Research Article

Second Harmonic of Laser Radiation for IR-Range in Mixed AgGa$_{0.6}$In$_{0.4}$Se$_2$ Crystals

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Received 20 November 2013; Accepted 6 January 2014; Published 27 February 2014

Academic Editors: K. Santhakumar and V. Srivastava

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The results of investigations of the influence of different parameters on conversion efficiency in mixed AgGa$_{0.6}$In$_{0.4}$Se$_2$ crystal in conditions of existing experiments are cited. The angular dispersion coefficients for three versions of crystal AgGa$_x$In$_{1-x}$Se$_2$, differing by a content of indium, have been calculated. A comparison was made of the obtained results on conversion efficiency with analogous results in case of other crystals and with corresponding experimentally measured values. The applied analytical method makes it possible to calculate the optimum parameters of both crystal-converter and a source of radiation for conditions of uncritical phase matching in the concrete experiment.

1. Introduction

Recently, the important achievements have been attained in application of nonlinear crystals in the numerous devices of IR-range. One can note among them the prospect CdGeAs$_2$ crystals, available ZnGeP$_2$ crystals, AgGaSe$_2$, and others [1–3]. Selection of nonlinear crystals is determined by requirement of obtaining the high conversion efficiency of pump radiation in a tuning wide spectral range. As one way of solving this problem, it is suggested to use chalcogenide crystals or chalcopyrite structures, for example, AgGa$_x$In$_{1-x}$Se$_2$, AgGa($Se_{1-x}S_x$)$_2$ [2]. In similar crystals, by changing parameter $x$, it is possible to realize uncritical 90° phase matching. This condition ensures the absence of walk-off of second harmonic energy, excluding decrease of second harmonic generation caused by birefringence.

We have chosen the perspective crystals AgGa$_x$In$_{1-x}$Se$_2$ as a subject of study among crystals of mixed type, owing to a number of its qualities. As showed in the result of experiment [4] by researchers, by choosing indium content, it is possible to perform the uncritical condition of 90° phase matching at second harmonic generation in near and middle IR-range. With this, a value of parameters $x$ on wavelength of CO$_2$ laser radiation $\lambda = 9.64$ mcm is equal to 0.6. The measured value of quadratic nonlinear susceptibility for AgGa$_{0.6}$In$_{0.4}$Se$_2$ crystal is equal to $d_{36} = 41$ pm/V [4]. For comparison, the analogous specified value for AgGaSe$_2$ crystal is equal to 39 pm/V [3, 4].

For the task of application in IR-range of spectrum CO$_2$ lasers (which present a powerful source of optical coherent radiation in the given region of spectrum) play a leading role. If in the middle IR-region of spectrum an efficient generation of radiation for this laser permits realizing tunable coherent radiation, then in the near IR-range it can succeed owing to radiation of second harmonics for CO$_2$ laser.

To consider a wide circle of the tasks in a theory for nonlinear waves, it is necessary to solve the system of coupled nonlinear differential equations. In most cases, solution of such system in a general form is not possible, so to analyze the complete system of reduced equation, different approximation methods have been used. Among them, constant-field approximation of basic radiation was widely used [5–7]. In this approximation, both the real amplitude and phase for wave are considered to be constant. The constant-field approximation depicts rightly only an initial stage of nonlinear interaction for waves until it is not generally possible to take into account the reverse reaction of generated or enforced waves to the intense pump wave. This leads to restricted consideration of wave interaction in real media and loss of information on features of the nonlinear process.
For analyzing of nonlinear process, it is possible to use the direct calculation of coupled equations. However, development of the analytical method will permit receiving the concrete analytical expressions and defining optimum parameters of the task for the purpose of obtaining the maximum efficiency of conversion.

To study the nonlinear optical properties of selected type of crystal, it is expedient to use the direct calculation of coupled equations. However, at frequency conversion in the conditions of phase mismatch, it is impossible to fulfill the complete transfer of energy to the second harmonic. In this paper the influence of different parameters of crystal refraction, and initial phase of pump wave at the entry of the medium. The ways of increasing conversion efficiency are presented on the example of AgGa\textsubscript{0.6}In\textsubscript{0.4}Se\textsubscript{2} crystal. For conversion efficiency of pump radiation to second harmonic, it is possible to receive optimum length of crystal. At this length the optimum length decreases. When we ignore the losses $\ell_{\text{opt}} = \lambda^{-1} \arctan(\lambda/\delta_2)$, where $\lambda^2 = 2\Gamma^2 + \Delta^2/4$. Whence it appears that in this case, in contrast to the results of the constant-field approximation, a coherent length of nonlinear medium depends on pump intensity $I_{10}$ and dissipation in a medium. By increasing in pump intensity and mismatch, the optimum length decreases. When we ignore the losses $\ell_{\text{opt}} = 0.5\pi/(2\Gamma^2 + \Delta^2/4)^{1/2}$ [8, 9].

We will solve the system with the following boundary conditions:

\[ A_1 (z = 0) = A_{10} \exp(i\varphi_{10}), \quad A_2 (z = 0) = 0, \]

\[ A_1 (z = \ell) = A_{1\ell}, \quad A_2 (z = \ell) = A_{2\ell}, \]

where $z = 0$ corresponds to the entry of crystal and $\varphi_{10}$ is an initial phase of pump wave at the entry of the medium.

\[ A_2 (\ell) = -i\gamma_2 A_{1\ell} A_{2\ell} \sin \ell \exp \left[ 2i\varphi_{10} - \frac{(\delta_2 + 2\delta_1 + i\Delta_2)}{2} \ell \right], \]

where

\[ \lambda^2 = 2\Gamma^2 - \frac{(\delta_2 - 2\delta_1 + i\Delta_2)^2}{4}, \]

\[ \gamma_1 = \frac{4\pi^2 d_{\text{eff}}}{\lambda_1 n(\omega_1)}, \]

\[ \gamma_2 = \frac{4\pi^2 d_{\text{eff}}}{\lambda_2 n(\omega_2)} \]

are nonlinear coefficients of waves.

2. Theory

Let us analyze the process of redoubling the frequency of CO\textsubscript{2} laser radiation (at frequency $\omega_1$) in AgGa\textsubscript{0.6}In\textsubscript{0.4}Se\textsubscript{2} negative uniaxial crystal in case of $\text{oo} \rightarrow \text{e}$ scalar phase matching for first type.

For nonlinear conversion, theoretical analysis of wave interaction is made by using the known system of the reduced equation describing second harmonic generation (at frequency $2\omega_1$) [5–7]:

\[ \frac{dA_1}{dz} + \delta_1 A_1 = -i\frac{8\pi^2 d_{\text{eff}}}{\lambda_1 n(\omega_1)} A_2 A_1^* \exp(-i\Delta z), \]

\[ \frac{dA_2}{dz} + \delta_2 A_2 = -i\frac{4\pi^2 d_{\text{eff}}}{\lambda_2 n(\omega_2)} A_1^* \exp(i\Delta z), \]

where $A_1, A_2$ are complex amplitudes of pump wave and their second harmonic at frequencies $\omega_{12}$ ($\omega_2 = 2\omega_1$), correspondingly, $k_1$ and $k_2$ are values of wave vectors for pump and second harmonic waves, $\Delta = k_2 - 2k_1$ stands for phase mismatch, $d_{\text{eff}}$ are efficient nonlinear coefficient for the case $\text{oo} \rightarrow \text{e}$ scalar phase matching, $\lambda_{12}$ signify the wavelength of pump and second harmonic waves, $n(\omega_{12})$ are the indices of crystal refraction, and $\delta_{12}$ are the coefficients of absorption for waves at frequencies of $\omega_{12}$, respectively.

System (1) does not include the members that are responsible for thermal effects. In the present work the investigation of frequency conversion for low value of pump intensity is explained.
basic radiation energy to second harmonic energy. In this case the spatial beatings of harmonic amplitudes are observed. This time, the minimums of harmonic intensity beatings, as an analysis shows in the constant-intensity approximation, depend on nonlinear susceptibilities of crystal [10]. This fact permits defining the nonlinear susceptibilities of substances by a simple way, more precise than in the constant-field approximation. With an increase in phase mismatch, spatial frequency increases, but at this time, maximum value of second harmonic intensity decreases.

In the experiment for real frequency converters, it is impossible to ensure a condition of phase agreement, that is, phase matching ($\Delta = 0$). An error in following the condition of phase matching determines the width of phase matching. Spectral width of pump radiation line, deviation from phase matching angle due to divergence of laser radiation, and instability of temperature for a crystal converter all contribute to mismatch. Hence receiving information, in particular, on angular width of phase matching, will permit calculating the maximum divergence of light beam for pumping. Moreover, determination of conditions for realizing uncritical phase matching at chosen length of pump wave is important for exclusion of taking down the influence of birefringent walk-off on generation efficiency. This fact allows one to take away the restriction of length of the used crystals [4, 7, 11].

Let us define an angular width of phase matching in uniaxial crystal for mixed type AgGa$_{1-x}$In$_x$Se$_2$ in case of second harmonic generation of CO$_2$ laser radiation on three wavelengths of pump: 9.64 mcm, 9.55 mcm, and 9.31 mcm (scalar phase matching of the first type for $\alpha \rightarrow \epsilon$ interaction). We will carry out the calculation of deviation angle from the direction of phase matching $\Delta \theta$ according to [7] for three values reflecting iodium content in crystal (0, 0.3, and 0.4). With this, Sellmeier coefficients were used cited in [4, 12].

The results of calculations are presented for angular dispersion coefficient of the first and second orders in Table 1.

3. Results and Discussion

To study the ways of increasing frequency conversion efficiency in AgGa$_{1-x}$In$_x$Se$_2$ crystal of CO$_2$ laser radiation in IR-range, we will make the numerous calculation of the analytical expression for conversion efficiency ($\eta_2$), received in the constant-intensity approximation; the parameters of the task are chosen according to conditions of existing experiments for the given crystal [4, 13]. In Figures 1–3 the dynamic process of frequency conversion is shown to the second harmonic in AgGa$_{0.6}$In$_{0.4}$Se$_2$ crystal.

In Figure 1, the dependencies of frequency conversion efficiency on length of crystal $\eta_2(l)$ are presented. These are considered as 3 versions of conversion differing by CO$_2$ laser pump intensities, radiating at wavelength in 9.55 mcm. From the behavior of curves differing by monotonous behavior in case of the constant-field approximation, it follows that there exists an optimum value of crystal length at which conversion efficiency is maximum. As far as pump intensity grows the maximum of conversion is reached at lower lengths of a crystal; that is, by increasing the pump intensity, the coherent length of crystal decreases.

In Figure 2, the dependencies on pump intensity for three lengths of crystals are cited. As is seen in the figure, CO$_2$ laser radiation, generating at wavelength in 9.55 mcm, maximum efficiency is converted to second harmonic wave at optimum value of pump intensity. From comparison of curves 1, 2, and 3 it is seen that optimum value of pump intensity falls down as crystal length increases. It is explained by the fact that at greater values of pump intensity the coherent length of crystal, where maximum conversion occurs, comes out at lower geometric dimension of crystal converters.

As is known in the mixed structures AgGa$_{1-x}$In$_x$Se$_2$ crystal properties undergo the influence of indium content [4]. In Figure 3 the results of the analysis for frequency conversion
Table 1: The angular dispersion coefficients for AgGa$_{1-x}$In$_{x}$Se$_2$.

| Crystal         | λ, mcm | $n^0_\omega$ | $n^0_\nu$ | $n^2_\omega$ | $n^2_\nu$ | $d_{\text{ang}}$ | Phase matching type | $\theta_1$, degree | Angular dispersion coefficient of first order, cm$^{-1}$·ang.min.$^{-1}$ |
|-----------------|--------|--------------|------------|--------------|------------|-------------------|---------------------|-------------------|---------------------------------------------|
| AgGaSe$_2$      | 9.31   | 2.597798     | 2.565611   | 2.616307     | 2.58463    | 39.0 [11]        | oo $\rightarrow$ e | 47.9 [4]         | 0.048353                                    |
| AgGaSe$_2$      | 9.55   | 2.596647     | 2.563607   | 2.616158     | 2.584178   | 39.0 [11]        | oo $\rightarrow$ e | 49.1 [4]         | 0.047611                                    |
| AgGaSe$_2$      | 9.64   | 2.596565     | 2.563902   | 2.615704     | 2.58401    | 39.0 [11]        | oo $\rightarrow$ e | 49.6 [4]         | 0.046432                                    |
| AgGa$_{0.6}$In$_{0.4}$Se$_2$ | 9.64 | 2.560749     | 2.584379   | 2.624120     | 2.602546   | 40.6 [4]        | oo $\rightarrow$ e | 63.5 [4]         | 0.026456                                    |
| AgGa$_{0.7}$In$_{0.3}$Se$_2$ | 9.55 | 2.583259     | 2.605674   | 2.626359     | 2.602059   | 40.6 [4]        | oo $\rightarrow$ e | 65.0 [4]         | 0.024782                                    |
| AgGa$_{0.6}$In$_{0.4}$Se$_2$ | 9.64 | 2.605264     | 2.582832   | 2.623489     | 2.60188    | 40.6 [4]        | oo $\rightarrow$ e | 66.0 [4]         | 0.023831                                    |
| AgGa$_{0.7}$In$_{0.3}$Se$_2$ | 9.31 | 2.609726     | 2.590959   | 2.626858     | 2.608989   | 41.0 [14]       | oo $\rightarrow$ e | 77.6 [4]         | 0.011648                                    |
| AgGa$_{0.6}$In$_{0.4}$Se$_2$ | 9.55 | 2.609726     | 2.589858   | 2.626432     | 2.608496   | 41.0 [14]       | oo $\rightarrow$ e | 83.4 [4]         | 0.006216                                    |

| AgGa$_{0.7}$In$_{0.3}$Se$_2$ | 9.64 | 2.608277     | 2.6081 [4] | 2.608438     | 2.626222   | 2.608315        | 41.0 [14]       | oo $\rightarrow$ e | 90.0 [4]                                   | 0.000919                                    |

Figure 3: Dependences of conversion efficiency of radiation energy of pump wave to energy of wave of second harmonic on AgGa$_{1-x}$In$_x$Se$_2$ crystal $\eta_2$ as a function of the phase mismatch calculated in the constant-intensity approximation at pump intensity of $I_{p0} = 0.6$ MW/cm$^2$ for $x = 0.7$—AgGa$_{0.7}$In$_{0.3}$Se$_2$ (groups of curves 1 and 4), 0.6—AgGa$_{0.6}$In$_{0.4}$Se$_2$ (groups of dashed curves 2 and 5), and 1—AgGaSe$_2$ (group of curve 3). In each group, the upper curve corresponds to wavelength of radiation equal to 9.64 mcm, the middle one corresponds to 9.55 mcm, and lower curve corresponds to 9.31 mcm. Here crystal length of $l = 1.05$ cm [4] (groups of curves 1 and 2), 0.8 cm [4] (group of curve 3), and 0.65 cm (groups of curves 4 and 5). For AgGaSe$_2$ crystal $\delta_1 = 0.09$ cm$^{-1}$, $\delta_2 = 0.15$ cm$^{-1}$ [4] and AgGa$_{0.7}$In$_{0.3}$Se$_2$ $\delta_1 = 0.06$ cm$^{-1}$, $\delta_2 = 0.08$ cm$^{-1}$ [4].

wavelengths of pump radiation at lengths of AgGa$_{0.7}$In$_{0.3}$Se$_2$ crystal $(x = 0.7)$, equal to 1.05 cm and 0.65 cm, respectively. Groups 2 and 5 (containing on three curves each) correspond to dependencies of $\eta_2(\theta)$ to three wavelengths of pump radiation at crystal lengths AgGa$_{0.7}$In$_{0.3}$Se$_2$ $(x = 0.6)$, equal to 1.05 cm and 0.65 cm. Group 3 (containing three curves) corresponds to dependencies $\eta_2(\theta)$ in case of 3 wavelengths of pump radiation at length of AgGaSe$_2$ crystal $(x = 1)$, equal to 0.8 cm. In each group, the upper curve corresponds to wavelength of radiation equal to 9.64 mcm, the middle one corresponds to 9.55 mcm, and lower curve corresponds to 9.31 mcm.

By comparing the behavior of curves 3, 4, and 5, it is seen that with increasing the concentration of indium in the mixed crystal from 1 to 0.6, the dependence $\eta_2(\theta)$ becomes more flat. It testifies the transition to the regime of uncritical character of crystal towards following condition of phase matching; thus, for instance, in AgGaSe$_2$ crystal the change of conversation efficiency by 0.036% takes place in the angular range of changes from $-0.6$ mrad to $+0.6$ mrad. Substituting some part of Ga for indium to content $x = 0.7$ (AgGa$_{0.7}$In$_{0.3}$Se$_2$) in a crystal leads to the analogous change of efficiency, but already in the angular range greater by 1.67 times (from $-1$ mrad to $+1$ mrad). The further replacement of indium to crystal to $x = 0.6$ (i.e., AgGa$_{0.6}$In$_{0.4}$Se$_2$) increases angular range 33 times ($-20$ mrad to $+20$ mrad) in comparison with a case of AgGaSe$_2$ crystal. Hence, in AgGa$_{0.6}$In$_{0.4}$Se$_2$ crystal, uncritical behavior towards following phase matching condition is observed in angular range greater than in AgGa$_{0.7}$In$_{0.3}$Se$_2$ and in AgGaSe$_2$. This question was studied experimentally in [4], but at the length of experimental sample for AgGa$_{1-x}$In$_x$Se$_2$ crystal equal to 1.05 cm. In our case, it corresponds to curves group 1 $(x = 0.7)$ and group 2 $(x = 0.6)$. By comparing the curves in groups 1 and 4, and those of groups 2 and 5, we see that the use of optimum length process are represented in case of 3 different concentrations of indium in crystal: 0, 0.3, and 0.4. Three versions of CO$_2$ laser wavelengths are considered: 9.31 mcm, 9.55 mcm, and 9.64 mcm. Groups 1 and 4 (containing three curves each) correspond to dependencies of $\eta_2(\theta)$ in case of three
of a crystal-converter may enable increasing the conversion efficiency three times from \( \eta_2 = 0.15 \) to 0.45.

Moreover, it is seen in Figure 3 that in conditions of a real experiment, the maximum expected conversion efficiency in case of AgGa\(_{0.6}\)In\(_{0.4}\)Se\(_2\) crystal is more than conversion efficiency in AgGaSe\(_2\) crystal by 1.225 times and only on 0.0378% conversion efficiency in AgGa\(_{0.7}\)In\(_{0.3}\)Se\(_2\) is more than in AgGa\(_{0.6}\)In\(_{0.4}\)Se\(_2\) crystal.

Thus, theoretical investigation of frequency conversion in mixed crystals with regard to phase effects enables finding out the ways of increasing conversion efficiency. And, namely, at the given value of crystal converter length it is possible to calculate the optimum value of pump intensity, as well as to calculate the coherent length of crystal-converter at chosen pump intensity of laser radiation. The analytical method also permits estimating an expected conversion efficiency at different wavelengths of lesser radiation. The angular width of phase matching at different concentrations of indium has been estimated for crystals of mixed type. The condition for increasing degree of uncritical angular phase matching is possible to choose.

4. Conclusion

The results of the studies carried out will permit elaborating the reliable highly efficient generators of second harmonic in CO\(_2\) lasers. The method of analysis for second harmonic generation in AgGa\(_{0.6}\)In\(_{0.4}\)Se\(_2\) crystals developed in the present work may be used for considering the process of frequency conversion in other perspective crystals of IR-range, as well as for examination of nonlinear optical waves in similar crystals.

Conflict of Interests

The author declares that there is no conflict of interests regarding the publication of this paper.
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