scheme of (a) quantum-controlled FWM interaction and (b) coupling geometry.  $\omega_4$  is the signal frequency,  $\omega_2$  is the idler, and  $\omega_1$  and  $\omega_3$  are the control fields.  $n(\omega_4) < 0$ .

waves  $E_{1,2}$  assumed constant along the slab, the equations describing the coupled waves at  $\omega_4$  and  $\omega_2$  are

$$da_4/dz = -i\gamma_4^{(3)}a_2^* \exp[i\Delta kz] + (\alpha_4/2)a_4, \quad (1)$$

$$da_2/dz = i\gamma_2^{(3)}a_4^* \exp[i\Delta kz] - (\alpha_2/2)a_2. \quad (2)$$

Here  $\gamma_{2,4}^{(3)} = (\sqrt{\omega_4\omega_2}/\sqrt{\epsilon_4\epsilon_2/\mu_4\mu_2})(4\pi/c)\chi_{2,4}^{(3)}E_1E_3$  are NLO coupling coefficients;  $\epsilon_j$  and  $\mu_j$  are the dielectric permittivities and magnetic permeabilities (which are negative at  $\omega_4$ );  $\Delta k = k_1 + k_3 - k_2 - k_4$ ; and  $\alpha_j$  are the absorption or amplification coefficients. Transmittance (amplification) at  $\omega_4$  is given by the factor  $T_4 = |a_4(0)/a_4(L)|^2$ , where  $L$  is the slab thickness. Note that the signs in Eq. (1) are opposite to those in ordinary media, which is due to the backwardness of the signal wave. The solution to a similar set of equations and its analysis are given in [7,8].

Calculations of the optical constants for embedded NLO centers (driven by the control fields) with account for constructive and destructive quantum interference are performed following the density-matrix technique described in [11]. In our simulations, we used the following representative values for relaxation rates: energy level relaxation rates  $\Gamma_n=20$ ,  $\Gamma_g=\Gamma_m=120$ ; partial transition probabilities  $\gamma_{gl}=7$ ,  $\gamma_{gn}=4$ ,  $\gamma_{mn}=5$ ,  $\gamma_{ml}=10$ ; all in  $10^6 \text{ s}^{-1}$ ; homogeneous transition half-widths  $\Gamma_{lg}=1$ ,  $\Gamma_{lm}=1.9$ ,  $\Gamma_{ng}=1.5$ ,  $\Gamma_{nm}=1.8$ ,  $\Gamma_{gm}=0.05$ ; and  $\Gamma_{ln}=0.01$ , all in  $10^{12} \text{ s}^{-1}$ . We assumed that  $\lambda_2=756 \text{ nm}$ ,  $\lambda_4=480 \text{ nm}$ . The results of numerical simulations for the optical coefficients entering Eqs. (1) and (2) are shown in

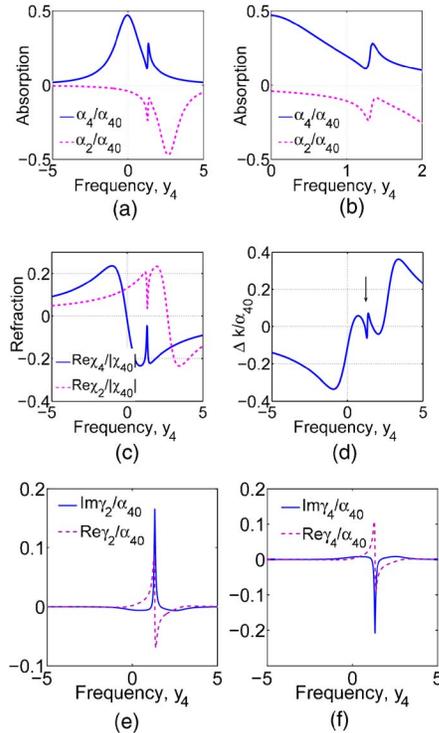


Fig. 2. (Color online) Nonlinear interference resonances induced by the control fields with  $G_1=G_3=50 \text{ GHz}$  and  $\Omega_1=\Omega_3=2.5\Gamma_{lg}$ . ( $y_4=\Omega_4/\Gamma_{mi}$ ).

Fig. 2. Here,  $\Omega_4=\omega_4-\omega_{mi}$ ; other resonance detunings  $\Omega_j$  are defined in a similar way. The coupling Rabi frequencies are introduced as  $G_1=E_1d_{lg}/2\hbar$  and  $G_3=E_3d_{nm}/2\hbar$ . The quantities  $\chi_{4,2}$  are effective linear susceptibilities, and  $\alpha_{40}$  and  $\alpha_{20}$  denote their resonant values when all the driving fields are turned off. Figures 2(a) and 2(b) depict changes in the absorption and amplification coefficients for the signal and idler, respectively, produced by the control fields. [Figure 2(b) shows the expanded interval corresponding to nonlinear interference resonances.] Figure 2(c) displays changes in refractive indices and Fig. 2(d) shows wave vector mismatch. The arrow in Fig. 2(d) points out the interval of  $y_4$ , roughly between 0.29 and 2.34, with five detunings  $y_4$  for which  $\Delta k=0$  (under the assumption that the host material does not introduce any additional phase mismatch). Figures 2(e) and 2(f) show narrow resonances in NLO coupling coefficients,  $\gamma_{4,2}^{(3)}$ , with widths on the order of other interference resonances. Figure 2 proves the feasibility of manipulating local optical parameters through nonlinear quantum interference induced by the control fields. For the used optical transition rates, the magnitude of  $G\sim 10^{12} \text{ s}^{-1}$  corresponds to control field intensities  $I$  of  $10\text{--}100 \text{ kW}/(0.1 \text{ mm})^2$ .

The optimization of the output signal at  $z=0$  is determined by the interplay between absorption, idler gain, and FWM, with the latter depending on the wave-vector mismatch and on the ratio of  $\text{Re } \gamma_{4,2}^{(3)}$  and  $\text{Im } \gamma_{4,2}^{(3)}$ . This is a multiparameter problem involving sharp resonance dependencies. Results of our numerical analysis of the steady state solutions [11] to the density matrix equations and Maxwell's equations for the slowly varying amplitudes in Eqs. (1) and (2) are shown in Fig. 3. The transmission of the host slab in the NI frequency domain has been set as 10%. Unlike conventional media, the output signal for the waves coupled in the NIM slab through OPA represents a set of *distributed feedback-type resonances* [7,8]. Such resonant behavior can be observed as a function of the intensity of the fundamental fields, the product of the slab length and the density of NLO centers, and the resonance offsets for the signal and fundamental fields. Figure 3(a) shows these narrow transmission resonances. Here, we introduce the scaled product of the slab length and the density number of embedded centers,  $L/L_{ra}$ , through the resonance absorption length,  $L_{ra}=\alpha_{40}^{-1}$ . Figure 3(b) displays the second peak in Fig. 3(a) with greater detail. It shows that the transparency window is on the scale of the narrowest (Raman, in this case) transition half-width as a function of detuning and on the scale of the resonant absorption length as a function of length. Figure 3(c) depicts the same dependence at the optimum resonance offset. Figure 3(d) shows that the intensity of the signal inside the slab may significantly exceed its output value at  $z=0$ , which depends on the ratio of the OPA and absorption rates. Here,  $\eta_2(z)=|a_2(z)/a_{4L}|^2$ . Remarkably, the transparency and amplification occur in the frequency range where magnitudes of  $|\gamma_{4,2}^{(3)}|$  are substantially less than their resonant values, which stems from the interplay of

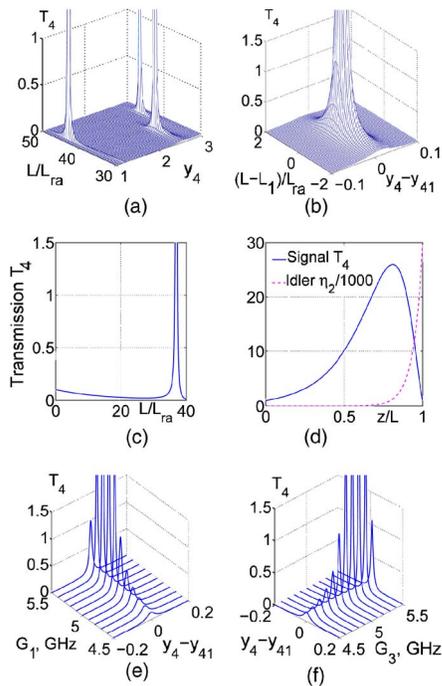


Fig. 3. (Color online) Laser-induced transmission resonances in the NI frequency domain.  $\Omega_1 = \Omega_3 = 2.5\Gamma_{lg}$ . (a)–(d)  $G_1 = G_3 = 50$  GHz. (a)–(c) and (e)–(f)  $z = 0$ . (c), (d)  $y_4 = y_{41} \equiv 2.5266$ . (c) maximum is at  $\alpha_{40}L = L/L_{ra} = L_1/L_{ra} \equiv 37.02$  and  $T_4 \approx 1$  at  $L/L_{ra} = 36.52$ . (d) intensity distribution inside the slab: signal, solid line; idler, dashed line;  $L/L_{ra} = 36.52$ . (e) and (f)  $L = L_1$ . (e)  $G_3 = 50$  GHz. (f)  $G_1 = 50$  GHz.

beneficial and detrimental resonant processes. Figures 3(e) and 3(f) display similar resonance dependence on the strength of the control fields. The amplification in the maximums in Fig. 3 reaches many orders of magnitude, which indicates the feasibility of *oscillations without a cavity* and, hence, the generation of counterpropagating left-handed signal and right-handed idler photons. It is known that even small amplification per unit length may lead to lasing provided that there is a high-quality cavity (or feedback resonance), which effectively increases the distance over which the amplification occurs.

According to Fig. 3(c), characteristic values of  $\alpha_{40}L \sim 10$  are required to ensure the transparency and gain. Assuming  $\sigma_{40} \sim 10^{-16} \text{ cm}^{-3}$  for the resonance absorption cross-section, which is typical for dye molecules, and  $N \sim 10^{19} \text{ cm}^{-3}$  for the density of molecules, we obtain that  $\alpha_{40} \sim 10^3 - 10^4 \text{ cm}^{-1}$ , and the required slab thickness is in the range of  $L \sim 10 - 100 \text{ } \mu\text{m}$ . At these values, the contribution of the nonlinear centers in the refraction index is esti-

mated as  $\Delta n < 0.5(\lambda/4\pi)\alpha_{40} \sim 10^{-2} - 10^{-3}$ , which essentially does not change the linear negative refractive index.

In conclusion, we propose the compensation of losses in strongly absorbing NIMs through embedded, tailored optical nonlinearities. Such a possibility is shown with a realistic numerical model. We have studied the resonant FWM-based OPA in such composite metamaterials with a negative refractive index at the frequency of the signal and a positive index for all other coupled waves. The strong nonlinear optical response of the composite is primarily determined by the embedded four-level nonlinear centers and hence, can be *adjusted independently*. In addition, we have shown the possibility of quantum control of the local optical parameters, which employs constructive and destructive quantum interference tailored by two auxiliary control fields. Frequency-tunable transparency windows in the negative-index frequency domain, *generation of entangled counterpropagating photons without a cavity*, and the feasibility of quantum switching in NIMs have been shown. Having attained basically the same features as predicted in [7,8] for three-wave off-resonant OPA, the given approach ensures engineering the strong nonlinear response of a NIM.

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## References

1. V. M. Shalaev, Nat. Photonics **1**, 41 (2007).
2. M. Lapine, M. Gorkunov, and K. H. Ringhofer, Phys. Rev. E **67**, 065601 (2003).
3. A. A. Zharov, I. V. Shadrivov, and Y. S. Kivshar, Phys. Rev. Lett. **91**, 037401 (2003).
4. M. W. Klein, M. Wegener, N. Feth, and S. Linden, Opt. Express **15**, 5238 (2007).
5. V. M. Agranovich, Y. R. Shen, R. H. Baughman, and A. A. Zakhidov, Phys. Rev. B **69**, 165112 (2004).
6. M. Scalora, G. D'Aguanno, M. Bloemer, M. Centini, N. Mattiucci, D. de Ceglia, and Yu. S. Kivshar, Opt. Express **14**, 4746 (2006).
7. A. K. Popov and V. M. Shalaev, Appl. Phys. B **84**, 131 (2006).
8. A. K. Popov and V. M. Shalaev, Opt. Lett. **31**, 2169 (2006).
9. N. M. Litchinitser, I. R. Gabitov, A. I. Maimistov, and V. M. Shalaev, Opt. Lett. **32**, 151 (2007).
10. A. I. Maimistov and I. R. Gabitov, <http://arxiv.org/abs/nlin/0702023>.
11. A. K. Popov, S. A. Myslivets, and T. F. George, Phys. Rev. A **71**, 043811 (2005).