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Conversion Efficiency in AgGa($Se_{1-x}S_x$)$_2$ Crystals

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Abstract: ***In the work there has been investigated second harmonic generation of CO_2 laser radiation in AgGa($Se_{1-x}S_x$)$_2$ crystals in the constant-intensity approximation of the fundamental radiation. This approximation permits to take into account the influence of phase effects on the process of frequency conversion. A comparison was made of the obtained results on conversion efficiency with the corresponding experimentally measured values. It is shown that by choosing the optimum parameters of the task, it is possible to increase efficiency of conversion to second harmonic in these crystals. The conditions for the uncritical angular phase matching regime in AgGa($Se_{1-x}S_x$)$_2$ have been investigated.***

Keywords: second harmonic generation, IR-range, mixed type crystal, constant-intensity approximation.

1. Introduction

Searching for the prospect materials for the tasks of modern nonlinear optics continues. Nonlinear crystals of the IR-range of spectrum take a particular place [1]–[7]. Recently the nonlinear crystals of mixed type have turned to be the subject of the researches. It is connected with that realization of the efficient tuning of radiation in a broad range of spectrum needs in the nonlinear crystals for which the condition of uncritical phase matching is kept in the chosen range of spectrum. In compounds of mixed type on the account of increasing the content of one element and decreasing of the other's content, the possibility of elaboration of crystals with uncritical phase matching on the chosen radiation wavelength is experimentally shown [8]-[10].
Among the crystals of mixed type we can cite $AgGa_xIn_{1-x}Se_2$ [8], AgGa($Se_{1-x}S_x$)$_2$ [9] and $Zn_{1-x}Mg_xSe$ [10]. In the cited works the optical properties of crystals have been investigated, the coefficients in Sellmeyer's equations by means of which the curves of phase matching were plotted for generation of second harmonic of laser, have been experimentally determined.
For the purpose of research of nonlinear properties of crystals it is expedient to work in the constant-intensity approximation [11], taking into account a change of phases of all interacting waves. The given approximation allows one to examine the features of the nonlinear process, which are impossible to expose at analysis in a widely spread constant-field approximation [12]-[14].

In the present article it is reported on the results of researches in the constant-intensity approximation for nonlinear interaction of waves in the process of generation of second harmonic in AgGa($Se_{1-x}S_x$)$_2$ crystals.

2. Theory

We solve the task for the case of plane waves of pump with complex amplitude A_1 at frequency ω_1 and second harmonic with complex amplitude A_2 at frequency ω_2 ($\omega_2 = 2\omega_1$). An analysis of harmonic generation in a medium could be described by the following known reduced equations (interaction oo→e) [12]-[14]:

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

In this equation $\delta_{1,2}$ are absorption coefficients, $d_{1,2eff}$ are efficient nonlinear coefficients for the case of oo→e scalar phase matching, $\lambda_{1,2}$ stand for wavelengths of pump and second harmonic waves, $n(\omega_{1,2})$ are crystal refraction indices, $\Delta = k_2 - 2k_1$ is phase mismatch, and $k_{1,2}$ are values of wave vectors at frequencies $\omega_{1,2}$, respectively. The investigation supposes the following boundary conditions:

$$A_1(z=0) = A_{10}\exp(i\varphi_{10}), \quad A_2(z=0) = 0\,, \qquad (2)$$

where $z = 0$ corresponds to an entry to the crystal, and φ_{10} is an initial phase of pump wave at entry to medium.
We solve the system of reduced equations (2), first by differentiating the second equation of harmonic amplitude and then by applying the constant intensity approximation $\left(\mathrm{I}_1(\mathrm{z}) = \mathrm{I}_1(\mathrm{z}=0) = \mathrm{I}_{10},\ \mathrm{I}_2(\mathrm{z}) = \mathrm{I}_2(\mathrm{z}=0) = \mathrm{I}_{20}\right)$. As a result, for efficiency of conversion to second harmonic at the exit of crystal ($z = \ell$) we obtain [11]:

$$\begin{aligned}&\eta_2(\ell) = I_2(\ell)/I_{10} =\\ &= \gamma_2^2 I_{10}\ell^2 \operatorname{sinc}^2\lambda\,\ell\ \exp\left[-\left(\delta_2 + 2\delta_1\right)\ell\right],\end{aligned} \qquad (3)$$

where

$$\lambda^2 = 2\Gamma^2 - \left(\delta_2 - 2\delta_1 + i\Delta_1\right)^2/4, \qquad \Gamma^2 = \gamma_1\gamma_2 I_{10},$$

$$\operatorname{sinc} y = \sin y / y, \qquad I_j = A_j A_j^*.$$

Here $\gamma_1 = \dfrac{8\pi^2 d_{1eff}}{\lambda_1 n(\omega_1)}$, $\gamma_2 = \dfrac{4\pi^2 d_{2eff}}{\lambda_2 n(\omega_2)}$ are the second order coupling coefficients of interacting waves.
From (3) the expression for efficiency (η_2) is obtained for $\gamma_1 = 0$, $\delta_j = 0$ in the constant-field approximation, i.e. the

result in the absence of pump depletion.

It is well-known that for maintaining the regime of uncritical phase matching it is necessary to exclude or decrease the amount of the impact of factors that affect the width of phase matching. For estimation of this let's calculate first the refraction indices for ordinary and extraordinary waves at frequencies ω_1 and ω_2 using the coefficients of Sellmeyer equation for the main values of the refraction indices [9], and also phase matching angle for the given compound at different values of x which presents the concentration of sulfur in crystal. Then we will define the angular dispersion coefficient of the first order for all the considered versions. The results of the calculations for $AgGa(Se_{1-x}S_x)_2$ crystal are presented in the Table for three values of wavelengths of IR-range radiation. The choice of the values of wavelengths is dictated by the experiment conditions [9], [15] where there was considered frequency conversion from 3 sources of pumping on laser transition of CO_2.

3. Results and discussion

The further analysis of second harmonic generation is performed for doubling frequency in $AgGa(Se_{1-x}S_x)_2$ crystals for three values $x = 0$, 0.2 and 1 as example. Such a choice of x values is made because we do not have experimental data for other values of concentration for this crystal at the present time. To investigate the ways of increasing the efficiency of frequency conversion of CO_2 laser radiation in the IR-range we've carried out numerical calculations for analytical expression (3).

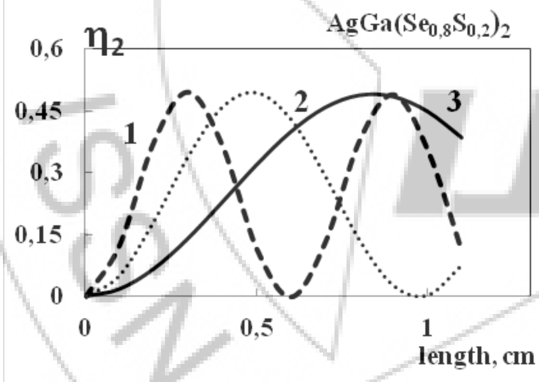

Figure 1: Dependences of conversion efficiency of radiation energy of pump wave (λ =9.55 mcm and 9.64 mcm) to energy of wave of second harmonic η_2 on lengths of ℓ for $AgGa(Se_{0.8}S_{0.2})_2$ crystal calculated in the constant-intensity approximation for $\delta_{1,2} =$ 0.02 cm^{-1} and $\Delta = 0.06$ cm^{-1} at pump intensity of I_{10} = 0,5 MW/cm^2 (curve 3), 1.5 (curve 2) and 4.0 (curve 1) [9], [15].

In Fig. 1 dependencies of frequency conversion efficiency on crystal length $\eta_2(\ell)$ are displayed. Three versions of conversion are considered in $AgGa(Se_{0.8}S_{0.2})_2$ crystal, differed by intensities of CO_2 laser pump, radiating at wavelength in 9.55 mcm and 9.64 mcm. The choice of these wavelengths of radiation is connected with an existence of the experimental data on the given crystal namely on these wavelengths. According to numerical account (3) the value of conversion efficiency at chosen wavelengths differs slightly, in order of one hundredth of one percent.

From behavior of curves, differed from monotonous behavior in case of the constant-field approximation, it follows that there exists an optimum value of a crystal length, at which conversion efficiency is maximum. Just as it was expected as far as increase of pump intensity, maximum of conversion is achieved at lesser crystal lengths, i.e., with increasing intensity of pump, coherent length of crystal decreases. For comparison, experimental value of conversion efficiency at crystal length of 0.2 cm, pump intensity equal to 0.5 MW/cm^2 and at wavelength in 9.55 mcm, makes up 4.1% [9], [15]. In our case the corresponding value is equal to 6.59% (see, curve 3). In case of wavelength in 9.64 mcm, experimental value of conversion efficiency at crystal length of 0.2 cm and at pump intensity equal to 30 MW/cm^2, makes up 1.5% [9], [15]. From Fig. 1 the corresponding value for efficiency is equal to 3.9 %. One of the reasons of existing difference in efficiency values may be lack of information on a number of the important parameters of the experiment.

In Fig. 2 there are cited the dependencies η_2 on pump intensity for two values of losses and three values of phase mismatch. As is seen in figure CO_2 laser radiation, generating at wavelength in 9.55 mcm, reaches maximum efficient conversion to wave of second harmonic at optimum value of pump intensity. By comparing of curves 1 and 3 (at identical losses) it is seen that the optimum value of pump intensity reduces as phase mismatch increases.

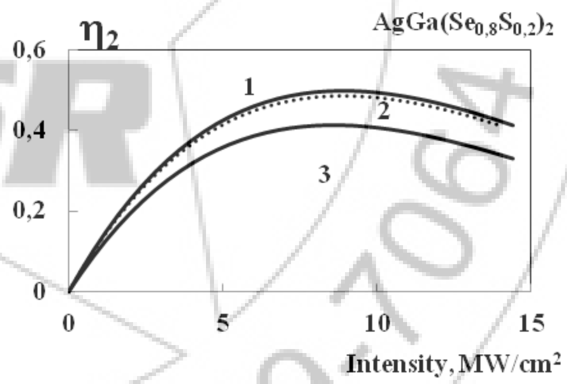

Figure 2: Dependences of conversion efficiency of radiation energy of pump wave (λ =9.55 mcm) to energy of wave of second harmonic in $AgGa(Se_{0.8}S_{0.2})_2$ crystal η_2 as a function of the pump intensity calculated in the constant-intensity approximation at crystal length of $\ell =$ 0.2 cm [9] for $\delta_2 = 2\delta_1 = = 0.02$ cm^{-1} (curves 1 and 3) and 0.07 cm^{-1} (curve 2) for $\Delta = 0.06$ cm^{-1} (curve 1), 1.8 cm^{-1} (curve 2) and 6.7 cm^{-1} (curve 3).

As is known, in mixed structures of $AgGa(Se_{1-x}S_x)_2$ type, crystal properties undergo the influence of a value of a parameter x, i.e. relative contents of initial $AgGaS_2$ and $AgGaSe_2$ crystals in the given compound [9].

In Fig. 3 there are shown the results of the analysis in the constant-intensity approximation of the process of frequency conversion $\eta_2(\vartheta)$ in case of three different values of content of sulphur relative to selenium, for x = 0, 0.2 and 1. The analysis of conversion in considering cases, for example, x = 0.1 and 0.4 and etc. has not been made because of the lack of experimentally measured values d_{36} (see table). The frequency conversions at the wavelength of CO_2 laser pump, equal to 9.55 mcm are considered.

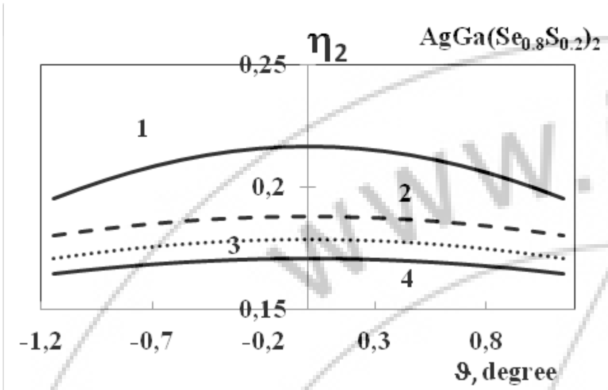

Figure 3: Dependences of conversion efficiency of radiation energy of pump wave (λ=9.55 mcm) to energy of wave of second harmonic in $AgGa(Se_{1-x}S_x)_2$ crystals η_2 as a function of the phase mismatch calculated in the constant-intensity approximation at $\delta_{1,2}$ = 0.1 cm^{-1} and pump intensity of I_{10} = 1,5 MW/cm^2 [15] for at crystal length of ℓ = 0.2 cm [9] for x = 0 (curve 3), 0.2 (curve 2) and 1 (curve 1).

From the behavior of curves 1-3 it is seen that dependency becomes smoother as the parameter x increases. If in case of $AgGaSe_2$ (x =0) angular dispersion coefficient of the first order is equal to 0.047185 ang. min^{-1} · cm^{-1}, then for $AgGaS_2$ (x = 1) this parameter is already equal to 0.079066 ang. min^{-1} · cm^{-1} (see Table). This fact confirms that the uncritical regime of crystal under the following condition of phase matching is fulfilled better with an increase of sulphur concentration. However, choosing this compound has the disadvantage of the decrease of nonlinear susceptibility as result. The further experimental investigations of the data of the perspective nonlinear crystals $AgGa(Se_{1-x}S_x)_2$ will permit to determine the components of tensor of nonlinear susceptibility for the various concentrations of sulphur. And this, in its turn, will allow one to calculate analytically in the constant-intensity approximation the optimum relative content of sulphur in the given prospect compound and to give the recommendations to the elaborators of laser devices.

4. Conclusion

Thus, the results of studying nonlinear interaction of waves with regard for changes of interacting wave phases in the process of second harmonic generation in $AgGa(Se_{1-x}S_x)_2$ crystals permit to state the following. By a choice of the optimum values of nonlinear medium length, pump intensity, phase mismatch and taking the influence of linear losses in a medium into consideration it is possible to increase efficiency of conversion to second harmonic in these crystals of mixed type and to select conditions for fulfilling or increasing a degree of uncritical angular phase matching.

The results of the studies carried out permit to state that further experimental investigations of the perspective $AgGa(Se_{1-x}S_x)_2$ crystals are necessary. Knowledge of tensor components for different content of Se will allow to give recommendations on working out the reconstructable laser frequency converters in the IR -range of spectrum.

The developed method can be used to study other nonlinear optical processes, e.g., sum frequency, parametric generation, etc. in other perspective crystals.

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Table 1: Calculated data for AgGa(Se1-xSx)2 crystal at SHG in the IR

x	λ, mcm	n_o^{ω}	n_e^{ω}	$n_o^{2\omega}$	$n_e^{2\omega}$	d_{36}, pm/V	Phase matching type	ϑ_s, degree	angular dispersion coefficient of first order, cm^1·ang.min.$^{-1}$
x =0	9.31	2.597798	2.564778	2.615886	2.583349	39.0[12]	oo→e	47.9672	0.062275
x =0	9.55	2.596647	2.563607	2.615434	2.582876	39.0[12]	oo→e	49.1893	0.047185
x =0	9.64	2.596208	2.56316	2.615267	2.582701	39.0[12]	oo→e	49.6679	0.046629
x =0.1	9.31					no data	oo→e		
x =0.1	9.55	2.57121	2.539504	2.591704	2.560677	no data	oo→e	54.1457	0.43463
x =0.1	9.64	2.570707	2.538984	2.591535	2.50502	no data	oo→e	54.7939	0,042725
x =0.2	9.31					33.5 [9]	oo→e		
x =0.2	9.55	2.551289	2.514641	2.626432	2.608496	33.5 [9]	oo→e	51.62306	0.052303
x =0.2	9.64	2.550734	2.514072	2.57347	2.537352	33.5 [9]	oo→e	52.2384	0.051536
x =0.4	9.31					no data	oo→e		
x =0.4	9.55	2.501129	2.46417	2.527275	2.490613	no data	oo→e	57.3614	0.050338
x =0.4	9.64	2.500465	2.463499	2.527078	2.490413	no data	oo→e	58.17535	0.049188
x =1.0	9.31					12.0	oo→e		
x =1.0	9.55	2.362347	2.308471	2.399047	2.345804	12.0	oo→e	55.70351	0.079066
x =1.0	9.64	2.361373	2.307477	2.398805	2.34556	12.0	oo→e	56.56404	0.077336

Author Profile

Rena J. Kasumova received the Ph.D. and Dr. of Sciences degrees in Quantum Electronics from Moscow Physical Engineering Institute in 1982 and from Baku State University in 2003, respectively. Since 1979 she is currently working at Physics Department of Baku State University, Azerbaijan. She investigates the optical wave interactions in nonlinear media.