

## ON THE THEORY OF FOUR-WAVE INTERACTION IN DISSIPATIVE METAMATERIALS

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An analytical expression is obtained for the threshold pumping amplitude under phase-matching conditions. It is shown that the threshold pump amplitude increases with increasing losses, the input intensity of the idler wave, and with increasing nonlinear coupling coefficients for pump waves. The conditions under which losses can be compensated in the case of propagation of a backward signal wave under parametric four-wave interaction are analyzed. It is shown that, in the presence of a parametric coupling between forward and backward waves, compensation of signal wave losses by losses of direct waves will allow the parametric amplification and generation of the backward wave to be realized at the threshold pump amplitude.

**Keywords:** metamaterials, constant intensity approximation, four wave mixing, compensation of losses.

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### 1. INTRODUCTION

An intense investigation of metamaterials is currently underway to compensate for losses in them. Since the localized plasmons in metamaterials accumulate enough electric fields around themselves the latest advances are observed in nanoplasmonics. It allows to reduce losses in such structures [1,2]. The introduction of plasmon metals, for example, silver [4] or gold [5], near the resonances of which the nonlinearity increases sharply, into the dielectric structure, leads to a significant concentration of the electromagnetic field around the plasmon nanoparticles. So in the case of a gold nanoparticle, silicon is used as a dielectric structure, because of the small work function of an electron from gold to silicon. This fact is successfully used to increase the sensitivity of photodetectors, solar photoconverters. The ability of plasmon nanoparticles to accumulate large electric fields around themselves allowed the authors of Refs. 1 and 2 to report on overcoming large losses in similar structures.

When three light waves at frequencies  $\omega_2$ ,  $\omega_3$  and  $\omega_4$  are incident onto cubic nonlinear medium the nonlinear polarization leads to the generation of a new electromagnetic with frequency  $\omega_1 = \omega_3 + \omega_4 - \omega_2$ . In a photon language the FWM is that two photons with initial frequencies are subjected to elastic scattering to produce two new photons. Here the law of conservation of energy and momentum have to be fulfilled in this process:  $k_1 = k_3 + k_4 - k_2$  where  $k_j$  – are the wave numbers at respective frequencies  $\omega_j (j = 1 - 4)$ . Conservation of momentum leads to the phase matching conditions. Boundary conditions of the described interaction of waves are given by  $A_{2,3,4}(z = 0) = A_{20,30,40}$  and  $A_1(z = l) = A_{1l}$ , where  $z = 0$  corresponds to the left input of the metamaterial,  $A_{20,30,40}$  are the initial amplitudes of the transmitted

weak wave ( $A_{20}$ ) at the frequency  $\omega_2$  and of the pump waves ( $A_{30,40}$ ) at the frequencies  $\omega_{2,3}$  and  $A_{1l}$  is the initial amplitude of the transmitted signal wave at the right input of the nonlinear medium at  $z = l$ . We consider a third-order nonlinear process connecting four waves at frequencies  $\omega_j (j=1\div 4)$ , of which two strong pump waves (at frequencies  $\omega_3$  and  $\omega_4$ ). We assume that the medium is "left-handed" only at the frequency of the signal wave  $\omega_1$ , i.e. the medium takes negative values of the real parts of the dielectric permittivity and magnetic permeability at this frequency and the positive values of the dielectric permittivity and magnetic permeability at frequencies  $\omega_2$ ,  $\omega_3$  and  $\omega_4$ . We assume that the energy fluxes of a wave at a frequency  $\omega_2$  and two pump waves, i.e.

$S_{2,3,4}$  fall normally on the left side surface of the metamaterial thickness  $l$  and propagate along the positive  $z$  axis direction. Hence the transfer of energy of the signal wave, for which the medium is "left-handed", occurs in the opposite direction.

Third-order nonlinear processes, resolved both in centrosymmetric and non-centrosymmetric media, are more often encountered, they are easier to implement. For their observation, apparently, there is no restriction on the symmetry of the medium. Therefore, in spite of the fact that cubic processes are an order of magnitude weaker than quadratic processes, they are widely used and have many applications [3].

The modulation instability that arises in most nonlinear wave systems has been extensively studied for metamaterials [4-8]. In the of the constant-field approximation, a theoretical study of nonlinear optical interaction in such artificial structures has been carried

out in a number of papers, of which we note [9-11]. The four-wave interaction in metamaterials is considered in [12-14]. In the constant-intensity approximation [15-17], we investigated the second and third harmonic generation, self-action effects and parametric interaction in metamaterials [2,18-20] four-wave interaction in conventional materials [3] and metamaterials.

Studies in work are conducted under conditions similar to those discussed in [12]:  $\omega_1$  is the highest frequency;  $\omega_2 = \omega_3 + \omega_4 - \omega_1$ ;  $\lambda_2 = 756$  nm, and  $\lambda_1 = 480$  nm, i.e. we are within the visible part of the optical range. Optical parametric amplification of an optical wave by means of four-wave mixing in a metamaterial doped with nonlinear optical centers is considered. As the authors of [12] note, the nonlinear optical response of a composite is mainly determined by embedded nonlinear optical centers and, as a consequence, can be independently controlled.

We follow the method used in [9], assuming that the nonlinear response of the medium is mainly related to the magnetic component of the waves. In a similar way, from Maxwell's equations, the corresponding equations for the electrical component can be obtained in a similar form, replacing the dielectric constant of the medium  $\epsilon_j$  by the magnetic permeability  $\mu_j$  and vice versa [9]. For the considered case of four-wave interaction in the metamaterial, the truncated equations have the form [12], which differs from the case of similar interaction in a traditional medium [3].

The purpose of this paper is to analytically determine the conditions under which it is possible to eliminate losses in the case of propagation of the backward-wave at a parametric four-wave interaction, i.e. propagation of a backward wave with a constant amplitude, and also with amplification and generation in a nonlinear medium.

## 2. THEORETICAL APPROACH AND DISCUSSIONS

For this consideration dielectric permittivity of the signal wave becomes negative that is reflected in the first equation of following set of reduced equations [ 20]:

$$\begin{aligned} \frac{dA_1}{dz} + \delta_1 A_1 &= -i\gamma_1 A_3 A_4 A_2^* e^{i\Delta z} , \\ \frac{dA_2}{dz} + \delta_3 A_3 &= i\gamma_2 A_3 A_4 A_1^* e^{i\Delta z} \\ \frac{dA_3}{dz} + \delta_2 A_2 &= i\gamma_3 A_1 A_2 A_4^* e^{-i\Delta z} , \\ \frac{dA_4}{dz} + \delta_4 A_4 &= i\gamma_4 A_1 A_2 A_3^* e^{-i\Delta z} \end{aligned} \quad (1)$$

where  $A_j$  -are the complex amplitudes of the magnetic fields of the transmitted quasi-monochromatic waves,  $\delta_i$  – dissipative losses in metamaterial medium ,  $\gamma_1 = 2\pi k_1 \chi_1^{(3)} / |\epsilon_1|$  and  $\gamma_{2,3,4} = 2\pi k_{2,3,4} \chi_{2,3,4}^{(3)} / \epsilon_{2,3,4}$  are the nonlinear wave coupling coefficients,  $\chi_j^{(3)}$  is the cubic susceptibility, and  $\Delta = k_3 + k_4 - k_1 - k_2$  is the phase detuning of the interacting waves. The corresponding equations for the electric components can be derived analogously with replacement of the dielectric permittivity of the medium  $\epsilon_j$  by the magnetic permeability  $\mu_j$  and *vice versa* [4] .

Differentiation of the first equation of system (1) yields to the following second order differential equation

$$\frac{d^2 A_1}{dz^2} - i\Delta \frac{dA_1}{dz} + (\gamma_1 \gamma_2 I_{30} I_{40} - \gamma_1 \gamma_3 I_{20} I_{40} - \gamma_1 \gamma_4 I_{20} I_{30} + (\sum_1^4 \delta_j - i\Delta)^2 / 4) A_1 = 0 \quad (2)$$

Solving equation (2) in the constant intensity approximation [10,11,14-17 ] of the fundamental radiation ,  $I_1(z) = I_1(z=0 = I_{10})$  under boundary conditions

$$A_1(z=l) = A_{1l} \quad \text{and} \quad A_{2,3,4}(z=0) = A_{20,30,40} \quad (3)$$

we obtain for the complex amplitude of the signal wave propagating in opposite direction with respect to other three waves ( $\delta_j = 0$ ) :

$$A_1(z) = (M_1 + iM_2) e^{\frac{\Delta z}{2}} \quad (4)$$

here

$$\begin{aligned} M_1 &= \frac{A_{1l} e^{-i\frac{\Delta l}{2}} \cos \lambda z + m(\Delta/2) \frac{\sin \lambda l}{\lambda} \frac{\sin \lambda z}{\lambda}}{\cos \lambda l - i \frac{\Delta}{2\lambda} \sin \lambda l} \\ M_2 &= \frac{m \sin \lambda l \frac{\cos \lambda z}{\lambda} - A_{1l} e^{-i\frac{\Delta l}{2}} (\Delta/2) \frac{\sin \lambda z}{\lambda}}{\cos \lambda l - i \frac{\Delta}{2\lambda} \sin \lambda l} - m \frac{\sin \lambda z}{\lambda} \end{aligned}$$

where  $\lambda = (\gamma_1\gamma_2I_{30}I_{40} - \gamma_1\gamma_3I_{20}I_{40} - \gamma_1\gamma_4I_{20}I_{30} + \Delta^2/4)^{1/2}$ ,  $m = \gamma_1A_{30}A_{40}A_2^*$   
In the input ( $z = 0$ ) to the medium amplitude of signal wave is simplified as

$$A_1(z) = (M_3 + iM_4) \quad (5)$$

where

$$M_3 = A_{1l}e^{-i\frac{\Delta l}{2}}/(\cos\lambda l - i\frac{\Delta}{2\lambda}\sin\lambda l)$$

$$M_4 = m\frac{\sin\lambda l}{\lambda}/(\cos\lambda l - i\frac{\Delta}{2\lambda}\sin\lambda l)$$

Taking into account the dissipative losses for complex amplitude of signal wave in the constant intensity approximation yields:

$$A_1(z) = e^{-\frac{a}{2}z} \left[ \frac{A_{1l}e^{\frac{a}{2}l} - \left( \frac{\delta_1}{\lambda} A_{1l} - i\frac{b}{\lambda} \right) \cdot \sin\lambda l}{\cos\lambda l + \frac{a}{2\lambda}\sin\lambda l} \left( \cos\lambda z + \frac{a}{2\lambda}\sin\lambda z \right) + \frac{\delta_1 A_{1l} - ib}{\lambda} \sin\lambda z \right] \quad (6)$$

$$a = \delta_2 + \delta_3 + \delta_4 - \delta_1 - i\Delta, \quad b = \gamma_1 A_{20}^* A_{30} A_{40},$$

$$\lambda = \sqrt{\gamma_1\gamma_2I_{30}I_{40} - \gamma_1\gamma_3I_{20}I_{40} - \gamma_1\gamma_4I_{20}I_{30} - \frac{\left( \sum_1^4 \delta_j - i\Delta \right)^2}{4}}$$

From the condition of equality of the initial value of the complex amplitude of the signal wave at the entrance to the medium on the right,  $A_1(z = l) = A_{1l}$ , and the final value at the exit from the medium on the left,  $A_1(z = 0)$ , i.e.

$$A_1(z = 0) \geq A_1(z = l) = A_{1l}, \quad (7)$$

under phase-matching conditions, the following relationships are obtained (to simplify further calculations, we assume  $I_{30} = I_{40} = I_{p0}$ ):

$$\delta_2 + \delta_3 + \delta_4 = \delta_1, \quad (8)$$

$$\lambda l = 0, 2\pi, \dots \quad (9)$$

(9) we determine the threshold pump intensity

$$I_{pump}^{thresh} = \frac{\gamma_3 + \gamma_4}{2\gamma_2} I_{20} + \sqrt{\left( \frac{\gamma_3 + \gamma_4}{2\gamma_2} I_{20} \right)^2 + \frac{\delta_1^2}{\gamma_1\gamma_2}}, \quad (10)$$

As can be seen from expression (10), the threshold intensity of the pump increases with increasing dissipative losses  $\delta_j$  ( $\delta_2 + \delta_3 + \delta_4 = \delta_1$ ).

After a single passage of a nonlinear medium as a result of parametric interaction of waves, it is possible to achieve the constancy of the amplitude of the signal wave when a certain threshold condition for the parameters of the problem is fulfilled.

The inequality sign in (7) assumes that in the process of nonlinear interaction of waves, in addition to the constancy of the amplitude of the backward signal wave, its amplification is also possible. Condition (8)

means that in the presence of a coupling between forward and backward waves, compensation of signal wave losses ( $\delta_1$ ) by losses of direct waves ( $\delta_2, \delta_3, \delta_4$ ) will allow the parametric amplification to be realized at the threshold pump intensity. In agreement with the condition (9) we can determine threshold value of pump intensity. However even without writing the analytical expression for the threshold pump it can be noticed that this value is a function of nonlinear coupling coefficient dissipation losses as well as the intensity of idler wave. It was

calculated that the threshold intensity of the pump increases with increasing dissipative losses  $\delta_j$  ( $\delta_2 + \delta_3 + \delta_4 = \delta_1$ ). The value of threshold pump intensity also increases with an increase in the input intensity of the idler wave  $I_{20}$ . In addition to these two parameters, the threshold intensity of the pump is influenced by a factor that takes into account the inverse effect of the excited waves on the phase of the pump wave ( $\gamma_{3,4} \neq 0$ ). The higher the nonlinear coupling coefficients, the higher this factor and the greater the threshold pump intensity. Also from expression for the threshold intensity of idler wave we obtain the result of the constant-field approximation:

$$I_{pump}^{CFA} = \frac{\delta_1}{\sqrt{\gamma_1 \gamma_2}}. \text{ The same result is obtained when}$$

there is no idler wave at the entrance to the metamaterial,  $A_2(z=0) = 0$ .

Below are the results of a numerical calculation of the analytical expression (6) obtained in the constant-intensity approximation. In Fig. 1, under phase-matching conditions, the dynamics of the parametric generation of a signal wave when at the input to the medium  $I_1(z=l) = 0$  and of the parametric amplification at  $I_1(z=l) = 0.4 \cdot I_{30}$  are

considered. With parametric generation, the signal wave is excited as three direct waves propagate in the medium as a result of transferring the energy of these waves to the energy of the signal wave generating in the opposite direction. Noticeable amplification of the signal wave is observed at considerable distances (curves 1-8). When condition (8) is satisfied, the losses of the interacting waves are compensated, while the intensity of the signal wave increases (compare solid curves 1 and 2-4). With an increase in the input value of the intensity of the idler wave, the intensity of the signal wave  $I_1(z)$  also increases (compare solid curves 3 and 4). Comparison of solid curves 2 and 3 shows that an increase in losses of 7 times from 5000 cm<sup>-1</sup> (solid curve 3) to 35000 cm<sup>-1</sup> (solid curve 2) leads to a decrease in the intensity of the signal wave, which leads to a settling of the dependence. In the process of parametric amplification, when the input value of the intensity of the signal wave  $I_1(z=l) \neq 0$ , with other equal parameters, the output intensity level  $I_1(z=0)$  is higher (compare the corresponding solid and dashed curves). There is a nonlinear increase in the level of the signal wave with parametric amplification at a rate higher than in the case of generation. The steepness of dashed curves is higher than that of solid curves. These two groups of curves differ in the value of the input intensity  $I_1(z=l)$

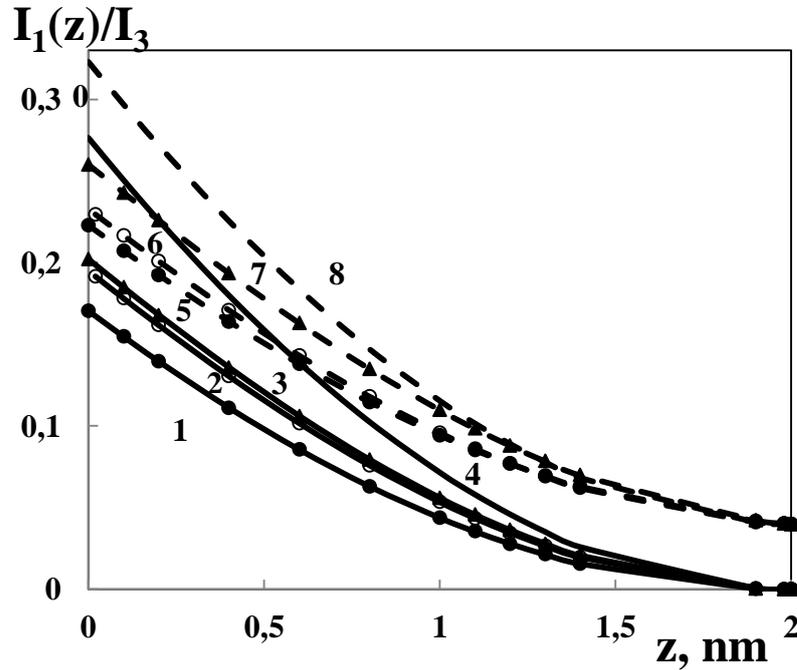


Fig. 1. Relative intensity of the signal wave  $I_1(z)/I_{30}$  as a function of the length of the metamaterial  $z$  at  $l = 20$  nm,  $\Delta = 0$ ,  $I_{30} = I_{40} = 8.848 \cdot 10^{13}$  W / cm<sup>2</sup>,  $I_1(z=l) = 0$  (solid curves 1-4),  $I_1(z=l) = 0.04 \cdot I_{30}$  (dashed curves 5-8),  $I_{20} = 0.3 \cdot I_{30}$  (curves 1-3 and 5-7),  $I_{20} = 0.5 \cdot I_{30}$  (curves 4 and 8),  $\delta_2 = \delta_3 = \delta_4 = 5000$  cm<sup>-1</sup> (curves 3-4 и 7-8), 35000 cm<sup>-1</sup> (curves 1 and 2),  $\delta_1 = \delta_2 + \delta_3 + \delta_4 = 35000$  cm<sup>-1</sup> (curves 1 and 5), 1050000 cm<sup>-1</sup> (curves 2 and 6), 15000 cm<sup>-1</sup> (curves 3-4 and 7-8).

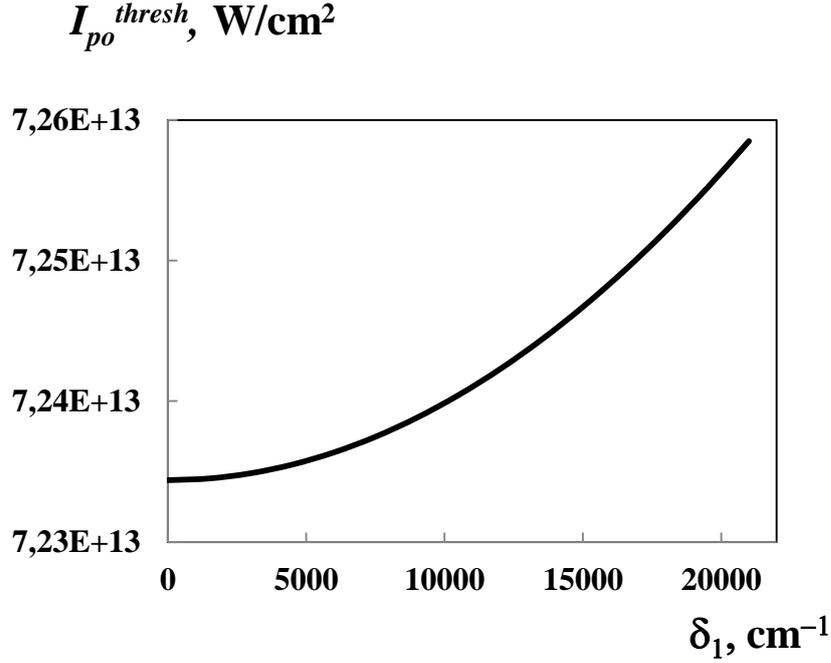


Fig.2. The threshold pump intensity as a loss  $\delta_1$  function, obtained in the constant-intensity approximation, at  $l = 20$  nm,  $\Delta = 0$ ,  $I_{30} = I_{40} = 8.848 \cdot 10^{13}$  W/cm<sup>2</sup>,  $I_{20} = 0.04 \cdot I_{30}$ ,  $\delta_2 = \delta_3 = \delta_4$ ,  $\delta_1 = \delta_2 + \delta_3 + \delta_4$ .

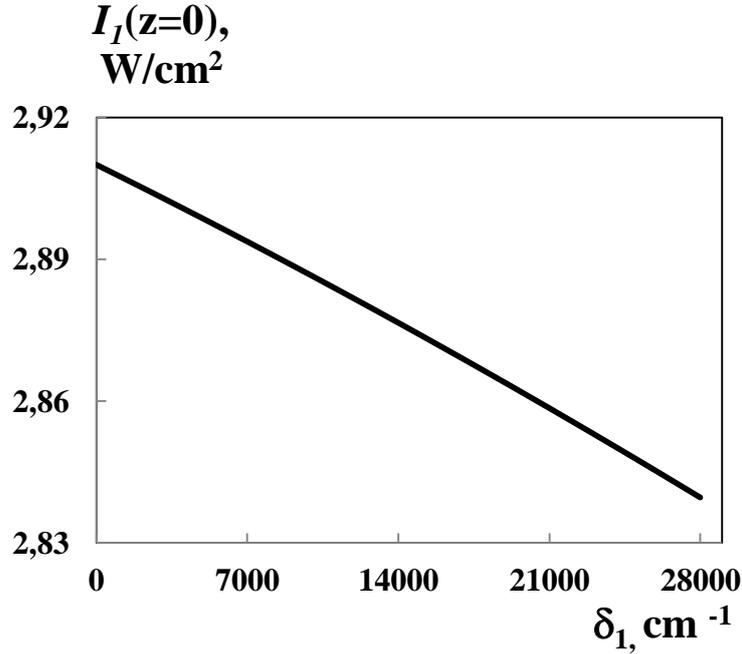


Fig. 3. Intensity of the signal wave at the output of the metamaterial as a loss  $\delta_1$  function, obtained in the constant-intensity approximation, at  $l = 20$  nm,  $\Delta = 0$ ,  $I_{30} = I_{40} = 8.848 \cdot 10^{13}$  W/cm<sup>2</sup>,  $I_{20} = 0.05 \cdot I_{30}$ ,  $\delta_2 = \delta_3 = \delta_4$ ,  $\delta_1 = \delta_2 + \delta_3 + \delta_4$ .

In Fig. 2 shows the dependence of the threshold pump intensity on the loss of interacting waves. The value of the threshold pump intensity increases, as expected, with increasing losses, which agrees with the analytical expression (10) obtained in the constant-

intensity approximation. Analysis (10) shows that as the intensity of the idler wave  $I_{20}$  increases, the threshold pump intensity  $I_{pump}^{thresh}$  necessary to amplify and generate the signal wave also increases.

This is because simultaneously there is an increase in two counter signal and idle waves due to the energy of the pump intensity.

In Fig. 3, under conditions of phase matching, the dependence of the intensity of the signal wave on the output from the metamaterial on the value of losses  $\delta_1$  is given. There is a near-linear decrease in intensity from losses in the case of interaction in the metamaterial, in contrast to the exponential decrease in ordinary materials. At compensation of losses, according to (8), in the expression (6), the intensity dependence on losses is determined by the term  $\frac{\delta_1}{\lambda} A_{II} \cdot \sin \lambda l$ , and also by terms with the parameter  $\lambda$ . The analysis showed that the main contribution to the practically linear dependence on losses,  $I_{pump}^{thresh}(\delta_1)$ , is made by a term that is absent in the case of a conventional material. This term  $(\frac{\delta_1}{\lambda} A_{II} \cdot \sin \lambda l)$  characterizes the dependence on the total length of the metamaterial arising from specific boundary conditions in the metamaterial.

Thus, in metamaterials, the possibility of propagating traveling waves in opposite directions makes it possible to realize a positive feedback between them through parametric interaction in the medium. This is provided by frequency dispersion in the "left-handed" nonlinear medium for the emerging backward wave. The metamaterial plays the role of distributed feedback and can compensate for losses. In this case, the backward wave propagates in a metamaterial with a constant amplitude and perhaps even its amplification. By varying the losses of direct waves, the input intensity of the idler wave and the coefficients of the nonlinear coupling, it is possible to control the

threshold pump intensity. From here it is possible to obtain significant amplification in the metamaterial, as well as parametric generation of the backward wave.

### 3. CONCLUSIONS

In work in the constant-intensity approximation, the threshold generation regime is studied for parametric interaction in a cubic nonlinear medium, which is the "left-handed" one for the signal wave. It is shown that, on the one hand, in the presence of losses in the medium, the amplitude of propagating waves decreases in the direction of energy transfer. On the other hand, the metamaterial, providing a positive feedback, through the nonlinearity of the medium, can compensate for losses at the parametric interaction. In this case, the backward signal wave will propagate with a constant amplitude and possibly even amplification and generation of this wave. An analytical expression is obtained for the amplitude of the pump wave in the case of phase matching in dissipative media. Conditions are determined that allow, in the process of nonlinear interaction of waves, in addition to the constancy of the amplitude of the backward signal wave, to obtain its amplification. It is shown that the threshold pump intensity increases with grown of dissipative losses and of the input intensity of the idler wave. In addition, the threshold pump amplitude is influenced by a factor that takes into account the inverse effect of the excited waves on the phase of the pump wave ( $\gamma_{3,4} \neq 0$ ). The higher the nonlinear coupling coefficients for pump waves, the higher this factor and the greater the threshold pump intensity  $I_{pump}^{thresh}$ . The method developed may be necessary in the development of frequency converters on the basis of nonlinear metamaterials.

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