Optical Parametric Amplification At 6.45 Mcm For Gas$_x$Se$_{1-X}$

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Abstract: In the present work, parametric interaction is considered when the phase change of interacting pump, signal and idler waves is studied in the materials of mid-infrared range of spectrum for GaS_xSe_{1-x} crystals. The values of refractive indices, angle of phase matching and angular dispersion coefficient of the first order have been calculated for $GaS_{0.4}Se_{0.6}$ crystal. It was shown that $GaS_{0.4}Se_{0.6}$ compound could be used for nanosecond/picoseconds pumping of optical parametric converters at 1.064 mcm (Nd:YAG laser systems) without considering two-photon absorption

Index Terms: constant-intensity approximation, mid-IR region, parametric interaction.

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1 INTRODUCTION

MORE recently, there have been reached the considerable achievements in application of tuning parametrical sources of optical coherent radiation in the mid-infrared spectral range. Among them it is possible to note the perspective GaS_xSe_{1-x} compounds. The observed strong reduction of the nonlinear coupling constant with increasing S content and deterioration of the crystal quality in comparison to the two parent compounds, GaS and GaSe obviously contributed to the lacking interest in further investigation of such solid solutions. As is known, two-photon absorption is an unwanted higher-order nonlinear process, which can be avoided at 1.064 mcm only in very few chalcogenide compounds, most of them with modest nonlinearity [1], [2]. Authors of [2] showed that $GaS_{0.4}Se_{0,6}$ or, in brief, GaSSe two-photon absorption coefficient at 1.064 mcm is 3.5 times lower than in GaSe, and its microhardness is increased by 30% relative to GaSe. Thus, more recent studies of the authors of [1], [2], [3], [4] demonstrated that GaS_xSe_{1-x} crystals is a promising nonlinear material for down conversion of pulsed 1.064 mcm to the mid-IR above 5 mcm without significant two-photon absorption. It is expectation that around x=0.4, a number of characteristics will make this compound suitable for nanosecond/picoseconds pumping of optical parametric oscillators or optical parametric generators/amplifiers at 1064 nm (Nd:YAG laser systems) without the onset of two-photon absorption. Therefore, based on these crystals we can produce parametric generation where technologically developed and popular Nd:YAG lasers could be used as pumping resource.

For study of nonlinear optical properties of the investigated type of crystal it is expedient to resort to the constant–intensity approximation [5], in contrast to the constant–field approximation [6], [7], permitting 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 GaS_xSe_{1-x} compounds with the account for phase changes of all interacting waves. The recommendations on increasing frequency conversion efficiency are offered. The results of calculation of the angles of phase matching, and angular dispersion coefficients for $GaS_{0.4}Se_{0.6}$ crystal on 6.45 mcm are presented.

2 THEORY

Let us consider the parametric amplification in GaS_xSe_{1-x} and GaSe compounds according to experimental scheme suggested in [2]. As a pump source authors employed a Nd:YAG laser at λ_p =1.064 mcm with pulse duration of 58 ps with pump intensity ranging from 0.1 to 1 GW/cm^2. The idler beam with energy in the 1÷10 mcJ range is collinearly mixed in GaSSe or GaSe with the collimated fundamental beam at 1.064 mcm through the bending mirror. In this experiment for optical parametric amplification of the idler wave, the eo-e process is chosen (e polarized pump wave and o polarized signal wave). In experiment the idler energy measured at a wavelength of 6.45 mcm, effective nonlinearities of 38 and 49 pm/V are estimated for GaSSe and GaSe, respectively. According to experience a GaSSe sample with 4.7 mm in length and GaSe sample with 3.9 mm in length are used. The beam diameter inside the crystals was 2 mm. The linear losses in GaSSe and GaSe were equal to 0,071 cm^{-1} and 0.29 cm^{-1}, respectively. The two-photon absorption coefficients were 0.415 cm/GW for GaSSe and 1.46 cm/GW for GaSe. Nonlinear transmission of GaSSe and GaSe calculated inside the crystals were estimated by us according to [3]. Let's analyze the process of parametric wave interaction in negative uniaxial GaS_xSe_{1-x} crystals in case of eo→e scalar phase matching for second type. For nonlinear conversion, theoretical analysis of wave interaction is made by using the known system of the reduced equations [6[, [8]

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$$\frac{dA_s}{dz}+\delta_s A_s+\frac{\gamma_n}{2}I^{n-1}A_s=-i\gamma_s A_p A_i^* \exp(i\Delta z),$$

$$\frac{dA_i}{dz}+\delta_i A_i+\frac{\gamma_n}{2}I^{n-1}A_i=-i\gamma_i A_p A_s^* \exp(i\Delta z), \qquad (1)$$

$$\frac{dA_p}{dz}+\delta_p A_p+\frac{\gamma_n}{2}I^{n-1}A_p=-i\gamma_p A_s A_i \exp(-i\Delta z).$$

Here $A_{s,i,p}$ are the complex amplitudes of the signal, idler and pump waves at respective frequencies $\omega_{s,i,p}$ in direction of axis z. The nonlinear coefficients and loss parameters for j-th wave ($j=s,\ i,\ p$) are labeled as γ_j and δ_j, respectively; γ_n is the constant of n photon absorption, $I=|A|^2$, $A=\sum_{j=s,i,p} A_j e^{i\beta_j}$; $\beta_j=\omega t-\vec{k}_j\vec{r}$. It is supposed that the crystal has linear and two-photon absorption (n =2). And phase mismatch between the interacting waves is given by $\Delta=k_p-k_s-k_i$. Here we used the standard technique [9] to theoretically investigate the parametrical three wave interaction for pass of crystal by pump and signal waves. 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 dissipation for all interacting waves include: effective dissipation in unit length of crystal while scattering is considered, dissipation of diffraction, losses on mirrors and Fresnel losses for each surface ($\alpha_{s,i,p}^{Fresnel}=\left(n_{s,i,p}-1\right)^2/\left(n_{s,i,p}+1\right)^2$ for perpendicular waves). In [2] experimental setup has highly reflective dichroic ZnSe mirror for pump wave at wavelength 1.0642 mcm and high reflectivity at 1.15-1.4 mcm and also 2.5 mcm cut-on filter which provides 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 the boundary conditions become as follows

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

where $\varphi_{po,io,so}$ are an 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 (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]

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

where

TABLE

Calculated data for $GaS_{0.4}Se_{0.6}$ crystal at parametric amplification of idler wave in the mid-IR

Crystal	λ, mcm	n_o	n_e	Phase matching type	ϑ_s, degree	Angular dispersion coefficient of the first order, cm^{-1}·ang. min^{-1}

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

As we know, GaS_xSe_{1-x} in mixed structure is affected by sulfur composition. Let's identify the wavelength of signal wave in $GaS_{0.4}Se_{0.6}$ crystal at the pump wavelength of λ_p =1.0642 mcm and idler wavelength of λ_i =6.45 mcm. By choosing x =0.4 for S concentration, the experimental value of wavelength of signal and idler waves are specified for this case which are presented in [1], [2]. Here the equation of $1/\lambda_p^e=1/\lambda_i^e+1/\lambda_s^o$ should be established for the sake of parametric three wave interaction of the radiations at pumping wavelength of λ_p =1.0642 mcm. From here we can get the proper value of 1.274479 mcm for wavelength of signal wave. Let's determine deviation angle from the direction of the phase matching $\Delta\theta$ for negative uniaxial $GaS_{0.4}Se_{0.6}$ typical mixed crystal in which parametric amplification of radiation occurs at approximate wavelength of 6.45 mcm (phase matching of the second type, eo-e interaction). For this crystal, the calculation was carried out using coefficients in the relation of Sellmeier for the main values of refractive indices [3]. To obtain the angular width of phase matching we'll calculate the angular disperse coefficient according to [7]. Below in the Table there are offered the values of refractive indices, the angle of phase matching and angular dispersion coefficient of the first order.

In the experiment for real frequency converters, it is impossible to ensure a condition of phase agreement (phase matching Δ = 0). An error that is followed from the condition of phase matching determines its width. Phase mismatching is affected by spectral width of pump radiation line, deviation from phase matching angle which is caused by divergence of laser radiation and instability of temperature for a crystal converter. Then the information that we have, particularly on angular width of phase matching will make possible to calculate the maximum divergence of light beam for pumping.

3 Results and discussion

To study the parametrical frequency conversion in middle IR-range, we will make the numerical calculation of the analytical expression (3) 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 $GaS_{0.4}Se_{0.6}$ and GaSe crystals [1], [2]. In Fig. 1 the dynamic process of parametrical amplification for idler wave $I_i(\ell)$ is depicted over the reduced pumping intensity I_{po} when the condition of phase mismatching is fulfilled at I_{so}/I_{po} =4.5·10^{-8}. Here three variants of initial value of idler wave intensity I_{io} are considered for $GaS_{0.4}Se_{0.6}$ (curves 4-6) and GaSe (curves 1-3) crystals. Here in addition to the numerical calculation of the expression (3) (curves 1-6) experimental points (squares-GaSSe and dots-GaSe) are used from [2]. Better agreement between theoretical and experimental results is observed at I_{io} =1÷2 mcJ in the range of experimental values of pump intensity (from 0.2 GW/cm^2 up to 1 GW/cm^2). From figure 1 we see (both experimentally and theoretically) that monotonous increase of $I_i(l)$ takes place as pump intensity grows.

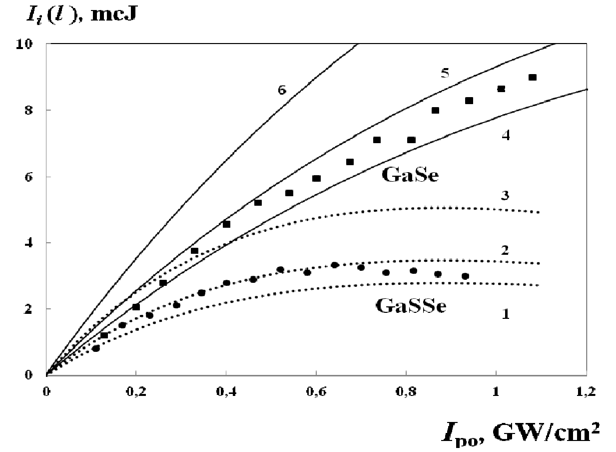

Fig. 1. Dependences of idler wave intensity in $GaS_{0.4}Se_{0.6}$ (curves 4-6) and GaSe (curves 1- 3) crystals $I_i(\ell)$ as a function of the reduced pump intensity I_{po} calculated in the constant-intensity approximation for I_{io} =1 mcJ (curves 1 and 4), 2 mcJ (curves 2 and 5) and 5 mcJ (curves 3 and 6) at phase mismatching and I_{so}/I_{po} =4.5·10^{-8}. For $GaS_{0.4}Se_{0.6}$ crystal length of ℓ =0.47 cm [2], $\delta_i = 0.07$ cm^{-1} and $\gamma_n = 0.415$ cm/GW, for GaSe crystal length of ℓ =0.39 cm [2], $\delta_i = 0.29$ cm^{-1} and γ_n =1.46 cm/GW. Here in addition to the numerical calculation of the expression (3) (curves 1-6) experimental points (squares-GaSSe and dots-GaSe) are used [2].

In the constant-intensity approximation at a pump intensity of 1 GW/cm^2, the idler energy $I_i(l)$ gets the following values: 8.1735 mcJ for GaSSe (curve 4), 2.753 mcJ for GaSe (curve 1) 9.331 mcJ for GaSSe (curve 5), and 3.442 mcJ for GaSe (curve 2) for initial values of I_{io} =1 mcJ (curves 1 and 4) and 2 mcJ (curves 2 and 5). Analogous values in experiment [2] at a pump intensity of 1.08 GW/cm^2 are equal to $I_i(l)$=9.1 mcJ (for GaSSe) and it is about 3 times higher than GaSe [1], [2]. As it is shown from our theoretical analysis, same decreasing conversion efficiency for GaSe (in comparison to GaSSe) can be explained by absorption influence, i.e. two-photon absorption at 1.0642 nm. This fact was observed earlier by the authors of the work [2], as confirmed experimentally. As is known GaS_xSe_{1-x} is a crystal without significant two-photon absorption. Thus, despite of a small nonlinear coefficient for GaSSe (in comparison to GaSe) absence of significant two-photon absorption makes this compound suitable for optical parametrical converters at 1.0642 mcm. By using the data provided in the table, dependencies of parametric conversion efficiency on angular mismatching $\Delta\theta$ are presented in Fig. 2 for $GaS_{0.4}Se_{0.6}$ crystal at three values of pump intensity (I_{so}/I_{po} =4.5∗10^{-8}). Just as one should expect, the dependencies are of no monotonous character. With the increase in pump intensity, conversion efficiency also increases (compare curves 1-3). From the numerical analysis of the analytical expression (3) for idler wave intensity, obtained in the constant-intensity approximation, it follows that the width of angular phase matching is being changed by pump intensity. To compare, when pump intensity increases 1.3 times, the idler intensity increases 2.665 times (curves 1 and 3) and at the same time the width of curve for phase matching $I_i(\Delta\theta)$ changes 3%.

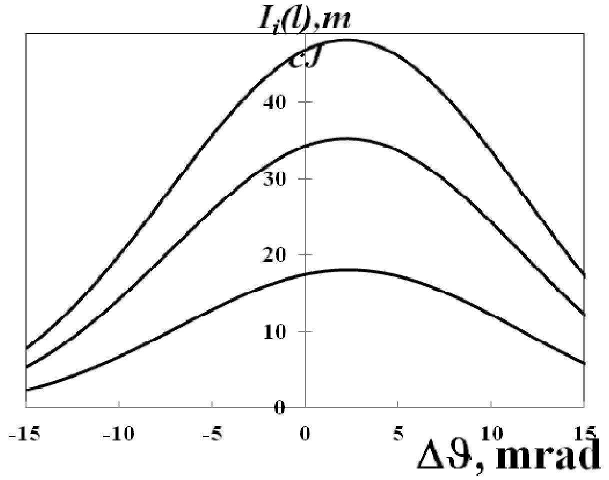

Fig. 2. Dependences of intensity of idler wave $I_i(l)$ in $GaS_{0.4}Se_{0.6}$ (curves 2, 4 and 6) and GaSe (curves 1, 3 and 5) crystals on angular mismatching $\Delta\theta$ for I_{io} =1 mcJ (curves 1-6) at pump intensity I_{po}=0.04 GW/cm^2 (curve 1), 0.07 GW/cm^2 (curve 3), 0.08 GW/cm^2 (curves 2 and 5), 0.13 GW/cm^2 (curve 4) and 0.15 GW/cm^2 (curve 6). Here $\delta_i = 0.07$ cm^{-1} and γ_n 0.415cm/GW for $GaS_{0.4}Se_{0.6}$ crystal with length of ℓ =0.47 cm [2], and for GaSe crystal with length of ℓ = 0.39 cm [2], $\delta_i = 0.29$ cm^{-1} and γ_n =1.46 cm/GW.

As is seen from dependence of $I_i(\Delta\theta)$ on angular width of phase mismatching chosen at 1/2 of maximum efficiency, angular width is equal to 1,13^0 when we calculate it for $GaS_{0.4}Se_{0.6}$ crystal with 4.7 mm length and pump intensity of 0.1 GW/cm^2. To compare, in case of 3 mm long CGA crystal, angular width of phase matching reaches 4.24^0 (see [10]) when pump intensity is equal to 0.0012 MW/cm^2. Also in case of $GaS_{0.4}Se_{0.6}$ crystal when pump intensity is equal to 0.1 GW/cm^2 (curve 1) maximum for idler intensity will be 18.12 mcJ. By comparing the behavior of curves 1, 2 and 3, it is seen that when pump intensity decreases the dependence becomes less sharp. So, slight change of efficiency dependence on angular mismatch from phase matching direction occurs at small values of pump intensity. Similar delicate changes of conversion efficiency under deviation from direction of phase matching (i.e. the uncritical regime of the considered crystals) can be obtained at low levels of intensity of pump wave.

4 CONCLUSION

Thus, from the results of the analysis of nonlinear interaction of optical waves in GaSSe 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 idler wave intensity I_{io} and I_{so}/I_{po}) efficiency of parametric conversion could be increased for the considered crystals of mid- IR range and the condition of increasing the degree of uncritical angular phase matching is possible to select. An absence of significant 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 resource. It will make it possible development of the efficient sources of coherent radiation and it could be achieved by converters of frequency tuning in the mid-IR region of spectrum.

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