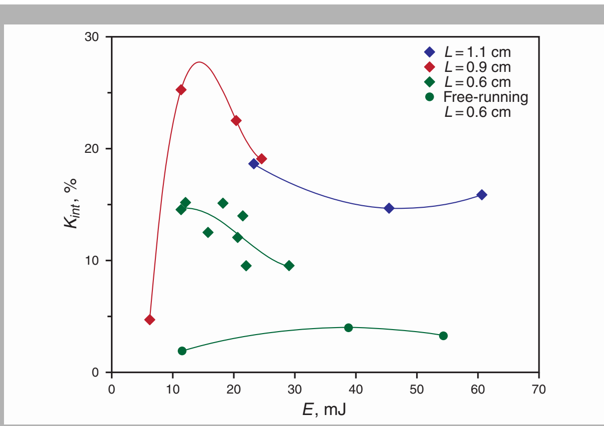
Abstract: Frequency doubling of mode-locked frequency-tunable CO laser emission in high quality 17-mm long $ZnGeP_2$ nonlinear optical crystal was studied. The internal efficiency of frequency doubling exceeded 25%, which was obtained by stabilization of mode-locking regime and increasing peak power of pump radiation in the scheme "master oscillator – power amplifier". A feasibility of difference frequency conversion of both fundamental and first overtone CO laser lines to cover ∼4.0 – 5.0 μm spectral range and a possibility of getting spectral range of ∼1.25 – 10 μm by mixing of all CO laser lines were estimated.



Internal efficiency of SHG in $ZnGeP_2$ crystal *versus* pump energy when the nonlinear crystal after focal plate. CO Laser transition 9→8 P(10) ($\lambda \approx 5.274$ μm)

Mode-locked CO laser frequency doubling in $ZnGeP_2$ with 25% efficiency

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1. Introduction

Extension of a gas laser emission range is attractive for application in laser spectroscopy, atmosphere sensing, laser media diagnostic, chemical reaction initiation, isotope separation, etc. Pulsed CO laser offers several advantages over other sources of infrared (IR) radiation. It is a highly efficient laser that provides an opportunity to control the output energy and pulse duration within wide dynamic range, and to ensure a high average power in a repetitively pulsed regime. CO laser can operate on hundreds of ro-vibrational transitions of both fundamental (wavelength range 4.6 – 8.2 μm) [1] and first-overtone (2.5 – 4.2 μm) vibrational bands [2–4]. In its turn, parametric frequency conversion of CO laser emission using a single nonlinear optical crystal is able to cover both mid- and far-IR spectral ranges [5,6].

Nonlinear crystal $ZnGeP_2$, or so-called mid-IR "standard" nonlinear crystal, is well known as a most efficient crystal in frequency conversion within mid-IR. The sec-

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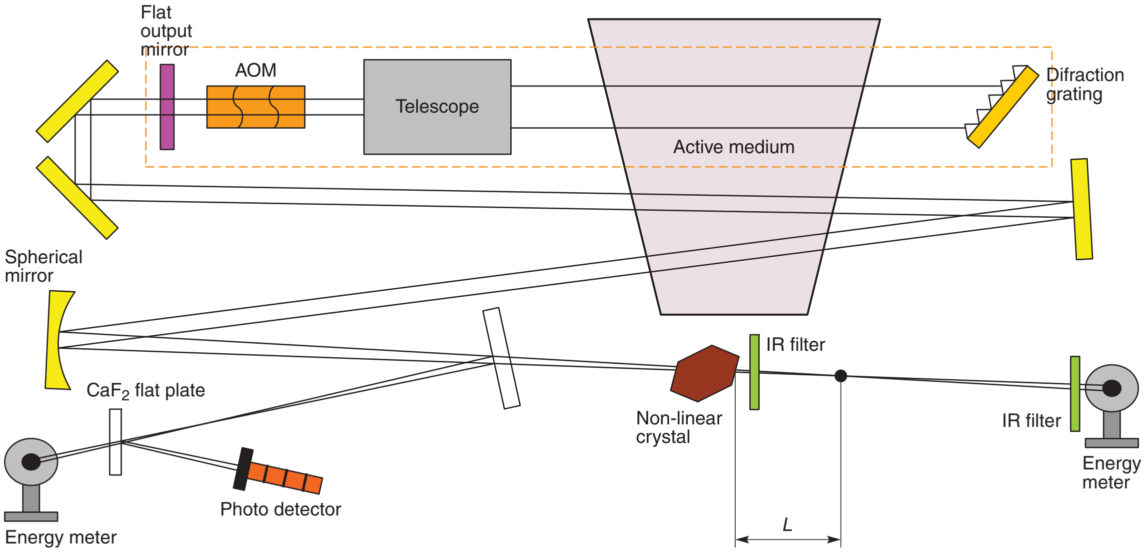

Figure 1 (online color at www.lphys.org) Optical schematic of the frequency converter of the pulsed EBSD CO laser MOPA system

ond harmonic generation (SHG) of CO_2 laser emission in $ZnGeP_2$ crystal was studied for the first time at the beginning of the 80-th [7]. It was third type of parametric frequency converter that had ever been realized with $ZnGeP_2$ crystal after up-converter [8] and THz emission generator [9]. Later $ZnGeP_2$ was applied for efficient SHG and fourth harmonic generation of different kinds of CO_2 lasers: from continuous wave (CW) to nanosecond pulsed lasers [10,11]. It was also applied for SHG generation of CW and Q-switched CO lasers [12–14], sum frequency generation of CO and CO_2 laser emissions [15,16], and far-IR or THz generation [17]. Application of $ZnGeP_2$ for frequency conversion of IR radiation appears to be advantageous over other nonlinear crystals such as $AgGaSe_2$ and others [18,19], which stipulated our choice for our experiments on frequency conversion of CO laser radiation. It should be pointed out that CO laser supplied with efficient $ZnGeP_2$ frequency converter can cover much wider spectral range in comparison with any all-solid-state laser system [20–24].

The results of our first experiments on SHG of repetitively pulsed Q-switched low-pressure CO laser radiation are presented in [5,6,25]. Output power, temporal and spectral parameters of frequency converted emission was studied in detail. External SHG efficiency of Q-switched multiline CO laser radiation exceeded 1% in 17-mm $ZnGeP_2$. Taking into account Fresnel losses, the internal efficiency was about $\sim 2\%$. The laser output consisted of ~ 80 emission lines in wavelength range 4.96 – 6.30 μm. Frequency converted spectrum contained more than 110 emission lines in 2.53 – 2.85 μm spectral range. The minimal wavelength 2.53 μm of the frequency converted lines was shorter than 2.65 μm reported in [14] and the most powerful SHG emission line was at the wavelength ~ 2.6 μm. The excess number of the frequency converted lines to those of the pump spectrum was due to the fact that frequency conversion run simultaneously for both SHG and sum frequency generation of different pairs of the CO laser lines. The bandwidth of SHG non-critical spectral phase matching was as wide as ~ 200 cm^{-1} [14], which resulted in SHG phase matching for all pump lines simultaneously.

The SHG of electron beam sustained discharge (EBSD) frequency-tunable pulsed CO laser radiation in $ZnGeP_2$ crystal for both mode-locked and free-running operation modes was studied in [5,6,25]. Maximal external efficiency of frequency conversion of 3.5 % which corresponds to $\sim 7\%$ internal efficiency was obtained for mode-locked CO laser. The external efficiency was limited by two factors. The first one was the fact that only the high intensity leading peak rather than of the pump pulse "tail" was efficiently frequency converted. The second reason was a deterioration of temporal structure of mode-locked CO laser emission which resulted in decrease of pump radiation peak power [5]. It was connected with cavity loss-level overshoot by gain coefficient of CO laser active medium and therefore acousto-optical modulator did not function effectively. Note that the $ZnGeP_2$ crystal was fabricated from the same boule as crystals applied in high efficiency short-pulse optical parametric oscillator (OPO) [26,27]. Thus, mode-locking is evidently a key point in the improvement of CO laser for frequency conversion.

In this paper we report about improved CO laser frequency conversion efficiency in high optical quality $ZnGeP_2$ crystal, which was obtained by stabilization of mode-locking regime and increasing peak power of pump radiation in the scheme "master oscillator – power amplifier" (MOPA).

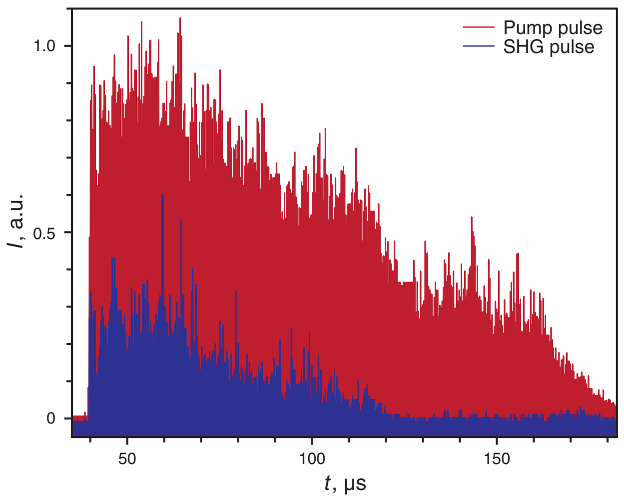

Figure 2 (online color at www.lphys.org) Temporal shape-form of pump and SHG pulses of frequency-selective mode-locked CO laser (identified in the figure inset). CO laser transition 9→8 P(11) ($\lambda \approx 5.286$ μm)

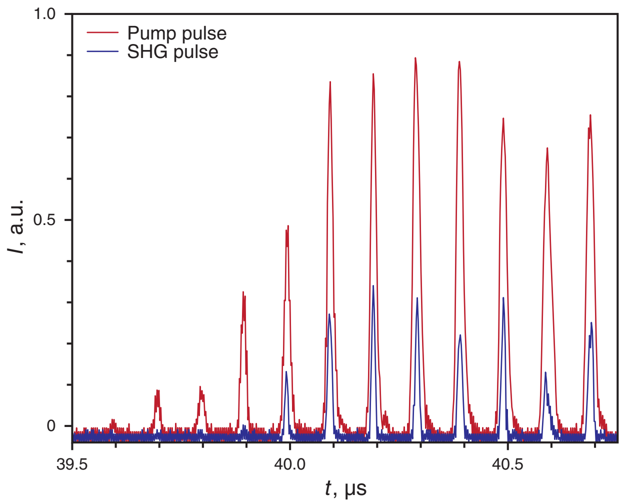

Figure 3 (online color at www.lphys.org) Pump and SHG pulses at the beginning of frequency-selective mode-locked CO lasing (identified in the figure inset). CO Laser transition 9→8 P(11) ($\lambda \approx 5.286$ μm)

2. Experimental techniques and results

The high optical quality 17-mm long not-antireflection coated $ZnGeP_2$ crystal was grown by V.G. Voevodin with co-workers from V.D. Kuznetsov Siberian Physical-Technical Institute at the Tomsk State University, Tomsk, Russia. Experiments were carried out at the Gas Lasers Laboratory of the P.N. Lebedev Physical Institute of RAS. In order to increase peak power of the spikes, and finally the SHG efficiency, in this study CO laser pulses were amplified in CO laser amplifier. Thus, the MOPA CO laser system was used. The optical schematic of our facility is presented in Fig. 1.

A cryogenically cooled EBSD CO laser applied for this study was described in detail in [28]. The length of the active medium was 1.2 m. The optical length of the laser cavity composed of diffraction grating (240 grooves/mm) installed in Littrow configuration and plane output mirror with reflectivity 30% in wavelength range 5.0 – 7.0 μm was 15 m. Concave spherical mirror with curvature radius of 1 m and convex spherical mirror with curvature radius of 0.3 m were used as a telescope. Germanium acousto-optical modulator (AOM) with aperture 8-mm and antireflection coating in spectral range of 4.5 – 5.5 μm was located close to the output mirror. To excite standing acoustic wave in the AOM, its piezoelectric transducer was fed by RF driver with quartz-controlled frequency 5 MHz and output power up to 7 W. Mode-locked line-selective CO laser emitted a train of $\sim$10 ns (full width at half maximum – FWHM) spikes with pulse repetition rate 10 MHz [29].

The CO laser was tuned to the powerful single ro-vibrational lines 9→8 P(11) ($\lambda \approx 5.286$ μm) or 9→8 P(10) ($\lambda \approx 5.275$ μm). The energy of a laser pulse on the crystal was varied in the range of 0.01 – 0.10 J by changing of the EBSD laser input energy.

After two passes through the active medium the amplified laser beam was directed to the spherical mirror (a curvature radius 0.6 m or 1.0 m). The crystal $ZnGeP_2$ was positioned at the distance L from focal plane of the spherical mirror to exclude optical damage of crystal input surface. One part of CO laser beam was reflected by first CaF_2 plate, split by second CaF_2 plate and then directed, respectively, onto sensitive areas of the reference energy meter OPHIR 3A-H and photoelectromagnetic detector PEM-L-3 (response time is below 0.5 ns) to control pulse shapeforms. Frequency converted emission was measured with the energy meter OPHIR 3A-SH. The residual pump radiation was blocked before both energy meter and detector by two IR filters. First one was 2 mm thick IR-quartz plate installed right after the crystal, and the second one was installed in front of the energy meter and detector. In such a way interference caused by the emission from the first quartz plate heated by pump radiation was blocked by the second quartz plate. Signals of the output of the detectors were recorded by oscilloscope Tektronix TDS5052B. Pump emission wavelengths were measured by IR spectrograph IKS-31 (LOMO PLC).

Due to the amplification the peak power of CO laser emission increased as much as 3 – 4 times up to $\sim$ 60 kW. Moreover, the temporal shape-form of the amplified spikes was much regular as compared to that of not-amplified CO laser spikes [30]. The time behaviors of pump and converted pulses are presented in Fig. 2. At the beginning of the train of pump and SHG spikes following with axial period of 100 ns (Fig. 3) one can see much regular spike

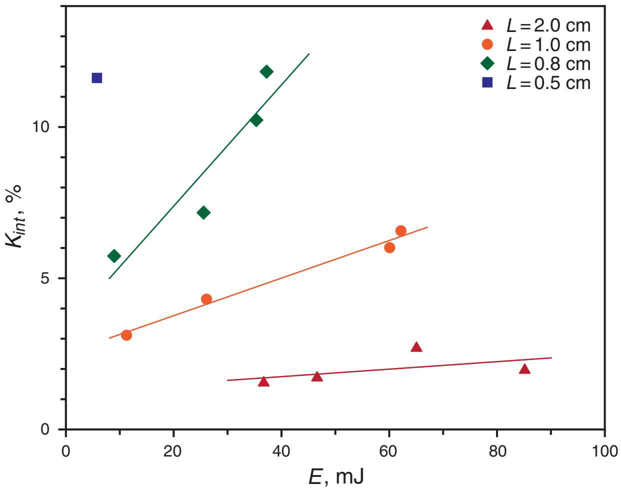

Figure 4 (online color at www.lphys.org) Internal efficiency of SHG in $ZnGeP_2$ crystal *versus* pump energy when the nonlinear crystal before focal plate. CO Laser transition 9→8 P(10) ($\lambda \approx 5.274$ μm), curvature radius of focusing mirror 0.6 m

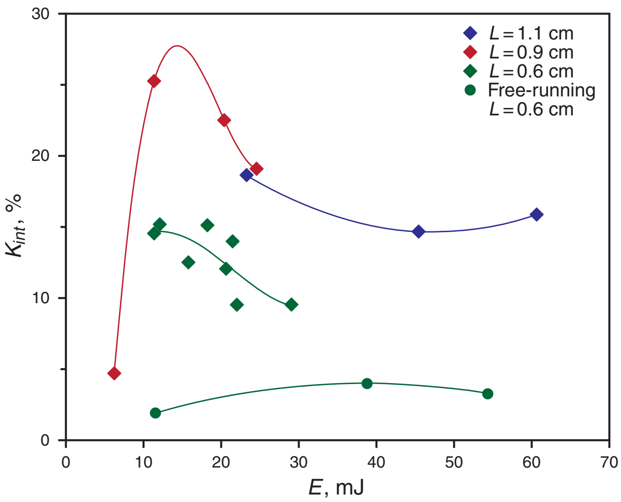

Figure 5 (online color at www.lphys.org) Internal efficiency of SHG in $ZnGeP_2$ crystal *versus* pump energy when the nonlinear crystal after focal plate. CO Laser transition 9→8 P(10) ($\lambda \approx 5.274$ μm), curvature radius of focusing mirror 1.0 m

structure as compared to that of not-amplified spikes. Energy contrast between spikes of ~ 10 ns (FWHM) duration and valleys was almost 100%. It should be noted that only the first part of the pump pulse (from 40 to 120 μs after the beginning of EBSD pulse ($t = 0$) is efficiently frequency converted (Fig. 2).

External SHG efficiency K_{ext} is the ratio of SHG energy to the pump energy E measured with the energy meters. Internal SHG efficiency K_{int} was calculated by the formula: $K_{int} = K_{ext}(1 - R_{pump})^{-1}(1 - R_{SHG})^{-1}$, where R_{pump} is the Fresnel reflection coefficient of pump energy by front surface of the crystal and R_{SHG} is the Fresnel reflection coefficient of SHG energy by output surface of crystal. Reflection loss was calculated using Sellmeier dispersion equations for $ZnGeP_2$ taken from [31]. Optimal incidence angle of the pump beam was estimated as 29° [5]. It turned out that internal SHG efficiency is related with external SHG efficiency as $K_{int} = 1.91 K_{ext}$. Fig. 4 illustrates internal efficiency of SHG in $ZnGeP_2$ *versus* pump energy for the nonlinear crystal located before focal plane. The internal efficiency K_{int} increases with L decrease due to pump intensity rising up. The maximal internal efficiency of SHG in $ZnGeP_2$ exceeded 11.7% at $L = 0.8$ cm and pump energy of 37 mJ and 11.9% at $L = 0.5$ cm and pump energy of 5 mJ. Further increase of the pump energy in every case was limited by optical damage of $ZnGeP_2$ output surface. This fact was caused by formation of a thermal lens in the crystal volume due to the positive sign of $dn/dT > 0$ (where n is index and T is the crystal temperature) for $ZnGeP_2$ crystal [32]. That is why the total intensity of the pump and SHG emissions on the output surface of $ZnGeP_2$ many times increased and damaged it. Simple solution of the problem is the nonlinear crystal installation after the focal plane of spherical mirror. In this case divergence of pump laser beam after the focal plane of spherical mirror compensates the effect of thermal lens. The result of such experiment is shown in Fig. 5. The internal SHG efficiency increased up to 25% or by two times as compared to previous experimental results. In the every series of the experiment, the further increase of the pump energy higher than energy corresponded to the right point of every curve shown in Fig. 5 leaded to the optical damage of $ZnGeP_2$ output surface. It is interesting to compare K_{int} for mode-locked and free-running CO lasers under the same other conditions ($L = 0.6$ mm). At pump energy of ~ 10 mJ, internal SHG efficiency for mode-locked CO laser was 7.5 times higher as to that of free-running one. At higher pump energy ~ 30 mJ this deference decreased by 3 times.

3. A feasibility of frequency tunable emission within the spectral range from 1.25 to 25 μm

Taking into account the high efficiency of CO laser generation both in fundamental and the first overtone bands, and high efficiency of frequency conversion of its radiation in nonlinear crystals $ZnGeP_2$ into mid-IR it can be proposed that frequency converted emission of CO laser can cover the wide spectral range from 1.25 to 25 μm. In particular, a coherent emission in 1.25 – 2.10 μm range can be obtained by SHG of overtone CO laser lines.

In Fig. 6 *oe-e* type phase matching diagram for difference frequency generation (DFG) of overtone and fundamental CO laser lines in $ZnGeP_2$ is presented. Solid lines are related to CO laser fundamental lines (λ_2) with

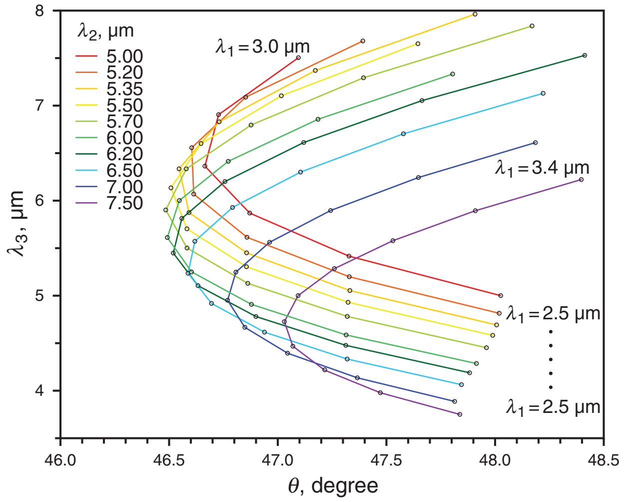

Figure 6 (online color at www.lphys.org) - type phase matching diagram for DFG of overtone and fundamental band CO laser lines in $ZnGeP_2$ crystal

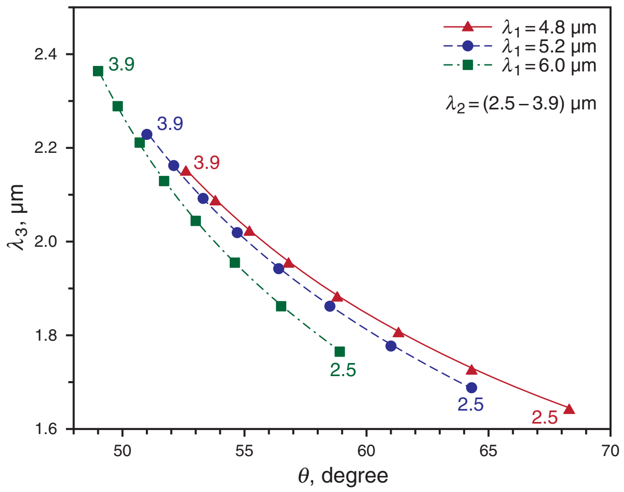

Figure 7 (online color at www.lphys.org) - type phase matching diagram for SFG of overtone and fundamental band CO laser lines in $ZnGeP_2$ crystal

wavelengths in microns shown in the figure. Markers on the solid lines following every 0.1 μm are related to overtone lines (λ_1) from 2.5 μm to the higher wavelength. These markers show converted wavelength (λ_3) and internal DFG phase matching angle θ. By this type of conversion of fundamental and overtone bands it is possible to cover interband range of 4.0 – 5.0 μm, which covers the atmospheric transparency bands. There are no powerful laser sources operating within this wavelength range. However, a frequency converter of CO laser to this wavelength range could be realized rather simply because CO laser can operate in both fundamental and first overtone bands simultaneously [2–4]. It must be noted that the maximal wavelength obtained with this type of conversion is up to 10 μm. This limitation of theoretical maximal wavelength (25 μm) is connected with high absorption of nonlinear crystal $ZnGeP_2$ at 10 μm and higher [17].

The model calculation of phase matching for collinear *ee-o* type sum frequency generation (SFG) of overtone and fundamental CO emission lines in $ZnGeP_2$ was also carried out. The results are presented in Fig. 7 in the same way as in Fig. 6. In this calculation three fundamental (λ_1) and eight overtone CO laser lines (λ_2) from 2.5 – 3.9 μm range were considered. Frequency converted emission covered 1.6 – 2.4 μm range that is rather well in coincidence with transparency window of the atmosphere. Sellmeier equations from [31] were applied in these calculations to estimate reflection losses.

4. Conclusion

In conclusion, in this paper we showed SHG of EBSD frequency-tunable mode-locked CO laser emission in $ZnGeP_2$ crystal with internal efficiency 25%. This result was obtained by stabilization of mode-locking regime and increasing peak power of pump radiation in the MOPA scheme. A feasibility of difference frequency conversion of both fundamental and first overtone CO laser lines to cover $\sim$4.0 – 5.0 μm spectral range and a possibility of getting spectral range of $\sim$1.25 – 100 μm by mixing of all CO laser lines were demonstrated.

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