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HIGH EFFICIENCY MIDDLE IR PARAMETRIC SUPERRADIANCE IN ZnGeP_2 AND GaSe CRYSTALS PUMPED BY AN ERBIUM LASERK.L. VODOPYANOV⁽¹⁾, L.A. KULEVSKII, A.I. GRIBENYUKOV* and K.R. ALLAKHVERDIEV***General Physics Institute, Vavilov str. 38, Moscow 117942, USSR***Siberian Branch Acad. Sci. USSR, Tomsk, USSR****The Institute of Physics, Acad. Sci. Azerbaijan SSR, Baku, USSR*

Abstract Parametric superradiant (resonator free) generation in nonlinear crystals of ZnGeP_2 and GaSe (type I phase matching) was obtained for the first time. As a pump source 110 ps pulses from the actively mode locked Er:YAG laser ($2.94 \mu\text{m}$) were used. The tuning range achieved was $4\text{-}10 \mu\text{m}$ (ZnGeP_2) and $3.5\text{-}18 \mu\text{m}$ (GaSe). In case of ZnGeP_2 (42 mm crystal) and type II phase matching the quantum efficiency reached 17.6% ,corresponding to the output peak power of 3 MW near $5\text{-}6 \mu\text{m}$.

1. Introduction

The distinctive feature of ZnGeP_2 and GaSe crystals proposed as materials for nonlinear optics is a wide transmission range ($0.74\text{-}12$ and $0.65\text{-}18 \mu\text{m}$ accordingly) and extremely high second order nonlinearities^{1,2}. The nonlinear figure of merit d^2/n^3 for ZnGeP_2 , for instance, is about 100 times higher than the corresponding value for LiNbO_3 and 20 times higher than that for AgGaS_2 crystal³.

2. Experimental set-up

Solid state erbium lasers with an i.r.generating wavelength near $3 \mu\text{m}$ are well suited as a pumping source for parametric devices based on ZnGeP_2 and GaSe crystals due to their good transparency in this range (absorption coefficient less than 0.1 cm^{-1}) and the existence of phase matching conditions for three wave interactions. We used in our experiment actively mode-locked cavity dumped Er:YAG ($\lambda=2.94 \mu\text{m}$)⁴ or Er:YSGG ($\lambda=2.79 \mu\text{m}$)⁵ lasers with typical pulse duration of 100 ps and output energy of 0.5-2 mJ.

The laser radiation was focused onto the nonlinear crystal using one or two CaF_2 lenses. The output PSR signal was monitored with the Ge-Au detector (77 K); an InAs filter was used to cut down the pump laser light.

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3. Experimental results

Fig.1 shows experimental and theoretical tuning curves for ZnGeP_2 and GaSe crystals. Most of other experimental data are given in the Table.

Fig.1 Angular tuning curves for $\lambda=2.94 \mu\text{m}$ pump. a) ZnGeP_2 , b) GaSe. Continuous curves - calculated from [3].

In case of ZnGeP_2 we had two crystals with different orientations, suited for type I and type II phase matching. For the second crystal the azimuthal angle $\varphi=31^\circ$ was not optimal - it should be 45° for getting maximum d_{eff} , according to the following equations³:

$$\text{Type I (o-ee)} \quad d_{\text{eff}} = d_{36} \sin 2\theta \cos 2\varphi$$

$$\text{Type II (o-eo)} \quad d_{\text{eff}} = d_{36} \sin \theta \sin 2\varphi$$

For the GaSe crystal, which was cleaved perpendicular to the z axis, we used Type I (e-oo) phase matching because smaller angles of incidence are needed, than for Type II. Here we have^{2,3}:

$$d_{\text{eff}} = d_{22} \cos \theta \cdot \sin 3\varphi.$$

In the case of ZnGeP_2 we have used $f=25 \text{ cm}$ for the focusing lens (Type I) or a 1:4 telescope formed by two lenses of 10 and 2.5 cm focal lengths (Type II). For GaSe we have used the combination of a 30 cm spherical lens and a 4 cm cylindrical lens to focus laser radiation mostly perpendicular to the $(\vec{k}\vec{z})$ plane because of the significant walk-off in the crystal, reaching 0.53 mm.

The calculated spectral width of PSR $\Delta\nu_{\text{PSR}}$, corresponding to the condition $\Delta k l = \pm\pi$ is also presented in the table. The experimentally measured $\Delta\nu_{\text{PSR}}$ value of 10 cm^{-1} in the case of ZnGeP_2 (Type II) is in good agreement with the calculated value. For Type I phase matching the PSR linewidth becomes larger, especially near degeneracy. The effective crystal length is calculated here taking into account the walk-off effect and linear absorption. The PSR threshold energy was as small as 0.35 GW/cm^2 in ZnGeP_2 ⁶ and an order of magnitude higher in GaSe⁷, mostly due to the small effective length. A properly designed geometry of the focusing optics can reduce the threshold to less than 1 GW/cm^2 for 12 mm long GaSe, since the nonlinear figures of merit differ only by a factor of 1.3 for these two crystals. It is known⁸ that the parametric gain increment is proportional to $(d_{\text{eff}}^2/n^3 l_{\text{eff}}^2 I_p)^{1/2}$. The close

values of the term $d_{\text{eff}}^2/n^3 l_{\text{eff}}^2 I_{\text{thr}}$ in the case of different crystals, especially for 11 mm long ZnGeP_2 and 12 mm long GaSe show that the experimental results are in good accordance with the values of nonlinear coefficients taken from literature. In the case of the 42 mm long ZnGeP_2 crystal the effective length l_{eff} is apparently still smaller than calculated due to possible stoichiometry (and birefringence) nonuniformity along the crystal.

Table

Crystal	ZnGeP_2 ($n_o < n_e$)		GaSe ($n_o > n_e$)
Interaction	Type I (o-ee)	Type II (o-eo)	Type I (e-oo)
Crystal orientation	$\theta=47^\circ$, $\varphi=0^\circ$	$\theta=84^\circ$, $\varphi=31^\circ$	$\theta=0^\circ$, $\varphi=90^\circ$
Effective nonlinearity, d_{eff} , cgs esu	$2.1 \cdot 10^{-7}$	$1.85 \cdot 10^{-7}$	$1.6 \cdot 10^{-7}$
Range of θ variation	$47 - 49.55^\circ$	$76 - 90^\circ$	$9.5 - 13^\circ$
Tuning range achieved, μm	4 - 10	5.2 - 5.6 6.2 - 6.7	3.5 - 18
PSR linewidth $\Delta\nu_{\text{PSR}}$, cm^{-1}	540 ($\lambda_i=5.9\mu\text{m}$) 80 ($\lambda_i=8\mu\text{m}$) 30 ($\lambda_i=10\mu\text{m}$)	7 ($\lambda_i=6.7\mu\text{m}$)	850 ($\lambda_i=5.9\mu\text{m}$) 63 ($\lambda_i=10\mu\text{m}$) 10 ($\lambda_i=15\mu\text{m}$) 6 ($\lambda_i=18\mu\text{m}$)
Pump beam waist parameter w_o , mm	0.28	0.1	Ellips 0.13 0.28
Crystal length l , mm	11	42	12
Walk-off, mm	0.14 (48°)	0.11 (84°)	0.53 (10°)
Effect. length l_{eff} , mm	11	19	3.2
PSR threshold intensity I_{thr} , GW/cm^2	0.5	0.35	6
$d^2/n^3 l_{\text{eff}}^2 I_{\text{thr}}$, a.u.	8.7	14.1	7.0
I_{max} used, GW/cm^2	6.5	30	12
Max. quant. efficiency	3%	17.6%	1%
I_{damage} , GW/cm^2	6.5	30	30

In the Fig.2 the energetic characteristics of PSR in the 42 mm long ZnGeP_2 crystal are shown ($\lambda_s = 5.6$, $\lambda_i = 6.2$ μm). The quantum efficiency had reached $\eta_{\text{PSR}} = 17.6\%$, although the crystal surfaces were not antireflection coated, the Fresnel losses being in the order of 45%

Fig.2.

Dependencies of the parametric superradiance (PSR) output energy E_{PSR} (1) and quantum efficiency η_{PSR} (2) on the laser pump intensity for ZnGeP_2 (42 mm)

4. Conclusion

Pumping of ZnGeP_2 and GaSe crystals with single picosecond erbium laser pulses ($\lambda \approx 3$ μm) generated intense continuously tunable radiation in the range of 4-10 μm (ZnGeP_2) and 3.5-18 μm (GaSe) with a frequency tuning practically up to crystals' longwave transmission limit. The quantum efficiency achieved (17.6 % for ZnGeP_2) reaches highest reported value for middle i.r. parametric superluminescent devices. The output peak power in the region of 5-6 μm reached 3 MW. This new sources of i.r. radiation can be successfully applied in the fields of molecular spectroscopy, atmosphere monitoring and different types of resonance interaction of radiation with matter.

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