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Research Article

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Group velocity mismatch at ultrashort electromagnetic pulse propagation in nonlinear metamaterials

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Abstract: The parametric interaction of optical wave pulses in metamaterials is considered in the first approximation of the theory of dispersion. The interaction between the quasi-monochromatic pump wave and the wave pulse at the total frequency with quadratic phase modulation is assumed. The results of calculation of the shape of the spectrum of an excited signal wave at a difference frequency are presented for low frequency pumping. It is shown that the effects of group mismatch in metamaterials lead to a narrowing of the spectrum of the excited wave. With an increase in the modulation degree of a weak exciting wave, the spectrum of the excited wave broadens.

Keywords: group velocity, metamaterial, ultrafast pulse

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1 Introduction

Metamaterials are artificial materials formed from microstructures (metaatoms), whose properties can be controlled. Such media are made from natural materials with a positive refractive index. It can be the smallest metal conducting wires and split-rings. They form an oscillating circuit with microscopic solenoids and capacitors that determine the electrical, magnetic, and optical properties of the structure. By changing the capacitance and inductance of such an oscillating circuit, it is possible to control and determine the optical properties of a material in a certain spectral range. It is possible to design a left-handed material in which an optical wave at a negative refraction frequency propagates with a phase velocity opposite to its group velocity. In such artificial structures, the usual optical phenomena change, new unusual effects appear. Thus, the metamaterial can be created from a composite material forming a dielectric matrix, with inclusions providing the resonant properties of the material. A similar approach was used in the development of solid-state lasers, when activator ions were introduced into the matrix, for example, of a crystal, which ultimately determined the physical properties of the laser medium. The electromagnetic properties of the metamaterial are also dictated by the characteristics of the microstructures that are embedded in the composite material. This opens up new prospects for their use [1–3]. For example, the modern development of photonics is associated, in particular, with the technology of developing metamaterials. As is known, in optical information processing systems photons are used as information carriers, and here the problem arises of controlling this data carrier. The discovery of metamaterials contributed to the possibility of controlling light by changing the optical properties of such meta-atoms. In optical switching systems using the phenomenon of bistability, it is proposed to use a sandwich structure. The use of thin films of material opens up new possibilities for the design of photonic crystals [4].

The current level of nanotechnology development allowed the development of materials with a negative refractive index already in the visible range [5, 6]. The study of the non-stationary mode of frequency conversion in metamaterials is of practical interest.

Ultrashort laser pulses are used in medicine [7], biology, for generation of terahertz radiation [8], for non-linear sounding of the atmosphere [9] and a number of other applications. A distinctive feature of ultrashort laser pulses, in particular femtosecond pulses, is the ability to concentrate huge energy in extremely small spectral, temporal and spatial intervals, the record intensity at the focus of the laser beam is ~$10^{21} \div 10^{22}$ W/cm^2. As is well known, for ultrashort pulses the nature of the interaction of modulation waves depends essentially on the dispersion properties of the medium. In a material with a positive refractive

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index at the interaction of an ordinary and extraordinary wave, the wave with a large group velocity is ahead of the second wave. At a certain point in time, the wave packets of the waves are spatially shifted relative to each other and the waves cease to interact. In left-handed media, the group velocities of the interacting waves are directed towards each other. At some point, the wave packets completely overlap and then there is an effective interaction between the waves. Then the waves move away from each other, propagate independently, *i.e.* interaction between them is excluded. Such overlap of wave packets can occur in different areas of the material. It depends on the ratio of group velocities. In addition, it is necessary to take into account the presence of different phase velocities of the frequency components of the pulse wave in the dispersing medium. This leads to the effect of dispersive spreading of an optical pulse. Therefore, consideration of the effect of group velocity mismatching of counter pro-pagating waves in the study of the nonlinear parametric interaction of ultrashort pulsed waves in metamaterials is of interest.

The study of nonlinear effects in artificial media and engineering the nonlinear response of such media are crucially important for the future success of the entire field of metamaterials [10]. Research is being conducted in various directions. The method of obtaining composite metamaterial from a mixture of two isotropic dielectric materials is considered by author in [11]. Ultrathin active chiral metamaterials of highly tunable chiroptical responses for exploit the metamaterials as ultrasensitive sensors have been investigated in [12]. In [13], the use of structural metamaterials for the spectral analysis of deformation is considered. Authors of [14] illustrate subtleties in the calculation of effective dielectric constant of the metamaterial for different forms of the dielectric. In [15], the concept of left-handed chiral metamaterials is analyzed by emphasizing their optical ability on the rotation of the plane of polarization of a wave.

The importance of left-handed materials led to the rapid study of nonlinear optical effects in metamaterials, both stationary effects of the second order (SHG, gene sum and different frequencies, opt pairs), and non-stationary effects of the third order nonlinearity [16–22]. In [23] authors reported on the recent progress on investigaton of ultrashort pulses propagation in metamaterials, including second-order and third-order nonlinear optical phenomena such as spatiotemporal instability.

IThe authors of this paper also investigated the nonstationary generation in the second approximation of the theory of dispersion [24]. It was shown that the effects of group-velocity dispersion of counterpropagating waves and dispersive spreading of pulses of interacting waves lead to a narrowing of the spectrum of the excited signal. The study of the nonstationary interaction of waves in the mixing of three waves in media with negative refraction is the goal of this work. The results of calculating the shape of the spectrum for the signal wave are presented at low-frequency pumping in the first approximation of dispersion theory.

2 Theory

Let's investigate the nonstationary process of parametric three-frequency nonlinear interaction of waves. Let's assume that the medium is "left" only at the frequency of the signal wave ω_1. The authors assume that the pump wave is a long pump pulse at a frequency ω_2, in contrast to a short idler wave pulse at a sum frequency $\omega_3 = \omega_1 + \omega_2$, while also ssuming that the energy fluxes of the pump wave and the idler waves at the sum frequency $S_{2,3}$ fall normally on the left side surface of the metamaterial of length l and propagate along the positive direction of the axis. Hence the transfer of energy of the signal wave, for which the medium is "left", occurs in the opposite direction.

Let's consider, in a first approximation, the theory of dispersion. In this case, the system of truncated equations has the following forms ($\delta_j = 0$) [4, 10, 25]

$$\left(\frac{\partial}{\partial z} - \frac{1}{u_1}\frac{\partial}{\partial t}\right) A_1 = i\gamma_1 A_3 A_2^{*} e^{i\Delta z}, \tag{1}$$
$$\left(\frac{\partial}{\partial z} + \frac{1}{u_2}\frac{\partial}{\partial t}\right) A_2 = -i\gamma_2 A_3 A_1^{*} e^{i\Delta z},$$
$$\left(\frac{\partial}{\partial z} + \frac{1}{u_3}\frac{\partial}{\partial t}\right) A_3 = -i\gamma_3 A_1 A_2 e^{-i\Delta z}.$$

Here $A_{1,2,3}$ are the complex amplitudes of the signal wave at the frequency ω_1, the pump wave at the frequency ω_2 and the idler wave at the sum frequency ω_3. $u_{1,2,3}$ are group velocities of the corresponding waves, $\gamma_{1,2,3}$ are coefficients of nonlinear coupling of waves at the corresponding frequencies, $\gamma_j = \frac{8\pi}{k_j c^2}\chi_{eff}^{(2)}\omega_j^2\varepsilon_j$, and $\Delta = k_3 - k_2 - k_1$ is the phase mismatching from the central frequency of the pump wave.

The authors solve the problem in the case when a pump wave and a wave at a frequency ω_3 are present at the entrance to a medium with negative refraction

$$A_1(z = l) = 0; \quad A_2(z = 0) = A_{20}; \quad A_3(z = 0) = A_{30} \tag{2}$$

It should be noted that in the case under consideration there is a counter interaction of the initial short pulse with the excited signal wave. Therefore, the group delay effect, which is the characteristic of the incident waves due to the different phase velocities of the waves in the medium, in the metamaterial leads to a group velocity mismatching of the waves.

The peculiarity of the parametric interaction of the pump wave, signal and idler waves in the metamaterial is determined by the conditions of propagation of these waves. The nature of the propagation of a signal wave in a left-handed medium differs from the case of propagation in a traditional medium, which is explained by the opposite directions of group and phase velocities. It should be noted that, according to the boundary conditions, a nonzero signal wave enters the medium on the right and this leads to the dependence of the complex amplitude of the signal wave at the output on the full length of the metamaterial. In the process of wave propagation in a nonlinear medium, as a result of nonlinear interaction, energy is exchanged between the direct pump wave, the idler wave and the reverse signal wave, and the pump wave energy is transferred into the signal wave energy. The effectiveness of this process depends on the phase relationship between the interacting waves.

The system (1) is solved in the constant-field approximation, *i.e.* $A_2(z,\ t) = A_{20} = const$. After a number of standard transformations using the spectral approach, the solution of the system (1) is found with respect to the signal wave. Taking into account the boundary conditions (2) for the complex amplitude of the signal wave at the output of the metamaterial, one obtains

$$A_1(z=0,\omega) = -i\gamma_1 A_{30} A_{20}^{*} \exp\left(i\frac{\omega+\Delta}{2}z\right) \times \tag{3}$$
$$\times \frac{\tan\lambda l}{\lambda - \frac{i}{2}(\Delta - \nu\omega)\tan\lambda l}$$

where

$$\nu = \frac{1}{|u_1|} + \frac{1}{u_3}, \quad \lambda = \sqrt{\frac{(\omega\nu+\Delta)^2}{4} - \Gamma_2^2},$$
$$\Gamma_2^2 = \gamma_1\gamma_3 I_{20}, \quad I_{20} = A_{20}\cdot A_{20}^{*}.$$

Hence, for the spectral power density of the output radiation of the signal wave, one will have

$$S_1(z=0,\omega) = \frac{cn_1}{8\pi} A_1 A_1^{*} = \tag{4}$$
$$= \frac{cn_1}{8\pi}\gamma_1^2 A_{30}^2 S_{20} \frac{\tan^2\lambda l}{\lambda^2 + \frac{(\Delta-\nu\omega)^2}{4}\tan^2\lambda l}$$

Suppose that the wave pulse at the total frequency at the input to the left of the metamaterial has a Gaussian shape with quadratic phase modulation, *i.e.* $A_3(t) = A_{30}e^{-\frac{t^2}{2\tau^2} - ik\frac{t^2}{2}}$ [26]. Then the expression for the spectral density of this wave will take the following form

$$S_{30}(\omega) = \frac{cn_3}{8\pi}\frac{A_{30}^2\tau^2}{2\pi}\cdot\frac{1}{\sqrt{1+k^2\tau^4}}e^{-\frac{\omega^2\tau^2}{1+k^2\tau^4}} \tag{5}$$

After substituting (5) into (4), one obtains

$$S_1(z=0,\omega) = C\frac{1}{\sqrt{1+k^2\tau^4}}e^{-\frac{\omega^2\tau^2}{1+k^2\tau^4}} \times$$
$$\times \frac{\tan^2\lambda l}{\lambda^2 + \frac{(\Delta-\nu\omega)^2}{4}\tan^2\lambda l},$$

where

$$C = \frac{cn_1\tau^2}{16\pi^2}\gamma_1^2 I_{20}\, I_{30}.$$

When studying the shape of the spectrum, it is convenient to operate with a ratio where the nonlinear length is $l_{NL} = 1/\Gamma_2$, and $l_\nu = \tau/\nu$.

3 Discussion

Analysis of the spectrum of the excited wave at the difference frequency will be carried out for the reduced spectral density $S'_1(z=0,\omega) = S_1(z=0,\omega)/C$. Taking into account only the group velocity mismatching, in the absence of the dispersive spreading effect, a symmetric course of the spectral dependences is observed.

Figure 1 shows how the shape of the spectrum S'_1 changes as the waves propagate in the metamaterial. The analysis is carried out for the Gaussian form of the input

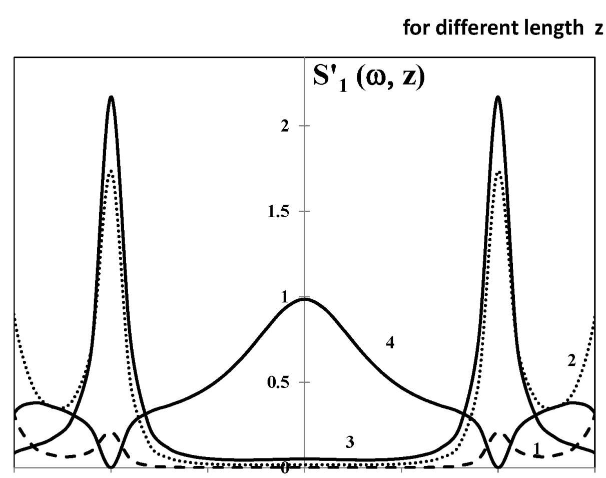

Figure 1: Dependence of the relative spectral density of the excited wave, S'_1, on the parameter $\omega\tau$ at the total thickness of metamaterial $\ell = 2$ cm, $k^2\tau^4 = 0$, $\Delta l_{NL} = 0$, $\frac{l_{NL}}{l_\nu} = 3$, $I_{20} = 2$W for different z:1.8 cm (dashed curve 1), 1.3 cm (dotted curve 2), 1 cm (solid curve 3), 0 (solid curve 4)

excitation pulse. As the waves propagate in the metamaterial, first the side maxima grow near $\omega\tau = \pm 1$, then the energy of the side components is transferred to the central maximum, *i.e.* there is a monochromatization of the spectrum of the excited pulse.

Figure 2 shows how the shape of the spectrum changes with different contributions of group velocity mismatching of the opposing signal wave and the wave at the sum frequency. As the parameter v, characterizing the group velocity mismatching, increases, the spectrum of the excited pulse gets narrowed.

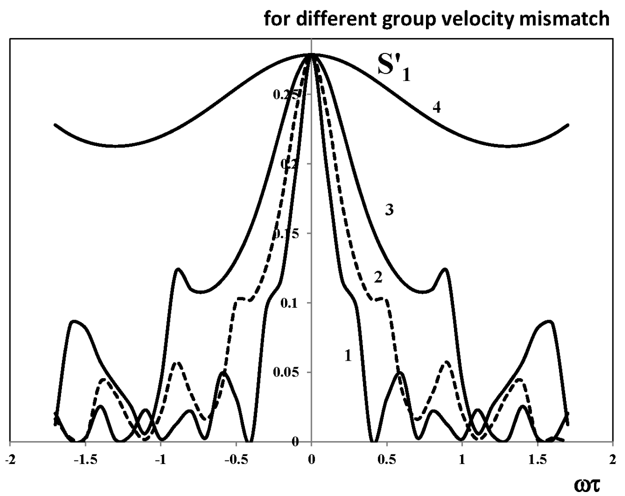

Figure 2: Dependence of the relative spectral density of the excited wave, S_1', on the parameter $\omega\tau$ at the total thickness of metamaterial ℓ=2 cm, $\Delta l_{NL} = 0$, $z = 0.2$ cm, $I_{20} = 2$W, $k^2\tau^4 = 3$ for different group velocity mismatch: $\frac{l_{NL}}{l_v} = 8$ (curve 1), 5 (dashed curve 2), 3 (solid curve 3) and 1 (solid curve 4)

In Figure 3 it can be seen how the shape of the spectrum of the signal wave is modified for different values of the modulation factor (k) for quadratic phase modulation of the pulse of the exciting wave. The effect of phase modulation leads to a broadening of the spectrum of the excited signal wave. With increasing the coefficient of phase modulation, the central maximum is converted to side maxima, *i.e.* the energy of the spectrum from the center is pumped to the lateral maxima. For large values of the phase modulation factor ($k\tau^2 > 1$), an increase in the number of lateral maxima is observed; fragmentation of the excited pulse is observed.

Thus, the influence of the phase modulation of the exciting pulse, the group detuning parameter on the width of the laser radiation spectrum, as well as the observed narrowing of the spectrum of the excited pulse as the wave passes through the metamaterial can be taken into account when developing frequency converters, as well as, for example, to realize a smooth change in the frequency of the output radiation.

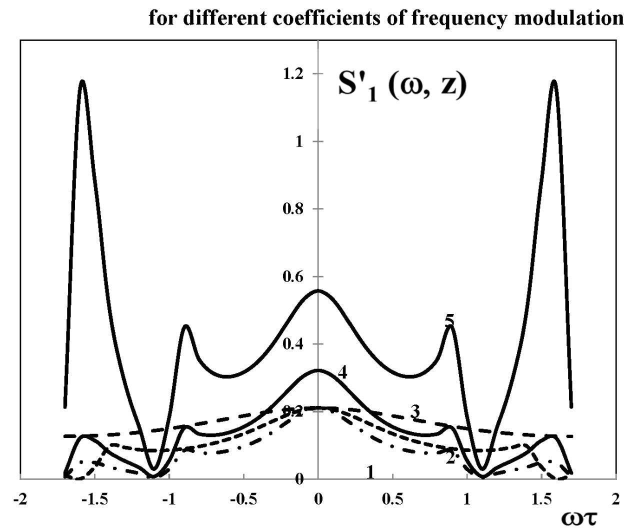

Figure 3: Dependence of the relative spectral density of the excited wave, S_1', on the parameter $\omega\tau$ at the total thickness of metamaterial $\ell = 2$ cm, $\Delta l_{NL} = 0$, $z = 1.8$ cm, $I_{20} = 2$W, $\frac{l_{NL}}{l_v} = 3$ (dotted curve 1, solid curves 4 and 5), 2 (dashed curve 2) and 1 (dashed curve 3) for different coefficients of frequency modulation $k^2\tau^4$: 6 (dotted curve 1, dashed curves 2 and 3), 2 (solid curve 4) and 0 (solid curve 5)

Due to the lack of experimental data on the spectrum of the excited wave in the metamaterial, the characteristic length of the dispersion of group velocities (quasistatic interaction length) for three-wave interaction, when $\omega_1 = \omega_2$ is estimated. In this study, the case for a medium with negative refraction and with nonlinearity as in $LiNbO_3$ crystals is considered. Let one's to calculate the quasistatic length when converting the radiation of a neodymium laser at a wavelength of $\lambda = 1.064$ µm with a pulse duration $\tau = 2$ ps and 7 ps into the second harmonic during $oo \to e$ interaction. According to calculations, in case of a reciprocal interaction in the metamaterial, the quasistatic interaction length l_v are equal to 0.32 mm and $l_v = 0.11$ cm, respectively, and at the wavelength of the main radiation λ =0.532 μm the quasistatic length l_v are equal to 0.53 mm for 2 ps and 0.18 cm for 7 ps.

For comparison, in the case of an ordinary medium with a passing geometry of wave propagation in a $LiNbO_3$ crystal for $oo \to e$ interaction at $\tau = 2$ ps and 7 ps and $\lambda = 1.064$ µm, the analogous values l_v are 0.22 mm and 0.77 mm, respectively, and at the fundamental radiation wavelength $\lambda = 0.532$ µm the quasistatic length l_v are equal to 0.154 mm for 2 ps and 0.54 mm for 7 ps.

It follows from the estimates that the effects of the group velocity mismatch must be taken into account in metamaterials whose dimensions are larger than the calculated values of the quasistatic interaction length.

It can be concluded that in the case of a pulsed mode of interaction of optical waves, the nature of the interaction of the waves, the emission spectrum of the excited wave are determined by the characteristics of both the pulse and the left-handed medium. Overlapping of wave packets of interacting waves leads to an increase in the efficiency of interaction. Such overlap is possible in different areas of the material. It depends on the ratio of group velocities. In addition, it is necessary to take into account the presence of different phase velocities of the frequency components of the pulse wave in the dispersing medium. The latter leads to the effect of dispersive spreading of an optical pulse. Hence, by controlling the characteristics of the left-handed medium and the pulse mode for the idler wave, one can achieve a high conversion efficiency by combining the wave packets of opposite waves.

4 Conclusions

Analysis of the spectrum of the signal wave, by taking into account only the group velocity mismatching, shows that the spectral dependence is symmetric. The energy of the lateral components decreases, and the central maximum increases if the input excitation pulse has a Gaussian shape. Also, as the group velocity mismatching increases, the spectrum of the excited pulse gets narrowed. At high values of the phase modulation coefficient, an increase in the number of lateral maxima is observed, *i.e.* fragmentation of the excited pulse is obtained. The established dependence of the width of the laser radiation spectrum on the phase modulation factor of the exciting pulse, as well as the narrowing of the spectrum of the excited pulse as the metamaterial passes and the group mismatching increases, can be taken into account when designing frequency converters. Also, for example, to perform a smooth tuning of the output radiation frequency. By selecting the parameters of the artificial structure that provide the resonant properties of the material, one can construct a metamaterial of the required quadratic nonlinearity. In such a structure, by varying the phase modulation factor of the exciting pulse and the detuning of the group velocities, it is possible to control and change the width of the spectrum of the output coherent radiation from the minimum to maximum value. The calculation model developed in the present work for quadratic nonlinear metamaterials can be applied to the case of cubic nonlinear metamaterials.

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