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ABSTRACT

of the dissertation for the degree of Doctor of Philosophy

NONLINEAR FREQUENCY CONVERSION OF LASER RADIATION IN METAMATERIALS AND MID-IR RANGE CHALCOGENIDE CRYSTALS

Speciality: 2211.01 – Solid state physics

Field of science: Physics

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Baku – 2021

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GENERAL CHARACTERISTICS OF WORK

Relevance and currency of the research topic. The modern development of photonics is associated with the technology of developing metamaterials. The successes achieved in the creation of these media with a negative refractive index in the super high frequency and optical ranges of the spectrum have revived interest in the processes of parametric interaction of meeting waves.

In optical information processing systems, photons are used as information carriers. However, this raises the problem of media management. The discovery of metamaterials made it possible to control light radiation by changing the optical properties of such artificial structures using nonlinear optics methods. As is known, a metamaterial is a composite material forming a dielectric matrix with inclusions providing 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. As a result, in such an inhomogeneous metamaterial medium, abrupt changes in the material parameters of the medium (dielectric ε and magnetic μ permeability) and a negative value of the refractive index in a certain frequency range are observed. As a consequence, in such a medium an electromagnetic wave at the frequency of negative refraction propagates with multidirectional phase and group velocities. A negative value of the phase velocity means that during propagation, the phase incursion of the wave occurs in the direction from the receiver to the source, while the energy transfer, in turn, occurs from the source to the receiver. With this geometry, for the signal wave, as it follows, the signal wave is a backward wave. Hence, the incident electromagnetic wave in the metamaterial undergoes unusual changes, which leads to new unconventional effects.

Recently, the most important directions in the development of laser technology are the elaboration and research of tunable radiation sources, in combination with frequency mixing effects, having extremely short pulse durations, which makes it possible to significantly expand the range of tunable laser radiation wavelengths. Such sources of the mid-infrared (IR) region of the spectrum are an indispensable tool for studying semiconductor elements in optical communication systems, the main elements of modern LIDARs, in particular for studying the Earth and the atmosphere, scientific and medical devices, are successfully used for spectroscopy problems. However, to accomplish these tasks, coherent radiation sources with the required energy parameters and efficiency are required.

However, the crystals used for these purposes have a number of disadvantages; for example, $CdGeAs_2$ crystals operating at low temperatures have high losses at the second harmonic wavelengths, while $ZnGeP_2$ and Te crystals have losses at the wavelengths of the fundamental radiation of a carbon dioxide laser. Tl_3AsSe_3 and $AgGaSe_2$ crystals have poor thermal properties, and the GaSe crystal has low mechanical properties This, in particular, leads to the problems of creating efficient and reliable harmonic generators of CO_2 lasers.

For this reason, the search for promising materials for solving the problems of modern nonlinear optics is still ongoing.

Optical nonlinear IR crystals, such as $ZnGeP_2$, $AgGaSe_2$, $AgGaS_2$, GaSe ($Hg_{1-x}Cd_xGa_2S_4$, GaS_xSe_{1-x}, $AgGa_{1-x}In_xSe$ and $AgGa(Se_{1-x}S_x)_2$) have acquired great interest for solving the above problems in the mid and far infrared region, due to their unique properties. As is known, the listed IR crystals have a large effective optical nonlinearity, which attracted the attention of researchers.

The current trend towards miniaturization of devices is accompanied by the technological development of nanoscale structures. This promoted the use of thin dielectric and semiconductor films instead of bulk materials. The study of semiconductors and dielectrics by the harmonic generation method provides information on the morphology and structure of these compounds.

The process of generating higher harmonics is attractive for solving a number of applied problems in the ultraviolet and extreme ultraviolet ranges of the spectrum, for obtaining high-energy photons. The main advantage of third harmonic generation (THG) is that, due to high frequencies, it exhibits purely coherent electronic nonlinearity and is not sensitive to thermal response.

In view of all the above, the study of nonlinear optical processes and the identification of the reasons that impede the increase in the efficiency of frequency conversion can be regarded as a very important and urgent problem.

Research objects and subjects. The object of the study is solid solutions of $AgGa_{1-x}In_xSe$, $AgGa(Se_{1-x}S_x)_2$, $Hg_{1-x}Cd_xGa_2S_4$ and GaS_xSe_{1-x} crystals, metamaterial and nanocomposite films of ZnO/PMMA. As a subject of research, we studied the phase effects occuring in the studied chalcogenide crystals, metamaterials, and nanocomposite films.

Aims and purpose of the research: The purpose of the dissertation is a theoretical study of the nonlinear optical conversion of laser radiation in metamaterials, chalcogenide crystals of the IR range and nanocomposite ZnO/PMMA films, as well as the search for ways to increase the conversion efficiency to obtain optimal values of the problem parameters (length of a nonlinear medium, phase mismatch, pump wave intensities and the idler wave in the parametric transformation).

To achieve this goal, the following tasks were set, developed in the constant intensity approximation (CIA):

1. Toa nalyze the phase changes of interacting waves during second harmonic generation (SHG) and THG in a ZnO/PMMA nanocomposite film. To carry out a theoretical analysis of the effect of the size and concentration of ZnO nanoparticles on the quadratic and cubic nonlinearities of ZnO/PMMA nanocomposite films. Comparison of theoretical and experimental results for ZnO/PMMA films with different concentrations of ZnO nanoparticles.

2. To theoretically investigate the effects of three-wave interaction during THG in a metamaterial. Investigate the effect of self-action of a light wave in a metamaterial and compare the results with a similar effect that occurs in a homogeneous cubic medium.

3. To study the meeting parametric interaction of optical waves in a metamaterial at low-frequency and high-frequency pumps in the CIA.

4. Theoretically investigate optical parametric effects in chalcogenide crystals $Hg_{1-x}Cd_xGa_2S_4$ and GaS_xSe_{1-x} with quadratic nonlinearities taking into account phase changes of all interacting waves. To compare the obtained results on the conversion efficiency with the experimentally measured values. Provide recommendations for improving the efficiency of frequency conversion.

5. Theoretically investigate the second harmonic of laser radiation in chalcogenide crystals $AgGa_{0.6}In_{0.4}Se_2$ and $AgGa(Se_{1-x}S_x)_2$ IR ranges in the conditions of existing experiments. Show ways to improve conversion efficiency.

Research methods. The constant intensity approximation was used as a theoretical research method in the dissertation work. In this approximation, nonlinear processes can be considered more accurately, taking into account the phase changes of all interacting waves.

Basic provisions for defense.

1. For the selected laser pump intensity, the coherent length of the ZnO/PMMA can be theoretically calculated. At high concentrations of ZnO nanoparticles, the films generate a stronger signal of the second and third harmonics due to the longer interaction length of the nonlinear medium, but on the other hand, the effective quadratic and cubic nonlinear susceptibilities decrease.

2. In the case of THG in a metamaterial, taking into account the phase changes of interacting waves leads to a threefold decrease in the efficiency of conversion to the third harmonic from 0.91 to 0.3 and to a change in the parameters of the phase matching curve depending on the pump intensity.

3. The propagation of waves in a metamaterial with cubic nonlinearity is accompanied by a self-action effect. With an increase in the phase mismatch, the absolute change in the phase of the fundamental radiation in the case of a metamaterial is higher than in an ordinary quadratic medium.

4. Parametric interaction of waves in a metamaterial with low-frequency and high-frequency pumps proceeds most effectively under optimal interaction conditions. At the same time,

compensation of the losses of the backward signal wave by the losses of direct waves is realized.

5. By choosing the optimal values for the length of the nonlinear medium 0.3 cm, pump intensity 4 MW/cm^2, phase mismatching 0.06 cm^{-1} and taking into account the influence of linear losses in the medium, it is possible to increase the conversion efficiency to the second harmonic to 0.5 in the $AgGa(Se_{0.8}S_{0.2})_2$ crystal and select the conditions for fulfilling or increasing the degree of noncriticality of crystals to the condition of angular phase matching.

6. In $Hg_{1-x}Cd_xGa_2S_4$ and GaS_xSe_{1-x} crystals, with an increase in the pump intensity and a decrease in phase mismatching and losses, parametric amplification of the idler wave takes place, and it is possible to select conditions for increasing the degree of noncritical angular phase matching.

7. It is possible to obtain nanocomposite thin films of ZnO/PMMA by electrochemical deposition.

Scientific innovations.

The scientific novelty of the work is determined by the fact that it has further developed the analytical method - CIA and in this approximation for the first time:

1. It has been analytically shown in PZI that the effective cubic nonlinearity in ZnO/PMMA nanocomposites decreases with an increase in the concentration of ZnO nanoparticles, while on the other hand, due to the long nonlinear interaction length, the nanocrystal generates a strong third harmonic wave. (This result was included in the “Annual scientific report of the National Academy of Sciences of Azerbaijan on important scientific results for 2015”, 2016, third issue). The same can be said about the quadratic susceptibility in ZnO/PMMA nanocomposites and about the appearance of a strong second harmonic wave.

2. The theory of THG and the effect of self-action of a light wave in metamaterials is developed, taking into account the change in the phases of all interacting waves.

3. The intensity of the third harmonic wave in metamaterials is theoretically investigated for various parameters of the problem

under consideration (reduced length of the metamaterial, phase mismatch at large and small values, intensity of the pump wave, total length of the metamaterial).

4. Phase effects have been studied for three-wave parametric interaction with low and high frequencies pumping in metamaterials in the case of negative refraction at the frequency of the signal wave.

5. The parametric interaction of optical pump waves, signal and idler, is considered taking into account the change in their phases in chalcogenide crystals in the mid-IR spectral range. The conditions for the producing of efficient parametric light generators based on $Hg_{1-x}Cd_xGa_2S_4$ and GaS_xSe_{1-x} crystals, where Nd: YAG lasers can be used as a pump source, have been determined.

6. The SHG process is analyzed in the case of nonlinear interaction of waves in chalcogenide crystals of a solid solution in the mid-IR range of the spectrum. The applied analytical method makes it possible to calculate the optimal parameters of both the converter crystal and the radiation source for the implementation of noncritical phase matching in a particular experiment.

Theoretical and practical significance of the research. The practical value of the work is:

• For the development of effective parametric frequency converters, frequency doublers and triplers, specific conversion parameters (pump wave intensity, phase mismatch, converter crystal length, total metamaterial length and idler wave intensity in the case of a parametric converter) obtained in the CIA are given.

• The "method of generation of the second and third harmonics" has been developed in the CIA for the study of ZnO/PMMA nanocomposite films.

• Crystals of the mid-IR range $Hg_{1-x}Cd_xGa_2S_4$, GaS_xSe_{1-x} $AgGa_{x-1}In_xSe$ and $AgGa(Se_{1-x}S_x)_2$ have been studied in detail to determine the conditions for a regime that is not critical to the phase matching angle.

• The method of electrochemical deposition of thin films using ZnO as an example has been developed and mastered with the aim of further investigating the nonlinear optical properties of the obtained experimental samples.

The developed CIA method, in addition to problems of nonlinear optics, can be applied in radio physics, nonlinear acoustics, interferometry, and plasma physics.

The results of the developed approach, the analysis of nonlinear wave interactions of light waves, make it possible to optimize the conversion process. Knowledge of the efficiency of frequency conversion of a nonlinear process makes it possible to find out the most favorable operating parameters and operating conditions of the optical system, thereby improving the parameters of this system.

Approbation and application. The results of the research included in the dissertation work were discussed and published at the following international and republican conferences: Materials digest of the X International Scientific and Practical Conference “Trends of modern science 2014”, Sheffield, May30-7June, 2014; Workshop, Tbilisi, 1-2 June 2015; Materials of 8th International Scientific Conference “Science and Society”, London, November, 2015; "Fizikanın aktual problemləri" VIII Respublika elmi konfransı, Bakı, 17 dekabr, 2015; Materials of the XI International Scientific and practical Conference “Conduct of modern science-2015” England,7 December, 2015; Materials digest of the IX International Conference "Science, Technology and Higher Education", Canada, December 23-24, 2015; “Opto, nanoelektronika, kondensə olunmuş mühit və yüksək enerjilər fizikası” Beynəlxalq konfrans, Bakı, 25-26 dekabr, 2015; Eastern-European Scientific Journal, Dusseldorf, Ausgabe 5 2016; "Fizikanın aktual problemləri" IX Respublika elmi konfransı, Bakı, 22 dekabr, 2016; “XƏZƏRNEFTQAZYATAQ”-2016 elmi-təcrübi konfrans, Bakı, 24-25 dekabr 2016; “Modern Trends in Physics” İnternational Conference, Bakı, 20-22 April 2017; "Fizika və asrtonomiya problemləri” beynəlxalq elmi konfransı. Bakı, 24-25 may, 2018.

The materials of the dissertation were published 22 scientific works, including 11 articles and 11 theses in local and foreign journals. Of these, 3 articles and 1 conference material were published in journals included in the Clarivate Analytics (Web of Science) database, 3 in the Scopus database.

Name of the organization where the dissertation work is executed. The dissertation work was accomplished at the departments of "Optics and Molecular Physics" and "Physical Electronics" of Baku State University.

Structure, volume and main content of dissertation work. Dissertation work is posted on 192 pages as a whole. It consists of an introduction, including 64 figures, 10 tables, 4 chapters, a conclusion, practical recommendations, references include 38 works in Russian, 140 works in English and 3 works in Azerbaijani. The volume of the dissertation (with the exception of gaps and pictures in the text, tables, graphs, appendices and list of reference) - 210819 characters (introduction - 23212, Chapter I - 40843, Chapter II - 38596, Chapter III - 29281, Chapter IV - 76844, result - 2043 characters).

CONTENT OF THE DISSERTATION WORK

In the introduction, the relevance of the topic is substantiated, the purpose of the work, scientific novelty, the main scientific provisions presented for defense are indicated, practical significance is given, a brief summary of the chapters of the dissertation is given separately.

In the first chapter of the dissertation work, first of all, a detailed description of domestic and foreign scientific literature devoted to modern problems encountered by researchers in the study of nonlinear optical effects in metamaterials, as well as in nonlinear chalcogenide crystals is given. The results of the work of the world's leading research centers in this field are analyzed.

The second chapter is devoted to the study of THG in nanocomposites and metamaterials in CIA. Within the framework of the used approximation, the results of a theoretical study are presented, with the help of which it is possible to more accurately describe the physical features of a nonlinear process in comparison with the widely constant field approximation (CFA).

In the investigated case of negative values of the permittivity and permeability at the pumping frequency ω_1 and positive values of the permittivity and permeability at the harmonic frequency ω_3, we consider the standard system of reduced equations under the following boundary conditions:

$$A_1(z=\ell)=A_{1\ell}\exp(i\varphi_{1\ell}),\quad A_3(z=0)=0\,, \tag{1}$$

here $A_{1,3}$- complex amplitudes of the pump wave and the third harmonic at frequencies $\omega_{1,3}$, respectively. We assume that the waves propagate along the z axis. Here $z=\ell$ corresponds to the entrance to the metamaterial on the right side for the pump wave (in the negative direction of the z axis), $\varphi_{1\ell}$ - is the initial phase of the pump wave at the entrance to the nonlinear medium.

For the complex amplitude of the harmonic wave in the CIA at the output of the nonlinear medium, we obtain

$$A_3(z)=i\gamma_3 A_{1\ell}^3\frac{\sin\lambda' z}{\lambda'\cos\lambda'\ell-\frac{i\Delta}{2}\sin\lambda'\ell}e^{-\frac{i\Delta}{2}(z+l)+i3\varphi_{1\ell}} \tag{2}$$

$$\lambda'^2=\frac{\Delta^2}{4}-3\Gamma^2\,,\qquad \Gamma^2=\gamma_1\gamma_3 I_{1\ell}^2\,,$$

here $\gamma_{1,3}$- nonlinear coupling coefficients, $\Delta=k_3-3k_1$- is the phase mismatch between the interacting waves, $k_{1,3}$ ($k_{1,3}>0$) - modules of wave vectors directed along the positive z axis.

The obtained analytical expression for the frequency conversion in the metamaterial (2), calculated within the boundary condition (1), directly depends on the total length of the metamaterial, which is why it differs from a similar analytical expression for frequency conversion in conventional materials.

According to the expression obtained for the amplitude of the harmonic wave (2), it can be seen that the amplitude depends on the factor that takes into account the reverse effect of the excited harmonic wave on the pump wave. In contrast to the CIA, the phase of the harmonic wave depends on the intensity of the pump wave. The analysis showed that the metamaterial plays the role of a mirror, namely, the excited harmonic radiation is directed towards the exciting pump wave. From a comparison of the curves, figure 1, of the intensities of the pump and third harmonic in the CIA and CFA, it was found that taking into account the inverse reaction of the excited harmonic wave to the fundamental radiation wave leads to a decrease in the reduced intensities of the pump wave by 2 and of the third-harmonic wave by 3 times.

This chapter also studies the effect of the self-action of a light wave in a metamaterial and compares it with a similar effect occurring in a conventional cubic medium (see figure 2).

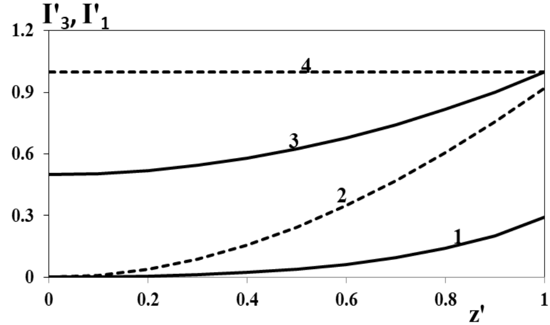

Figure 1. Dependences of the reduced intensities of the pump wave (curves 3 and 4) and the third harmonic (curves 1 and 2) on the reduced length of the metamaterial, calculated in the CIA (curves 1 and 3) and CFA (dashed lines 2 4).

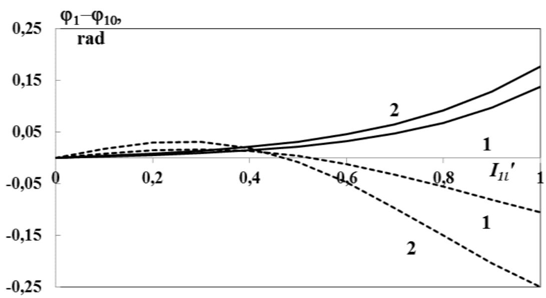

Figure 2. Dependences of the phase shift of the pump wave phase on the intensity of the fundamental radiation in a metamaterial (solid curves 1 and 2) and in a homogeneous medium (dashed curves 1 and 2) at $\Gamma\ell = 1$ for the reduced phase mismatch Δ = 0.5 (solid and dashed curves 1) and 1 (solid and dashed curves 2).

Next, the THG process in a ZnO/PMMA nanocomposite film was considered. Since THG occurs in any materials, an analytical calculation was carried out for each layer of the film and substrate. Sequential calculations of the nonlinear interaction in the CIA from layer to layer in the structure were carried out, similar to that carried out in the CFA in work[1]. Two expressions for the complex amplitude of the third harmonic in the film and film + substrate layers are obtained.

From the expressions obtained for the efficiency of frequency conversion to a third harmonic wave ($\eta_3(\ell_2) = I_3(\ell_2)/I_{10}$) at the output of the nanocomposite structure , we have

$$\eta_3(\ell_2) = \left(t_{sa}^{3\omega}\right)^2 \left(t_{fs}^{3\omega}\right)^2 \eta_3(\ell_1)\left[\left(\cos\lambda_2\ell_2 + c_1\frac{\sin\lambda_2\ell_2}{\lambda_2}\right)^2 + b^2\frac{\sin^2\lambda_2\ell_2}{\lambda_2^2}\right]\exp(-2\delta_3\ell_2), \qquad (3),$$

here $c_1 = \frac{\gamma_3^{sut}}{\gamma_3^f}\lambda_1 ctg\lambda_1\ell_1$, $b = \left(\frac{\gamma_3^{sut}}{\gamma_3^f}\Delta_f - \Delta_{sub}\right)\Big/2$, γ_j^f - the nonlinear coupling coefficients of the waves, δ_j^f are the absorption coefficients of the medium for the ZnO/PMMA film, at the corresponding frequencies (j = 1,3), $t_{fs,sa}^{3\omega}$ are the transmission coefficients according to the Fresnel formula at the film-substrate and substrate-air interface for the third harmonic wave, $\eta_3(\ell_1) = I_3(\ell_1)/I_{10}$ the conversion efficiency signal to the third harmonic at the output of the first layer. According to expression (3), obtained for the conversion efficiency, the dynamic process of frequency conversion to the third harmonic in ZnO/PMMA structures is analyzed.

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[1]Tagiev, Z. H., Kasumova, R.J. Safarova, G.A. Third-harmonic generation in regular domain structures // Journal of Russian Laser Research, − 2010. 31 №4, − p.319-331.

It was found that at higher ZnO concentrations, due to the longer interaction length of the nonlinear medium, the films generate a stronger third harmonic signal (figure 3).

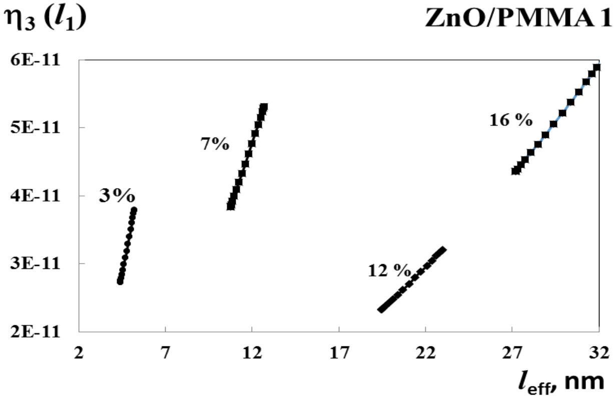

Figure 3. Dependences of conversion efficiency of the pump wave to wave of third harmonic on the effective length at a pump intensity of 20 GW/cm^2 for $\delta_3 = 3\delta_1$ = 0.06 · 10^{4}cm^{-1} (curves 1 and 3), 0.076 · 10^{4}cm^{-1} (curves 2 and 4) and 0.13 · 10^{4}cm^{-1} (curves 5 and 6).

The third chapter of the dissertation is devoted to parametric processes of nonlinear interaction of optical waves, due to induced nonlinear polarization in metamaterials, as a result of which the parameters of the nonlinear medium change.

In this chapter, the parametric interaction of waves in the quadratic medium of a metamaterial is discussed, taking into account phase effects at low-frequency and high-frequency pumps, where the frequencies of the pump wave and the idler waves lie in the region of positive values of the refractive index, and the signal wave - in the region of negative values.

The boundary conditions corresponding to this geometry of the considered waves can be represented in the form:

$$A_{2,3}(z=0) = A_{20,30}\exp(i\varphi_{20,30}),\ A_1(z=\ell) = A_{1\ell}\exp(i\varphi_{1\ell}), \qquad (4)$$

Here z=0 corresponds to an input in a metamaterial from the left side, $A_{20,30}$, $\varphi_{20,30}$ are the initial amplitudes and phases of idler and pump waves at the input from the left side of the nonlinear medium, $A_{1\ell}$, $\varphi_{1\ell}$ are the initial amplitude and phase of signal wave at the input from the right side of the nonlinear medium ($z=\ell$).

Taking into account the boundary conditions (4), we obtain expressions for the complex amplitudes of the backward wave at low frequency and high frequency pumps. Next, two important parameters are considered that determine the conversion efficiency and the dynamics of the amplification process in the metamaterial with low-frequency and high-frequency pumps.

The efficiency of the frequency conversion of the signal wave in the case of low frequency pumping has the form

$$\eta_1(z=0)=\frac{\Gamma_3^2\sin^2\lambda\ell}{\lambda^2\cos^2\lambda\ell+\frac{\Delta^2}{4}\sin^2\lambda\ell}, \qquad (5)$$

where $\lambda=\sqrt{\frac{\Delta^2}{4}-\Gamma_3^2-\Gamma_2^2}$, in the case of high frequency pumping

$$\eta_1=\frac{\Gamma_3^2\sin^2\lambda\ell}{\left(\frac{\Delta}{2}\right)^2\sin^2\lambda\ell+\lambda^2\cos^2\lambda\ell}, \qquad (6)$$

where $\lambda=\sqrt{\frac{\Delta^2}{4}+\Gamma_3^2-\Gamma_2^2}$ $\quad \Gamma_3^2=\gamma_1\gamma_2 I_{30}, \quad \Gamma_2^2=\gamma_1\gamma_3 I_{20}, \quad I_j=A_j\cdot A_j^*.$

The signal wave amplification coefficient in the case of low-frequency pumping is determined by the expression:

$$\eta_{yc}=\frac{1}{\cos^2\lambda\ell+\left(\frac{\Delta}{2\lambda}\right)^2\sin^2\lambda\ell}, \qquad (7)$$

for high-frequency pumping, the expression for the amplification coefficient of the signal wave is:

$$\eta_{yc} = \left| \frac{\sin \lambda z + ib \cos \lambda z}{\sin \lambda \ell + ib \cos \lambda \ell} \right|^2 , \qquad (8)$$

here $b = \frac{2\lambda}{\Delta}$.

On the basis of analytical expressions for the amplification coefficient and conversion efficiency in the metamaterial under low-frequency pumping, graphs of dependences are constructed that give a clear picture of the interaction.

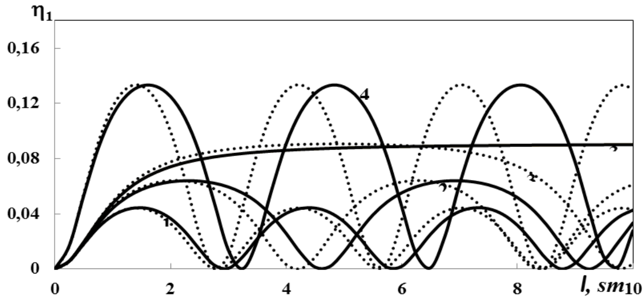

Figure 4. Dependence of the conversion efficiency into a signal wave on the total length of the metamaterial ℓ, obtained in CIA, for $I_{1\ell}$= 0, Γ_3= 1 cm^{-1} at Δ= 3 cm^{-1} (solid curves 1 and 4), 2.5 cm^{-1} (solid curve 2) and 2.097688 cm^{-1} (solid curve 3), I_{20} / I_{30} = 0.1 (solid curves 1-3) and 0.3 (solid curve 4). The results obtained in the CFA are given by dotted curves.

It can be seen from Figure 4 that for each value of the phase mismatch, there is an optimal value of the total length of the metamaterial at which the conversion efficiency is maximum. In this case, the optimal value of the total length of the metamaterial plays the role of the coherent total length of the nonlinear medium, a concept that in a normal medium is characteristic of the current length of the material. With the increase of the phase mismatch, the amplitude of the oscillations decreases, and the frequency of the oscillations increases (compare curves 1 and 2).

The results for the conversion efficiency into a signal wave for two different levels of the input signal wave intensity $I_{20}/I_{30} = 0.1$ and 0.2 are shown in figure 5.

As follows from the behavior of the dependencies, with an increase in the input (on the right into the medium) value of the signal wave intensity $I_{1\ell}$ by 2 times, the amplification coefficient at the output on the left of the metamaterial increases by 2 times (curves 2 and 4), while the optimal value of the pumping power is 2.85 W. Hence, by choosing a higher level of the backward wave at the input, it is possible to realize a more intense signal wave at the output of the metamaterial.

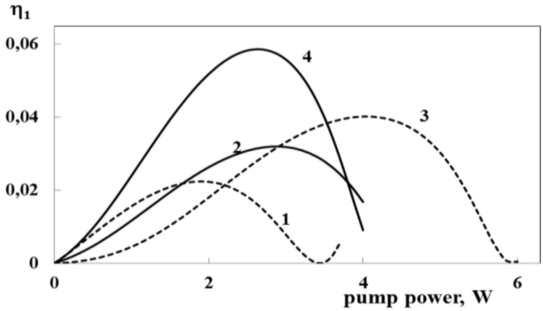

Figure 5. Dependence of the conversion efficiency into a signal wave on the pump power for $I_{1\ell} = 0$ at $I_{20}/I_{30} = 0.1$ (curves 1-3) and 0.2 (curve 4) and Δ = 5 cm^{-1} (curve 3), 5.5 cm^{-1} (curves 2 and 4) 6 cm^{-1} (curve 3).

As a result of the analysis of the expression for the amplification coefficient, in the case when all three waves are present at the entrance to the metamaterial (see figure 6), the increase in the amplification coefficient of signal wave depends on the ratio of the intensity levels of the idler and signal waves at the entrance to the metamaterial. The greater this ratio, the higher the signal wave amplification, for example, compare curves 2 and 7, where the intensity level of the idler wave to the signal wave differs by 5 times.

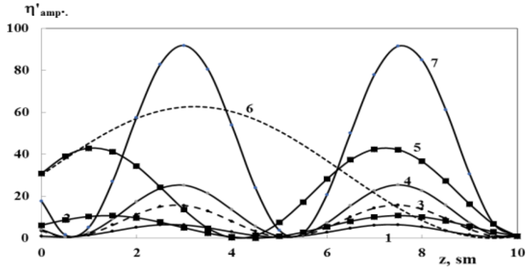

Figure 6. Dependences of the amplification coefficient on the length of the metamaterial for Γ_3 =1 cm^{-1}, ℓ =10 cm, Δ=2.5 cm^{-1} at I_{20} / I_{30} = 0.1 (curves 1, 3, 4 and 6), 0.3 (curves 2 and 5) and 0.5 (curve 7), $I_{20} / I_{1\ell}$ = 1 (curve 1), 5 (curve 3), 10 (curves 2 and 4), 50 (curves 5, 6 and 7)

Hence, according to the analytical expressions obtained in the work in the CIA, it is possible to calculate the expected values for the amplification coefficient and conversion efficiency in the metamaterial in low-frequency pumping for each specific experimental condition.

In the case of high frequency, infinitely large values of the conversion efficiency into a signal wave and the amplification coefficient of the signal wave are observed at choosing the optimal value of the total reduced length of the metamaterial (see figure 7).

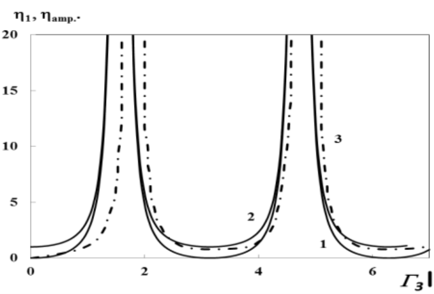

Figure 7. Dependences of the conversion efficiency into a signal wave (curve 1) and the amplification coefficient (curves 2 and 3) on the total reduced length of the metamaterial for Δ=0,

$I_{1\ell} / I_{20} =$**0.1. Curves 1 and 2 are calculated in CIA, and curve 3 in CFA.**

The results for the conversion efficiency to a signal wave for three different levels of the input intensity of the signal wave are shown in Figure 8. As follows from the behavior of the dependencies, with an increase in the input (on the right side of the metamaterial) value of the intensity of the signal wave by 5 times, the amplification coefficient at the output on the left of the metamaterial increases by almost 2 times (while the optimal value of the pumping power is 4.205 W). Hence, by choosing a larger input value of the backward (signal) wave, it is possible to realize a more intense reverse wave signal at the output of the metamaterial.

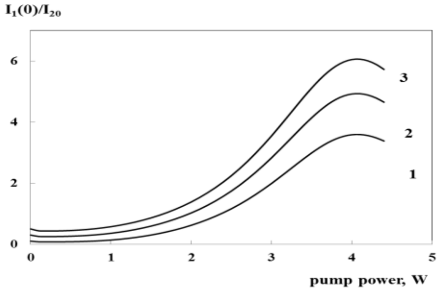

Figure 8. Dependence of the conversion efficiency in the signal wave on the pump power for the total length of the metamaterial ℓ **= 2 cm,** $\Delta\ell / 2$ **= 2.6 at** $I_{1\ell} / I_{20} =$ **0.1 (curve 1), 0.3 (curve 2), and 0.5 (curve 3).**

In the fourth chapter, SHG and the parametric interaction of electromagnetic waves in nanocomposites and chalcogenide crystals of the mid-IR range in CIA are theoretically considered.

This chapter discusses phase effects in GaS_xSe_{1-x}, $Hg_{1-x}Cd_xGa_2S_4$, $AgGa(Se_{1-x}S_x)_2$ and $AgGa_xIn_{1-x}Se_2$crystals. In these compounds, by increasing the content of one element and decreasing the content of the other element, the possibility of developing crystals with noncritical phase matching at the interesting radiation wavelength has been theoretically shown.

For nonlinear conversion, the theoretical analysis of the interaction of waves in negative uniaxial GaS_xSe_{1-x} crystals in CIA is carried out using a system of reduced equations[2]

$$\begin{aligned}
&\frac{dA_s}{dz} + \delta_s A_s + \frac{\gamma_n}{2} I^{n-1} A_s = -i\gamma_s A_p A_i^* \exp(i\Delta z),\\
&\frac{dA_i}{dz} + \delta_i A_i + \frac{\gamma_n}{2} I^{n-1} A_i = -i\gamma_i A_p A_s^* \exp(i\Delta z),\\
&\frac{dA_p}{dz} + \delta_p A_p + \frac{\gamma_n}{2} I^{n-1} A_p = -i\gamma_p A_s A_i \exp(-i\Delta z).
\end{aligned} \qquad (9)$$

Here $A_{s,i,p}$ are the complex amplitudes of the signal, idler and pump waves propagating in the direction of the z axis at the corresponding frequencies. The nonlinear coupling and loss coefficients for the j-th wave (j = s,i,p) are denoted as γ_j and δ_j, respectively, γ_n is the n-photon absorption constant.

It is assumed that the crystal has linear and two-photon absorption (n = 2). And the phase mismatch between the interacting waves has the form $\Delta = k_p - k_s - k_i$.

The problem was investigated in the general case, when all three waves with the corresponding frequencies $\omega_{p,i,s}$ are present at the input; therefore, the boundary conditions take the following form:

$$A_{p,i,s}(z=0) = A_{p0,i0,s0} \exp\left(i\varphi_{p0,i0,s0}\right) \qquad (10)$$

where $\varphi_{p0,i0,s0}$ are the initial phases of the pump wave, idler and signal waves at the entrance to the medium, $z=0$ correspond to the entrance to the crystal.

Solving the system of equations (9) with respect to the complex amplitude of the idler wave A_i in the CIA, using the boundary conditions (10) for the intensity of the idler wave at the crystal

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[2]Тагиев, З.А., Касумова, Р.Дж., Салманова, Р.А. Теория вырожденного четырёхволнового взаимодействия в приближении заданной интенсивности // − Санкт-Петербург: Оптика и спектроскопия, − 1999, 87, №1, − с. 94-97.

output, which is determined as $I_i(\ell_1) = A_i(\ell_1)\cdot A_i^*(\ell_1)$, we obtain the following expression

$$I_i(\ell_1) = I_{io}\exp(-2\delta_i\ell_1)\left[\cosh^2 q\,\ell_1 + \left(\frac{\Delta}{2} + \frac{\gamma_i A_{so}^* A_{po}}{A_{io}}\right)^2 \frac{\sinh^2 q\,\ell_1}{q^2}\right], \quad (11)$$

where $q^2 = \Gamma_p^2 - \Gamma_s^2 - \frac{\Delta^2}{4}$, $\Gamma_s^2 = \gamma_i\gamma_p I_{so}$, $\Gamma_p^2 = \gamma_s\gamma_i I_{po}$.

According to the analytical expressions obtained by us for the conversion efficiency of the fundamental wave into the second harmonic wave and the amplification coefficient, the dynamics of the processes is studied on the basis of the plotted dependences.

As can be seen from the curves in figure 9, at certain values of the problem parameter, a stronger idler signal can be obtained in a GaSSe crystal than in GaSe. Experimental points (squares-GaSSe and points- GaSe)[3] are also shown here.

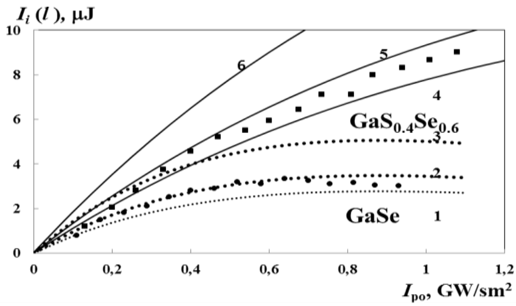

Figure 9. Dependences of the idler wave intensity in $GaS_{0.4}Se_{0.6}$ crystals (curves 4-6) and GaSe (curves 1-3) depending on the reduced pump intensity I_{p0}, calculated in CIA at I_{io} = 1 µJ (curves 1 and 4), 2 µJ (curves 2 and 5) and 5 µJ (curves 3 and 6), I_{so}/I_{po} = 4.5 10^{-8}. For the following parameters of $GaS_{0.4}Se_{0.6}$ crystal: ℓ = 0.47 cm [3], δ_i = 0.07 cm^{-1} and γ_n = 0.415 cm/GW and GaSe crystal: ℓ = 0.39 cm [3], δ_i = 0.29 cm^{-1} and γ_n = 1.46 cm/GW.

[3] Miyata, K., Picosecond mid-IR optical parametric amplifier based on the wide-bandgap $GaS_{0.4}Se_{0.6}$ pumped by a Nd:YAG laser system at 1064 nm / K.Miyata, G.Marchev, A.Tyazhev [et al.] // Optics Letters, − 2011, 36, − p. 1785-1787

Table 1 below shows the calculated values of the phase matching angle and the angular dispersion coefficient of the second order for the $Hg_{0.7}Cd_{0.3}Ga_2S_4$ crystal for the case of parametric generation of light at an idler wave.

Table 1. Data on the $Hg_{0.7}Cd_{0.3}Ga_2S_4$ crystal for the case of parametric generation of light at an idler wave.

Crystal	λ, mcm	n_o^ω	n_e^ω	Type of synchronism	d_{eff}, pm/V	θs, degree	the angular dispersion coefficient of the second order ,sm^{-1}degr. min.$^{-1}$
$Hg_{0.7}Cd_{0.3}Ga_2S_4$	1.064 pump	2.430288	2.39899	e→oo	24.94	90	0.0000634955
	1.5772 signal	2.405065	2.367763				
	3.27 idler	2.386262	2.35255				

Next, we considered the effect of second harmonic generation in promising $AgGa(Se_{1-x}S_x)_2$ and $AgGa_xIn_{1-x}Se_2$ crystals.

The task was solved for the case of plane waves: pump wave with complex amplitude A_1 at frequency ω_1 and second harmonic with complex amplitude A_2 at frequency ω_2. The analysis of harmonic generation in a medium can be described by the well-known reduced equations of the form (interaction oo-e)[4]

[4]Дмитриев, В.Г. Прикладная нелинейная оптика: Генераторы второй гармоники и параметрические генераторы света / В.Г.Дмитриев, Л.В.Тарасов. – Москва: Физматлит, – 2004, – 512 с.

$$\frac{dA_1}{dz} + \delta_1 A_1 = -i\frac{8\pi^2 d_{1eff}}{\lambda_1 n(\omega_1)} A_2 A_1^* \exp(-i\Delta z),$$
$$\frac{dA_2}{dz} + \delta_2 A_2 = -i\frac{4\pi^2 d_{2eff}}{\lambda_2 n(\omega_2)} A_1^2 \exp(i\Delta z) \qquad (12)$$

here $\delta_{1,2}$ are the absorption coefficients, $d_{1,2eff}$ are the effective nonlinear coefficients for the case oo→e of scalar phase matching, $\lambda_{1,2}$ are the wavelengths, $\Delta = k_2 - 2k_1$ is the phase mismatch, $n(\omega_{1,2})$ are the refractive indexes of the crystal, and k_1, k_2 are the values of the wave vectors at frequencies $\omega_{1,2}$, respectively.

The study was carried out under the following boundary conditions

$$A_1(z=0) = A_{10}\exp(i\varphi_{10}), \quad A_2(z=0) = 0, \qquad (13)$$

where z = 0 corresponds to the entrance to the crystal, φ_{10} is the initial phase of the pump wave at the entrance to the medium.

We solve the system of reduced equations (12) by differentiating the second equation with respect to the amplitude of the harmonic and applying the CIA. As a result, for the conversion efficiency to the second harmonic at the output of the crystal ($z = \ell$), we obtain[5]:

$$\eta_2(\ell) = I_2(\ell)/I_{10} = \gamma_2^2 I_{10} \ell^2 \mathrm{sinc}^2 \lambda\ell \exp[-(\delta_2 + 2\delta_1)\ell], \qquad (14)$$

where $\lambda^2 = 2\Gamma^2 - (\delta_2 - 2\delta_1 + i\Delta)^2/4$, $\Gamma^2 = \gamma_1\gamma_2 I_{10}$, $\mathrm{sinc}\, x = \sin x/x$, here γ_1, γ_2 are the nonlinear coupling coefficients of waves of the second order.

[5] Tagiev, Z.H., Constant-intensity approximation in a nonlinear wave theory / Z.H.Tagiev, R.J.Kasumova, R.A.Salmanova [et al.] // Journal of Optics B: Quantum and Semiclass, − 2001.3, − p. 84-87.

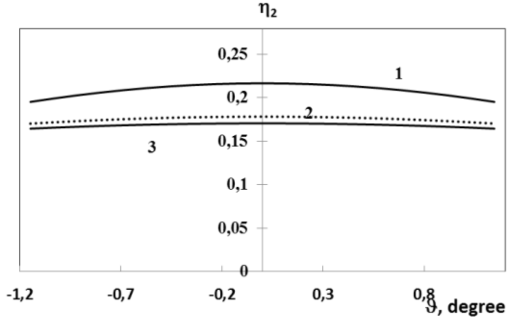

Figure 10. Dependence of the second harmonic conversion efficiency in the $AgGa(Se_{1-x}S_x)_2$ crystal on the phase mismatch, at $\delta_{1.2}$ = 0.1 cm^{-1}, I_{10} = 1.5 mW/cm^2, crystal length l = 0.2 sm for three values of the parameter x = 0 (curve 3), 0.2 (curve 2) and 1 (curve 1).

It can be seen from the behavior of curves 1-3 in figure 10 that the dependence becomes smoother with increasing parameter x. This fact confirms the fact that the regime of non-criticality of the crystal to the fulfillment of the phase matching condition is better fulfilled with an increase in the sulfur concentration. However, in this case, the nonlinear susceptibility of the structure under study decreases. Further experimental studies of the data on promising nonlinear crystals $AgGa(Se_{1-x}S_x)_2$ will make it possible to determine the components of the nonlinear susceptibility tensor for different sulfur concentrations. And this, in turn, will allow to analytically calculate in CIA the optimal relative sulfur content in this promising compound and give recommendations to the developers of laser devices.

Figure 11 shows the results of the CIA analysis of the frequency conversion process in the case of three different indium contents in the $AgGa_xIn_{1-x}Se_2$ crystal: 0, 0.3, and 0.4. Three versions of the CO_2 laser pump wavelengths are considered: 9.31 mm, 9.55 mm, and 9.64 mm.

From a comparison of the behavior of the group of curves 3, 4, and 5, it can be seen that with an increase in the concentration of

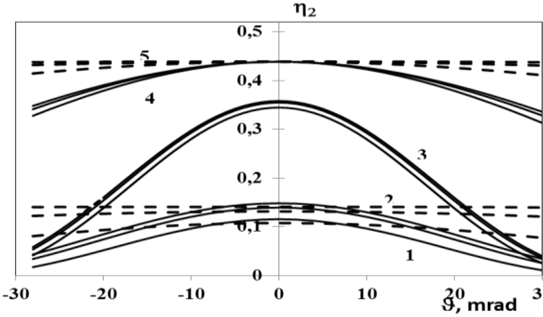

Figure 11. Dependences of the conversion efficiency of the second harmonic wave in the $AgGa_xIn_{1-x}Se_2$ crystal on the phase mismatch at I_{10} = 0.6 MW/cm² for x = 0.7 (groups of curves 1 and 4), 0.6 (groups of curves 2 and 5), 1 (groups of curves 3). In each of the groups, the upper curve corresponds to a wavelength of 9.64 μm, the middle one - 9.55 μm, and the lower curve - 9.31 μm. Crystal length[6] *l* = 1.05 cm (groups of curves 1 and 2), 0.8 cm (curve 3), and 0.65 cm (groups of curves 4 and 5). For $AgGaSe_2$ crystal δ_1 = 0.09 cm^{-1}, δ_2= 0.15 cm^{-1} and $AgGa_xIn_{1-x}Se_2$ δ_1= 0.06 cm^{-1}, δ_2= 0.08 cm^{-1}.

indium (i.e., the parameter x) from 1 to 0.6, the dependence flattens and becomes more and more flat. This indicates a transition to the crystal noncriticality regime and the fulfillment of the phase matching condition. For example, in the $AgGaSe_2$ crystal, the conversion efficiency changes by 0.036% in the angular range of values from –0.6 mrad to + 0.6 mrad. Substitution of indium for a part of Ga in the crystal up to x = 0.7 ($AgGa_{0.7}In_{0.3}Se_2$) leads to a similar change in efficiency, but already in an angular range that is 1.67 times larger (from - 1 mrad to + 1 mrad). Further incorporation of indium into the crystal up to x = 0.6 ($AgGa_{0.6}In_{0.4}Se_2$) increases the angular interval 33 times (- 20 mrad + 20 mrad) as compared with the case of the $AgGaSe_2$ crystal. Hence, in $AgGa_{0.6}In_{0.4}Se_2$

[6] Андреев, Ю.М., Удвоение частоты CO_2-лазеров в новом нелинейном кристалле $AgGa_xIn_{1-x}Se_2$ / Ю.М.Андреев, И.С.Батурин, П.П.Гейко [и др.] // Квантовая электроника, − Москва: − 1999. 29, №1, −с. 66-70.

crystals, the noncriticality to the fulfillment of the phase matching condition is fulfilled in a wider angular range than in $AgGa_{0.7}In_{0.3}Se_2$, and even more so in $AgGaSe_2$. This issue was experimentally investigated[6], but with the length of the experimental sample of the $AgGa_xIn_{1-x}Se_2$ crystal equal to 1.05 cm. In our case, this corresponds to the group of curves 1 (x = 0.7) and 2 (x = 0.6). Comparison of curves of groups 1 and 4 and groups 2 and 5 shows that the use of coherent length, i.e. the optimal length of the crystal-converter can allow 3 times to increase the conversion efficiency from 0.15 to 0.45.

In addition to nonlinear three-frequency processes in chalcogenide crystals, this chapter also discusses SHG in ZnO/PMMA nanocomposite films. ZnO nanocrystals were obtained by electrochemical deposition on the surface of PMMA films.

The study of the surface morphology of the films was carried out using a scanning electron microscope. In addition, X-ray diffraction analysis was carried out, the results of which are shown in figure 12.

The given diffraction pattern illustrates films deposited at -1.0 V. According to the figure, the films are polycrystals with a hexagonal phase and have a strong peak in the (002) plane.

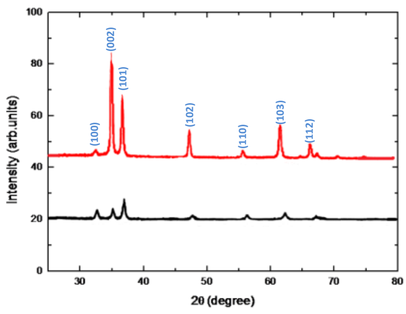

Figure 12. X-ray diffraction pattern of ZnO films

In figure 13 shows the optical transmission spectra of zinc oxide films deposited at different cathode potentials.

For a film deposited at –1.0 V, the optical transmittance is about ~ 85%. The band gap of the film is 3.3 eV.

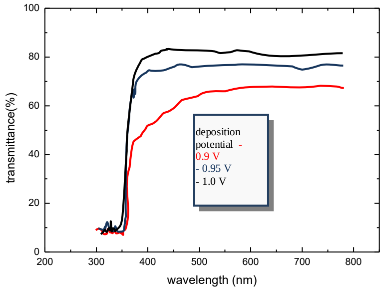

Figure 13. Optical transmission spectra

This chapter also shows various methods for calculating the refractive indexes for ZnO. The empirical Cauchy absorption model was used in the analysis[7] from the work

$$n(\lambda) = A + B/\lambda^2 + C/\lambda^4$$

for ZnO/PMMA2 (table 2) and for ZnO/PMMA1 (table 3)

Table 2. Refractive indexes for ZnO/PMMA2 calculated by the Cauchy method

λ, nm	A	B, nm^2	C, nm^4	$n_{\omega,2\omega}$
1064	1.9960	-0.024	0.01	1.9960
532	1.9960	-0.024	0.01	1.9960

Table 3. Refractive indexes for ZnO/PMMA1 calculated by the Cauchy method

λ, nm	A	B, nm^2	C, nm^4	$n_{\omega,2\omega}$
1064	1.984	-0.023	0.011	1.984
532	1.984	-0.023	0.011	1.984

Due to the theoretically obtained results, a decrease in the quadratic susceptibility with an increase in the ZnO concentration was found, in contrast to similar bulk materials. However, due to the long optical wavelength interaction, ZnO/PMMA films generate a strong second harmonic signal. This conclusion is consistent with the experimental results of work[8].

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[7] Tompkins, H. G. Spectroscopic Ellipsometry and Reflectometry and / H.G.Tompkins, W.A.McGahan, – New York: John Wiley & Sons, – 1999. – p. 93.
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MAIN RESULTS

1. At a pump intensity of 20 GW/cm^2, for a 16% concentration of ZnO, the conversion efficiency at THG is maximum at a coherent length of 0.64 μm. As the ZnO concentration increases from 3% to 16%, the frequency conversion efficiency increases by almost 1.6 times during THG, and in the case of second harmonic generation, an increase in the concentration from 5% to 15% leads to an almost twofold increase in the conversion efficiency.

2. Taking into account the reverse reaction of the third harmonic wave to the phase of the fundamental radiation in materials with a negative refractive index leads to the fact that, with an increase in the intensity of the fundamental radiation in the interval $\Gamma\ell = 0.7 \div 1$, the conversion efficiency decreases three times.

3. As a result of considering the effect of self-action in a metamaterial, it was found that a change in the phase mismatch from 0.1 to 0.5 leads to an increase in the pump wave phase by approximately five times, and in the case of a homogenous cubic medium, this change is 26.4 times smaller.

4. In a metamaterial under low frequency pumping at a level of input intensity of the idler wave exceeding the input intensity of the signal wave by 5 times, an increase in the amplification of the signal wave by almost 20 times is observed. In the case of high-frequency pumping at the optimal value of the total reduced length of the metamaterial, infinitely large values of the conversion efficiency and signal wave amplification coefficient are observed.

5. As the pump intensity increases, the maximum of the parametric conversion is reached at shorter coherent crystal lengths. The noncriticality regime of $AgGa(Se_{1-x}S_x)_2$ and $AgGa_{1-x}In_xSe_2$ crystals to the fulfillment of the phase matching condition is better fulfilled with an increase in the parameter x in these compounds. In the $AgGa_{0.6}In_{0.4}Se_2$ crystal, the conversion efficiency changes by 0.036% in an angular interval 33 times larger than in the $AgGaSe_2$ crystal.

6. In a GaSSe crystal, at a pump intensity (λ= 1.0642 μm) of 0.7 GW/cm^2 and a loss of 0.07 cm^{-1}, the idler wave (λ= 6.45 μm) is amplified from 5 mJ to 10 mJ. The regime of non-criticality of the crystals under consideration to the accuracy of setting the phase matching angle can be realized at low intensities of the pump wave and signal wave.

7. In nanocomposite thin ZnO/PMMA films obtained by electrochemical deposition, the hexagonal phase of the films was confirmed by X-ray diffraction analysis.

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The defense will be held on __11__ __November_____2021 at __15^{00}___ at the meeting of the Dissertation council ED 2.19 of Supreme Attestation Commission under the President of the Republic of Azerbaijan operating at

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Abstract was sent to the required addresses on_08_ _October_ 2021.

Signed for print: 05.10.2021

Paper format: A5

Volume: 37982

Number of hard copied: 20