TUNABLE PARAMETRIC SUPERLUMINESCENCE IN $ZnGeP_2$ NONLINEAR CRYSTALS

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Temperature tunability of parametric superluminescence in $ZnGeP_2$ *crystal has been investigated. This crystal is promising for development of coherent radiation sources overlapping all atmospheric optical windows in the middle IR region. Q-switched, actively mode-locked and damped* Er^{3+}:YSGG *laser* (2.79 μm) *was used as a pumping source. Wavelength gap* 5.3 ~ 5.9 μm *near degeneracy in II type of parametric interaction, which was not covered by angular tuning, was partially covered by crystal temperature rise. The result was explained by changes in phonon absorption, birefringence, and thermal optical coefficient with the temperature rise. Temperature tuning eliminated beam work-off (displacement) which always exists in angular tuning.*

1. INTRODUCTION

$ZnGeP_2$ is a positive uniaxial crystal with $42m$ point symmetry group having the third largest figure of merit compared to other infrared crystals such as Te, $CdGeAs_2$, Tl_3AsSe_3, $AgGaSe_2$, GaSe, etc. $ZnGeP_2$ crystals have large dimensions (38 mm in diameter and 210 mm in length) and good quality. Typical value of the absorption coefficient at the maximum transparency range is 0.01~0.05 cm^{-1} for as-grown crystals. Typical absorption coefficient spectra for as-grown and annealed $ZnGeP_2$ crystals are presented in Fig. 1.

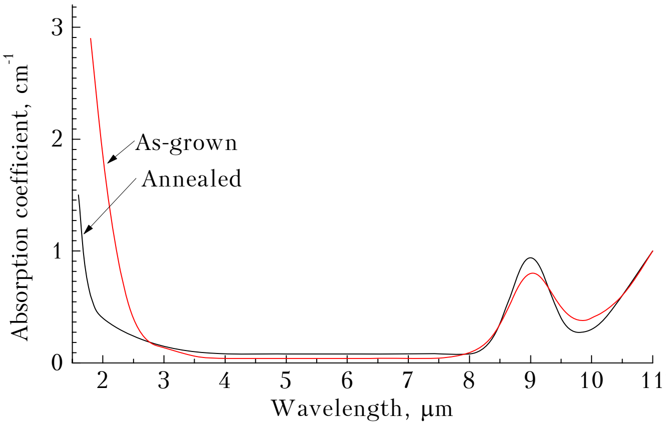

FIG. 1. Dependence of the absorption coefficient on wavelength for as-grown and annealed $ZnGeP_2$ *crystals.*

Annealing reduces the absorption coefficient in the shortwave range of the transparency spectrum approximately by a factor of four at λ = 2 μm up to ~0.4 cm^{-1} for the non-polarized beam. Therewith it is well known that the absorption coefficient for an ordinary wave is at least twofold lower than for extraordinary one. We found that this difference may achieve three or four times. The absorption coefficient for the maximum transparency range often increases insignificantly, but sometimes up to 2–3 times.

The changes in the 9-μm three-phonon absorption peak area are always insignificant. This crystal is thermally resistant and can be immersed from boiling water into liquid nitrogen and back many times. It is harder than the above-mentioned crystals; its thermal conductivity is an order of magnitude higher; it is insoluble in acids and alkali even under boiling, etc.

This crystal was widely used earlier in a number of parametric devices for efficient generation of the second and fourth harmonics, as well as sum and difference frequency generation in the middle[1–5] and far[6] infrared range. The highest quantum efficiency and the lowest threshold of picosecond parametric superluminescence have also been reported.[7] Due to sufficiently high absorption in the shortwave spectral range (0.75 to 2.5 μm) observed for as-grown $ZnGeP_2$ crystals such efficient lasers like Nd:YAG and Ho:Tm:ILF cannot, as a rule, be used as pumping sources for parametric generation. In this case the most short-wavelength pumping may be a radiation of erbium lasers that are working at the 3-μm region.

The efficiency and spectral tuning of the parametric superluminescence have been investigated using various crystals,[8] including $ZnGeP_2$, but phase matching was always done by angular tuning. The latter demands continuous realignment of the optical path. Moreover, such tuning is impossible in $ZnGeP_2$ at wavelength of 5.3–5.9 μm close to the degeneracy

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region when pumping with 3-μm radiation at II type of parametric interaction due to insufficient birefringence value at room temperature. However, $ZnGeP_2$ crystal birefringence increases with temperature,[9,10] that may give a possibility to extend the range of superluminescence to the range of 5.3–5.9 μm unattainable for angular tuning.

Previously the temperature phase matching in $ZnGeP_2$ crystal was shown only for the second harmonic generation of CO_2 laser radiation.[1,11,12]

The purpose of this work is experimental studying of temperature tuning of parametric superluminescence in as-grown $ZnGeP_2$ crystals and, in particular, feasibility to overlap the 5.3–5.9 μm range with superluminescence spectrum at II type of parametric interaction and pumping with 2.79-μm erbium laser radiation. Thus, we study the possibility to develop a corresponding parametric light oscillator.

2. EXPERIMENTAL EQUIPMENT

To study parametric superluminescence frequency tuning by changing crystal temperature, the experimental setup was constructed with the block diagram shown in Fig. 2. This setup comprises of a pumping laser, a thermostat with the crystal, and an optoelectronic system for control over the pumping laser and recording of parametric superluminescence signals.

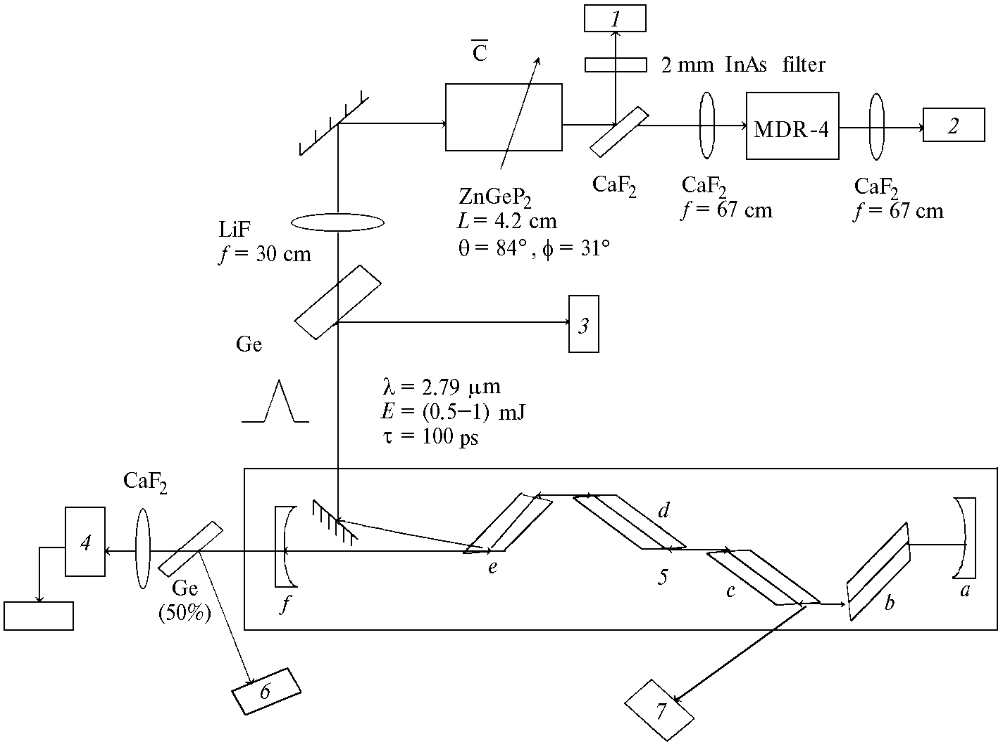

FIG. 2. Block diagram of the experimental setup: Ge:Au *photoresistors (1, 2, 3, and 4); cavity-damped, mode-locked, Q-switched erbium laser (5): 100% reflector – bulk copper mirror (a),* $LiNbO_3$ *crystal for mode locking (b),* $LiNbO_3$ *crystal for Q-switching (c),* Er:YSGG rod (*d*), $LiNbO_3$ *crystal for cavity damping (e), output dielectric mirror (f);* Ge *avalanche diodes (6 and 7).*

The Er:YSGG crystal rod 73.5 mm in length and 5 mm in diameter alloyed with chromium to increase pumping efficiency was used as an active element of the pumping laser. The lasing wavelength of 2.79 μm can be shifted a little to 2.94 μm when using YAG matrix. The optical cavity comprises of a bulk copper mirror 2.5 m in radius, and a dielectric-coated mirror with curvature radius of 2.5 m and the reflection coefficient of 99.9%. Since all crystals were cut at the Brewster angle, beam interference in the cavity was eliminated. Moreover, beam expansion at crystal input reduced their intensity. In the free running laser mode, the oscillation threshold corresponded to ~ 10 J pump energy. However, realization of the picosecond single-pulse mode required a great number of optical elements in the cavity, what resulted in the increase of pump energy up to 500–600 J at inductance of 93 μH in the pump circuit charged to 500 V. Three $LiNbO_3$ crystals were placed into the laser cavity to realize the picosecond laser mode. Q-switching was done by a $LiNbO_3$ crystal (*c*, see Fig. 2) 5 × 10 × 20 mm in size when light propagated along Z-axis of the crystal. The half-wave voltage was 3 kV. The electro-optic mode-locker (*b*, Fig.2) was made of 3 × 6 × 3 mm crystal. The cavity length was adjusted by a micrometer drive with an accuracy of ± 10 μm to set the modulating frequency of the crystal RF driver at 120 MHz.

To separate a single pulse, we applied a control step voltage of 5 kV and leading-edge time of 8.3 ns to the $LiNbO_3$ crystal (*e*, Fig. 2) from a pulse generator triggered by the output signal of the Ge avalanche photodiode placed behind the dielectric mirror (*f*, Fig. 2). This step voltage changed the beam polarization in the crystal by 90°, so the beam left the crystal and, hence, the cavity at an angle of 6° to the original direction. The single pulse obtained in this laser had duration of 100 ps and energy of 0.5~1.0 mJ.

The laser radiation was focused by a LiF lens with 30-cm focal length to the $ZnGeP_2$ crystal. A nitrogen-cooled Ge:Au photoresistor recorded the parametric superluminescence radiation (PSR) at the input and after passing through a monochromator MDR–4. A 2-mm-thick InAs filter blocked the residual pump radiation in the first case. As the PSR divergence at half-intensity was broad (6.2°) for this crystal, a CaF_2 lens was used to collect the generated radiation. To realize a temperature tuning of parametric superluminescence, we placed the crystal in the simplest thermostat: the crystal was wrapped with a thin copper foil three times longer than the crystal to provide for a homogeneous volume temperature. Wrapped crystal was thermally and electrically isolated with Kaolin cotton and SiO_2 cloth. Then Nichrome wire used as a heater was wrapped around it. A set of two Pt-Pt/Ro alloy thermocouples in close contact with Cu foil was used to control the crystal temperature. Then all this set was thermally and electrically isolated again. The temperature stability attained was ± 0.1°C.

Superluminescence phase matching was achieved by crystal heating through slow increase of heater current and long-time withstanding to get a heat balance. The experiment was repeated at reverse temperature change.

3. EXPERIMENTAL RESULTS

So, we obtained the parametric oscillation without mirrors by providing for conditions for parametric superluminescence. Its intensity threshold was 0.35 GW/cm^2 for 42-mm $ZnGeP_2$ crystal used under conditions close to degeneracy. The pump beam diameter was 0.27 mm, its displacement was 0.11 mm, working pump intensity was 5–10 GW/cm^2 at damage threshold of 35 GW/cm^2. Oscillation efficiency was 10% and spectral half-width of the emission line was 25 cm^{-1} at the half-maximum. The crystal position did not change at temperature tuning as opposed to angular one.

An interesting fact was the observed gradual decrease of the parametric signal intensity with temperature. This fact restricted our experiments from above to the temperature range up to 325°C, when the signal was barely perceptible. Note that the initial signal level was two orders of magnitude higher. This fact was observed at least in three previous experiments, in particular, in second harmonic generation of CO_2 laser radiation.[1,11,12] In the experiments described in Ref. 1, the optimum temperature for crystal with the length of 9 mm and the absorption coefficient of 0.35 cm^{-1} was 160°C. In Ref. 12, the maximum was observed at 200°C in temperature dependence of the second harmonic oscillation efficiency. Later it has been found that optimum depends on the crystal effective length and, hence, its optical quality. In the third case, the optimum was noticed at 250°C. In all the cases, the growth in losses was related to the growth and the corresponding spectral broadening of the three-phonon absorption peak by virtue of its temperature dependence.

In a similar manner, we can say that decrease in the parametric superluminescence signal at higher temperatures is a result of increase in the crystal absorption with growth of temperature and spectral broadening of this peak to the range $\lambda \cong 5$ μm. For the crystal 42 mm long the absorption at about 5.6 μm increases to several dozens of cm^{-1} resulting in strong signal attenuation found.

The experimental data on spectral tuning of the superluminescence signal wave are shown in Fig. 3.

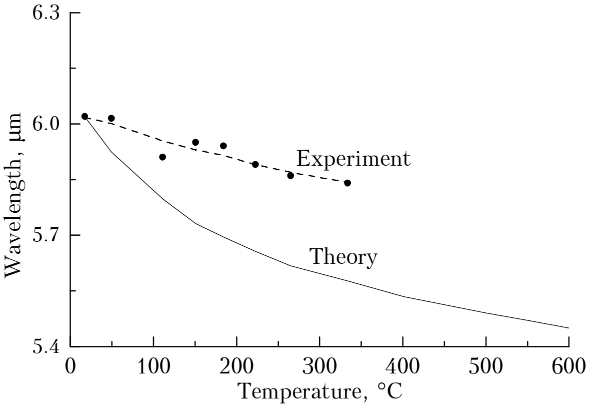

FIG. 3. Dependence of parametric superluminescence wavelength in $ZnGeP_2$ *on crystal temperature under* 2.79 μm Er^{3+}:YSGG *laser pumping.*

At temperature tuning, the signal wavelength falls off lower than 5.9 μm, while the idle mode wavelength not shown here rises from 5.3 μm. We tried to explain the temperature tuning using the data on temperature dependence of the refractive indices. The theoretically predicted tuning curve is based on measurement of dn_o/dT and dn_e/dT using spline interpolation as a smoothing procedure. It is seen that observed frequency tuning is far less than the predicted one. Under the condition of tuning linearity due to its small range, we can say that the experimental values are 2.5–3 times less than the predicted ones. The wave variance of the thermo-optical coefficient should be thoroughly considered,[10] but, undoubtedly, to estimate accurately the temperature tuning, it is necessary to determine its accurate value. In estimations, we have not taken care of any changes in the thermo-optical coefficient with temperature, but assumed that they are constant over the whole temperature range studied. Spectral range of the temperature tuning of superluminescence, calculated under the condition that n_o and n_e grow linearly with temperature, gave overestimation (see Fig. 3).

4. CONCLUSIONS

1. Temperature tuning of parametric superluminescence in the nonlinear $ZnGeP_2$ crystal was experimentally realized and investigated for the first time.

2. Magnitude of temperature tuning of parametric superluminescence in $ZnGeP_2$ crystal with pumping by Er^{3+}:YSGG laser radiation at 2.79 μm is 2.5~3 times less than estimations made from the now available data on temperature dependence. It means that corresponding changes of n_o and n_e are less than predicted (calculated) ones[10] at temperatures above the room temperature. The possible reason of narrow tuning range may be temperature dependence of the thermo-optical coefficient that is to be investigated.

3. Decrease in the parametric superluminescence signal by two orders of magnitude at temperature rise from the room temperature to 325°C can be explained by increase in phonon losses.

4. Both effects gave us no possibility to cover the 5.3 to 5.9 μm spectral gap in II type phase matching in $ZnGeP_2$ crystal pumping at 2.79 μm by temperature tuning. This disadvantage can be overcame by using annealed crystals with low absorption at 2.0 μm (≤ 0.15 cm^{-1} for an ordinary wave) pumping with Ho:Tm:ILF laser radiation ($\lambda = 2.01$ μm).

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