DOI 10.1007/s11182-018-1572-6

Russian Physics Journal, Vol. 61, No. 9, January, 2019 (Russian Original No. 9, September, 2018)

OPTICS AND SPECTROSCOPY

FOUR-WAVE MIXING IN METAMATERIALS

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Interaction of four counterpropagating waves in a cubic medium being "left" for the signal wave is considered in the constant-intensity approximation. Analytical expression for the signal wave intensity is derived for the general case of four-wave interaction in a metamaterial. The influence of different parameters on the signal wave amplification coefficient, efficiency of conversion into the signal wave, and the reflection coefficient of the mirror whose role is played by the metamaterial is analyzed. It has been obtained for the first time that the determining role in the backward signal wave amplification is played by the total length of the metamaterial and the intensities of all three forward waves. An analysis has shown that the optimal thickness of the metamaterial depends not only on the phase mismatch and the strong coherent pump field intensity, as in the constant-field approximation, but also on the intensity of the weak wave at the frequency ω_2. It has been demonstrated for the first time that the efficiency of frequency conversion in metamaterials depends on the total metamaterial length, input intensities of all four interacting waves, phase mismatch, and losses in the medium. It is established that the maxima of the reflection coefficient of the metamaterial depend on the total metamaterial length and the intensities of all three forward waves.

Keywords: four-wave interaction, metamaterial, negative refraction, constant-intensity approximation.

INTRODUCTION

Various approaches to the development of such perspective artificial structures, as metamaterials have been used [1–4]. The main problems for introduction of these developments remain the big losses inherent in them. In a composite structure, the nonlinear response is determined by the dielectric permittivity of two components: the matrix and the inclusions consisting of the smallest metal elements – conductive rods and rings with gaps [2]. In [5] the composite structures with the third-order dielectric susceptibilities varying in the interval $\chi^{(3)} \sim 10^{-9}$–10^{-8} CGSE units were reported. The four-wave interaction in metamaterials was studied in a number of works, among which [6–8] should be mentioned. According to [8], the four-wave interaction in the medium with a negative refractive index was realized experimentally in a layered metal-dielectric-metal nanostructure. Whereas in the gold metal layer 20 nm thick the generation efficiency during four-wave mixing was 10^{-8}, in the layered metal-dielectric-metal structure the record conversion efficiency of the order of 10^{-6} was obtained. In the constant-intensity approximation [9], the parametrical interaction of optical waves in metamaterials was studied in [10, 11]. A study of the interaction of nonlinear counter-propagating optical waves during four-wave mixing in metamaterials is the purpose of the present work.

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Translated from Izvestiya Vysshikh Uchebnykh Zavedenii, Fizika, No. 9, pp. 10–17, September, 2018. Original article submitted February 19, 2018; revision submitted July 2, 2018.

1. THEORY

In the quasistatic approximation applicable for pulse durations ~10^{-12} s and greater [12], we consider the third-order nonlinear process of interaction of four optical waves with frequencies ω_j ($j = 1-4$) with two strong pump waves of frequencies ω_3 and ω_4. We assume that the medium is "left" only at the signal wave frequency ω_1, that is, the metamaterial medium has simultaneously negative values of the real components of the dielectric permittivity, ε_j, and magnetic permeability, μ_j, at the frequency ω_1 ($\varepsilon_1 < 0$ and $\mu_1 < 0$) and positive values of real components of the dielectric permittivities and magnetic permeabilities at frequencies $\omega_{2,3,4}$ ($\varepsilon_{2,3,4} > 0$, $\mu_{2,3,4} > 0$). The energy fluxes of waves at the frequency ω_2 and of the two pump waves ($\boldsymbol{S}_{2,3,4}$) are normally incident on the left side surface of the metamaterial with thickness ℓ and propagate along the positive direction of the z axis. From here it follows that the energy of the signal wave for which the medium is "left" is transferred in the opposite direction. It is well known that under these conditions, the wave vectors of all four waves $\boldsymbol{k}_{1,2,3,4}$ are oriented along the positive direction of the z axis, that is, their directions coincide with the directions of energy fluxes $\boldsymbol{S}_{2,3,4}$ of three interacting waves. Thus, seven vectors during the four-wave interaction ($\boldsymbol{k}_{1,2,3,4}$ and $\boldsymbol{S}_{2,3,4}$) are opposite to the Pointing vector $\boldsymbol{S}_1$ of the backward signal wave. In accordance with the method used in [13], we consider that the nonlinear response of the medium is mainly caused by the magnetic component of the waves. Then the truncated equations for the examined case of four-wave interaction in the metamaterial will have the form [7] that differs from the case of similar interaction in a traditional medium [6, 12]. In the case under consideration, the interaction in the metamaterial in the first equation of the system is taken into account first, by the fact that the signal wave transfers the energy in the direction opposite to that of other three waves, and second, that the dielectric permittivity at the signal wave frequency takes a negative value:

$$\frac{dA_1}{dz} - \delta_1 \mathrm{A}_1 = -\mathrm{i}\gamma_1 A_3 A_4 A_2^* e^{\mathrm{i}\Delta z}, \quad \frac{dA_2}{dz} + \delta_2 A_2 = \mathrm{i}\gamma_2 A_3 A_4 A_1^* e^{\mathrm{i}\Delta z},$$
$$\frac{dA_3}{dz} + \delta_3 A_3 = \mathrm{i}\gamma_3 A_1 A_2 A_4^* e^{-\mathrm{i}\Delta z}, \quad \frac{dA_4}{dz} + \delta_4 A_4 = \mathrm{i}\gamma_4 A_1 A_2 A_3^* e^{-\mathrm{i}\Delta z}. \tag{1}$$

Here A_j are the complex amplitudes of the magnetic fields of the transmitted quasi-monochromatic waves, δ_j are the absorption coefficients of the medium at the corresponding frequencies ω_j ($j = 1-4$), $\gamma_1 = 2\pi k_1 \chi_1^{(3)} / |\varepsilon_1|$ and $\gamma_{2,3,4} = 2\pi k_{2,3,4} \chi_{2,3,4}^{(3)} / \varepsilon_{2,3,4}$ 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 mismatch of the interacting waves. The corresponding equations for the electric components can be derived analogously with replacement of the dielectric permittivity of the medium ε_j by the magnetic permeability μ_j and *vise versa* [13]. For the chosen scheme of wave interaction, it is generally possible to present the corresponding boundary conditions in the form $A_{2,3,4}(z = 0) = A_{20,30,40}$ and $A_1(z = \ell) = A_{1\ell}$. Here $z = 0$ 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 ω_2 and of the pump waves ($A_{30,40}$), and $A_{1\ell}$ is the initial amplitude of the transmitted signal wave at the right input of the nonlinear medium at $z = \ell$. We note that these initial values of the transmitted waves at the input of the nonlinear medium are related with the incident wave amplitudes through the corresponding reflection and refraction coefficients.

Solving system of equations (1) within the limits of the constant-intensity approximation, that is, when the equality $I_{2,3,4}(z) = I_{2,3,4}(z = 0) = I_{20,30,40}$ is satisfied, and taking into account the boundary conditions, we derive the following expressions for the complex amplitude of the signal wave propagating from the right to the left in the nonlinear medium:

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

where

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

$$\lambda = \sqrt{\gamma_1\gamma_2 I_{30}I_{40} - \gamma_1\gamma_3 I_{20}I_{40} - \gamma_1\gamma_4 I_{20}I_{30} - \left(\sum_1^4 \delta_j - i\Delta\right)^2 / 4},\ I_{j0} = A_{j0}\cdot A_{j0}^*.$$

From the above expression it follows that taking into account the change of the phases of all interacting waves in the constant-intensity approximation leads to the appearance of additional terms under the radicand for the parameter λ in comparison with the result obtained in the constant-field approximation. In addition, the complex signal wave amplitude depends not only on the distance traveled by the wave in the medium, total metamaterial length, phase mismatch, strong pump wave intensities, as in the constant-field approximation, but also on the intensity of the weak wave at the frequency ω_2 through the parameter λ. The effective tunable parametrical frequency converter based on the metamaterial can be provided with the sufficient level of the pump wave and the weak wave at the frequency ω_2. Let us now determine the signal wave amplification coefficient in the metamaterial.

To study the dynamics of variations of the signal wave amplification coefficient $\eta_{\text{ampl}}(z)$ in the process of wave propagation in a dissipative medium under phase matching conditions, that is, at $\Delta = 0$, from Eq. (2) we obtain the following dependence on the spatial coordinate z ($A_{20} = 0$):

$$\eta_{\text{ampl}}(z) = e^{-a_1 z}\left[e^{a_1\ell/2}\left(\cos\lambda_1 z + \frac{a_1}{2\lambda_1}\sin\lambda_1 z\right) - \frac{\delta_1}{\lambda_1}\sin\lambda_1(\ell - z)\right]^2 / \left(\cos\lambda_1\ell + \frac{a_1}{2\lambda_1}\sin\lambda_1\ell\right)^2, \quad (3)$$

where $\lambda_1 = \sqrt{\gamma_1\gamma_2 I_{30}I_{40} - \left(\sum_1^4 \delta_j\right)^2 / 4}$. From Eq. (3) we obtain for amplification coefficient at the output of the metamaterial $\eta_{\text{ampl}} = \left(e^{a_1\ell/2} - \frac{\delta_1}{\lambda_1}\sin\lambda_1\ell\right)^2 / \left(\cos\lambda_1\ell + \frac{a_1}{2\lambda_1}\sin\lambda_1\ell\right)^2$. Neglecting losses, we obtain $\eta_{\text{ampl}} = 1/\cos^2\lambda_1'\ell$ ($\lambda_1' = \sqrt{\gamma_1\gamma_2 I_{30}I_{40}}$), that is, the amplification coefficient in the metamaterial undergoes periodic resonances with maxima at $\eta_{\text{ampl}}(z) = I_1(z)/I_{1\ell}$, $k = 0, 1, 2, \ldots$, whereas in the conventional medium without losses, the exponential increase of the signal wave intensity is observed. For arbitrary phase mismatch, from Eq. (2) we obtain the following dependence for the signal wave amplification coefficient at the output of the metamaterial $(\delta_j = 0)$:

$$\eta_{\text{ampl}} = \frac{I_1(z=0)}{I_{1\ell}} = \left[\left(\cos\frac{\Delta\ell}{2}\cos\lambda_2\ell + \frac{\Delta}{2\lambda_2}\sin\lambda_2\ell\cdot\sin\frac{\Delta\ell}{2} - \frac{\Delta b}{A_{1l}}\frac{\sin^2\lambda_2\ell}{\lambda_2^2}\right)^2\right.$$

$$\left. + \left(\cos\frac{\Delta\ell}{2}\frac{\Delta}{2\lambda_2}\sin\lambda_2\ell - \sin\frac{\Delta\ell}{2}\cos\lambda_2\ell + \frac{b}{A_{1\ell}}\frac{\sin 2\lambda_2\ell}{2\lambda_2}\right)^2\right] \Big/ \left[\cos^2\lambda_2\ell + \left(\frac{\Delta}{2\lambda_2}\right)^2\sin^2\lambda_2\ell\right]^2, \tag{4}$$

where $\lambda_2 = \sqrt{\gamma_1\gamma_2 I_{30} I_{40} - \gamma_1\gamma_3 I_{20} I_{40} - \gamma_1\gamma_4 I_{20} I_{30} + \frac{\Delta^2}{4}}$.

For $A_{20} = 0$ when phase matching conditions are satisfied, we obtain $\eta_{\text{ampl}} = \cos^2\lambda_1' z / \cos^2\lambda_1'\ell$. Before proceeding to an analysis of the expression for the reflection coefficient R of the metamaterial that serves as a nonlinear mirror at close values of the frequencies ω_1 and ω_2, we can easily find the frequency conversion efficiency. The graphical analysis of the given parameter allowed us to track the dynamics of spatial variations of $\eta_1(z) = I_1(z)/I_{20}$ in the metamaterial (Fig. 4). At the left output of the metamaterial ($A_{1\ell} = 0,\ \delta_j = 0$), we obtain for the given parameter $R = \frac{I_1(z=0)}{I_{20}} = \left(\frac{\gamma_1}{\lambda_2}A_{30}A_{40}\sin\lambda_2\ell\right)^2 / \left[\cos^2\lambda_2\ell + \left(\frac{\Delta}{2\lambda_2}\right)^2\sin^2\lambda_2\ell\right]$. To optimize the phase mismatch, we differentiate the given expression with respect to the parameter Δ. As a result, we have $\operatorname{sinc}\lambda_2\ell = \cos\lambda_2\ell$, whose solutions are found by the graphic method. From here we obtain the following values of the parameter: $\lambda_2\ell = 0,\ 3\pi/2,\ 5\pi/2$. To obtain the central maximum of the reflection coefficient R, the optimal phase mismatch Δ, according to the expression for λ_2, is determined by the expression $\Delta_{\text{opt},1} = 2\sqrt{\gamma_1\gamma_3 I_{20} I_{40} + \gamma_1\gamma_4 I_{20} I_{30} - \gamma_1\gamma_2 I_{30} I_{40}}$, and for the subsequent maxima we have $\Delta_{\text{opt},2}\ell = 2\sqrt{4.71^2 - (\gamma_1\gamma_2 I_{30} I_{40} - \gamma_1\gamma_3 I_{20} I_{40} - \gamma_1\gamma_4 I_{20} I_{30})\ell^2}$. As follows from these expressions, $\Delta_{\text{opt}}\ell$ are equal to the corresponding solutions to within the radicands $\lambda_2\ell$ comprising the intensities of the pump waves and of the wave with frequency ω_2. It should be noted that the lateral maxima are observed for strong pump waves without saturated pump field.

2. DISCUSSION OF THE RESULTS OBTAINED

To calculate numerically the above analytical expressions, the problem parameters were chosen from the available experiments: the metamaterial thickness was varied in the range 20–80 nm, the cubic susceptibility of the metamaterial was $\chi_j^{(3)} = 10^{-9} - 1.5\cdot 10^{-10}$ CGSE units, the pump wave intensities were varied in the range 10^8–10^{14} W/cm^2, and the losses in the medium were altered in the range $(1-2)\cdot 10^4$ cm^{-1} [7, 8]. Figure 1 shows values of η_{ampl} at the output of the metamaterial versus the total thickness of the metamaterial ℓ given that the conditions $\Delta = 0$ and $I_{20} = 0$ are satisfied. From a comparison of the curves for $\eta_{\text{ampl}}(\ell)$ for the indicated losses, it can be seen that the steepness of resonant curves is higher for smaller losses in the metamaterial. From a comparison of resonances for ℓ in the range from 40 to 65 nm, it follows that the width of the resonance increases with losses, that is, by analogy with the resonant circuit, the Q-factor of the resonant curve is deteriorated. A qualitatively analogous character of the curves is also observed in the constant-field approximation.

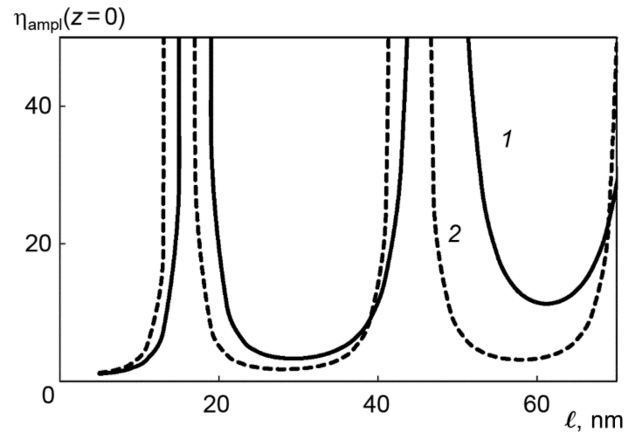

Fig. 1. Dependences of the signal wave amplification coefficient $\eta_{ampl} = I_1(z=0)/I_{1\ell}$ on the total metamaterial thickness ℓ under conditions $\Delta = 0$ and $I_{20} = 0$ for losses in the medium $\delta_j = 2 \cdot 10^4$ cm^{-1} (solid curve *1*) and 10^4 cm^{-1} (dashed curve *2*).

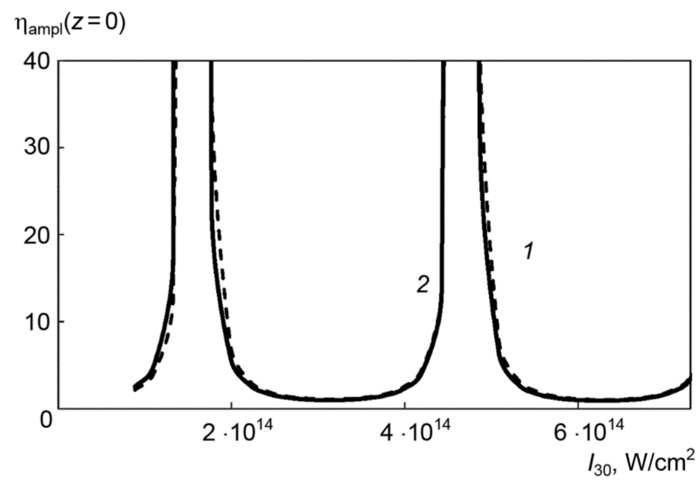

Fig. 2. Dependences of the signal wave amplification coefficient $\eta_{ampl} = I_1(z=0)/I_{1\ell}$ on the pump wave intensity I_{30} under phase matching conditions and $I_{20} = 0$ for losses in the medium $\delta_j = 3 \cdot 10^4$ cm^{-1} (dashed curve *1*) and 10^3 cm^{-1} (solid curve *2*).

Figure 2 shows curves for η_{ampl} versus the strong coherent pump field intensity under conditions of phase synchronism in the dissipative medium. The resonances are observed at the examined values of the pump field intensity. From here the driving field intensity corresponding to a resonant growth of the amplification coefficient for the given parameters of the problem can be calculated. The dependences obtained allow us to conclude that the observed resonances of the amplification coefficient can be regulated by adjusting the problem parameters, in particular, the losses of the coherent pump field. In addition, the change in the total plate thickness and in the coherent pump field

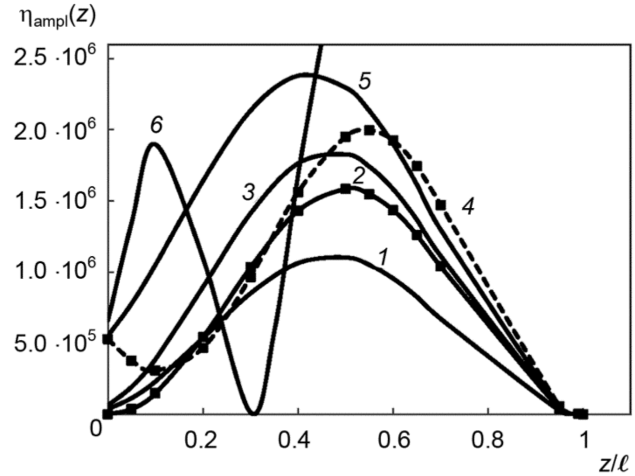

Fig. 3. Dynamics of the amplification coefficient $\eta_{\text{ampl}}(z) = I_1(z) / I_{1\ell}$ during wave propagation in the medium at $I_{1\ell} = 10^{-7} I_{30}$, total metamaterial thickness $\ell =$ 20 nm (curves *1–5*) and 60 nm (curve *6*), $\Delta\ell / 2 =$ 2.5 (curves *1* and *5*), 2.8 (curve *3*), 3 (curves *2* and *4*), and 8.4 (curve *6*), and $I_{20} = 0.3 \cdot I_{30}$ (curve *1*) and $0.5 \cdot I_{30}$ (curves *2–6*). Here solid curves *1–3* and *5–6* were calculated in the constant-intensity approximation, and dashed curve *4* was calculated in the constant-field approximation.

intensity leads to the corresponding change of the signal wave amplification coefficient and hence, to the variation of the wave intensity distribution versus the coordinate z in the metamaterial. As a result, the output η_{ampl} value also changes (Fig. 2). As demonstrated our analysis, the observable resonances testify to the amplification exceeding the losses, which can cause signal wave generation without optical resonator. As can be seen from the dependences obtained, the observable resonances are narrow, that is, the losses are large outside of resonances. From here it follows that under conditions of phase matching a stable signal at the signal wave frequency is obtained for the constant intensities of the fundamental coherent pump waves and the total metamaterial thickness.

Figure 3 illustrates the dynamics of the amplification coefficient $\eta_{\text{ampl}}(z)$ in the process of wave propagation in the nonlinear medium. The amplitude of $\eta_{\text{ampl}}(z)$ oscillations decreases with increasing phase mismatch (compare curves *2*, *3*, and *5*). Here the dashed curves show the results of calculations in the constant-field approximation (curve *4*). The different dependences (curves *2* and *4*) in both approximations are explained by nonzero nonlinear coefficients γ_3 and γ_4 in the constant-intensity approximation that considers the reverse reaction of the pump wave on the signal wave. Our analysis demonstrates that it is possible to amplify significantly the backward signal wave by varying the input intensities of the three forward waves. The conversion efficiency (compare curves *1* and *2*) and the displacement of the oscillation maxima and minima in the constant-intensity approximation when $\gamma_3, \gamma_4 \neq 0$ increase with the input intensity of the weak wave at the frequency ω_2. The distance between two neighboring minima as well as the oscillation period can easily be determined. The displacement is not observed in the constant-field approximation when the parameters $\gamma_3, \gamma_4 = 0$; in this case, the maxima and minima of the corresponding curves coincide. The maximum of the conversion efficiency increased by a factor of 1.43 as the input intensity of the wave at the frequency ω_2 increased from 0.3 I_{30} to 0.5 I_{30}.

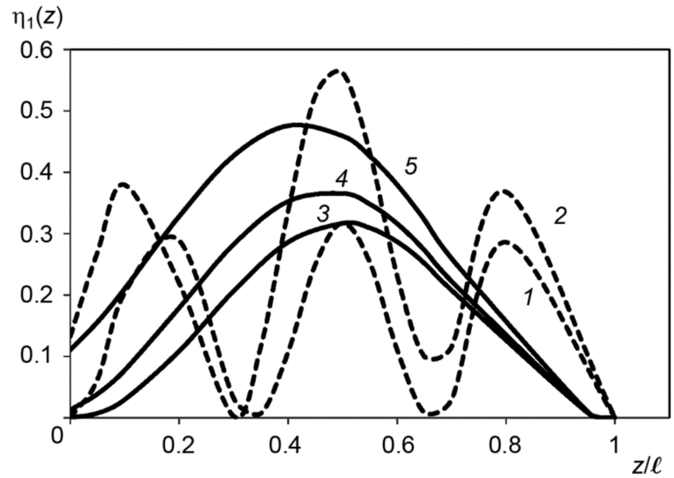

Fig. 4. Dynamics of the frequency-conversion efficiency $\eta_1(z) = I_1(z) / I_{20}$ during wave propagation in the medium at $I_{1\ell} = 10^{-7} I_{30}$, $I_{20} = 0.5 \cdot I_{30}$, total metamaterial thickness $\ell =$ 20 nm (solid curves *3–5*) and 60 nm (dashed curves *1–2*), $\Delta\ell / 2 =$ 2.5 (solid curve *5*), 2.8 (solid curve *4*), 3 (solid curve *3*), 8.4 (dashed curve *2*), and 9 (dashed curve *1*).

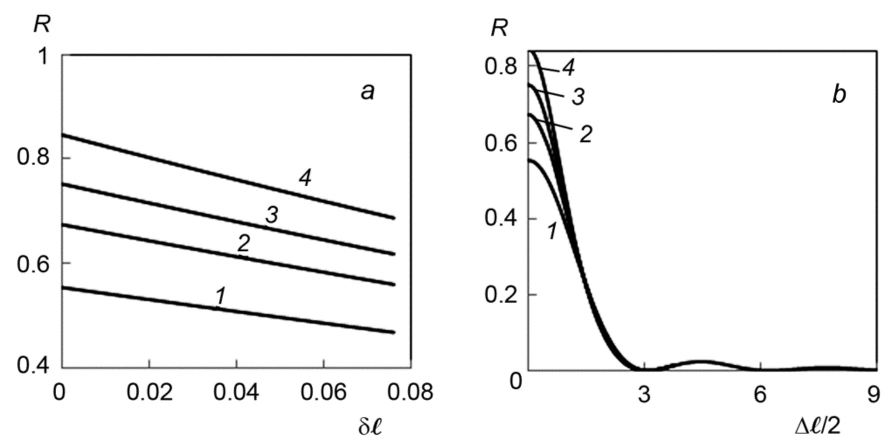

Fig. 5. Dependences of the reflection coefficient $R = I_{1\ell} / I_{20}$ on the linear losses $\delta\ell$ in the nonlinear medium at $\Delta = 0$ (*a*) and on the phase mismatch $\Delta\ell / 2$ at $\delta_j = 0$ (*b*) for $\ell = 20$ nm, $I_{20} = 0.5 \cdot I_{30}$ (curve *1*), $0.3 \cdot I_{30}$ (curve *2*), $0.2 \cdot I_{30}$ (curve *3*), and $0.1 \cdot I_{30}$ (curve *4*).

Figure 4 shows the dependence of the frequency-conversion efficiency $\eta_1(z) = I_1(z)/I_{20}$. Here oscillations of the efficiency of conversion into the signal wave are seen. A comparison of curves *1*, *2* and *3*, *4*, and *5* demonstrates that the increase in the reduced phase mismatch $\Delta\ell/2$ leads to the increase of the oscillation frequency.

From Fig. 5*a* it can be seen that a decrease in the signal wave intensity at the left output leads to the decrease of the reflection coefficient R at $z = 0$. It should be taken into account that the observable dependence of *R* on the losses is nonlinear, since in addition to the exponential dependence on the losses, terms with losses arise under the radicand for the parameter λ_1 (see Eq. (3)). Figure 5*b* shows that the maxima of the reflection coefficient R are observed at the output of the metamaterial for the phase mismatch values being solutions of the equation derived above. The maxima of the reflection coefficient R are found at theoretically calculated values of Δ_{opt}. The main maximum is located in the vicinity of $\Delta = 0$ at the distance determined by the terms under the radicand depending on the intensities of the pump waves and the wave at the frequency ω_2. Thus, the increase of the signal wave amplification coefficient depends primarily on the ratio of the intensities of the wave at the frequency ω_2 and the pump waves at the input of the metamaterial, that is, on $I_{20} / I_{30,40}$.

It should be noted that the main conditions of observation of lateral maxima are the strong pump fields, that is, non-exhausted pumping during nonlinear four-wave interaction. The efficiency of energy conversion of the strong wave at one frequency into the energy of the wave at another frequency can be controlled by changing the total metamaterial length, the intensities of all three interacting forward waves, as well as the input intensity of the signal wave, phase mismatch, and losses in the medium. Based on the experimental data presented in [8] on four-wave parametrical interaction in the gold-based metamaterial, we numerically estimated the expected efficiency of frequency conversion of the pump wave energy into the signal wave energy. Results of the corresponding calculations for $\eta = I_1 / I_{30}$ at the pump wave intensity $I_{30} = 5 \cdot 10^7$ W/cm^2 yield $\eta \sim 10^{-7}$–10^{-8} for the phase mismatch $\Delta\ell / 2 = 3.5$ and 10^{-6}–10^{-7} for $\Delta\ell / 2 = 1.68$. Analogous conversion efficiency was experimentally obtained in [8]. The increase of the pump wave intensity by an order of magnitude causes the conversion efficiency to increase. From here it follows that a more intensive signal wave at the output of the metamaterial can be realized by choosing higher levels of the driving pump wave and of the forward weak wave at the input of the medium as well as of the optimal values of the phase mismatch and metamaterial thickness.

CONCLUSIONS

In this work, the parametrical interaction in the cubic nonlinear medium being “left” for the signal wave has been investigated. Analytical expressions for the signal wave amplitude were obtained both for the conditions of phase matching in dissipative media and of the phase mismatch in non-dissipative media. For the first time it has been obtained that the determining role in the signal wave amplification in metamaterials is played by the following factors: total metamaterial thickness and the intensities of all three forward waves, optimal metamaterial thickness that depends not only on the phase mismatch and the high coherent pump field intensity, as in the case of analysis in the constant-field approximation, but also on the intensity of the weak wave at the frequency ω_2. The efficiency of strong wave energy conversion at one frequency into the energy of the wave at another frequency is directly proportional to the total metamaterial thickness, input intensities of all four interacting waves, and the phase mismatch and losses in the medium. The maxima of the reflection coefficient of the metamaterial depend on the total metamaterial length and the intensities of all three forward waves. The suggested method can be used to develop frequency converters based on nonlinear metamaterials.

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