Influence of $ZnGeP_2$ crystal temperature on broadband cascaded frequency conversion (numerical study)

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Abstract. Broadband three-stage frequency conversion of multiline CO laser radiation in a single sample of $ZnGeP_2$ crystal was numerically studied at various crystal temperatures. Decrease of the $ZnGeP_2$ crystal temperature resulted in enhancement of efficiency for each frequency conversion stage due to phase-matching shifting. The third stage efficiency at liquid-nitrogen crystal temperature was 1.7-fold higher as compared to one at the room temperature. Also, the crystal temperature decrease resulted in spectral broadening of frequency converted radiation.

1. Introduction

The mid-IR range is the "molecular fingerprint" region. Therefore, a development of mid-IR coherent sources is a very important for gas analysis, laser chemistry, medicine and other applications. Such application as remote multi-component spectroscopy of the atmosphere requires a broadband mid-IR sources with a lot of narrow high brightness spectral lines [1]. A broad spectrum of discrete evenly spaced narrow-band and high-brightness lines is referred to as an optical frequency comb which is traditionally generated by means of mode-locked broadband lasers [2]. It was shown in [3] that both Q-switched multiline CO laser and its sum-frequency (SF) spectra are also broadband reproducible mid-IR optical frequency combs numbering hundreds narrow lines. The latter is attractive for the atmosphere sensing by both differential absorption technique and absorption line profile spectroscopy [3]. It should be noted that multiline Q-switched CO laser can emit more than 200 spectral lines simultaneously within the wavelength interval of 5–8 μm in a single pulse [4]. CO laser sum frequencies radiation covers spectral interval of 2.5–4 μm, respectively.

In addition, a broadband cascaded frequency conversion of multiline CO laser radiation in a single nonlinear crystal is possible under noncritical phase matching. For example, broadband three-stage frequency conversion of multiline sub-microsecond CO laser radiation in a single sample of $ZnGeP_2$ crystal was performed [5]. Such a laser system emitted more than 200 narrow spectral lines within 2.4–6.2 μm spectral range. The broadband cascaded frequency conversion involves following processes: 1) second-harmonic and sum frequency generation for CO laser spectral lines (the 1^{st} stage); 2) difference frequency generation between the 1^{st} stage and residual pump radiation (the 2^{nd} stage); 3) sum frequency generation of the 2^{nd} stage and residual pump radiation (the 3^{rd} stage). Thus, broadband multiline CO laser comb experienced several sum and difference frequency mixing stages both in parallel and simultaneously in a single nonlinear crystal with spectral enrichment and expansion at each stage. This fact significantly enhances capability of such a laser system for multicomponent gas analysis application. It should be pointed out that such hybrid laser system (gas laser and nonlinear crystal) can be robust and compact one due to a slab design of laser device and intracavity scheme of frequency conversion [6, 7].

In paper [8], temperature phase-matching tuning of $ZnGeP_2$ crystal around noncritical spectral phase matching was studied. It was demonstrated that $ZnGeP_2$ crystal temperature changing results in spectral acceptance shifting. It means that the spectral band and efficiency of broadband cascaded CO laser frequency conversion (which was performed in [5]) can be improved by $ZnGeP_2$ crystal temperature changing. Therefore, to analyze this assumption, it is numerically studied in the present paper. Furthermore, the results of the study can be extended and applied for other broadband mid-IR laser sources based on $ZnGeP_2$ crystal like in [9].

2. Numerical simulation and results

At the first, phase-matching angles θ, and spectral phase-matching bandwidth (acceptance) $\Delta\lambda$ were calculated for second-harmonic generation in $ZnGeP_2$ crystal at different temperatures (Figure 1). The X-axis indicates pump radiation wavelength λ. Such calculation is a "standard" procedure which can be found in handbook [10]. The calculations were performed with verified dispersion equation from [8] based on experimental data from [11].

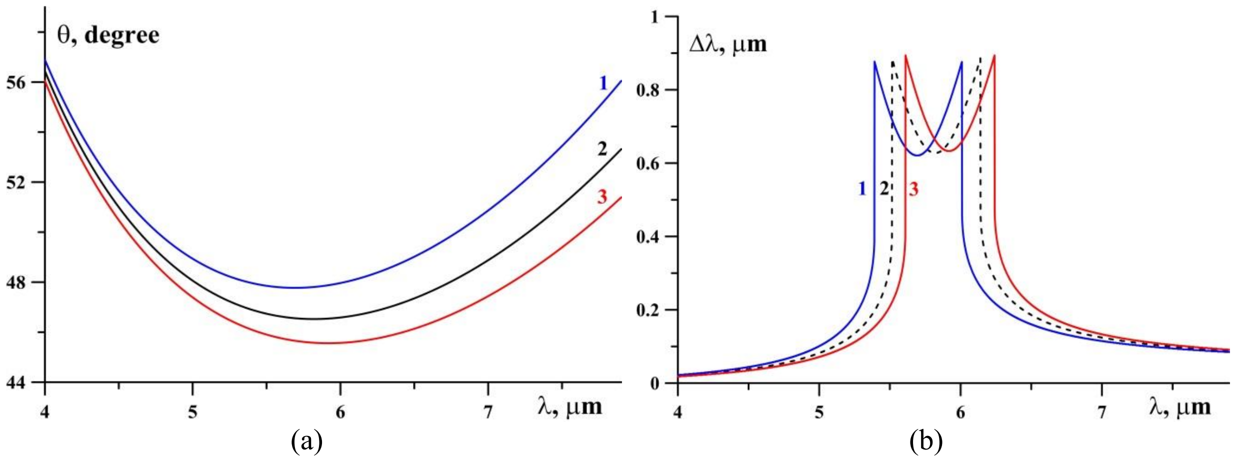

Figure 1. Calculated phase-matching angles (a) and spectral phase-matching bandwidth (b) at $ZnGeP_2$ crystal temperature of -196 °C (1), 23 °C (2) and 200 °C (3).

The Figure 1(a) shows that $ZnGeP_2$ crystal heat decreases phase-matching angles; the curve minimum is shifted to longer wavelengths. A reverse effect is for a crystal cooling. The curve minimum shifting to shorter wavelengths results in shifting of the broadest spectral acceptance to the shortwave domain (Figure 1(b)). That is an advantage for a broadband cascaded frequency conversion of multiline CO laser radiation. It should be noted that the value of the spectral acceptance almost does not change with temperature and is even slightly less at low temperature.

To find out an effect of $ZnGeP_2$ crystal temperature on broadband cascaded frequency conversion of multiline CO laser radiation, a numerical simulation was performed. The simulation was carried out under the plane-wave fixed-field approximation that is described in handbook [10] in details. The simulation was carried out with a CO laser spectrum (Figure 2(a)) from experimental study of three-stage frequency conversion in $ZnGeP_2$ crystal [4]. It consisted of 63 spectral lines within the wavelength range from 4.9 μm to 6.2 μm with 3 kW total peak power. The maximal peak power of individual CO laser line was 163 W. A radius of the laser beam spot was 0.1 mm. $ZnGeP_2$ crystal of 10-mm length was cut for type-I of mixing ($\varphi = 0°$). $ZnGeP_2$ second-order nonlinear coefficient d_{36} was equal 75.4 pm/V. The phase-matching angle θ was optimized for second-harmonic generation of 4.93 μm line as in paper [5]. The phase-matching angles θ at different temperatures were as follows: 49.2° (-196 °C), 48.4° (23 °C), and 47.7° (200 °C).

The 1st stage of the frequency conversion was broadband sum-frequency generation in the spectral range from 2.45 μm to 3.1 μm. The spectra of the 1st stage were calculated at temperatures of -196 °C, 23 °C, and 200 °C (Figure 2(b)). Maximum of SF peak power was near 2.47 μm wavelength. Figure 2(b) shows that crystal cooling resulted in SF power increase. The SF peak power integrated over spectrum was 573 W, 410 W, and 328 W at temperatures of -196 °C, 23 °C, and 200 °C, respectively. The conversion efficiency was calculated as a ratio of integral SF peak power to integral CO laser peak power. The conversion efficiencies were 19.0%, 13.6%, and 10.9% at temperatures of -196 °C (liquid nitrogen temperature), 23 °C (the room temperature), and 200 °C, respectively.

Next, spectra for the 2nd stage of the frequency conversion were calculated (Figure 2(c)). In this case, the 1st stage output radiation was mixed with the residual pump radiation in the same crystal, leading to parallel difference frequency generation within 4.1–5.2 μm wavelength range. The maximal 2nd stage efficiency of 1.3% was obtained at -196 °C temperature. The conversion efficiency at 23 °C and 200 °C was 0.9%, and 0.7%, respectively. As the 3rd stage of the frequency conversion, we considered SF generation between the 2nd stage radiation and the residual pump radiation. The spectral range of the 3rd stage radiation was within 2.35–2.60 μm wavelength range (Figure 2(d)). The maximal 3rd stage efficiency of 0.7% was also obtained at -196 °C temperature. The 3rd stage efficiency at 23 °C and 200 °C was 0.4%, and 0.3%, respectively.

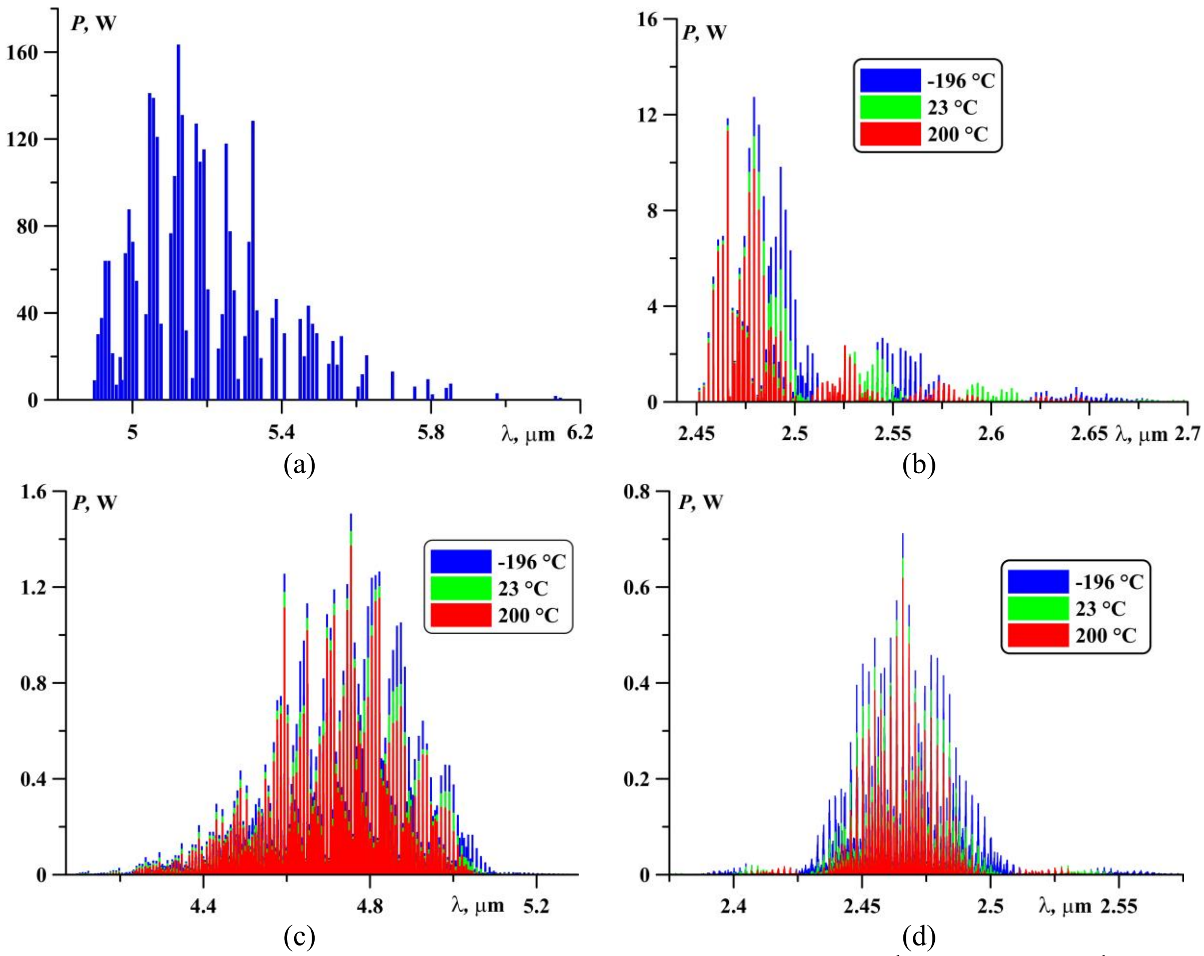

Figure 2. CO laser spectrum (a) and calculated spectra for 1st stage (b), 2nd stage (c), and 3rd stage (d) of broadband cascaded frequency conversion of multiline CO laser radiation in $ZnGeP_2$ crystal at different temperatures.

Conversion efficiencies of the 1st stage, the 2nd stage, and the 3rd stage at different temperatures normalized to its value at the room temperature are shown in Figure 3. Figure 3 demonstrates that the crystal cooling resulted in conversion efficiency enhancement. $ZnGeP_2$ crystal cooling down to liquid nitrogen temperature allows one to increase the 1st, 2nd and 3rd stage efficiency by 1.4, 1.45 and 1.7 times, respectively. Improving integral conversion efficiency is associated with expansion of the spectral phase-matching bandwidth for the process under consideration. Figure 2(d) demonstrates that the broadest 3rd stage spectrum was obtained at -196 °C temperature.

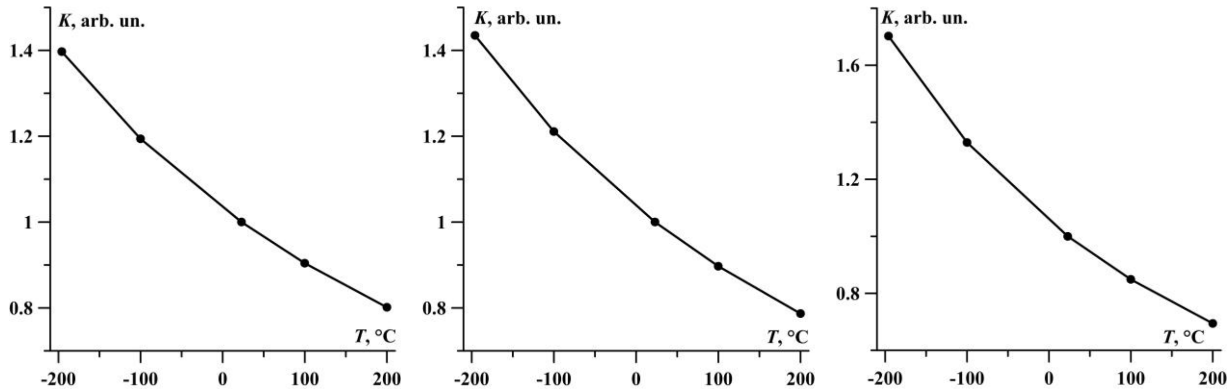

Figure 3. Calculated dependence of maximum peak power on $ZnGeP_2$ crystal temperature: the 1st stage (a), the 2nd stage (b), the 3rd stage (c)

3. Conclusions

Broadband three-stage frequency conversion of multiline CO laser radiation in a single sample of $ZnGeP_2$ crystal was numerically studied at various crystal temperatures. Our numerical simulation demonstrated that $ZnGeP_2$ crystal temperature falling down to the liquid nitrogen temperature leads to 1.4-, 1.4- and 1.7-fold increase for efficiencies of the 1st, the 2nd, and the 3rd stages, respectively, as compared to one at the room temperature. Also, crystal cooling resulted in spectrum broadening of frequency converted radiation. At higher temperatures, maximal peak powers and conversion efficiencies were lower. Also, it should be noted that one can expect for crystal cooling to diminish crystal absorption losses and enhance optical damage threshold.

Acknowledgments

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