Sum frequency conversion of multiline CO laser emission in BaGa$_2$GeSe$_6$ crystal of one cm long

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Abstract. Sum frequency conversion of multiline Q-switched CO laser radiation was studied in 10.4 mm-long BaGa$_2$GeSe$_6$ crystal under noncritical spectral phase-matching conditions. Spectrum of the CO laser consisted of 51 emission lines in the wavelength range of 4.95-6.20 μm. The frequency converted spectrum contained 85 spectral lines in the wavelength range of 2.5-2.8 μm. The internal energy conversion efficiency (taking into account optical losses on uncoated facets of the crystal) was up to 1.7%.

1. Introduction
Since the first observation of optical second harmonic generation [1] up to present time, different nonlinear optical phenomena, materials and techniques have been under intensive research. Particularly, there was reported in Ref [2] about discovery and synthesis of new promising mid-IR nonlinear optical crystals: BaGa$_2$GeSe$_6$ (BGGSe) and BaGa$_2$GeS$_6$. The BGGSe and BaGa$_2$GeS$_6$ crystals of high quality and long (several centimeters) size were grown and characterized at the Kuban State University [3]. A numerical study of these crystals [4] indicated that the BGGSe was very attractive for frequency conversion of the mid-IR radiation, i.e. CO- and CO$_2$-laser radiation. Authors of Ref [4] noted that the BGGSe crystal may be more efficient or, at least, competitive with the “standard” mid-IR crystal – ZnGeP$_2$ crystal.

Experimental study of second harmonic generation in BGGSe crystal was performed with CO$_2$ laser [3] and CO laser [5] radiation. The main object of the experiments [3, 5] was research and examination of linear and nonlinear properties of the new crystal. Therefore, a thin (of 4.7 mm length) sample of the crystal was applied. To get high efficiency of the frequency conversion, a crystal length should be significantly longer (at least 10 mm) because the conversion efficiency quadratically increases with the crystal length (see e.g. [6]). The object of this paper is the experimental study of broadband sum frequency generation (SFG) including second harmonic generation as a particular case of multiline Q-switched CO laser radiation in the longer (10.4 mm-long) BGGSe crystal.

2. Experimental set-up
The optical scheme of the experiment is shown in Figure 1. We used a cryogenically cooled low-pressure DC discharge CO laser, operated in Q-switching mode. Active media of this CO laser was gas mixture of He:N$_2$:CO = 70:6:1 at total gas pressure of 7.7 Torr with a little additive of air (~0.1 Torr). DC discharge voltage on the tube was 10 kV at DC current of 7 mA. An optical resonator
was formed by a rear spherical mirror (curvature radius 9.0 m) and flat output mirror with reflectivity of 70% in the wavelength range from 5.0 to 6.0 μm. A flat rotating mirror in the resonator provided Q-switching mode.

Figure 1. Optical scheme of the experiment.

A part (~6%) of laser beam was split off to control power and duration of the radiation pulses. A main part of the laser emission (~94%) was focused into BGGSe crystal with CaF$_2$ lens (the focal length 11.5 cm). The uncoated BGGSe crystal of 6.4x6.9x10.4 mm$^3$ size was grown in the Kuban State University and cut for I type of frequency conversion: polar angle $\theta=22.0\pm0.5$ degree and azimuthal angle $\varphi=30$ degree. A radiation coming out from the crystal was collimated by CaF$_2$ lens (the focal length 11.5 cm) and directed to the IR spectrometer (IKS-31, LOMO Ltd.). To measure SFG radiation power (or pulse waveforms) IR silica plane-parallel filter and power meter (or photodetector) were installed in front of the spectrometer. Power was measured by thermopile-based laser power/energy sensor Ophir-3A, pulse waveforms were measured by photoelectromagnetic detector PEM-L-3 (VIGO System).

3. Experimental results
The CO laser operated in pulse periodic mode at 102±1 Hz pulse repetition rate. Average radiation power was 0.18 W, pulse duration – 0.35 μs (Figure 2a). Peak radiation power was about 5 kW.

![Figure 2a](image1)

**Figure 2.** a) Waveforms of CO laser (channel 1) and SFG pulses (channel 2). b) Internal SFG efficiency versus the distance between the front crystal facet and the lens.

Optimal crystal position, relative to the focusing lens, was experimentally found by crystal moving along the laser beam (Figure 2b) and slight (±1 degree) tuning of the $\theta$ angle. Maximal SFG power (2.2 mW) was observed at 127 mm distance between a front crystal facet and the lens, where the focus of CO laser beam was located. Maximal external conversion efficiency reached 1.2% that corresponded to internal efficiency (taking into account the reflection losses on uncoated crystal facets) of 1.7% (Figure 2b). SFG radiation pulse duration was 0.27 μs that was 1.3 times less than...
pump pulse duration. This fact is due to SFG nonlinear process when conversion efficiency for intensive pulse maximum is significantly higher than for weak pulse wings. Another reason of shorter SFG pulse was related to some features of multiline Q-switched CO lasing. Spectral lines belonging to a Q-switched CO laser pulse have a time-spread which results in diminishing SFG efficiency, especially for weak lines at the beginning and end of the pulse [7]. Internal conversion efficiency estimated by peak power, i.e. considering the difference between pump and converted pulses duration, was 2.2%.

In the next experimental series, spectra of CO laser and SFG radiation were measured. The CO laser spectrum consisted of 51 lines in the wavelength range from 4.9 to 6.2 μm with maximum peak power at 5.1 μm (Figure 3a).

![Figure 3. Spectra of CO laser (a) and SFG (b) radiation.](image)

The SFG spectrum measured under normal incidence of CO laser beam onto the crystal consisted of at least 85 lines in the spectral range from 2.5 μm to 2.8 μm (Figure 3b). The SFG spectra measured under tuning of the incidence angle by ±1 degree were pretty the same due to high spectral phase-matching bandwidth of BGGSe crystal for multiline CO laser radiation.

4. Conclusions
Broadband sum frequency conversion of multiline Q-switched CO laser radiation was experimentally studied in the 10.4mm-long BaGa2GeSe6 crystal. Maximal internal energy conversion efficiency was 1.7% that 1.7 times one obtained in 4.7mm-long BaGa2GeSe6 crystal [5]. SFG pulse duration was 1.3 times less than CO laser pulse duration. Maximal internal peak power conversion efficiency was 2.2%. Thus, the conversion efficiency for 10.4mm-long BaGa2GeSe6 crystal was 1.7 times that of the 4.7mm-long one. We expected that two-fold increase of the crystal length should significantly (about 4 times) enhance SFG efficiency. The reasons of the lower efficient gain may be connected with a walkoff effect and narrower phase-matching spectral bandwidth for longer crystal.

Acknowledgments
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5. References
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