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Alexander K. Popov, Sergey A. Myslivets, "Remote sensing with nonlinear negative-index metamaterials," Proc. SPIE 9157, 23rd International Conference on Optical Fibre Sensors, 91573B (2 June 2014); doi: 10.1117/12.2059728

Event: OFS2014 23rd International Conference on Optical Fiber Sensors, 2014, Santander, Spain

Remote sensing with nonlinear negative-index metamaterials

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ABSTRACT

A concept of the remotely actuated and interrogated four-wave mixing photonic sensor, which employs the negative-index photonic materials, is described. Unique electromagnetic properties of such a metamaterial enable enhancement of frequency conversion and redirection of the signals which carry important information for environmental probing. Four-wave mixing process allows for independent engineering of resonantly enhanced optical nonlinearities and negative refractive index.

Keywords: Optical remote sensing, optical frequency up-conversion and parametric amplification, negative refractive index metamaterials, backward electromagnetic waves, infrared nonlinear optical microsensors.

1. INTRODUCTION

Infrared (IR) radiation plays a critically important role in pollution, chemical and image sensing. This work is to develop a concept and the underlying theory of all-optically controlled, remotely actuated and interrogated, ultra-compact sensor that can be employed for environmental probing in remote or hostile locations. Numerical simulations have been carried out to identify anticipated operational properties of the proposed sensor in the short-pulse regime, which utilizes nonlinear-optical (NLO) negative-index (NI) metamaterials (NIMs). Extraordinary properties of the electromagnetic propagation processes in NIMs stem from the fact that energy flow and phase velocity of electromagnetic waves become counter-directed inside the NIM slab. The direction of the wave-vector $\mathbf{k}$ with respect to the energy flow (Poynting vector $\mathbf{S}$) depends on the signs of electrical permittivity ϵ and magnetic permeability μ: $\mathbf{S} = (c/4\pi)[\mathbf{E} \times \mathbf{H}] = (c^2\mathbf{k}/4\pi\omega\epsilon)H^2 = (c^2\mathbf{k}/4\pi\omega\mu)E^2$. At $\epsilon < 0$ and $\mu < 0$, refractive index becomes negative, $n = -\sqrt{\mu\epsilon}$, and vectors $\mathbf{S}$ and $\mathbf{k}$ become contra-directed, which is in striking contrast with the electrodynamics of ordinary, positive index (PI) media. Such backward electromagnetic waves (BWs), do not exist in naturally occurring materials but become achievable in metamaterials. Current mainstream in fabricating optical NIMs grounds itself on crafting of plasmonic electromagnetic media composed of the metallic building blocks (metaatoms). The latter are nanoscopic LC circuits deliberately engineered to enable the negative optical magnetic response. Due to the resonant properties of the mesoatoms, the negative index can be engineered only within a certain frequency bandwidth and remains positive outside of it. The possibility of phase matching of the ordinary and BWs with contra-directed energy fluxes, (whereas all wave-vectors are co-directed), leads to counter-intuitive effects and requires novel theoretical and computational approaches to the corresponding problems of nonlinear optics. Extraordinary features of coherent NLO energy conversion processes in NIMs that stem from wave-mixing of the ordinary and backward electromagnetic waves were predicted. Particularly, they enable many-order increase of frequency-conversion efficiency through second[1,2] and third[3] harmonic generation as well as through three-wave mixing.[4–8] The latter is accompanied by the parametric amplification of the signals. The notable application is the frequency up- and down-converting NLO metamirror[4,9] in which a nonlinear NIM emits the generated frequency towards the source of the pump, which can be used for sensing.[10,11] Crafting of bulk nanostructured optical NIMs remains a challenging task.

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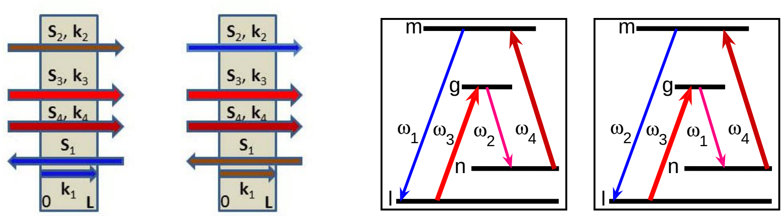
23rd International Conference on Optical Fibre Sensors, edited by José Miguel López-Higuera, Julian Jones, Manuel López-Amo, José Luis Santos, Proc. of SPIE Vol. 9157, 91573B

· doi: 10.1117/12.2059728

Recently, such a metamirror was simulated for the microwaves making use of a set of macroscopic electronic devices.[12] The possibility of independent nanoengineering of negative refraction index and quantum engineering of cubic nonlinearity through embedded four-levels centers was considered in.[13] Specific properties pertinent to resonantly enhanced four-wave mixing (FWM) of continuous waves were investigated. It appeared that in many cases the required intensity of the pump field falls above that achievable in cw regime. Investigation of FWM of ordinary and contra-propagating backward waves in short-pulse regime requires special approach, which is the goal of this work.

2. NONLINEAR-OPTICAL SENSOR UTILIZING FOUR-WAVE MIXING IN DOUBLE DOMAIN POSITIVE/NEGATIVE-INDEX METAMATERIAL: ALTERNATIVE COUPLING SCHEMES

Figure 1 depicts two options for phase matched FWM coupling of the ordinary and backward waves. Here, quantum four-level centers embedded in a NI host material, such as quantum dots, ions or molecules, introduce resonantly enhanced higher-order $[\chi^{(3)}]$ NLO response to the composite metamaterial. Frequency up-conversion and amplification of the signals rely on the FWM process $\omega_1 + \omega_2 = \omega_3 + \omega_4$. Amplification of the waves at ω_1 and ω_2 is controlled by the coherent energy transfer from the control fields at ω_3 and ω_4. The research challenge is that the resonant centers make the problem of losses even more severe and the system is strongly driven by the control fields, so that all local material parameters become intensity dependent. Investigations show that there exist such optimum conditions that the introduced NLO energy conversion process may become dominant which counter-intuitively leads to compensating losses and even to amplification of the signal and the idler. Consider the example depicted in panels (a,c). Assume that the IR wave at ω_2 with the wave-vector $\mathbf{k}_2$ traveling along the z-axis is a PI ($n_1 > 0$) signal and the higher-frequency idler at $\omega_1 = \omega_3 + \omega_4 - \omega_2$ falls in the NI domain ($n_1 < 0$). Then, the phase-matching condition $\Delta k = k_4 + k_3 - k_2 - k_1 = 0$ can be fulfilled for the frequency up-converted idler at ω_1 travelling against other beams. The idler contributes back into the wave at ω_2 through the same type of FWM interaction and thus enables optical parametric amplification (OPA) at ω_2 by converting the energy of the control fields into the signal. All of the coupled waves have their wave-vectors co-directed along z, whereas the energy flow of the idler wave, $\mathbf{S}_1$, is counter-directed to the energy flows of all the other waves, which are co-directed with their wave-vectors. In the second option, Fig. 1(b,d), control fields and the NI IR signal at ω_1 ($n_1 < 0$) enter the sensor from the opposite sides. The amplified IR signal propagates against the control beams, whereas up-converted idler travels along the control beams. Such coupling schemes are in strict contrast with the conventional phase-matching scheme for OPA in ordinary materials where all energy-flows and phase velocities are co-directed. It appears that operational properties for these option are essentially different.



(a) (b) (c) (d)

Figure 1. Two different options of FWM sensors. Here $\mathbf{S}_1$, $\mathbf{k}_1$ and ω_1 are energy flux, wave-vector and frequency for the backward wave $[n(\omega_1) < 0]$, whereas $\mathbf{S}_{2,3,4}$, $\mathbf{k}_{2,3,4}$ and $\omega_{2,3,4}$ are corresponding values for the ordinary waves. Here, $\omega_{3,4}$ correspond to control pump fields. Panels (a,c) and (b,d) depict alternative schemes of quantum controlled four-wave mixing in the embedded resonant nonlinear-optical centers with different ratio of the signal and the idler absorption rates and nonlinear susceptibilities. (a,c) - FWM sensor amplifies IR signal at ω_2, up-converts its frequency and sends up-converted radiation at ω_2 back against the control beam, towards a detector. Amplified IR signal propagates along the control beams towards the remote IR detector. (b,d) - FWM sensor amplifies IR signal traveling against the control beam and frequency up-converts it to the beam propagating along the control beams towards the remote detector.

The slowly varying amplitudes of the coupled waves are given by the equations

$$E_j = (1/2)\mathcal{E}_j(z,t)e^{ik_jz-i\omega_jt} + c.c., j = 1,2,3,4, \quad (1)$$

$$\frac{\partial a_1}{\partial z} - \frac{1}{v_l}\frac{\partial a_1}{\partial t} = -iX_1a_2^*a_3a_4\exp(i\Delta kz) + (\alpha_1/2)a_1, \quad (2)$$

$$\frac{\partial a_2}{\partial z} + \frac{1}{v_2}\frac{\partial a_2}{\partial t} = iX_2a_1^*a_3a_4\exp(i\Delta kz) - (\alpha_2/2)a_2, \quad (3)$$

$$\frac{\partial a_3}{\partial z} + \frac{1}{v_3}\frac{\partial a_3}{\partial t} = iX_3a_1a_2a_4^*\exp(-i\Delta kz) - (\alpha_3/2)a_3, \quad (4)$$

$$\frac{\partial a_4}{\partial z} + \frac{1}{v_4}\frac{\partial a_4}{\partial t} = iX_4a_1a_2a_3^*\exp(-i\Delta kz) - (\alpha_4/2)a_4. \quad (5)$$

Here, $\Delta k = k_4 + k_3 - k_2 - k_1$ is phase mismatch, v_j and α_j^{-1} are group velocity and effective photon mean free path at the corresponding frequencies; $a_j = \sqrt{|\epsilon_j/k_j|}\mathcal{E}_j$, $X_{1,2} = \sqrt{|k_1k_2/\epsilon_1\epsilon_2|}2\pi\chi_{1,2}^{(3)}$, $X_{3,4} = \sqrt{|k_3k_4/\epsilon_3\epsilon_4|}2\pi\chi_{3,4}^{(3)}$. Note, that the signs in Eq. (2) and the boundary conditions are opposite to those in Eqs. (3)-(5) because $n_1 < 0$ and, consequently, energy flux is directed against axis z. The computation challenge is that, in general case of resonant or near-resonant coupling, all local parameters here become dependent on the intensity of the coupled fields and, therefore, can be tailored by the means of quantum control. Quantum density-matrix technique is the most convenient for the computations and analysis of the local optical parameters of the metamaterial. For example, for the scheme Fig. 1(d), the linear and nonlinear polarizations at ω_1 and ω_2 are calculated as

$$P_1(z,t) = (1/2)\{P_{01}^L\exp(ik_1z) + P_{01}^{NL}\exp[i(k_3+k_4-k_2)z]\}\exp(-i\omega_1t) + c.c. = N(\rho_{ng}d_{gn} + \rho_{gn}d_{ng}); \quad (6)$$

$$P_2(z,t) = (1/2)\{P_{02}^L\exp(ik_2z) + P_{02}^{NL}\exp[i(k_3+k_4-k_1)z]\}\exp(-i\omega_2t) + c.c. = N(\rho_{ml}d_{lm} + \rho_{lm}d_{ml}). \quad (7)$$

Here, ρ_{ij} are the density matrix elements, d_{ij} are the transition dipole elements and N is number density of the embedded centers. For the scheme in Fig. 1(c), they can be defined in a similar way. The set of equations for off-diagonal (coherence) and diagonal (energy level populations) density-matrix elements are bulky. Analytical solutions to these equations can be found only in some ultimate cases.[14] Effective linear, $\chi_{1,2}$, and NLO, $\chi_{1,2}^{(3)}$, susceptibilities dependent on the intensities of the driving control fields E_3 and E_2 are defined as

$$P_{01}^L = \chi_1\mathcal{E}_1, \quad P_{01}^{NL} = \chi_1^{(3)}\mathcal{E}_3\mathcal{E}_4\mathcal{E}_2^*; \quad P_{02}^L = \chi_2\mathcal{E}_2, \quad P_{02}^{NL} = \chi_2^{(3)}\mathcal{E}_3\mathcal{E}_4\mathcal{E}_1^*. \quad (8)$$

The linear susceptibilities determine the intensity-dependent contributions to absorption and to the refractive indices of the composite attributed to the embedded centers, while the NLO susceptibilities determine the FWM. Here, $\omega_1 + \omega_2 = \omega_3 + \omega_4$, and $k_j = |n_j|\omega_j/c > 0$. Absorption and refraction attributed to the host material are assumed intensity and frequency independent. Linear and NLO local parameters attributed to the embedded centers are calculated by the density-matrix method as described in.[14] Analytical solution to the equations can be found for the ultimate case of the control fields constant across the NIM slab. Usually, they are a set of bulky equations dependent on many material and field parameters, such as optical thicknesses at corresponding frequencies, intensity of the control fields, input intensity of the signal, nonlinear susceptibilities, refraction indices at the coupled frequencies and phase mismatch, and some others. The outcomes, described by these equations, such as distribution of the signal and the idler across the slab, tailored transparency and reflectivity or enhanced opaqueness, oscillation threshold, etc. are sensitive to the indicated parameters.[6] The optimization allows deriving the range of optimum input fields' and material parameters and the range of the expected outcomes. Shape of the fundamental pulses of duration t_p was chosen nearly rectangular: $\mathcal{E}_{3,4} = (1/2)\mathcal{E}_{3,4}^0\{\tanh[(t_0 + t_p - t)/t_f] - \tanh[(t_0 - t)/t_f]\}$. Basic properties of the sensor for the ultimate case of constant pump amplitude can be analyzed and in the coordinate frame locked to the pump pulse. Then the equations for $a_{1,2}$ inside the fundamental pulses take the form:

$$K_1^{-1}da_1/d\xi = -iX_1a_2^*a_3a_4\exp(i\Delta k\xi) + (\alpha_1/2)a_1, \quad (9)$$

$$K_2^{-1}da_2/d\xi = iX_2a_1^*a_3a_4\exp(i\Delta k\xi) - (\alpha_2/2)a_2, \quad (10)$$

where $\xi = z - v_lt$, $v_3 = v_4 = v_l$, $K_1 = v_1/(v_1 + v_l)$, $K_2 = v_2/(v_2 - v_l)$. Equations (9) - (10) are similar to those describing continuous wave three-wave mixing,[4,5] except for the boundary conditions. They correctly describe possible huge amplification of the IR signal and the up-converted radiation in the BW regime until the related depletion of the strong input laser beam can be ignored. Boundary conditions must be determined at the edges of the fundamental pulses $\xi = 0$ and $\xi = l_p = v_l t_p$. In the cases of Fig. 1(a,c) and Fig. 1(b,d) the boundary conditions are $a_2(\xi = 0) = a_2^0, a_1(\xi = l_p) = 0$ and $a_1(\xi = l_p) = a_1^0, a_2(\xi = 0) = 0$, respectively. Equations (9)-(10) are similar to those describing huge enhancement of stimulated Raman scattering on backward optical phonons in the short-pulse regime.[11] Hence, the similar effect can be anticipated for the proposed sensors.

In summary, a theoretical approach towards a novel concept for photonic sensing is reported, which utilizes unparallel properties of four-wave mixing in negative-index metamaterials. Proposed process opens the possibility for independent nanoengineering of the resonantly enhanced nonlinearity and negative refraction index. Short-pulse regime allows for mitigating of detrimental accompanying effects peculiar to cw regime.

ACKNOWLEDGMENTS

This work was supported in parts by the Air Force Office of Scientific Research (Grant No FA950-12-1-298), by the National Science Foundation (Grant No ECCS-1346547) and by the Presidium of the Siberian Branch of the Russian Academy of Sciences (Grants No. 43 and No. 101).

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