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Ternary wide-bandgap chalcogenides $LiGaS_2$ and $BaGa_4S_7$ for the mid-IR

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Abstract

In this work we study the parametric interaction at the presence of the phase change of interacting pump, signal and idler waves. It is considered in the materials of mid-infrared range of spectrum for $LiGaS_2$ and $BaGa_4S_7$ crystals. The threshold intensity of pumping is analyzed for these crystals under the conditions of the experiments. The values of refractive indices and angle of phase matching have been calculated for these crystals. It is shown that $LiGaS_2$ and $BaGa_4S_7$ compounds could be used for nanosecond/picoseconds pumping of optical parametric converters at 1.064 mcm (Nd:YAG laser systems) without considering two-photon absorption.

Keywords: parametric interaction, mid-IR region, constant-intensity approximation.
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Introduction

At present, searching of crystals, for which efficient down-conversion in the mid-IR is satisfied, continues where the wavelength of pump wave varies near 1 mcm. As we known, coherent radiation that are tunable according to frequency has a wide range of application, for instance, on the basis of parametric frequency converters, LIDARs, could be constructed, which are used to investigate the Earth and atmosphere. Most promising candidate seems to be parametric generators near 6 мсм, which is also interesting for medical applications by using computer technology. Despite the importance of applications, nonlinear crystals, which provide for smooth tuning of laser radiation in an entire mid-IR optical diapason has not been elaborated yet.

In the works [1-8] triple chalcohenides is suggested to solve these issues. $LiGaS_2$ and $BaGa_4S_7$ non-oxide nonlinear optical crystals are transparent above ~5 mcm (up to 12 mcm) [2, 7] in the mid-IR. Simultaneously $LiGaS_2$crystal possess the largest band – gap energy E_g ~4.15 eV [2] (3.76 eV [1]) at room temperature among all the existing nonlinear crystals for the mid-IR. Analogous value for $BaGa_4S_7$ equals to 3.54 eV [1]. Therefore, at a pumping of relatively short wavelengths (1 mcm) for these crystals, processes of two-photon absorption can't be considered (without two-photon absorption). Despite the fact that these crystals have the lowest nonlinear coefficients, e.g. d_{eff}=5.5 pm/V (for $LiGaS_2$, eo-e type II phase matching in the xy plane)and d_{eff}=5.1 pm/V (for $BaGa_4S_7$, oo-e type I phase matching in the xz plane) these chalcohenides possesses highest damage threshold at 1.064 mcm with nanosecond long pulses [1, 5]. This peculiarity can allow to obtain more large conversion efficiency in these crystals (owing to highest damage threshold) that is obtained experimentally in Refs. 3-8. Based on these crystals we can produce parametric interaction where technologically developed and popular Nd:YAG lasers could be used as pumping resource. Thus, $LiGaS_2$ and $BaGa_4S_7$ crystals are very interesting for optical parametric oscillations of frequency conversion in the mid-IR.

To study the nonlinear optical properties of the investigated type of crystals, it is preferred to resort to the constant–intensity approximation [9-10], instead of the constant–field approximation [11, 12], which permits to take into account the influence of phase effects on the process of frequency conversion of laser radiation in the given crystals of mixed type.

In this work, the conditions of optimum frequency conversion in mid-infrared spectral range are considered for case of optical parametric amplification in ternary chalcogenides $LiGaS_2$ and $BaGa_4S_7$ with the account for phase changes of all interacting waves. An analysis of threshold intensity of pumping is given. The recommendations on increasing frequency conversion efficiency are offered. The results of calculation of the angles for phase matching are presented for $LiGaS_2$on 5.457 mcm and $BaGa_4S_7$ on 6.217 mcm.

Theory

Let us consider first the parametric amplification in biaxial $LiGaS_2$ and $BaGa_4S_7$compounds according to experimental scheme suggested in [5, 8]. As a pump source a Nd:YAG laser functioning at λ_p=1.064 mcm was used with pulse duration of 14 and 8 ns with pump energy ranging from 0 to ~90 mJ. For $BaGa_4S_7$ and $LiGaS_2$ the cross section areas of pump beam inside crystal were 0.38 cm^2 and 0.28 cm^2, respectively. The linear dissipations in $LiGaS_2$ and $BaGa_4S_7$ were equal to 0.06÷0.1 $cm^{-1}$and 0.3 cm^{-1}, respectively. Nonlinear transmission of $LiGaS_2$ and $BaGa_4S_7$, calculated inside the crystals, were estimated by authors according to Refs. 2 and 6.

In these experiments the eo-e process is chosen for optical parametric generation of the idler wave for $LiGaS_2$ (e polarized pump and idler waves) in the xy plane while for $BaGa_4S_7$ the oo-e process is carried out (e polarized pump wave) in the xz plane. In the experiment, for the idler energy measured at wavelengths of 5.457[8] and 6.217 mcm [5], effective nonlinearities of 5.5 and 5.1 pm/V are estimated for $LiGaS_2$ and $BaGa_4S_7$, respectively. In the experiment, a $LiGaS_2$ sample with length of 8 mm and $BaGa_4S_7$ with length of 14.05 mm are used. The two-photon absorption coefficients were neglected according to [1-8].

Let's analyze first the process of parametric wave interaction in crystals without taking two-photon absorption into consideration. For nonlinear conversion, theoretical analysis of wave interaction could be carried out using the known system of the reduced equations [11, 13]

$$\frac{dA_s}{dz}+\delta_s A_s=-i\gamma_s A_p A_i^* \exp(i\Delta z),$$
$$\frac{dA_i}{dz}+\delta_i A_i=-i\gamma_i A_p A_s^* \exp(i\Delta z),$$
$$\frac{dA_p}{dz}+\delta_p A_p=-i\gamma_p A_s A_i \exp(-i\Delta z). \qquad (1)$$

Here, $A_{s,i,p}$ are the complex amplitudes of the signal, idler and pump waves at respective frequencies $\omega_{s,i,p}$ in z direction. γ_j and δ_j signify the nonlinear coefficients and loss parameters for j-th wave ($j=s,\ i,\ p$), respectively, and $\Delta=k_p-k_s-k_i$ stands for phase mismatch between the interacting waves.

Here we used the standard technique [9, 10] to theoretically investigate the parametrical three wave interaction. The singly-resonant optical parametric oscillation of idler waves is considered for the case in which pump passes crystal two times. So we solved the reduced system of equations in the constant-intensity approximation with corresponding boundary conditions while the conditions of experiments were taken into account. Strictly speaking there are a couple of reasons for dissipation of all interacting waves including: effective dissipation in unit length of crystal while scattering is considered, dissipation of diffraction, losses on mirrors and Fresnel losses for each surface ($\alpha_{s,i,p}^{Fresnel}=\left(\frac{n_{s,i,p}-1}{n_{s,i,p}+1}\right)^2$ for perpendicular waves).

At parametric interaction in $LiGaS_2$ crystal, the dichroic ZnSe mirror served as input coupler, which transmitted 94% at the idler wavelength and reflected 70% at the signal wavelength. In experimental setup mentioned in [5] Ag mirror is used as a total reflector for all three waves. The pump beam reaches the $BaGa_4S_7$ crystal after reflecting from dichroic ZnSe mirror and passes through the flat output coupler with 82% transmition (or 55% like in [8]). This output coupler has a transmission of 18-22% at the signal wave and 73-84% at the idler wave, which cause in the minimal dissipation for output radiation at idler wavelength.

We carry on the task in general case, when at the entry all three waves with frequencies of

$\omega_{p,i,s}$ are present, so for the boundary conditions we will have the following

$$A_{p,i,s}(z=0) = A_{po,io,so}\exp(i\varphi_{po,io,so}) , \tag{2}$$

where $\varphi_{po,io,so}$ stand for initial phases of pump, idler and signal waves at the entry of the medium and $z = 0$ corresponds to the entry of crystal.

Now we solve the system of Eqs. (1) for the complex amplitudes of the idler wave A_i using constant–intensity approximation in the standard way by applying the boundary conditions (2). Then for the idler wave intensity at the output of crystal (which is determined by $I_i(\ell_1) = A_i(\ell_1)\cdot A_i^*(\ell_1)$) we obtain the following [9]

$$I_i(\ell_1) = I_{io}\exp(-2\delta_i\ell_1)\left[\cosh^2 q\,\ell_1 + \left(\frac{\Delta}{2} + \frac{\gamma_i A_{so}^* A_{po}}{A_{io}}\right)^2 \frac{\sinh^2 q\,\ell_1}{q^2}\right] , \tag{3}$$

where

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

Let's identify the wavelengths of signal wave in $LiGaS_2$ and $BaGa_4S_7$crystals at the pump wavelength of λ_p =1.0642 mcm and an idler wavelengths of λ_i =5.457 [2] and 6.217 mcm [5], respectively. Two conditions of phase matching should be carried out, one of them is the equality $\frac{1}{\lambda_p} = \frac{1}{\lambda_i} + \frac{1}{\lambda_s}$. From here we can get the proper value of 1.322013 mcm (in case of $LiGaS_2$) and 1.28399 mcm (in case of$BaGa_4S_7$) for wavelength of signal wave. The main values of refraction indices for wavelength of waves under study are derived from Sellmeyer equation [2, 7] and are n_x^p =2.281532, n_y^p =2.3010447, n_z^p =2.3217470 at λ_p =1.0642 mcm; are n_x^s =2.270057, n_y^s =2.28948932, n_z^s =2.3101926 at λ_s =1.28399 mcm are n_x^p =2.227538, n_y^p =2.24288419, n_z^p =2.2635723 at λ_i =6.217 mcm. Then we calculated refractive indices $n_{p,s,i}^{o,e}$ (see Table) and the angle Ω between the optic axes and the principal z-axis (in case $n_x < n_y < n_z$). Our result differs by 8% from experimental result obtained in Ref. 7.

As is known, the parametric process has threshold-dependent character. From (3) it is possible to determine threshold value of pumping wave amplitude in the constant – intensity approximation. At greater powers of pumping and lengths of interaction, the threshold amplitude of pumping ($\delta_s = \delta_p + \delta_i$) looks like [13]

$$A_{po,thresh}(\ell) = \sqrt{\frac{\Gamma_s^2 + \delta_i^2 + \Delta^2/4}{\gamma_s\gamma_i}} . \tag{4}$$

For investigation of threshold intensity of pumping we'll make the numerical account by using the analytical expression (4). With this, we choose the task parameters according to the conditions of experiment [5, 8]. To be exactly correct, dissipation for all interacting waves include: effective dissipation in unit length of crystal while scattering is considered; dissipation of diffraction ($\alpha_{s,i,p}^d = \frac{1}{L}\ln(\frac{a^2 - \lambda_{s,i,p}L}{a^2 - 2\lambda_{s,i,p}L})$ where a and L are the radius of circular flat-parallel mirror and length of optical resonator respectively); losses on mirrors due to reflection from mirrors and Fresnel losses for each surface. In experiment, reflection coefficients that are mentioned above for laser mirrors will make the resonator to have high quality for signal wave than idler wave. The high value of transmission coefficient in output laser mirror for idler wavelength signifies the minimal dissipation for output radiation at idler wavelength.

Table. Calculated data for $LiGaS_2$ and $BaGa_4S_7$ at parametric generation of idler wave in the mid-IR.

Crystal	λ, mcm	n_o	n_e	Phase matching type and plane	ϑ,φ, Ω degree
$LiGaS_2$,	1.0642(pump)		2.124105/ 2.122864	eo→e, XY	ϑ=90°, φ=40.6°[8]/ $φ_s$=42.40°
$LiGaS_2$	1.322013(signal)	2.134473		eo→e, XY	
$LiGaS_2$	5.457(idler)		2.076045/ 2.074852	eo→e, XY	
$BaGa_4S_7$	1.0642(pump)		2,319966	oo→e, XZ	ϑ=12° Ω^{theory}=46.3° [7] Ω^{exp}=45.6° [7] Ω=44.80°
$BaGa_4S_7$	1.28399 (signal)	2.289489		oo→e, XZ	
$BaGa_4S_7$	6.217 (idler)	2.242884		oo→e, XZ	

Results and discussion

To study the parametrical frequency conversion in middle IR-range, we will make the numerical calculation of the analytical expressions (3) and (4) that is derived from the constant-intensity approximation. To choose the parameters for calculation we use the information that we have from the experiment for $LiGaS_2$ and $BaGa_4S_7$ crystals [8, 5].

In Fig. 1, the dependencies of threshold intensity of pumping versus total losses for $LiGaS_2$ and $BaGa_4S_7$ crystals are given for two versions of initial signal wave intensity I_{s0}. From figure we see that by increasing the value of losses and signal wave intensity, the condition of parametric gain ($I_s(l) \geq I_{s0}$) is satisfied at greater values of pumping amplitude, that is the value of threshold intensity of pumping increases (compare curves 1 and 2 or 3 and 4). This fact was analytically found above following the analysis of Eq. (4).

The parametric generation in $BaGa_4S_7$ takes place at greater values of threshold intensity of pumping than in $LiGaS_2$(compare curves 1 and 3 or curves 2 and 4). So, from the numerical analysis (4) for $\Delta/2\Gamma_p$ = 0 and δ= 0.5 cm^{-1} it follows that in case of parametric interaction in $LiGaS_2$ and $BaGa_4S_7$ crystals, threshold intensity of pumping at I_{s0} = 0.001 mJ is equal to I_{po}^{thresh} = 0.531 mJ and 0.842 mJ, respectively. At I_{s0} = 0.5 mJ the threshold intensity reaches a value of I_{po}^{thresh} = 1.223 mJ and 1.526 mJ, respectively.

In Fig. 2, the dependencies of idler intensity (λ_i =5.457 mcm) on pump intensity for $LiGaS_2$ crystal are given at three values of nonlinear length. With an increase of pumping intensity there is an observed first horizontal section of dependence, i.e., parametric reinforcement is absent. Then at the definite value of pumping intensity, i.e., at threshold amplitude of pumping, the notable raising of dependence begins. With the decrease of crystal length, the growth of dependence takes place at greater values of threshold intensity of pumping (compare curves 1–3). Here in addition to the numerical calculation of the expression (3) (curves 1-3) experimental points are used from Ref. 8. The best agreement between theoretical and experimental results is observed at next values of task parameters: Γ_s/Γ_p =0.00001, $\Delta/2\Gamma_p$ =0.01 (at experimentally used values of crystal length 0.8 cm). In experiment, at pump wave energy of ~86 mJ, maximum of the idler energy gets the value of $I_i(l)$ =134 mcJ. Analogous value for idler energy calculated in the constant-intensity approximation is equal to 138.6 mcJ (curve 1).

As is seen in Fig. 1 it is observed both theoretically and experimentally that monotonous increase of $I_i(l)$ occurs when pump intensity grows.

Analogous behavior of dependences are presented in Fig. 3 for $BaGa_4S_7$ crystal (λ_i =6.217 mcm).

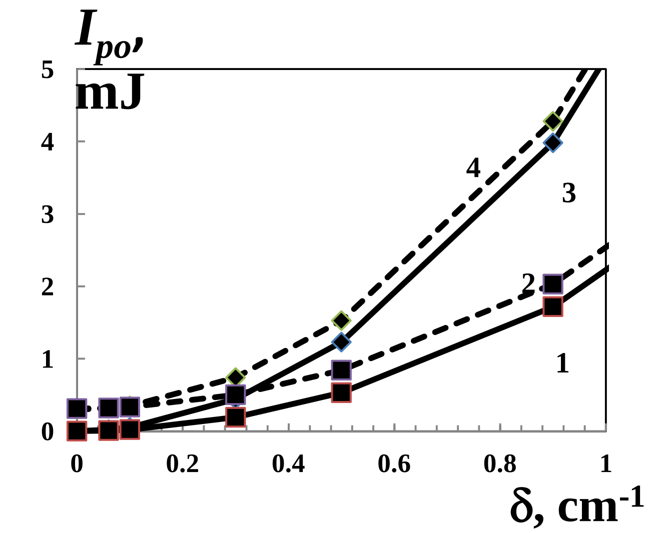

Fig. 1. Dependences of threshold of pump wave energy in $LiGaS_2$ (curve 1 and 2) and $BaGa_4S_7$ (curves 3 and 4) crystals I_{po}^{thresh} as a function of the total losses δ, ($\delta_p = \delta_i = \delta_s$) calculated in the constant-intensity approximation at phase mismatching for I_{so} =0.001 (curves 1 and 3), 0.5 (curves 3 and 4).

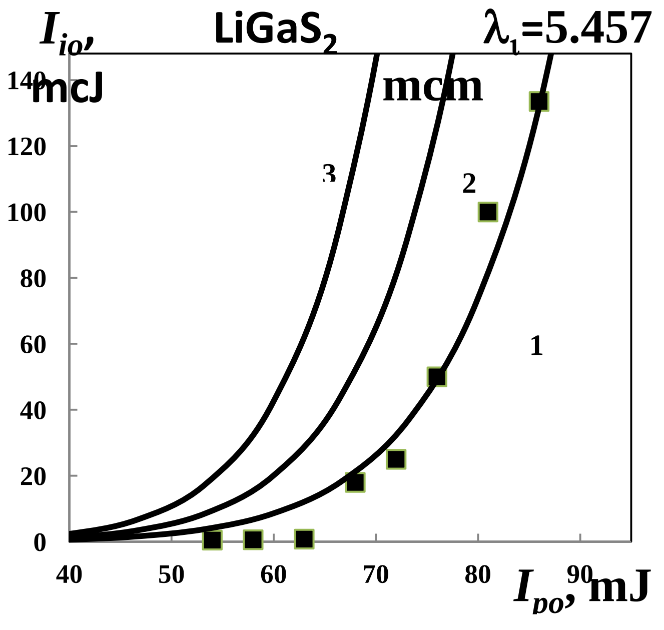

Fig. 2. Dependences of idler wave intensity in $LiGaS_2$ crystal (curves 1- 3) $I_i(\ell)$ as a function of the pump intensity I_{po} calculated in the constant-intensity approximation at phase mismatching, Γ_s / Γ_p =10^{-4} and $\delta_i = 0.06$ $cm^{-1}$for ℓ =0.8 cm [8] (curve 1), 0.85 cm (curve 2), 0.9 cm (curve 3). Here in addition to the numerical calculation of the expression (3) (curves 1-3) experimental points are used [8].

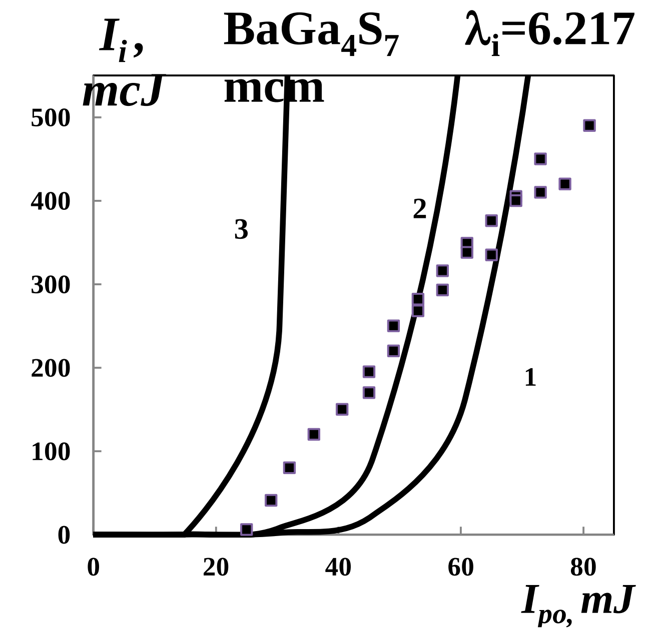

Fig. 3. Dependences of idler wave intensity in $BaGa_4S_7$ crystal (curves 1-3) $I_i(\ell)$ as a function of the pump intensity I_{po} calculated in the constant-intensity approximation at phase mismatching, Γ_s / Γ_p =10^{-3} and $\delta_i = 0.3$ cm^{-1} for ℓ =1.405 cm [5] (curves 1 and 2), 2 cm (curve 3). . Here in addition to the numerical calculation of the expression (3) (curves 1-3) experimental points are used [5].

The idler energy gets the maximum of value $I_i(l)$ =483 mcJ in experimental pump wave energy of ~ 81 mJ. For obtaining analogous value for idler energy calculated in the constant-intensity approximation, the pump wave energy is equal to 70 mJ (curve 1). By comparing these values, it follows that the maximum of the idler energy of $I_i(l)$ achieving in $BaGa_4S_7$ crystal is about 3.6 times higher than in $LiGaS_2$. This fact was observed experimentally earlier by the authors of the works [5, 8] and confirmed by us.

Thus, despite of small nonlinear coefficients for $LiGaS_2$ and $BaGa_4S_7$ absence of two-photon absorption makes these crystals suitable for optical parametrical converters at 1.0642 mcm.

Conclusion

Thus, from the results of the analysis for nonlinear interaction of optical waves in $LiGaS_2$ and $BaGa_4S_7$ crystals with account for phase effects and comparison of them with existing experimental data, it is possible to define optimal regime, which means that by choosing optimum values of parameters (pumping intensity I_{po}, an initial signal wave intensity I_{so}) efficiency of parametric conversion could be increased for the considered crystals of mid- IR range. The absence of two-photon absorption makes this compound suitable for optical parametrical converters at 1.0642 nm. Therefore, based on these crystals we can produce parametric generation where technologically developed and popular Nd:YAG lasers could be used as pumping source. It will make the development of the efficient sources of coherent radiation possible and it could be achieved by converters of frequency tuning in the mid-IR region of spectrum.

References

1. V. Petrov. Parametric down-conversion: The coverage of the mid-infrared spectral range by solid-state laser sources. Optical Materials. **34** 536-554, (2012).
2. L. Isaenko, A. Yelisseyev, S. Lobanov, P.Krinitsin, V. Petrov, J.-J. Zondy. Ternary chalcogenides $LiBC_2$ (B=In, Ga; C=S, Se, Te) for mid-IR nonlinear optics. J. of Non-rystalline Solids **352** 2439-2443, (2006).
3. V. Petrov, A. Yelisseyev, L. Isaenko, S. Lobanov, A. Titov, J.-J. Zondy. Second harmonic generation and optical parametric amplification in the mid-IR with orthorhombic biaxial crystals $LiGaS_2$ and $LiGaSe_2$. Applied Physics B, **78** 543-546 (2004).
4. V. Badikov, D. Badikov, G. Shevyrdayeva, A. Tyazhev, G. Marchev, V. Panyutin, F. Noack, V. Petrov, and A. Kwasniewski. $BaGa_4S_7$: wide-bandgap phase-matchable nonlinear crystal for the mid-inrfafed. Opt. Mater. Express **1** 316-320 (2011).
5. A. Tyazhev, D. Kolker, G. Marchev, V. Badikov, D. Badikov, G. Shevyrdayeva, V. Panyutin, and V. Petrov. Midinfrared optical parametric oscillator based on the wide-bandgap $BaGa_4S_7$ nonlinear crystal. Optics Letters **37** No.20, 2141-2143 (2012).
6. X. Lin, G. Zhang, and N. Ye. Growth and characterization of Ba Ga4S7: A new crystal for mid-IR nonlinear optics. Crowth& Design **9** No. 2, 1186-1189 (2009).
7. V. Badikov, D. Badikov, G. Shevyrdayeva, A. Tyazhev, G. Marchev, V. Panyutin, and V. Petrov. OSA/CLEO: Science and Innovations. Baltimore, Maryland US, $BaGa_4S_7$: Wide-bandgap phase-matchable nonlinear crystal for the mid-inrfafed. May 1-6, - (2011).
8. A. Tyazhev, V. Vedenyapin, G. Marchev, A. Yelisseyev, L. Isaenko, M. Starikova, S. Lobanov, and V. Petrov. Mid-IR optical oscillator based on $LiGaS_2$. CLEO/Europe and EQEC 2011 conference digest, OSA Technical Digest (CD), 2011, paper CD_P15.
9. Z.H. Tagiev, and A.S. Chirkin, Fixed intensity approximation in the theory of nonlinear waves, Zh. Eksp. Teor. Fiz. **73** 1271-1282, (1977);
10. Z. H. Tagiev, R. J. Kasumova, R. A. Salmanova, and N. V. Kerimova, Constant-intensity approximation in a nonlinear wave theory, J. Opt. B: Quantum Semiclas. Opt. **3** 84-87 (2001).
11. S. A. Akhmanov, R. V. Khokhlov, ProblemyNelineynoyOptiki [The Problems of Nonlinear Optics] (VINITI, Moscow, 1965).
12. V.G. Dmitriev, and L.V. Tarasov, PrikladnayaNelineynayaOptika [Applied NonlinearOptics] (Radio I Svyaz, Moscow, 1982).
13. Z.A. Tagiev, Sh.Sh. Amirov, On the efficiency of the optical parametric oscillation in the prescribed intensity approximation, SovietJournal of Quantum Electronics **16** 2243-2247 (1989).