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Energy and spectral characteristics of a parametric generator based on a nonlinear $ZnGeP_2$ crystal pumped by a Ho : YAG laser

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***Abstract.* We have experimentally realised the average generation power of a parametric generator based on a $ZnGeP_2$ single crystal, which constituted ~ 1.5 W at an efficiency of ~ 28.6 % in the wavelength range of 3.5–4.8 μm with an average pump radiation power of ~ 5.5 W and its energy density of ~ 0.47 J cm^{-2}. It is shown that there are threshold conditions for the average pump power, providing the realisation of parametric generation. In this case, an increase in the generation efficiency with an increase in the energy density of pump radiation to ~ 0.4 J cm^{-2} is observed; with its further increase, the output power increases due to a decrease in the parametric generation threshold. Parametric generators (PGs) based on $ZnGeP_2$ single crystals with such characteristics are promising for solving many applied problems, including generation of terahertz radiation when nonlinear crystals are pumped by radiation from these PGs at difference frequency.**

Keywords: *parametric generation, $ZnGeP_2$, nonlinear crystals, mid-IR radiation.*

1. Introduction

The sources of coherent radiation from the mid-IR range (λ = 3–8 μm) are widely used today in many fields of science and technology, including the processing of glasses, ceramics, semiconductor materials using the technology of thermal splitting and scribing [1, 2]. High-power sources of laser radiation with a discrete set of wavelengths and/or their smooth tuning over the spectral range are of considerable interest for sounding the atmosphere and remote determination of the composition of substances [3, 4], monitoring the ecological situation and determining the sizes of finely dispersed objects [5, 6]. Pulsed and repetitively pulsed mid-IR lasers with a pulse energy up to 1 mJ and a pulse duration of 10–100 ns possess great potential for medical applications, including resonant ablation of bone tissues [7–12] and low-invasive eye surgery [13, 14].

Laser emitters tunable in the wavelength range of 3–8 μm [15, 16] are of special interest for the implementation of high-efficient generation of THz radiation in the frequency range of 0.1–2 THz with an average output power up to 10 mW. To develop compact THz radiation sources operating on the principle of generating a difference frequency in nonlinear optical crystals (including $ZnGeP_2$), tunable narrow-band two-frequency lasers with a wavelength of 3–4 μm, similar to those developed for pumping GaSe crystals, are required [17]. One of the most effective ways of obtaining coherent radiation that meets the requirements of the above applications is the use of a parametric generator (PG) with a set of selective elements narrowing the lasing band.

To attain effective generation in PGs, it is necessary to use crystals possessing high optical transparency within a given wavelength range, large quadratic nonlinear susceptibility, high optical breakdown threshold, and good spatial homogeneity. In terms of a set of characteristics, the $ZnGeP_2$ single crystal is one of the most suitable for parametric generation [13, 18]. The absorption coefficient of this single crystal does not exceed 0.1 cm^{-1} in the spectral range of 2.5–8.3 μm [19]. In the range of radiation wavelengths that are generated by solid-state lasers (λ = 2.05–2.39 μm), the absorption coefficient of o-polarised radiation can be reduced to a level not exceeding 0.05 cm^{-1} (see Refs [20–25]). The radiation resistance of $ZnGeP_2$ at the wavelength of pump laser radiation (λ = 2.05 μm), with a pulse duration of ~ 10 ns and a pulse repetition rate of 10 kHz, constitutes ~ 0.074 GW cm^{-2} [26], and for erbium laser radiation (λ = 2.94 μm, pulse duration of 0.11 ns, pulse repetition rate of 1 Hz), the radiation resistance is ~ 30 GW cm^{-2} [27]. The $ZnGeP_2$ single crystal has a relatively large nonlinear susceptibility [$d = (70–85.4) \times 10^{-12}$ pm V^{-1}] and good thermal conductivity, which is especially important for the implementation of high average and peak radiation powers.

The aim of this paper is to determine the energy and spectral characteristics of a PG based on $ZnGeP_2$ single crystals pumped by a Ho : YAG laser, with the assessment of a possibility of the development of effective sources of radiation tunable in the wavelength range of 3–4 μm for the implementation of THz radiation in the $ZnGeP_2$ crystal pumped at difference frequency.

It should be noted that repetitively pulsed Ho : YAG lasers with resonant optical pumping by radiation from fibre or crystalline thulium lasers, for example, Tm : YLF lasers, are widely used as radiation sources with λ = 2.1 μm for pumping PGs based on the $ZnGeP_2$ single crystal. The conversion efficiency of the thulium laser radiation in such a system reaches ~ 50%, the average radiation generation power with λ = 2.1 μm constituting ~ 15 W [28–30].

2. 2. Experimental setup for PG investigation

The scheme of the experimental setup is shown in Fig. 1. The source of pumping PG based on the $ZnGeP_2$ single crystal is

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a repetitively pulsed Ho : YAG laser, which is pumped by a Tm : YLF laser light ($\lambda = 1.908$ μm) focused into the Ho : YAG crystal by the lens L1. The Ho : YAG laser resonator is formed by a dichroic mirror M1 with a reflection coefficient of ~ 99% at the laser wavelength and a transmittance of ~ 99% at the pump radiation wavelength, a highly reflecting mirror M2, an output spherical mirror M3 with a curvature radius of ~ 300 mm and a dielectric coating with a reflection coefficient of ~ 80% at the laser wavelength. The narrowing of the Ho : YAG laser generation spectrum is conducted by an interference-polarisation filter (IPF) placed in the resonator between the output (M3) and dichroic (M1) mirrors; in this case, the active *Q*-switching is performed using an acousto-optic modulator (AOM). Optical decoupling between the PG and Ho : YAG laser resonators is performed by an optical isolator (OI). In the experiments, we used a Ho : YAG laser with the following characteristics: the maximum average radiation power in the pulsed regime is 15 W, the pulse repetition rate is 10 kHz, and the FWHM pulse duration is 26 ns. The PG resonator is formed by mirror M4 with a transmittance coefficient of ~ 99% at the pump radiation wavelength and a reflection coefficient of ~ 99% at the laser wavelength, and mirror M5. This mirror has an antireflection coating with a transmittance of ~ 99% at the pump radiation wavelength and a beam-splitting coating with a reflection coefficient of 50% at the laser wavelength, which ensures implementation of single-pass pumping. The $ZnGeP_2$ single crystal with a length of 20 mm (LOK Ltd, Tomsk) was placed in a resonator formed by mirrors M4 and M5 with antireflection coatings applied to the working end-faces for wavelengths of 2.097 and 3.5 – 4 μm.

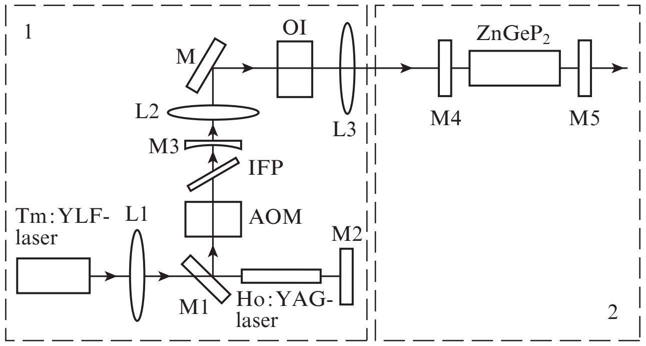


Figure 1. Scheme of the parametric generator based on the $ZnGeP_2$ single crystal pumped by a Ho : YAG laser beam. Part 1 of the setup: (L1, L2, L3) lenses; (M1) dichroic mirror; (M2) highly reflecting mirror of the resonator; (M3) output mirror of the resonator; (AOM) acoustooptical modulator; (IPF) interference-polarisation filter; (M) highly reflecting mirror; (OI) optical insulator. Part 2 of the setup: (M4) input mirror of the resonator; (M5) output mirror of the resonator.

3. Experimental results and their discussion

During the experiments, we measured the average pump power P_p at a wavelength of $\lambda = 2.097$ μm, the average pump radiation power $P_{2.097}$ released from the PG resonator (not absorbed by the $ZnGeP_2$ crystal) and the total radiation power P_{sum} at the PG resonator output, which allowed us to determine the average power of PG radiation ($P_{PG} = P_{sum} - P_{2.097}$) in the range $\lambda = 3.5 - 4.8$ μm. Based on the data obtained, we calculated the PG efficiency [$(P_{PG}/P_p) \times 100\%$] as a function of the average power and energy density *W* of pump radiation. The main experimental results are shown in Fig. 2. A significant increase in P_{PG} and in parametric generation efficiency is observed when the average power and the energy density (at the end face of the nonlinear crystal) of pump radiation exceed 2 W and 0.16 J cm^{-2}, respectively. Thus, the dependence of the average radiation power generated by a single crystal on the average pump radiation power indicates the existence of threshold generation conditions. The PG differential efficiency is ~ 3.9% for $P_p < 2.0$ W and ~ 49.8% for large values of the average pump power. The maximum values of the average PG radiation power and the efficiency coefficient obtained in the experiment were ~ 1.5 W and ~ 28.6% for $P_p \sim 5.5$ W and $W \sim 0.47$ J cm^{-2}. In this case, the pulse duration of PG radiation (~ 26 ns) is comparable with the pulse duration of pump radiation. The experiments were conducted with the pump beam diameter at the end face of the $ZnGeP_2$ single crystal, equal to 385 μm (at the 1/e^2 level).

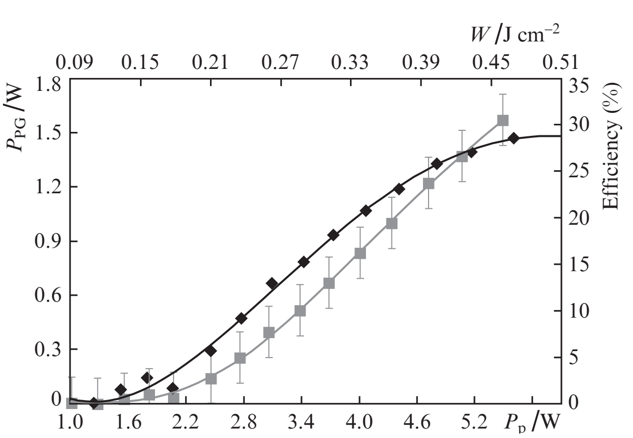
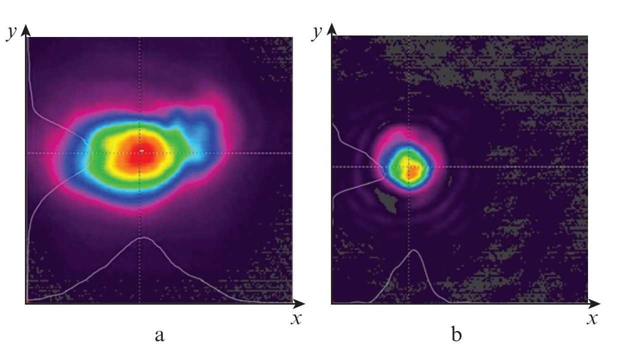


Figure 2. (■) Radiation power and (◆) efficiency of the parametric generator as functions of the energy density and average power of pump radiation.

The intensity distributions in the cross sections of PG beams and pump beams (Fig. 3), and also the PG radiation and pump radiation divergences were measured by the method [31] using the Pirocam III camera and an interference filter that reflected ~ 99% of pump radiation and transmitted 40% – 60% of PG radiation. Initially, the PG radiation beam diameter d_1 was measured in the far zone, then Pirocam III was moved away for a distance *L*, and the diameter d_2 was measured. The divergence angle was calculated by the for-



a b

Figure 3. Distributions of (a) PG radiation intensity and (b) pump radiation intensity in the absence of ZnGeP2 single crystal in the resonator in the beam cross sections.

mula $\theta = \arctan(d_2 - d_1)/L$. The pump radiation divergence was determined in a similar manner.

The divergence angle of pump radiation was about 9 mrad. The divergence angles of PG radiation constituted 20 mrad along the x axis and 16 mrad along the y axis. The parameter $M^2_{x,y}$ for pump radiation beam was ~2.6, and $M^2_x \sim 3.5$, $M^2_y \sim 2.8$ for the PG beam.

The PG emission spectrum was recorded in accordance with the scheme shown in Fig. 4, with a pump radiation power of ~4 W, the width of the input and output slits of an MDR-204 diffraction grating monochromator equal to 100 μm, and with the use of a diffraction grating with 300 lines mm^{-1} and a FR-XM0009 photoresistor made of PbS as a photodetector at the monochromator output slit. The photodetector spectral sensitivity range was 0.8–3.9 μm, which allowed spectral measurements only for the signal wave of the generated radiation. The residual radiations from the pump laser and the PG radiation reflected from the aluminium spherical mirror SM with a focal length of 20 cm were directed to the germanium filter F_{Ge} by the mirror M. The monochromator entrance slit was located in the focal plane of the aluminium spherical mirror. Focusing of PG radiation onto the monochromator entrance slit made it possible to substantially increase the signal coming to the photodetector and the filling area of the diffraction grating, which increased the monochromator resolving power. The filtration of PG radiation from residual pump radiation ($\lambda = 2.097$ μm) was performed owing to F_{Ge} location at the Brewster angle, since the polarisations of these radiations are mutually perpendicular. As a result, the residual radiation power with $\lambda = 2.097$ μm behind the germanium plate did not exceed 200 mW with an average PG radiation power of about 400 mW. The additional interference filter F completely reflected the residual pump radiation and transmitted ~70% of PG radiation.

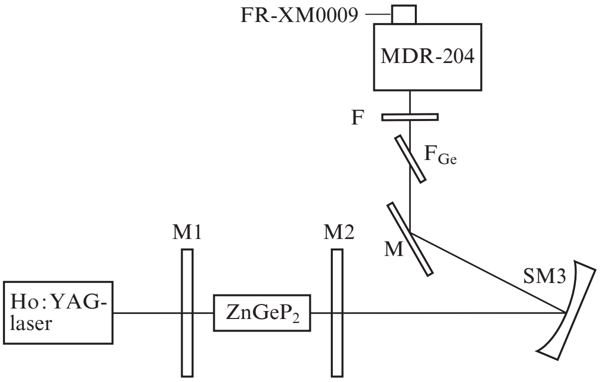


Figure 4. Scheme of the setup for recording the PG radiation spectrum: (M1, M2) resonator mirrors; (SM) spherical mirror; (M) aluminium mirror; (F_{Ge}) germanium filter located at the Brewster angle to the direction of radiation propagation; (F) interference filter.

Figure 5 shows the spectrum of the signal wave generated in the PG at an angle $\theta = 53.35°$ between the crystallographic axis C and the propagation direction of pump radiation.

The signal wave spectrum represented bands with a width of ~200 nm, including several lines with a width of 20–30 nm, with band centres at $\lambda = 3.43$ and 3.765 μm. This structure of the radiation spectrum is typical for PGs with a two-mirror resonator [18, 28].

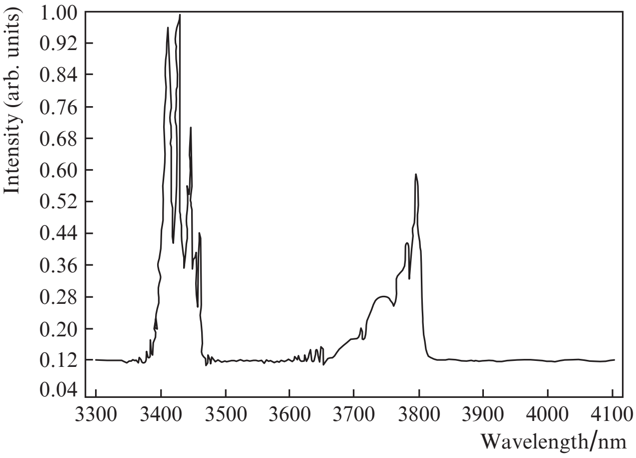


Figure 5. PG radiation spectrum measured with a step $\Delta\lambda = 1$ nm.

In work [18], the parametric generation efficiency is presented as a function of the pump radiation power with a pump beam diameter of ~900 μm ($1/\text{e}^2$ level), obtained on a single ZnGeP_2 element with a length of 18 mm and a tandem of such elements with a length of 18 mm each in accordance with a scheme of radiation drift compensation, which makes it possible to obtain the efficiency dependence on the energy density of pump radiation. Lippert et al. [28] present the PG radiation power dependence on the pump radiation at a beam diameter of ~400 μm ($1/\text{e}^2$ level). The PG consisted of two ZnGeP_2 elements with a length of 14 mm each, also arranged according to the scheme of radiation drift compensation, which allows obtaining the generation efficiency dependence on the energy density of pump radiation. The efficiency dependence on the energy density of pump radiation and the results of works [18, 28] show that a further increase in the pump energy density (above ~0.4 J cm^{-2}) does not lead to an increase in the generation efficiency, and the output power increases only at the expense of a decrease in the parametric generation threshold. Accordingly, the average power and energy of the generation pulse can be increased by increasing the pump power, while maintaining the pump energy density at a level of ~0.4 J cm^{-2} with a corresponding increase in the beam diameter of pump radiation, which may lead to a decrease in the radiation beam divergence and at the same time deteriorate its mode composition. This approach allows the use of a cheaper material with lower radiation resistance to implement parametric generation, providing the limiting efficiency and output energy characteristics, but deteriorating the generated beam quality. The average PG radiation power can be increased if the pump energy density exceeds 0.4 J cm^{-2}. In this case, the generated beam quality will not deteriorate, and the output power will increase due to a reduction in the parametric generation threshold.

In modern studies, fibre-optic channels are often used. The beam astigmatism has a significant effect on the efficiency of the radiation input into the multimode fibre-optic channels, whereas the beam propagation factor M^2 does not significantly affect this parameter. The M^2 value obtained in the present study was sufficient for the efficient coupling of radiation into the optical fibre. As can be seen from Fig. 3, the beam of generated radiation has a significant astigmatism, which may be due to errors in the processing of the element's working surfaces (wedging and astigmatism of the surface), the contribution of which is enhanced by a large refractive index of the $ZnGeP_2$ crystal (~ 3). In this regard, the surface treatment quality of a single crystal should provide high efficiency of the radiation input into a single crystal. In addition, the appearance of astigmatism is possible if a two-mirror resonator is used [13]. One of the solutions to this problem may be the use of a ring resonator [13].

4. Conclusions

We have experimentally implemented parametric generation with an average power of 1.5 W in the spectral range of 3.5 – 4.8 μm at a pulse repetition rate of 10 kHz. The generation pulse duration constituted ~ 26 ns, the pulse energy was 0.15 mJ. The studies have shown that at a pump power density of ~ 0.4 J cm^{-2}, it is possible to attain a high efficiency of the pump radiation conversion into the generation radiation, which will allow an efficient parametric generation to be obtained at a pump energy density below the $ZnGeP_2$ destruction threshold, when a single crystal is pumped by the radiation from a repetitively pulsed Ho : YAG laser. Spectral measurements have shown that in the PG based on the $ZnGeP_2$ single crystal, it is possible to implement the radiation tuning using interference filters together with Lyot filters [32]. This makes it possible to develop tunable dual-frequency pump sources based on the $ZnGeP_2$ single crystals for generation of THz radiation (pumped at the difference frequency) with frequency tuning in the range of 0.1 – 2 THz.

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