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SHG in IR region in mixed $Zn_{1-x}Mg_xSe$ crystals

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Abstract: In the present work the results of investigations of the influence of different parameters on conversion efficiency in mixed $Zn_{1-x}Mg_xSe$ crystal in conditions of existing experiments are cited. The comparison of the obtained results on conversion efficiency with the analogous experimentally measured values has been made. The results of calculation of the angles of phase matching and angular dispersion coefficients for $Zn_{0.52}Mg_{0.48}Se$ crystal on wavelengths of the IR range are cited. The angular dispersion coefficients for $Zn_{1-x}Mg_xSe$ have been calculated. The angular width of phase matching has been estimated.

Keywords: Second Harmonic Generation, IR Region, Constant-Intensity Approximation

1. Introduction

The current level of progress of society is determined by volume of transmitted information. Advancement of information technologies, creation of global computer net of Internet favor to it. One of the most prospect and actively developing orientations of science and technology for implantation of these tasks is elaboration of relatively inexpensive compact sources of frequency tunable radiation throughout the infrared, visible, and ultraviolet spectral regions [1].

Up to present there have been reached the considerable achievements in application of nonlinear crystals in the numerous facilities in the infrared range. Among them it is possible to note the perspective single-crystalline $A^{II}B^{VI}$ compounds. For instance, hexagonal single crystals of CdS and CdSe compounds and their solid solutions $CdS_{1-x}Se_x$ and ZnSe, $Zn_{1-x}Mg_xSe$ crystals [1-6]. As is known by choice of the content of particular elements (parameter x) in like compounds of mixed type it is possible to tune of band gap energies, crystalline structure and lattice constants in a wide range of values [3]. On the base of mixed crystals there are worked out tunable by frequency lasers which are actual, in particular at remote sounding of atmosphere and the tasks of highly sensitive spectroscopy.

For study of nonlinear optical properties of the investigated type of crystal it is expedient to resort to the constant–intensity approximation [7-8] 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 the present work the conditions of optimum frequency conversion in the IR range of spectrum for case of second harmonic generation in $Zn_{1-x}Mg_xSe$ 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 $Zn_{0.52}Mg_{0.48}Se$ crystal on different wavelengths of the IR range are presented of the angles of phase matching.

2. Theory

Let us consider the situation in which optical waves at frequencies ω_1 and ω_2 interact in a dissipative nonlinear optical medium to produce an output wave at the second harmonic frequency $\omega_2=2\omega_1$, i.e. a laser beam (ω_1) is incident upon a crystal for which the second -order $\chi^{(2)}$ is nonzero. For simplicity, we assume that the interacting waves are the plane waves for the second harmonic generation involving collimated, monochromatic, continuous -wave beams propagating in the +z direction. Here theoretical analysis of the condition of scalar phase matched second harmonic generation (ee-o interaction) in positive uniaxial single-crystalline $Zn_{1-x}Mg_xSe$ has been considered.

The coupled -amplitude equations describing second harmonic generation have the form [9-10]

$$\frac{dA_1}{dz} + \delta_1 A_1 = -i\frac{8\pi^2 d_{1eff}}{\lambda_1 n(\omega_1)} A_2 A_1^* \exp(-i\Delta z),$$
$$\frac{dA_2}{dz} + \delta_2 A_2 = -i\frac{4\pi^2 d_{2eff}}{\lambda_2 n(\omega_2)} A_1^2 \exp(i\Delta z), \quad (2.1)$$

the $A_{1,2}$ are complex amplitudes of laser pump wave and its second harmonic wave at frequencies $\omega_{1,2}$ correspondingly, $\delta_{1,2}$ are absorption coefficients, k_1 and k_2 are known as the modulus's of wave vectors at frequencies $\omega_{1,2}$. $\Delta=k_2-2k_1$ is phase mismatch, $d_{1,2eff}$ denote the effective nonlinear coefficients for ee-o scalar phase-matched, $\lambda_{1,2}$ are wavelengths. $n(\omega_{1,2})$ are refractive indexes in this crystal at frequencies $\omega_{1,2}$ correspondingly.

We assume that the ω_2 field is not present at the input, only the amplitude A_1 of the input ω_1 field must be taken nonzero, so that boundary conditions become

$$A_1(z=0) = A_{10}\exp(i\varphi_{10}), \quad A_2(z=0) = 0, \quad (2.2)$$

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

We next find the solution that satisfies the appropriate boundary conditions. These equations (2.1) in the constant-intensity approximation for the boundary conditions (2.2) possess the next solution for complex amplitude of second harmonic at the output the crystal ($z=l$) [7-8].

$$A_2(\ell) = -i\gamma_2 A_{10}^2 \ell \,\mathrm{sinc}\lambda\ell \exp\left[2i\varphi_{10} - (\delta_2 + 2\delta_1 - i\Delta_1)\ell/2\right], \quad (2.3)$$

where we have introduced the quantities

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

$\mathrm{sinc}x = \sin x / x$, $I_j = A_j A_j^*$,

$\gamma_1 = \frac{8\pi^2 d_{1eff}}{\lambda_1 n(\omega_1)}$, $\gamma_2 = \frac{4\pi^2 d_{2eff}}{\lambda_2 n(\omega_2)}$ are nonlinear coupling coefficients.

As it follows from (2.3) there takes place the basic and harmonic fields energy interchange periodically, as a result of spatial variations of the second harmonic field is observed. This time, the minimums of harmonic intensity beating, as an analysis shows in the constant-intensity approximation, depend on nonlinear susceptibilities of crystal [11]. Thus fact permits to define the nonlinear susceptibilities of substances by a simple way, more precise than in the constant-field approximation.

From (2.3) the optimum length of crystal-converter may be received. According to the theoretical analysis made in the constant -intensity approximation in contrast to the results of the constant-field approximation, an optimum length of crystal, i.e. coherent length of nonlinear medium depends on pump intensity I_{10} and losses in a medium [7-8].

At γ_1=0 and δ_j=0 from (2.3) we have an expression for conversion efficiency in the constant-field approximation.

The efficiency $\eta_2(l)$ for conversion of power from the ω_1 wave to the ω_2 wave can be defined by $\eta_2(\ell) = I_2(\ell)/I_{10}$ and from Eq. (2.3), we see that for the values given above, conversion efficiency is

$$\eta_2(\ell) = \gamma_2^2 I_{10}\ell^2 \,\mathrm{sinc}^2\lambda\,\ell \;\exp\left[-(\delta_2 + 2\delta_1)\ell\right]. \quad (2.4)$$

To study the ways of increasing frequency conversion efficiency in $Zn_{1-x}Mg_xSe$ crystal of laser radiation in IR range, we'll make the numerous calculation of the analytical expression for conversion efficiency (2.4), received in the constant-intensity approximation. The parameters of the task are chosen according to conditions of existing experiments for the given crystal [1-2].

3. Results and Discussion

In Figs. 1-3 the dynamic process of frequency conversion is shown to the second harmonic in $Zn_{0.67}Mg_{0.33}Se$ crystal. The value of quadratic susceptibility for this crystal is experimentally measured in the work [1] and is equal to $\chi_{SHG}^{(2)}$ =0.86 ×10^{-8} esu. The thickness of the investigated samples was about 1mm [1]. Experimental value of pump intensity was changed in the range 0 ÷ 1 GW/cm^2 [1].

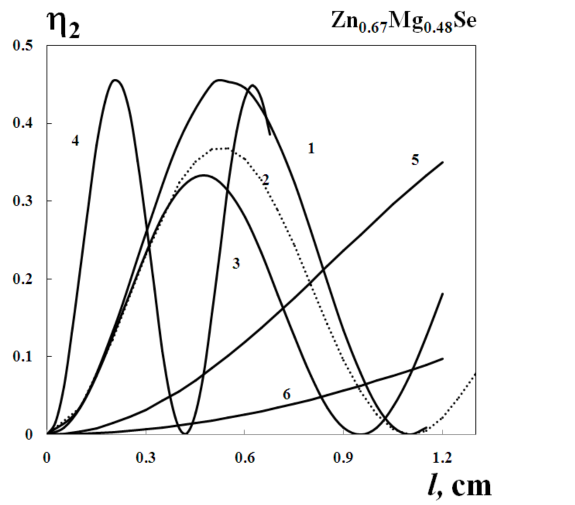

Fig. 1. Dependences of conversion efficiency of radiation energy of pump wave (λ=1.06 mcm) to energy of wave of second harmonic η_2 on lengths for $Zn_{0.67}Mg_{0.33}Se$ crystal l calculated in the constant-intensity approximation for $\Delta/2\Gamma$=0,5 (curves 1 and 2), 1 (curve 3) and 1.5 (curves 4-6), $\delta_{1,2}$=0 (curves 1, 3-4), 0.1 cm^{-1} (dashed curve 2) and pump intensity of I_{10}= 3.5 GW/cm^2 (curve 4), 0.5 GW/cm^2 (curves 1-3), 0.05 GW/cm^2 (curve 5), 0.01 GW/cm^2 (curve 6).

In Fig. 1 the dependencies η_2 on length of $Zn_{0.67}Mg_{0.33}Se$ are displayed. There are considered six versions of conversion differed by pump intensity of Nd:YAG laser (on wavelength in 1.064 mcm), losses and phase mismatch. From behavior of curves differed from monotonous increase in case of the constant-field approximation it follows that there exists optimum value of crystal length at which conversion efficiency is maximum. With growth of losses the conversion efficiency falls down (compare curves 1 and 2). With increase in phase mismatch between interacting waves two times an efficiency, as was expected, decreases by 20 %, as well optimum length of crystal decreases (compare curves 1 and 3). From comparison of curves 4-6 it is seen that with decrease of pump intensity the efficiency decreases on one order.

In addition, the period of spatial beatings and optimum length of crystal increase. The latter is explained by the fact that at lower values of pump intensity for achievement of maximum conversion longer geometrical lengths of crystal are required. The length of crystal sample in experiment (1 mm) corresponds to the initial part of considered theoretical dependence $\eta_2(l)$, where efficiency is low. As is seen in figure, an increase in length of a sample approximately twice would permit to increase conversion efficiency several times.

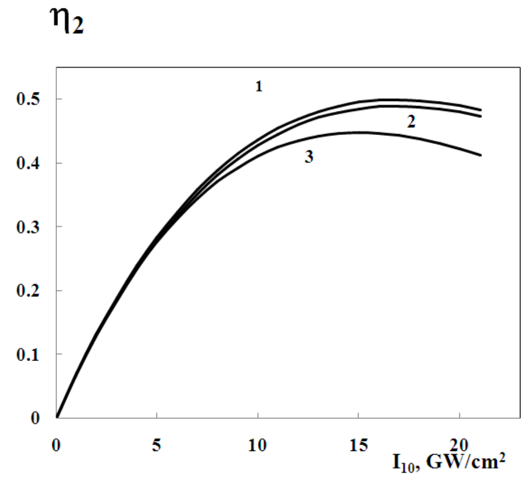

Fig. 2. *Dependences of conversion efficiency of radiation energy of pump wave (λ=1.06 mcm) to energy of wave of second harmonic η_2 on intensity I_{10} for $Zn_{0.67}Mg_{0.33}Se$ crystal l calculated in the constant-intensity approximation for $\delta_{1,2}=0$ and $\Delta/2\Gamma=0{,}5$ (curve 3), 0.05 (curve 2) and 0.005 (curve 1) at crystal length of l =0.1 cm [6] (curves 1-3).*

In Fig. 2 the dependencies η_2 on pump intensity for three values of phase mismatch of interacting waves are presented. As is seen in figure, laser radiation maximum efficiently converts into wave of second harmonic at optimum value of pump intensity. From comparison of curves 1, 2 and 3 it is seen that an optimum value of pump intensity falls down with increase of phase mismatch. It should be noted that analysis in the constant-intensity approximation shows that on crystal l=1 mm long the maximum conversion might be received at pump intensity of greater value on one order (10 GW/cm^2) than in case realized in the experiment. An increase of crystal length two times to 2 mm would permit to receive the maximum efficiency at pump intensity (3.5 GW/cm^2) exceeding the experimental one approximately 3 times (see curve 4). For comparison, in work [4] there has been obtained 15 % the slope-efficiency of conversion with respect to the absorbed energy in $Zn_{1-x}Mg_xSe$ of 5 mm length.

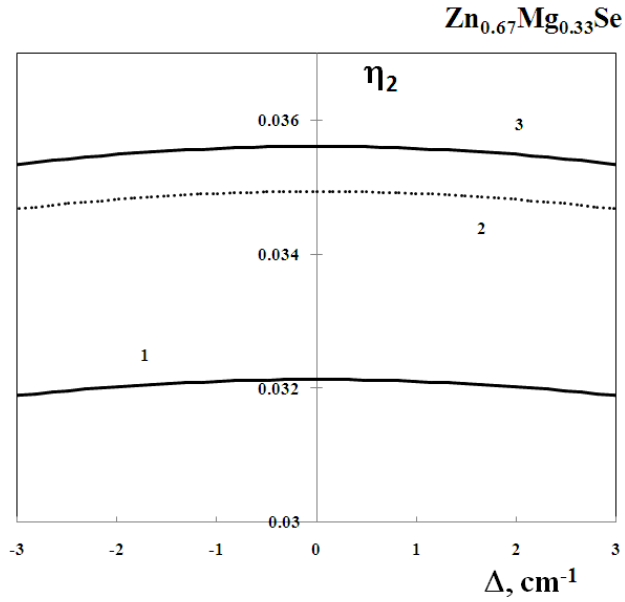

Fig. 3. *Dependences of conversion efficiency of radiation energy of pump wave (λ=1.06 mcm) to energy of wave of second harmonic for $Zn_{0.67}Mg_{0.33}Se$ crystal η_2 as a function of the phase mismatch Δ calculated in the constant-intensity approximation at pump intensity of I_{10}=0,45 GW/cm^2 (curve 1) and 0.5 GW/cm^2 (curves 2-3) for $\delta_2=2\delta_1=0$ (curves 1 and 3) and 0,05 cm^{-1} [6] (curve 2) and crystal length of l=0.1 cm [6] (curves 1-3).*

In Fig. 3 cited are the dependencies of conversion coefficient to second harmonic on phase mismatch of interacting waves. As is seen in figure an increase of conversion efficiency is observed at rise in pump intensity (curves 1 and 3) and decrease of losses (curves 2 and 3).

To estimate an angular width of phase matching $\Delta\theta$ in $Zn_{1-x}Mg_xSe$ crystals let's use the results of the following experiment. In Ref. 5 for 0.63 mcm, 1.06 mcm and 10.6 mcm wavelengths Sellmeier's coefficients are experimentally established for $Zn_{0.52}Mg_{0.48}Se$ crystal. Using the values of these coefficients we'll calculate the angles of phase matching in case of second harmonic generation for this crystal (case x=0.48) in the range of wavelength of pump radiation 5 mcm ÷ 12 mcm [2]. To determine the angular width of phase matching we'll calculate the angular disperse coefficient according to [12].

Below in table the results of calculations are cited.

Table. *The angular dispersion coefficients for $Zn_{0.52}Mg_{0.48}Se$.*

Crystal	λ, mcm	n_o^ω	n_e^ω	$n_o^{2\omega}$	$n_e^{2\omega}$	$\chi^{(2)}_{SHG}$, esu	Phase matching type	θ_s, degree	Angular dispersion coefficient of first order, cm^{-1} $ang.min.^{-1}$
$Zn_{0.52}Mg_{0.48}Se$	5.3	2.15627	2.165399	2.154113	2.18779	No data	ee→o	86.157111	0.007469
$Zn_{0.52}Mg_{0.48}Se$	6.0	2.151281	2.168459	2.164986	2.182786	No data	ee→o	63.447038	0.038765
$Zn_{0.52}Mg_{0.48}Se$	9.5	2.148363	2.164994	2.152627	2.171426	No data	ee→o	35.698993	0.028474
$Zn_{0.52}Mg_{0.48}Se$	10.6	2.147977	2.164994	2.152627	2.16987	No data	ee→o	31.684177	0.02401

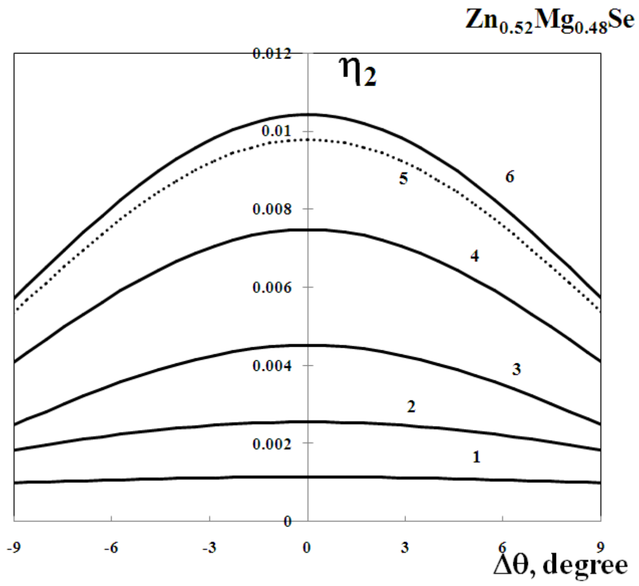

Fig. 4. *Dependences of conversion efficiency of radiation energy of pump wave (λ=10.6 mcm) to energy of wave of second harmonic for $Zn_{0.52}Mg_{0.48}Se$ crystal η_2 as a function of the phase mismatch $\Delta\theta$ calculated in the constant-intensity approximation for $\delta_2=2\delta_1=0.1$ cm^{-1} at pump intensity of I_{10}= 1.5 GW/cm^2 (curves 1-3), 2.5 GW/cm^2 (curve 4) and 3.5 GW/cm^2 (curves 5-6) and crystal length of l=0.1 cm [1] (curve 1) and 0.15 cm (curve 2) and 0.2 cm (curves 3-6).*

Using the results from table in Fig. 4 the dependencies of conversion efficiency on deviation angle from direction of phase matching $\eta_2(\Delta\theta)$ are given. Because of lack of experimentally measured of a value of quadratic susceptibility for $Zn_{0.52}Mg_{0.48}Se$ crystal at calculations it was supposed that for crystals with parameter x=0.33 and 0.48 these values are of one order. As is seen in figure an increase of conversion efficiency approximately two times an increase of pump intensity twice is observed (curves 2 and 3). The analysis of the analytical expression (2.4) for conversion efficiency in the constant-intensity approximation, showed that the width of angular phase matching depends on pump intensity and length of crystal.

As is seen in figure an increase in conversion efficiency approximately two times is observed at increasing pump intensity almost twice (curves 3 and 4). From comparison of curves 1, 2 and 3 it follows that with increase in length of nonlinear medium efficiency rises.

The width of angular phase matching is reduced with increase in pump intensity (compare curves 3, 4 and 6) and length of nonlinear medium (compare curves 2 and 3). As is seen from dependence $\eta_2(\Delta\theta)$ at angular mismatch from direction of phase matching for some degrees at experimentally considered values of parameters (pump intensity 1 GW/cm^2 and the length of crystal 0.1 cm [1]) the changes of conversion efficiency are inconsiderable and make up a value in 0.17 % (curve 1).

Hence, from the numerical analysis of the analytical expression (2.4) for conversion efficiency obtained in the constant-intensity approximation it follows that angular phase matching width depends on pump intensity and the length of crystal.

4. Conclusion

Thus, theoretical investigation of frequency conversion in $Zn_{1-x}Mg_xSe$ crystal with account for phase effects allows one to revel the ways of increasing conversion efficiency. Namely at the given values of the length of crystal - converter it is possible to calculate the optimum value of pump intensity. As well at chosen pump intensity of laser - radiator it is possible to calculate the coherent length of a crystal-converter. The analytical method also permits to estimate an expected conversion efficiency on different wavelengths of laser radiation. The results of calculation of the angles of phase matching and angular dispersion coefficients for $Zn_{0.52}Mg_{0.48}Se$ crystal on wavelengths of the IR range have been obtained.

The results of carried out researches will be useful at elaboration of the tunable highly efficient generators of second harmonic of lasers of the IR-range. Developed in the present work method of analysis of second harmonic generation in $Zn_{1-x}Mg_xSe$ crystals may be used for investigation of parametric and intracavity interaction of nonlinear optical waves.

PACS

42.65.-k; 42.70.Mp; 42.79.Nv

References

[1] B. Derkowska, Z. Essaidi, B. Sahraoui, A. Marasek, F. Firszt, M. Kujawa. Nonlinear optical properties of $Zn_{1-x}Mg_xSe$ and $Cd_{1-x}Mg_xSe$ crystals, Optical materials, 31 (2009) 518-522.

[2] N. Kovalenko. $Zn_{1-x}Mg_xSe$: A promising material for non-linear optics. J. of Nonlinear Optical Physics and Materials, 20 (2011) 123-127.

[3] N.R. Grigoryeva, R.V. Grigoryev, E.P. Denisov, B.A. Kazennov, B.V. Novikov, D.L. Fedorov. X-ray diffraction and optical studies of $CdS_{1-x}Se_x$ solid solutions with stacking faults. Solid State Physics, 42 (2000) 1570-1578.

[4] M.E. Doroshenko, H. Jelikova, P. Koranda, J. Sulc, T.T. Basiev, V.V. Osiko, V.K. Komar, A.S. Gerasimenko, V.M. Puzikov, V.V. Badikov, and D.V. Badikov. Tunable mid-infrared laser properties of Cr^{2+}:ZnMgSe and Fe^{2+}:ZnSe crystals. Laser Phys. Lett., 7 (2010) 38-45.

[5] V.M. Puzikov, Yu.A. Zagoruiko, O.O. Fedorenko, T.T. Basiev, and N.O. Kovalenko. Optical and electro-optical properties of single ZnMgSe. Crystallography, 49 (2004) 272-273.

[6] B. Derkowska, B. Sahraoui, X. Nguyen Phu, G. Glowacki, and W. Bala. Study of linear optical properties and two-photons absorption in $Zn_{1-x}Mg_xSe$ thin layers. Optical materials, 15, Issue 3, (2000) 159-236.

[7] Z.H. Tagiev, and A.S. Chirkin, Fixed intensity approximation in the theory of nonlinear waves, Zh. Eksp. Teor. Fiz., 73 (1977) 1271-1282.

[8] 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 (2001) 84-87.

[9] S.A. Akhmanov, and R.V. Khokhlov, Problemy Nelineynoy Optiki [The Problems of Nonlinear Optics] (VINITI, Moscow, 1964).

[10] N. Blombergen, Nonlinear Optics (W.A. Benjamin, New York, 1965).

[11] Z.A. Tagiev, and R.J. Kasumova, Determination of Nonlinear High-Order Susceptibilities, Optics and Spectroscopy, 80 (1996) 848-850.

[12] V. G. Dmitriev, and L.V. Tarasov, Prikladnaya Nelineynaya Optika [Applied Nonlinear Optics] (Radio I Svyaz, Moscow, 1982)