Letter

Influence of multi-line CO laser focusing on broadband sum-frequency generation

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Abstract

The influence of a multi-line CO laser focusing on spectral characteristics of broadband sum-frequency generation in ZnGeP$_2$ nonlinear crystal was experimentally and numerically studied. Maximal frequency conversion was experimentally observed under a tight focusing laser beam of a multi-line CO laser rather than a single-line one. The tight focusing resulted in a broadening sum-frequency generation spectrum and an increasing total, i.e. integrated over the spectrum, frequency conversion efficiency. These effects were due to the increasing phase-matching bandwidth and angular dispersion. The maximal conversion efficiency of the multi-line CO laser was numerically demonstrated to take place at a focal length of 0.4 times that required for the single-line one.

Keywords: nonlinear crystal, CO laser, mid-IR range, sum frequency generation

(Some figures may appear in colour only in the online journal)

1. Introduction

Nowadays, broadband mid-IR laser sources are actively being developed because they are required for different applications such as the investigation of ultrafast phenomena [1], optical frequency combs [2], high harmonic generation [3], spectroscopy [2, 4] and other fields. Frequency conversion in nonlinear crystal of laser radiation, including broadband lasers, has become the main element of modern laser systems. To obtain high frequency conversion efficiency, phase matching is required. The most commonly used method for frequency conversion is birefringent, applying phase-matched nonlinear crystals. However, it is difficult to find a nonlinear crystal with the dispersion and the birefringence to satisfy the phase-matching condition for broadband radiations. Broadband phase-matching frequency conversion in mid-IR was theoretically considered for widely-known ZnGeP$_2$, GaSe, AgGaSe$_2$, LiInS$_2$ and other nonlinear crystals which were suggested for the generation and amplification of ultra-short mid-IR laser pulses [5–7]. Experimentally, broadband cascaded frequency conversion at noncritical spectral phase-matching was obtained in ZnGeP$_2$ [8, 9] and AgGaSe$_2$ [10] crystals with a multi-line carbon monoxide laser (CO laser).

The phase-matching bandwidth of nonlinear crystal can be even more enhanced by introducing spectral angular dispersion. In this case, the different spectral components propagate under different phase matching angles in nonlinear crystal [11–13], i.e. under so-called achromatic phase-matching. Achromatic phase-matching was applied for broadband second harmonic generation of both multi-line CO$_2$ laser radiation [11] and femtosecond pulses [12, 13]. A simple way to introduce spectral angular dispersion can be to use a tight focusing laser beam in nonlinear crystal. The influence of beam focusing on frequency conversion is not a new subject and was clearly investigated in [14, 15] for single-line radiation. For broadband (multi-line) radiation it is intuitively clear that stronger focusing should lead to a higher spectral band of crystal, but this question was investigated insufficiently.

In this letter we report on our investigation regarding the influence of a multi-line CO laser beam focusing on spectral characteristics of broadband sum-frequency generation (SFG) in ZnGeP$_2$ crystal. We selected a CO laser because it
is a unique gas laser operating in mid-IR on hundreds of rotational-vibrational transitions in the wavelength interval from 4.6 [16] to 8.7 µm [17] in a single-line or multi-line mode. A multi-line Q-switched CO laser can emit dozens (up to 230 [18]) of spectral lines in a single microsecond pulse [8–10, 18, 19]. The rich spectrum and high power of the CO laser make it very attractive for covering the mid-IR range by frequency conversion of the CO laser radiation in different nonlinear crystals [19]. And noncritical spectral phase-matching in ZnGeP₂ takes place for multi-line CO laser radiation. The obtained results may be useful for the generation and amplification of ultra-short broadband mid-IR laser pulses as well.

2. Experimental setup and results

Our experiment was carried out at the Gas Lasers Laboratory of the P.N. Lebedev Physical Institute. The optical scheme of the experiment presented in figure 1 was the same as in [9].

Low-pressure cryogenic CO laser tube 1 was pumped by a DC discharge. A laser cavity of ~3 m length was formed by spherical mirror 2 (radius of curvature 9 m) and output mirror 3. The laser operated in Q-switch mode by rotating mirror 4. The FWHM pulse duration of the CO laser equaled 0.5 µs at the Q-switch frequency of 100 Hz. The peak power of the laser radiation reached 4 kW at an average power of 0.2 W. A spectrum of the CO laser consisted of about 115 spectral lines in the wavelength range from 4.9 µm to 6.5 µm (figure 2). The CO laser operated on the fundamental transverse mode due to the diaphragm (16), which was 10 mm in diameter and installed near the output mirror.

The multi-line CO laser radiation was focused by a CaF₂ lens (5), with the focal length (f) of 20 cm or 12 cm, onto uncoated ZnGeP₂ crystal (6), of 15 mm length. The crystal was installed at 47.2° phase-matching angle which corresponded to maximal SFG efficiency. To measure the parameters of the CO laser radiation, a small part of the laser beam (~5%) was split off by flat CaF₂ plates (7 and 8) and directed onto photodetector PEM-L-3 (9) and laser power meter Ophir-3A (10), with an additional spherical mirror (15). The radiation coming out of the crystal was collimated by a spherical mirror (11) and directed to an IR spectrometer (12) (IKS-31, LOMO PLC), equipped by a low-noise cryogenic Ge: Au photo-detector or THORLABS PDA20H-EC photo-detector. Measured spectra were displayed by oscilloscope (Tektronix TDS5052B).

To select and measure the power of SFG radiation, the fused silica filter (13) and laser power meter (14) Ophir-3A were installed in front of the spectrometer (12). The transmittance in the SFG spectral range of the applied 2 mm fused silica filter is presented in figure 3(a) (right Y-axis).

When the focal length of lens 5 was 12 cm (numerical aperture ~0.04), the SFG spectrum consisted of 110 lines in the wavelength range from 2.52 to 2.85 µm (figure 3(a)). The external conversion efficiency (the ratio of the SFG radiation power to CO laser power) was 1.8%. When the focal length of the lens was 20 cm (numerical aperture 0.025) the SFG spectrum consisted of 50 lines in the wavelength range from 2.55 to 2.67 µm. The external conversion efficiency was 1.2%. A comparison of SFG spectra obtained under different focusing shows that increasing the numerical aperture by a factor of ~1.7 broadens the SFG spectrum by a factor of ~2.5 and enhances the number of spectral lines twofold. The peak

Figure 1. Optical scheme of the experiment. See text for details.

Figure 2. Spectrum of CO laser.

Figure 3. Measured (a) and calculated (b) SFG spectra for the focal lengths of 20 and 12 cm. Dotted lines are envelope lines. The upper line in figure (a) is transmittance of fused silica filter.
power of the strongest spectral lines (with $\Delta k \approx 0$) falls down about twofold, which corresponds to a twofold decrease in conversion efficiency for a single-line CO laser radiation. But the total, i.e. integrated over the whole laser spectrum, conversion efficiency is 1.5 times higher. The simulation carried out below predicts the maximal conversion efficiency at 8 cm focal length, but in our experiment the shortest focal length of 12 cm was limited by the laser induced surface damage of the ZnGeP$_2$ crystal facet.

It should be noted that in the SFG of the multi-line CO laser in the similar set-up carried out in [20] the focal length of the lens was 4.5 cm. So matched tight focusing was possible due to less peak power of radiation and the very high quality polish of the facets. The tighter focusing in [20] resulted in higher conversion efficiency of up to 4.9% which confirms the advantage of tight focusing.

3. Calculations and discussion

To explain the obtained results, a simplified numerical simulation of the experiment was carried out. Calculated SFG spectra for the focal lengths of 20 cm and 12 cm are presented in figure 3(b). Under a simplified approximation of a plane wave and low conversion efficiency, the SFG radiation power $P_{SF}$ can be calculated by expression [21]

$$P_{SF} = \frac{8\pi^2 d_{eff} L_{d} P_{1} P_{2}}{\varepsilon\varepsilon_0 n L_{SF} A} \sin^2 \left( \frac{|\Delta k| L}{2} \right)$$  \hspace{1cm} (1)

where $d_{eff}$ is the effective nonlinear coefficient; $n$ is refractive index; $L_{SF}$ is SFG wavelength; $\varepsilon_0$ is the dielectric constant; $P_{1}$ and $P_{2}$ are the pump laser radiation power for two CO laser lines; $\Delta k$ is the wave mismatch, $L_{a}$ is effective the crystal length; $A$ is a cross-section of the laser beam. The dispersion equations and effective nonlinear coefficient were taken from [21]. Placing a lens into the optical path results in changing both $L_{a}$ and $A$ which affects conversion efficiency. Also, focusing affects phase-(mis) matching between different CO laser lines through the sinc function.

Intensity distribution in the crystal for a tightly focused laser beam should be taken into account. The exact solution of parametric interaction of a Gaussian light beam for single-line radiation was presented in [14, 15]. Our simplified model is close to the theory of second harmonic generation by focused laser beams considered by Bjorkholm in [14]. We considered a Gaussian beam $w(z)$ focused in the middle of ZnGeP$_2$ crystal ($z = 0$) of 15 mm length (figure 4). The spot radius $w(z)$ of the laser beam is given by

$$w(z) = w_0 \sqrt{1 + \left( \frac{\lambda \cdot z}{n \cdot \pi \cdot w_0^2} \right)^2}$$  \hspace{1cm} (2)

where $w_0$ is the waist radius. For simplicity, the Gaussian beam was calculated for the wavelength $\lambda = 5.5$ $\mu$m and refractive index $n = 3$. The solid and dashed lines in figure 4 correspond to laser beams with and without nonlinear crystal. The laser beam waist in the crystal was lengthening by a factor corresponding to the refractive index of the ZnGeP$_2$ crystal.

The calculation of SFG spectra was carried out only for the beam waist because it is the most intensive and essential part of the interaction. Unlike [14], we did not take into account walk-off effect because of a low ZnGeP$_2$ birefringence ~0.04. In other words, we considered the effective length of crystal $L_e$ to be equal to the waist length $b$, calculated as:

$$b = 2n \frac{\pi \cdot w_0^2}{\lambda}.$$  \hspace{1cm} (3)

The waist edge was defined by expression (2): $w(b/2)$ was $\sqrt{2}$ larger than $w_0$. The cross-section of the laser beam $A$ was calculated as $\pi w_0^2$. Local angular convergence and divergence of the laser beam were calculated as a derivative of the $w(z)$ function in the points of the waist edge. To take into account the angular spread of wave vectors, i.e. angular dispersion, the laser beam was divided into small slices in such a way that an angular aperture of the slices was less than the angular phase-matching bandwidth. These slices propagated in the crystal under different angles relative to the optical axes of both the laser beam and nonlinear crystal (see figure 4). The SFG spectrum for such a slice of the multi-line CO laser beam was calculated by expression (1), taking into account the experimental conditions, as was done in [8]. The resulting SFG spectrum was calculated as a sum of spectral contributions of these small beam slices.

The calculated SFG spectra for the focal length of 20 cm (which corresponded to equality of the waist and crystal length) and 12 cm (which corresponded to tight focusing) are presented in figure 3(b). For 12 cm focal length, the waist radius of the CO laser beam equalled 40 $\mu$m; the waist length and angular spread were 5.5 mm and $\pm 0.59^\circ$, respectively. Angular tolerance for SFG (angular phase-matching bandwidth) equalled $\pm 0.3^\circ$. The calculated SFG spectrum consisted of 939 lines in the wavelength range from 2.52 to 2.76 $\mu$m at 0.1 level of maximal power. For 20 cm focal length, the waist radius of the CO laser beam was 65 $\mu$m; the waist length and angular spread were 15 mm and $\pm 0.35^\circ$, respectively. Angular tolerance for SFG was $\pm 0.1^\circ$. The calculated SFG spectrum consisted of 507 lines in the wavelength range from 2.56 to 2.69 $\mu$m at 0.1 level of maximal power.

In our simulation an enhancement of the numerical aperture by a factor of ~1.7 broadened the SFG spectrum by a factor of ~2, enhanced the number of spectral lines by a factor of ~2, and decreased the peak power of the strongest spectral lines also by a factor of ~2, which correspond to our experimental
The frequency conversion of multi-line CO laser radiation in ZnGeP$_2$ nonlinear crystal differs significantly from that of a single-line laser. For multi-line radiation tight focusing appeared to be more efficient. The tight focusing resulted in the broadening of the SFG spectrum and the increase in the total frequency conversion efficiency (integrated over the spectrum). The SFG spectrum broadening under tight focusing was caused by two factors: by broader phase-matching bandwidth at lesser effective crystal length and by angular dispersion enhancement. The adequate agreement between envelopes of the experimental and calculated spectra demonstrates the applicability of our simple simulation model.

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