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CO 激光在非线性晶体 $ZnGeP_2$ 和 GaSe 中的混频效应

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摘要：为了获得 2.15～1 500 μm 的相干光源，研究了 CO 激光在高质量非线性晶体 $ZnGeP_2$ 和 GaSe 中的混频效应。为了提高转换效率，在激光锁模方式下对 CO 激光器的二次谐波、和频和差频的产生进行了研究。结果显示，利用 GaSe 晶体和 $ZnGeP_2$ 晶体，调 Q 多谱线 CO 激光辐射的谱线内倍频效率分别大于 0.3％和 1.1％。采用 $ZnGeP_2$ 晶体进行倍频时，可调谐锁膜 CO 激光器的转换效率为 12.5％。模拟结果显示，二次谐波与和频产生的输出光谱相同。相邻谱线下，和频和差频的产生过程中，基波和一次谐波可以分别在 4.0～5.0 μm 和 100～≥1 200 μm（太赫兹范围）形成振荡。利用锁模 CO 激光器在 $ZnGeP_2$ 晶体中的混频效应，可以得到 2.15～≥1 500 μm 的相干光源，同时转换效率可达到甚至高于 12.5％。

关　键　词：非线性晶体；$ZnGeP_2$ 晶体；GaSe 晶体；混频效应；脉冲 CO 激光；锁模
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CO laser frequency mixing in nonlinear crystals $ZnGeP_2$ and GaSe

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Abstract: The CO laser frequency mixing in high-quality $ZnGeP_2$ and GaSe crystals is studied to obtain the 2.15－1 500 μm coherent sources. Secondary Harmonic Generation(SHG), sum- and difference

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frequency generations are considered as a method for the CO laser frequency mixing and the mode-locking as an efficient way to improve the mixing efficiency. Results show that the internal SHG efficiency of Q-switched multiline CO laser radiation has exceeded by 0.3% for GaSe crystal and has reached by 1.1 % for $ZnGeP_2$ crystal. When the SHG is in $ZnGeP_2$, the internal efficiency of electron beam sustained discharge frequency-tunable mode-locking CO laser is up to 12.5 %. Simultaneous SHG and sum frequency generation show the same output spectrum. It is shown by modeling that the sum and difference frequency generations of neighboring lines of both fundamental and first overtone bands can allow one to get the oscillation, respectively, at 4.0—5.0 μm and 100—≥1 200 μm (THz). In conclusions, the frequency mixing of mode-locked CO laser emission lines in $ZnGeP_2$ crystals allows some one to design 2.15—≥1 500 μm coherent sources with the power frequency conversion efficiency up to or over 12.5%.

Key words: nonlinear crystal; $ZnGeP_2$ crystal; GaSe crystal; frequency conversion; pulsed CO laser; mode-locking

1 Introduction

Extension of a gas laser emission range is attractive for application in the laser spectroscopy, atmosphere sensing, laser media diagnostics, chemical reaction initiations, isotope separations, *etc.* Pulsed CO laser offers several advantages over other sources of IR radiation. CO laser has a high efficiency, which provides the opportunity to vary the energy and duration of pulses within broad range, and ensure a high average power in a pulse-periodic regime. This laser can operate in both fundamental (4.6—8.2 μm) [1] and first-overtone (2.5—4.2 μm) bands[2-4]. The frequency conversion of CO laser emission using single nonlinear crystal can produce radiation in mid- and far-IR spectral ranges.

Nonlinear crystal $ZnGeP_2$, or so called mid-IR "standard" nonlinear crystal, is well known as most efficient crystals in frequency conversion within mid-IR including gas laser frequency conversions. CO_2 lasers SHG in $ZnGeP_2$ for the first time were studied in 1983[5]. It was third type of parametric frequency converter that had ever been realized in $ZnGeP_2$ after up-converter [6] and THz emission generator[7]. Later $ZnGeP_2$ was applied for efficient SHG and fourth harmonic generation of different type CO_2 lasers from CW to nanosecond pulsed laser[8]. It was also applied to SHG generation of CW and Q-switched CO lasers[9-10] and sum frequency generation of CO and CO_2 laser radiation[11-12].

The p-type of ε-polytype GaSe crystals is the second attractive crystal for frequency conversion within and into mid-IR due to extra wide transparency range of 0.62—20 μm and extra large birefringence $B=0.35$ that allows phase matching almost all over the transparency range, and due to a number of other physical properties that are responsible for efficiency of frequency conversion processes. Unfortunately, former time growth technology state-of-the-art did not allow somebody to realize significant SHG efficiencies and other type of parametric frequency converters in GaSe[13]. Nowadays the technology is noticeably improved, but GaSe crystals are applied mainly in frequency conversion into THz range.

In this paper, we report about CO laser frequency conversion in high optical quality $ZnGeP_2$ and GaSe crystals. Output energy, temporal and spectral parameters of frequency converted emission is studied in detail.

2 Frequency conversion of Q-switched CO-laser emission

The optical layout of the experiment on SHG of radiation of CW Q-switched CO laser is presented in Fig. 1. There was used a low-pressure CW CO laser with cryogenically cooled active medium 1 under DC discharge pump, which operated in the Q-switched mode.

Q-switched laser cavity consisted of totally reflecting concave mirror (radius of curvature 9 m), flat output mirror (reflection $R \geqslant 90\%$ in wavelength range λ: 5.0－6.5 μm), and rotating flat mirror. The diaphragm with aperture 10 mm in diameter was located before the output mirror. The length of the laser resonator was 2.7 m. The laser operated in TEM00 mode. Part of the CO laser radiation was reflected by CaF_2 flat plate (1) to the spherical mirror (1) (radius of curvature 0.25 m) and then was directed to the power meter (1) (OPHIR 3A-SH). The small part of the laser radiation was reflected by CaF_2 flat plate (2) to the photodetector (1) (VIGO-system PEM-L-3, Hg-Cd-Zn-Te, sensitivity ～ 3×10^6 cm · $Hz^{1/2}$ · W^{-1}, response time was about 0.5 ns) that was located near the focal plane of spherical mirror (1). The main part of the CO laser radiation (～95%) was focused by CaF_2 focusing lens (CaF_2, focal length ～60 mm) in nonlinear crystal. The similar CaF_2 collimating lens was used to collimate defocusing radiation passed through the nonlinear crystal. Pump radiation was cut off by IR quartz plates (1) with thickness 2 mm. Converted radiation was directed by turning mirror and spherical mirror (2) (radius of curvature 0.25 m) onto the photodetector (2) (PEM-L-3) or the power meter (2) (OPHIR 3A-SH) through IR quartz plate.

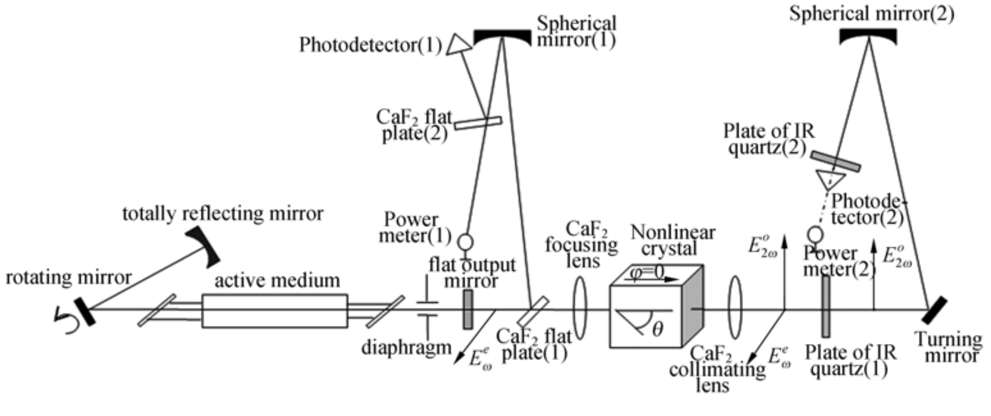

Fig. 1　Optical layout of experiment

The time behavior of pump and converted pulses were measured with oscilloscope Tektronix TDS 2014 (Fig. 2). The nonlinear crystal $ZnGeP_2$ with high optical quality ($\alpha < 0.1\ cm^{-1}$) and without antireflection coating was used in our experiments. The length of the crystal along laser beam was 12 mm. The crystal was placed before focal plane of CaF_2 focusing lens to decrease the intensity of pump laser radiation less than the surface damage threshold.

The full width half maximum (FWHM) pulse duration of pump CO laser radiation decreased from 1.5 to 0.2 μs with increasing pulse repetition rate (PRR) from 20 to 150 Hz. The pulse duration of SHG radiation was two times shorter than the pump pulse.

Average power of pump CO laser radiation reached 100 mW at PRR about 100 Hz, and peak power in this case was nearly 3 kW. Fig. 3 illustrates dependences of average power of SHG and external converted efficiency on PRR. Maximum of average SHG power reached 0.4 mW at ～

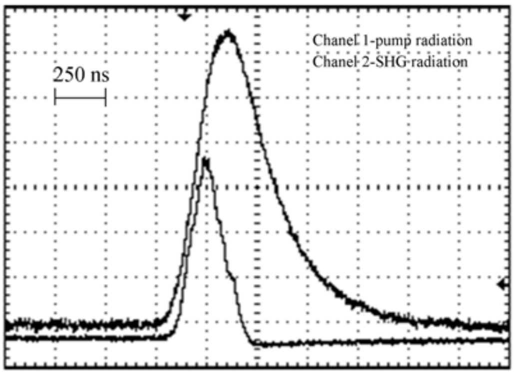

Fig. 2　Characteristic oscillogram of pump and SHG pulses in $ZnGeP_2$. Pulse repetition rate (PRR) is 120 Hz.

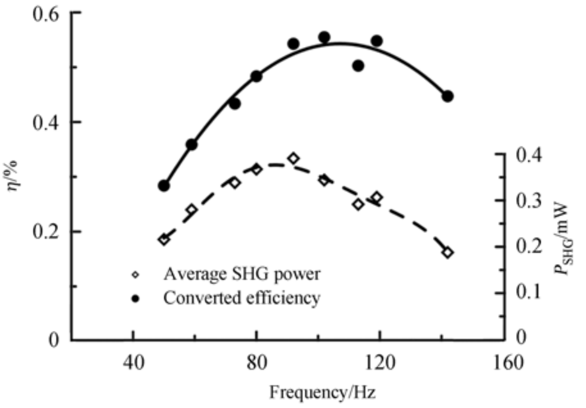

Fig. 3　Average power of SHG and external converted efficiency for nonlinear crystal $ZnGeP_2$ versus PRR.

90 Hz, and maximum of external converted efficiency reached 0.55% at 100 Hz. The external converted efficiency was calculated as the ratio of average SHG power to average power of pump laser radiation. External converted efficiency reached 1%, when nonlinear crystal $ZnGeP_2$ was close to the focal plane of the lens. Taking into account Fresnel losses in optics and crystal surfaces, the internal efficiency was about 2%.

The spectral composition of converted emission was studied in detail at multiline pump by emission of CO laser operating in non-selective mode. Spectrum of converted and pump emission was measured by IR spectrograph IKS-31 (LOMO PLC, spectral resolution 5×10^{-4} μm for wavelength range of 2.0—8.0 μm). The laser output contains about 80 emission lines in 4.96—6.3 μm (see Fig. 4). Spectrum of emission converted in nonlinear crystal $ZnGeP_2$ is shown in Fig. 5.

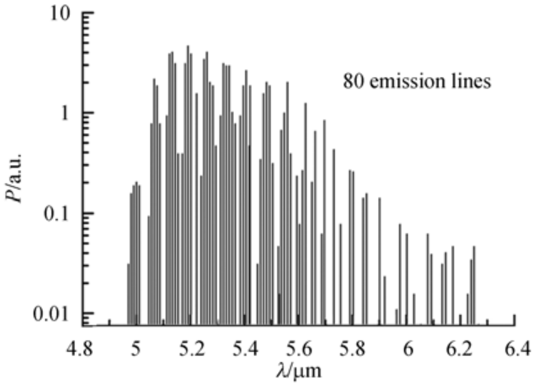

Fig. 4　Spectrum of Q-switched CO laser emission at 100 Hz of PRR

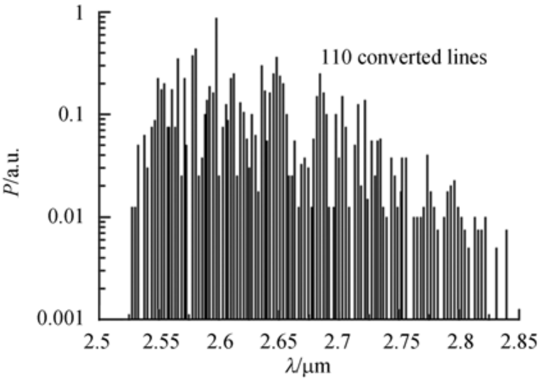

Fig. 5　Spectrum of converted of Q-switched CO laser emission in nonlinear crystal $ZnGeP_2$. PRR is 100 Hz.

One can see more than 110 emission lines within 2.53—2.85 μm spectral range when the crystal $ZnGeP_2$ was pumped by a 80 CO laser with emission lines of 80. The shortest-wavelength line of converted emission was shifted to much shorter wavelength range in comparison with the spectrum in[14] where the shortest-wavelength SHG emission line was limited at 2.65 μm. In our spectrum, the most powerful SHG emission line was one with wavelength 2.6 μm. The excess number of output lines to the number of pump lines was due to the fact that frequency conversion process run simultaneously for both SHG and sum frequency generation at different pairs of CO laser emission lines. The width of non-critical spectral phase matching was about 200 cm^{-1}[14] which gives phase matched SHG for all pump lines simultaneously. Besides that, wide SHG angular bandwidth of about 1.8° was wide enough to cover angular phase matching conditions for sum frequency generation with a large number of different pairs of CO laser emission lines[14].

In our experiments on CO laser frequency conversion in high optical quality GaSe crystal, the external converted efficiency came up to 0.15%. The length of this crystal along laser beam was only 4 mm. It shows that high quality GaSe crystals can be in competition with $ZnGeP_2$ crystal but their low mechanical properties: almost zero hardness in Mohs scale and easy cleavage might be improved, for example, by impurity doping.

3 Frequency conversion of mode-locked selective CO laser emission

Experimental results on SHG in $ZnGeP_2$ crystal with the laser emission of electron-beam-sustained-discharge (EBSD) frequency-tunable CO laser operated both in mode-locking and free-running mode are discussed in this section.

The experiments were carried out with cryogenic pulsed EBSD CO laser installation described in detail in [15]. In actively mode-locked regime, the CO-laser produced a train of about 10 ns (FWHM) pulses with pulse repetition rate of 10 MHz in single-line mode of operation[16]. The optical layout of our experiments is presented in Fig. 6. The length of active medium was 1.2 m. The optical length of the laser resonator was 15 m and it consisted of diffraction grating (240 grooves/mm) operated under near-Littrow configuration and flat output mirror with reflectivity 75% for the wavelength range 5.0 — 7.0 μm. Diaphragm with diameter 40 mm was placed between diffraction grating and active medium. Concave spherical mirror with radius of curvature 1 μm and convex spherical mirror with radius of curvature 0.3 m were used as a collimator. An acousto-optical modulator (AOM) made of Germanium was located close to flat output mirror. The aperture of the AOM with antireflection coating in wavelength range 4.5—5.5 μm was 8 mm in diameter. To excite standing acoustic wave in the AOM, its piezoelectric transducer was fed by RF driver with frequency of 5 MHz and output power up to 7 W.

Polarization plane of laser emission was changed by flat turning mirrors (1), (2) and then laser beam was directed to spherical mirror (radius 0.6 m). Crystal $ZnGeP_2$ was installed before focal plane of spherical mirror in order to reduce the intensity of pump CO laser radiation smaller than surface damage threshold. Part of CO laser radiation was reflected by CaF_2 flat plate 12 to the power meter (1) (OPHIR 3A-SH). Pump radiation was absorbed by IR quartz plates (1) with the thickness of 2 mm. Converted radiation was measured by power meter (2) (OPHIR 3A-SH). In order to measure the time behavior of pump and converted pulses, we changed power meters (1) and (2) by the photodetectors (1) and (2) (PEM-L-3). Influence of pump radiation diffusing from optical elements of scheme and heating by IR radiation of quartz plates (1) was cut off by second IR quartz plates (2), which was installed in front of the power meter (2) or photodetector (2). Signals from the photodetectors (1) and (2) were detected by oscilloscope Tektronix TDS5052B. Spectrum of the pump emission was measured by IR spectrograph IKS-31 (LOMO PLC). The optical tract was adjusted using radiation of He-Ne laser (1), which was directed to the scheme throw the the mirror with a hole. Control of turning the angle of diffraction grating was realized by He-Ne laser (2), flat turning mirrors (3) and the scale.

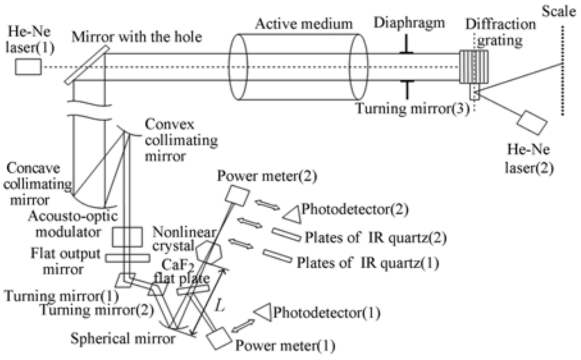

Fig. 6　Optical layout of experiment on frequency conversion of pulsed EBSD CO laser emission.

Frequency-tunable EBSD CO laser operated on single ro-vibrational line 8→7 P(12) ($\lambda=5.224\ \mu$m). The energy of laser pulse on the crystal was in the range of 0.1 to 0.5 J. It could be veried by changing of EBSD input energy. The time behaviors of pump and converted pulses are presented in Fig. 7 (free-running mode) and Fig. 8 (mode-locking).

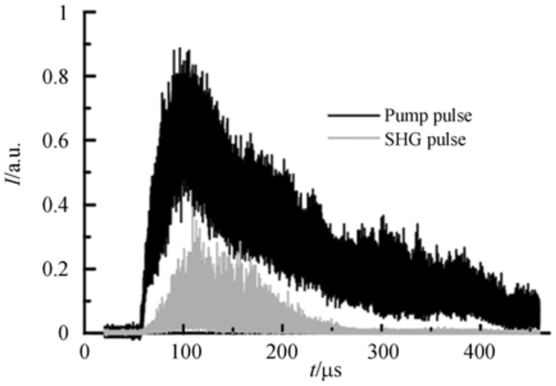

Fig. 7　Temporal shape-form of pump and converted pulses in free-running mode of selective CO laser. Transition 8-7 P(12) ($\lambda=5.224\ \mu$m).

In both cases, SHG was efficient in the first half of the pump pulse when there was maximal peak power of laser emission. But if one looks at pump and converted pulses with high temporal resolution, one can see the difference between them. At the beginning of pump pulse in free-running mode (Fig. 9), one can see spike structure due to intracavity mode beating. Note that laser power was non-zero between the maxima of intensity. Spikes of the converted radiation were correlated with the most powerful spikes of pump radiation. At the beginning of pump pulse in mode-locking (Fig. 10), one can see spike structure with strong periodical structure with axial period of 100 ns. Spikes had a duration about 15 ns (FWHM) and laser power dropped down to zero between the maxima of intensity.

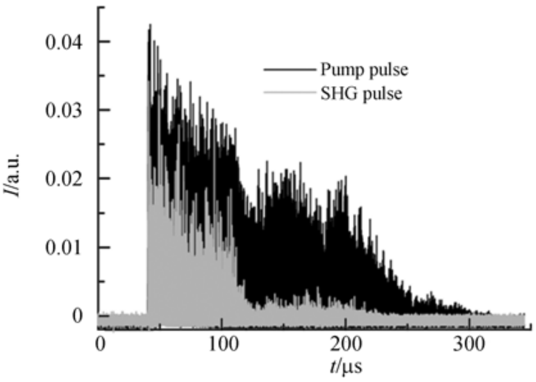

Fig. 8　Temporal shape-form of pump and converted pulses of selective mode-locked CO laser. Transition 8-7 P(12) ($\lambda=5.224\ \mu$m).

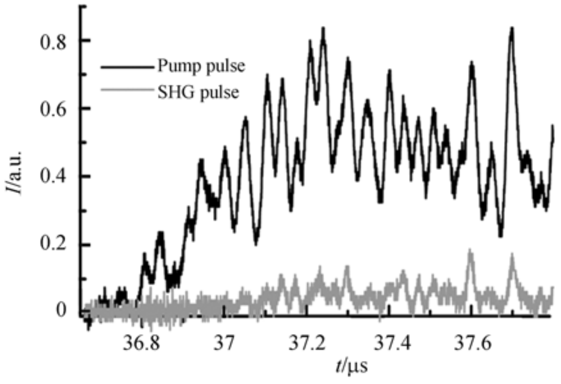

Fig. 9　Pump and converted pulses at the beginning of frequency-selective free-running mode CO lasing. Transition 8-7 P(12) ($\lambda=5.224\ \mu$m)

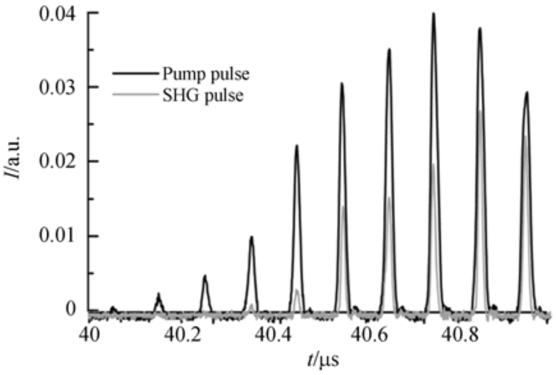

Fig. 10　Pump and converted pulses at the beginning of frequency-selective mode-locking CO lasing. Transition 8-7 P(12) (λ=5.224 μm)

Dependences of the external SHG efficiency from incidence angle of a pump beam on the surface of nonlinear crystal $ZnGeP_2$ are presented in Fig. 11.

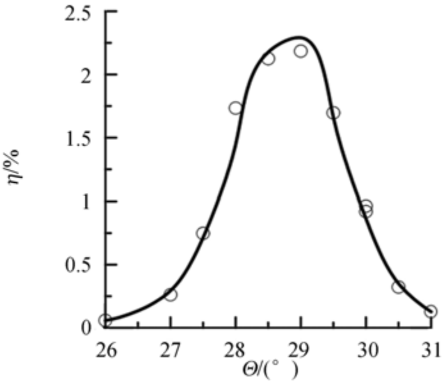

Fig. 11　Dependences of external SHG efficiency from incidence angle of pump beam on surface of nonlinear crystal $ZnGeP_2$. Incidence pump energy is 370 mJ.

According Fig. 11 the wide SHG angular bandwidth is about 2° (FWHM) that matches to the results of [14]. As external SHG efficiency, we used the ratio of SHG energy to the energy of pump CO laser radiation on the crystal. Fig. 12 illustrates dependences of external SHG efficiency on the latter for mode-locked and free-running EBSD CO laser.

The curve corresponding to the mode-locking case is higher than the free-running mode curve due to more powerful spikes of pump radiation. It should be noted that we expected this difference to be higher according to the results obtained for SHG with mode-locked CO_2 laser radiation[17]. It can be related to two factors. The first one is the fact that only the first part of the pump pulse takes part in the frequency conversion (Fig. 8). The second one is the fact of deterioration of quality of mode-locking (Fig. 13).

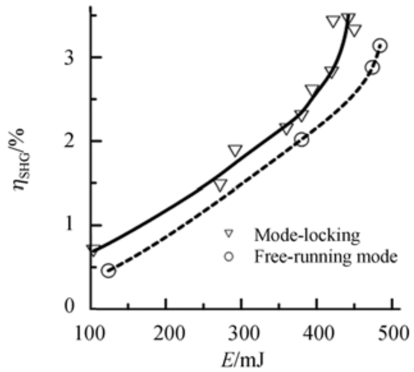

Fig. 12　External SHG efficiency versus the energy of pump CO laser radiation on crystal $ZnGeP_2$

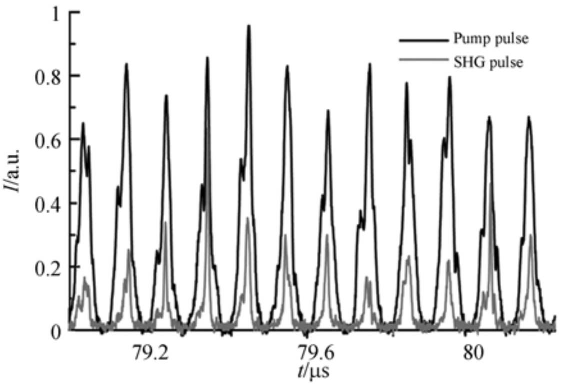

Fig. 13　Time behavior of pump and converted pulses of frequency-selective mode-locked CO laser, 40 μs after beginning of pump pulse.

4　Possibility of frequency tunable emission within extra wide range from 1.25 to 3 000 μm

Taking into account high efficiency of frequency conversion of CO laser radiation, i. e. emission within wide first overtone range 2.5 to 4.2 μm, it can be proposed that frequency converted emission of CO laser can cover extremely wide spectral range from 1.25 to 3 000 μm. In particular, by difference frequency generation or down-conversion of fundamental and overtone bands, it is possible to cover interband range 4.0−5.0 μm. There are no powerful laser sources operating within this spectral range. But such source based on the CO laser frequency converter can be realized rather easily because of CO laser can operate in both fundamental and first overtone bands simultaneously [2,4]. In turn, by difference frequency generation of close emission lines in both bands, it is possible to cover a wide part of THz range at least up to 3 000 μm. For example, in Fig. 14 and Fig. 15 estimated phase matching diagrams for oe-type difference frequency generation in ZnGeP$_2$ of overtone and fundamental CO emission lines into THz range are pictured (one line is fixed, other lines is chosen one by one from the spectral interval shown in the picture inset).

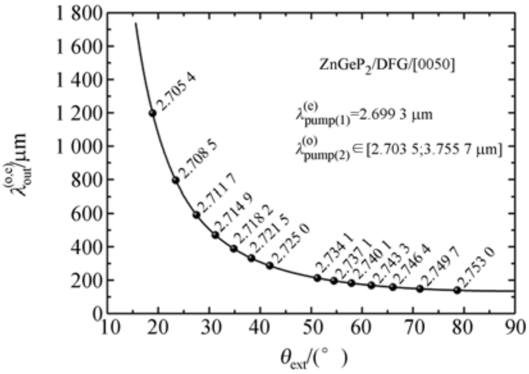

Fig. 14 THz wavelength versus external phase matching angle at pump wavelengths of CO overtone emission lines

Sellmeier equations used in the estimation are from [18]. Here, at phase matching angles 20°−80°, THz wavelength within 200−1 200 μm can be generated. One ZnGeP$_2$ crystals allow realizing so wide spectrum generation. Further extension of THz generating lines can be achieved by mixing of other emission lines of CO laser.

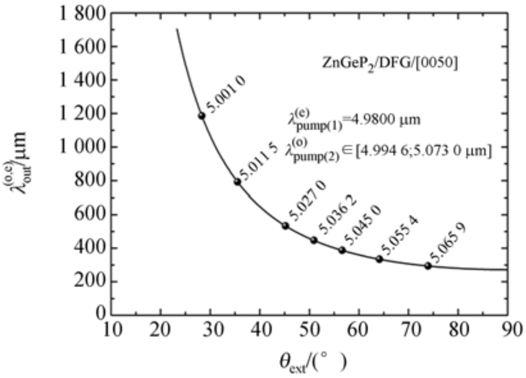

Fig. 15 THz wavelength versus external phase matching angle at pump wavelength of CO fundamental emission band

The same phase matching diagrams for oe-e type difference frequency generation of overtone and fundamental CO emission lines into THz range in GaSe crystal are presented in Fig. 16 and Fig. 17. It is important to note that all 1.15 to 3 000 μm range can be realized in one GaSe sample due to extremely high birefringence $B=0.35$. Sellmeier equations used in the estimation are from [19].

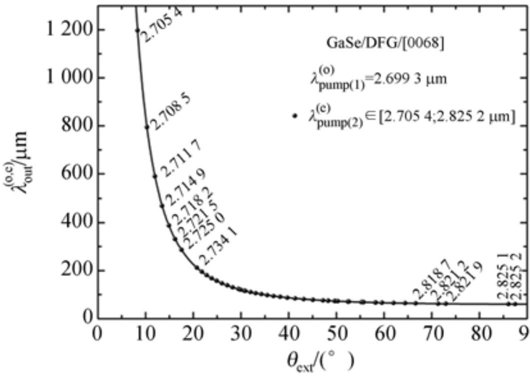

Fig. 16 THz wavelength versus external phase matching angle pumped by CO overtone emission lines

Further improvement in frequency conversion efficiency of CO laser can be achieved by antireflection coating of optical parts and nonlinear crystals, optimization of the pump beam, optics parts and schematic diagram, and crystal parameters, and also by shortening of pump pulses, for example, by mode-locking [16]. It is also proposed to use GaSe crystals with modified physical properties by doping with S, In or Te.

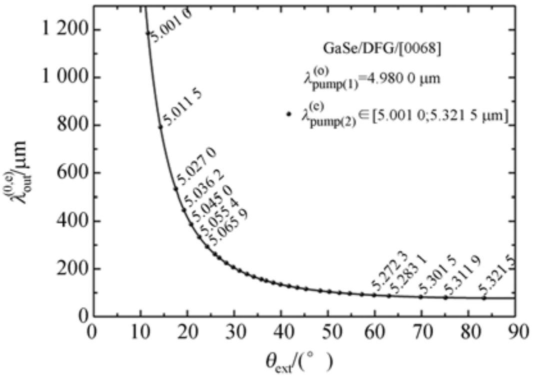

Fig. 17　THz wavelength versus external phase matching angle pumped by CO fundamental emission lines

5 Conclusions

The CO laser SHG observed in a high-quality $ZnGeP_2$ and GaSe crystals are presented. External SHG efficiency of Q-switched multiline CO laser radiation has exceeded 0.15% in GaSe and reached 1 % in $ZnGeP_2$. The SHG in $ZnGeP_2$ of radiation of electron beam sustained discharge frequency-tunable mode-locking CO laser with external efficiency 3.5 % was realized. A possibility of difference frequency conversion of fundamental and first overtone CO laser lines to cover about 4.0—5.0 μm spectral range is discussed. It is shown by calculation that the difference frequency conversion of neighboring lines of both fundamental and first overtone bands can allow one to get in oscillation THz spectral range of about 100—1 200 μm and wider range.

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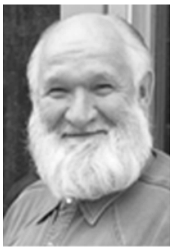

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