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Sum-frequency generation of Q-switched CO laser radiation in $BaGa_2GeSe_6$ and GaSe nonlinear crystals

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Abstract Sum-frequency generation of multiline Q-switched CO laser radiation is studied in the $BaGa_2GeSe_6$ (BGGSe) crystal and in the GaSe crystal. The maximal external conversion efficiency obtained in the BGGSe crystal is 0.7%, which corresponded to the internal conversion efficiency of 1% taking into account optical losses under reflection from uncoated facets of the crystal. The maximum external conversion efficiency in the GaSe crystal is 0.15% with internal efficiency 0.31%. Effective nonlinearity of BGGSe crystal is determined through the ratio between measured efficiency in BGGSe and GaSe crystals. Effective nonlinearity for sum-frequency generation of the CO laser emission in the BGGSe crystal is $d_{eff} = 42 \pm 14$ pm/V, and nonlinear coefficient of this crystal is $d_{11} = 49 \pm 15$ pm/V.

Keywords Sum-frequency generation · Multiline CO laser · Nonlinear crystals · Nonlinear coefficient

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1 Introduction

One of the most effective sources of laser radiation in the mid-IR is a CO laser. This laser can generate on a number of rotational–vibrational lines. The number of these lines can reach many hundreds for fundamental vibrational band in the wavelength range from 4.7 µm (Ionin 2007) to 8.7 µm (Ionin et al. 2017a) with efficiency of ~ 50% and for first-overtone vibrational band in the wavelength range of 2.5–4.2 µm with efficiency ~ 16% (Ionin et al. 2010). One of the procedures for expanding and enriching the CO laser spectrum is a frequency conversion of its radiation in nonlinear crystals. At present, sum-frequency generation (SFG) of multiline CO laser radiation in $ZnGeP_2$ (Andreev et al. 2013), $AgGaSe_2$ (Budilova et al. 2016) and $PbIn_6Te_{10}$ (Ionin et al. 2016) with internal conversion efficiency of 6.5, 1, and 0.01%, respectively, are reported. Our estimations of the phase-matching (PM) angle show that GaSe and $BaGa_2GeSe_6$ (BGGSe) crystals can be also potentially effective for the same type of conversion (Ionin et al. 2018).

The second harmonic generation (SHG) of CO_2 laser (single 100-ns pulses at 10.6 µm) was already realized in the BGGSe crystal (Badikov et al. 2016). The authors of this paper demonstrated that damage threshold in terms of peak values was 11 J/cm^2 (110 MW/cm^2) for BGGSe. That is significantly higher than for the "standard" mid-IR crystal $ZnGeP_2$, 78 MW/cm^2 under the similar conditions (Nikogosyan 2005).

The purpose of this paper is the experimental study of broadband SFG (including SHG as a particular case) of multiline Q-switched CO laser radiation in GaSe and new nonlinear crystal BGGSe. Also, we estimate nonlinear coefficient of the latter.

2 Experimental setup

The optical scheme of the experiments is shown in Fig. 1. We used cryogenically cooled low pressure DC discharge CO laser, operated in Q-switch mode, time-resolved spectral characteristics of which are studied in detailed in (Ionin et al. 2017b). For the active media 1 of the CO laser we used mixture of He:N_2:CO = 70:6:1 at total gas pressure of 7.7 Torr with little additive of air (~ 0.1 Torr) at the voltage on the tube ~ 10 kV and DC current ~ 7 mA. Optical resonator is formed by total reflective spherical mirror 2 (curvature radius 9.0 m), flat output mirror 3 and flat rotating mirror 4 providing Q-switching mode. In contrast to previous studies (Andreev et al. 2013; Budilova et al. 2016; Ionin et al. 2016)

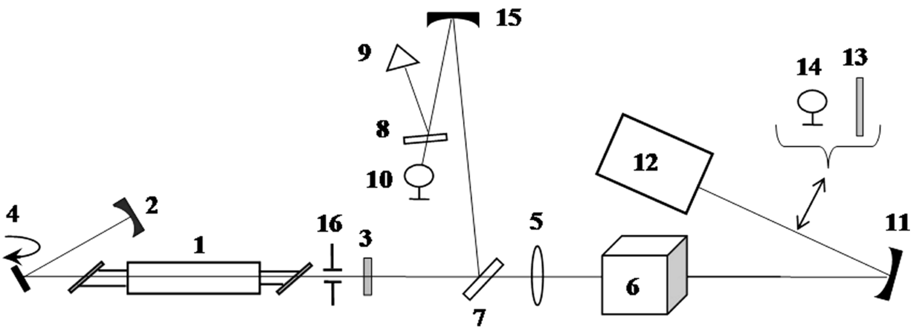


Fig. 1 Optical scheme of the experiments

this laser used an output mirror 3 with reflectivity of 70±3% in the wavelength range from 5.0 to 6.0 µm. In this case, by changing Q-switching frequency in the range from 60 to 160 Hz, the laser pulse duration varies from ~ 0.6 to ~ 0.2 µs, and the laser peak power—from ~ 5.5 to ~ 3.5 kW (see Fig. 2).

A part of CO laser emission (~ 6%) is sent to power meter 10 (Ophir-10A) and photodetector 9 (PEM-L-3) with BaF_2 plane-parallel plates 7 and 8. The main part of the laser emission (~ 94%) is focused with CaF_2 lens 5 onto nonlinear crystal 6. Behind the crystal the beam is collimated with spherical mirror 11 (curvature radius 50 cm). The SFG emission (in 2.4–3.0 µm spectral range) is separated from the pump pulse with IR silica plane-parallel plate 13 with the width of 2 mm and is directed to power meter 14 (Ophir-3A).

Spectral characteristics of CO laser pulses and of converted emission in the nonlinear crystals are measured using a scanning monochromator of IR spectrometer 12 (IKS-31, LOMO Ltd., spectral resolution is 0.1 nm for 2.5 µm and 0.3 nm for 5 µm). The CO laser spectrum consisted of 66 lines in the wavelength range from 4.9 to 6.2 µm with maximum peak power at 5.2 µm (see Fig. 3).

3 Experimental results

In the experiments, SFG is studied with the same sample of BGGSe crystal of 4.7 mm thickness, cut angle θ = 36.0 ± 0.5° and φ = 30°, as in paper (Badikov et al. 2016). The crystal structure belongs to the space group R3 of the trigonal system. Several series of measurements are performed using different lenses with the focal lengths F = 20, 11.5, and 6 cm. Maximal SFG efficiency is observed at the incident angle to the input crystal face of ~ 37°, which correspond to the PM angle of ~ 22° (Fig. 4).

Under focusing by the lens with the focal distance of F = 11.5 cm the efficiency of SFG reach 0.7% that corresponded to internal efficiency of 1% (Fig. 4) taking into account the reflection losses on uncoated crystal facets. The PM angular width at a half of the

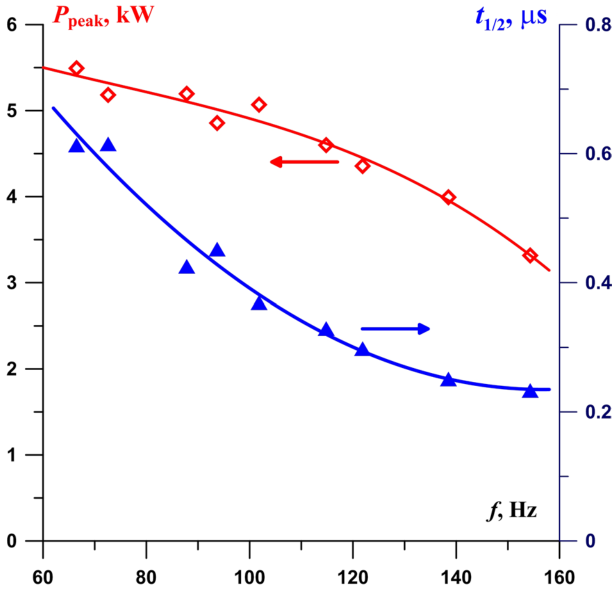


Fig. 2 Peak power and pulse duration of CO laser in dependence of Q-switching repetition rate

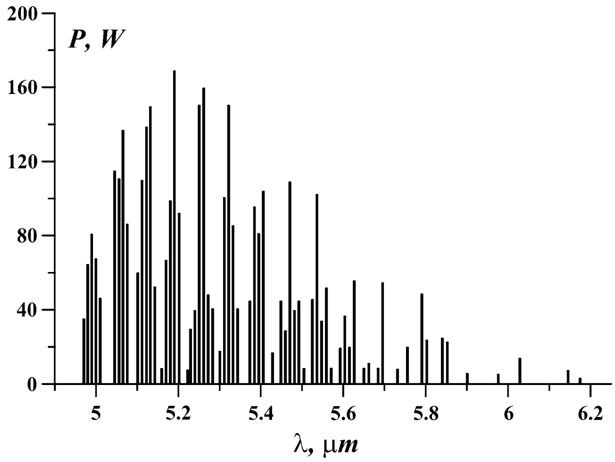


Fig. 3 CO laser emission spectra at Q-switching repetition rate of ~ 100 Hz

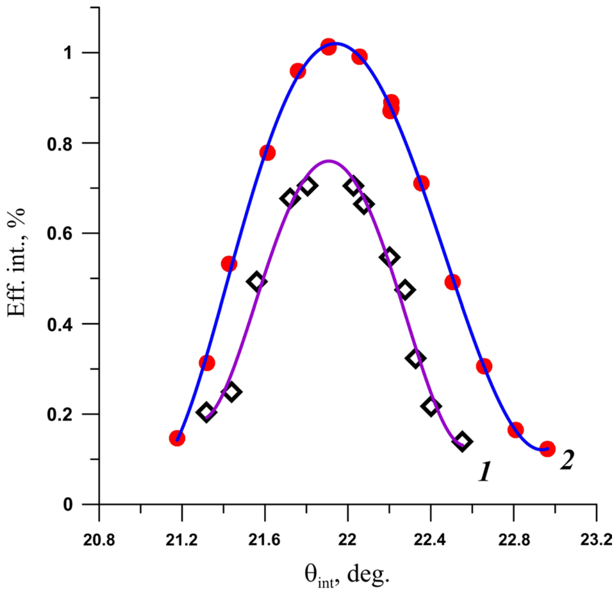


Fig. 4 Internal efficiency of SFG in BGGSe crystal in dependence on phase matching angle at F = 20 cm (1) and F = 11.5 cm (2)

maximum is 1.1 degrees. For lens with the longer focal distance F = 20 cm internal conversion efficiency and PM angular width of SFG is 0.7% and 0.8°, respectively. For the lens with the focal distance of F = 6 cm internal conversion efficiency is 0.46%.

Measured spectra of SFG emission under focusing with the lenses with focal lengths of F = 20 cm and F = 11.5 cm are presented in Fig. 5a, b respectively.

When we use the lens with focal length of F = 20 cm, the SFG spectrum consisted of 110 lines in the spectral range from 2.48 to 2.8 μm. With the lens of F = 11.5 cm the SFG spectrum contain 209 lines. Comparison of the results obtained with different lenses shows that more tight focusing leads not only to an increase of the conversion efficiency, but also to an increase of the angular and spectral width of phase-matching. However, if focusing is too tight, the conversion efficiency begins to decrease because of the short effective length of the crystal.

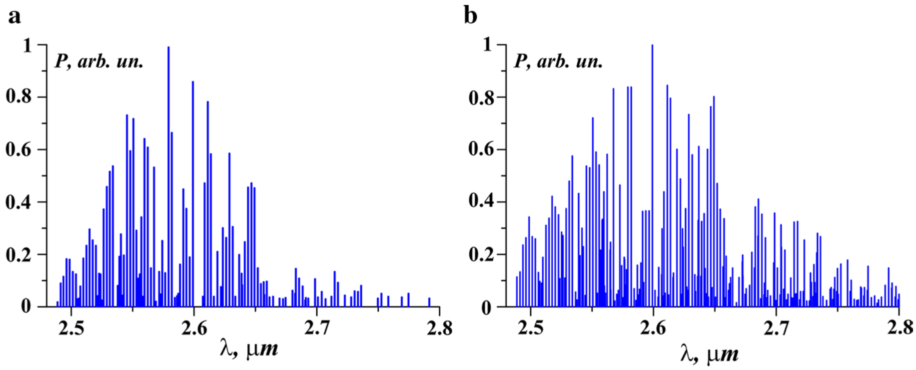


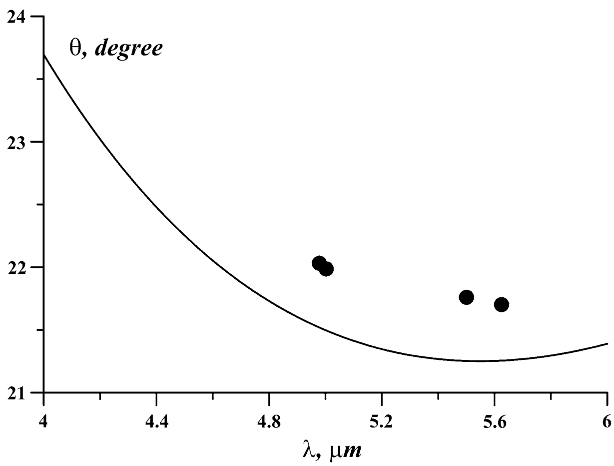
Fig. 5 SFG in BGGSe emission spectra under focusing with the lens with focal length of F = 20 cm (**a**) and F = 11.5 cm (**b**)

Comparison of calculated values of PM angle for SHG (as a partial case of SFG) and experimentally obtained values of PM angle for several SFG spectral lines is presented in Fig. 6. The dispersion equations for BGGSe crystal used in the calculation are introduced in Badikov et al. (2016). The PM angles for several SFG spectral lines are measured by finding maximal amplitude of the signal after the monochromator by tuning the angle of the crystal.

The PM angular measurement error of SHG is ± 0.05°. Herewith the difference between measured and calculated angles is ~ 0.5° degrees, which may be due to an inaccuracy both the cut angle and the dispersion equations. It is worth noting that in (Badikov et al. 2016) the discrepancy between measured and calculated PM angles of SHG of CO_2 laser was also obtained and amounted 0.3°. Therefore an additional test of the dispersion equations given in (Badikov et al. 2016) is required.

The SFG efficiency in the GaSe crystal with the length of 6 mm is measured to determine nonlinear coefficient of BGGSe crystal. For that purpose CO laser emission is focused with the lens of 20 cm focal length. Maximal conversion efficiency is observed at PM angle of ~ 10.8° and reached 0.15%, hence internal efficiency is 0.31%. The SFG

Fig. 6 Calculated and measured PM angle values



spectra contained more than 140 lines in the wavelength range from 2.48 to 2.8 μm (Fig. 7).

4 Estimation of nonlinear coefficient of BGGSe crystal

The SHG efficiency in the approximation of plane waves and low conversion efficiency is given by the formula (Dmitriev et al. 1999):

$$K = \frac{2\pi^2 d_{eff}^2 L_{eff}^2 P}{\varepsilon_0 c n_1^2 n_2 \lambda_2^2 A} sinc^2\left(\frac{|\Delta k| L_{eff}}{2}\right) \tag{1}$$

where d_{eff} is the effective nonlinear nonlinearity; n is the refractive index; λ_2 is sum-frequency wavelength; ε_0 is the dielectric constant; P is power of CO laser lines; Δk is the wave mismatch, L_{eff} is effective crystal length; A is a cross-section of the laser beam.

Effective nonlinearity of BGGSe crystal can be determined through the ratio between measured efficiency in BGGSe and GaSe crystals. However, experiments are conducted at multi-frequency emission conversion regime, which is why PM spectral width determined by a *sinc* function in (1) plays significant role. Nevertheless, we obtained the close spectral characteristics of SFG emission (wavelength range from 2.48 to 2.8 μm with maximum at 2.6 μm and full width at a half of the maximum of a spectra envelope was ~ 0.15 μm) for two cases. One of them is SFG in BGGSe crystal under the focusing with the lens of F = 11.5 cm and the second is SFG in GaSe crystal with the lens of f = 20 cm. Therefore *sinc* function can be reduced in an efficiency ratio (K_1 for GaSe crystal and K_2 for BGGSe) and effective nonlinearity is given by:

$$\frac{K_1}{K_2} = \frac{d_{eff\_1}^2}{d_{eff\_2}^2} \cdot \frac{L_{eff\_1}^2}{L_{eff\_2}^2} \cdot \frac{n_{1\_2}^2 n_{2\_2}}{n_{1\_1}^2 n_{2\_1}} \cdot \frac{\omega_{0\_2}^2}{\omega_{0\_1}^2}, \tag{2}$$

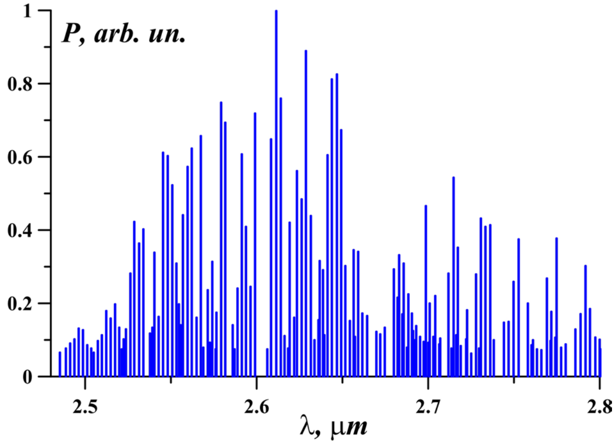


Fig. 7 The SFG spectrum in GaSe crystal under focusing with the lens of the focal length of 20 cm

where ω_{0_x}—radius of the beam waist. Effective interaction length of the crystal L_{eff} was taken as a minimum of the real crystal length L_c, the aperture length L_a and the waist length L_f (Boyd and Kleinman 1968).

The refractive indices of GaSe crystals are calculated using dispersive equations from (Nikogosyan 2005).

The radius of the beam waist for the lens of F = 20 cm measured with Foucault's knife is ω_{0_1} = 90 μm. The diameter of the beam waist for lenses with the focal distances of F = 11.5 cm and F = 6 cm is calculated according to the expression:

$$\omega_{0\_x} = \frac{l_x}{l_1}\omega_{0\_1}, \tag{3}$$

where l_x is the distance from the waist to the main plane of the lens. Waist size is ω_{0_2} = 51 μm and ω_{0_3} = 27 μm for F = 11.5 cm and F = 6 cm respectively. The length of the focal waist L_f (double Rayleigh length) taking into consideration refractive index of the crystal is calculated from the formula (Boyd and Kleinman 1968):

$$L_f = \frac{2\pi \cdot n \cdot \omega_0^2}{\lambda} \cdot \frac{\pi}{2}, \tag{4}$$

where n_o (for GaSe at 5.2 μm) = 2.724, n_e (for BGGSe at 5.2 μm, θ = 22°) = n_o (for BGGSe at 2.6 μm) = 2.491.

The influence of spectrum width (multiline) is not taken into account.

The walk-off angle of the Poynting vector for e-wave of the emission is calculated according to the expression:

$$\rho = -\frac{1}{n_e}\frac{\partial n_e}{\partial \theta}, \tag{5}$$

where for GaSe *e*-wave corresponds to 2.6 μm, for BGGSe *e*-wave corresponds to 5.2 μm.

Aperture length of the crystal L_a (the length of the radiation drift) is calculated in accordance with the expression (Boyd and Kleinman 1968):

$$L_a = \frac{\sqrt{\pi} \cdot \omega_0}{\rho}. \tag{6}$$

Since the aperture length L_a in both crystals is smaller than the crystal length L_c and the waist length L_f, the effective interaction length L_{eff} in the crystal is determined as the geometric mean between the real crystal length and the aperture length (Boyd and Kleinman 1968):

$$L_{eff} = \sqrt{L_a L_c}. \tag{7}$$

The real crystal length L_c including crystal inclination angle and refraction of laser beam is 6.1 mm and 4.8 mm for GaSe and BGGSe crystals respectively.

Effective nonlinearity of GaSe crystal is calculated from the formula (Nikogosyan 2005):

$$d_{eff\_1} = d_{22}\cos(\theta)\sin(3\phi) = 53 \text{ pm/V}, \tag{8}$$

where d_{22} = 54 ± 11 pm/V (Nikogosyan 2005), PM angle θ = 10.8°, φ = 30°. Therefore effective nonlinearity of SFG of the CO laser emission in GaSe crystal is equal to 53 ± 10 pm/V.

Effective nonlinearity of BGGSe crystal is calculated from the formula (Badikov et al. 2016):

$$d_{eff\_2} = (d_{11}\sin(3\phi) + d_{22}\cos(3\phi))\cos^2(\theta) = d_{11}\cos^2(\theta) = 0.86d_{11}, \tag{9}$$

where d_{11}—required value of nonlinear coefficient, PM angle $\theta = 22°$, $\varphi = 30°$.

The parameters used for the estimation of the BGGSe crystal nonlinear coefficient according to (2) are presented in a Table 1.

Effective length of conversion is 2.7 and 4.5 mm for GaSe crystal and BGGSe crystal, respectively. The radius of the beam waist is 90 and 51 μm respectively.

$$d_{eff\_2} = d_{eff\_1}\sqrt{\frac{n_2^3}{n_1^3} \cdot \frac{K_2}{K_1} \cdot \frac{L_{eff\_1}}{L_{eff\_2}} \cdot \frac{\omega_{0\_2}}{\omega_{0\_1}}} \tag{8}$$

$$d_{eff\_2} = 53 \cdot \sqrt{\frac{2.491^3}{2.724^3} \cdot \frac{1.02}{0.31} \cdot \frac{4.06}{4.65}} \cdot \frac{51}{90} = 0.785 \cdot 53 = 42 \tag{9}$$

From the equations above nonlinear coefficient can be derived to be equal to $d_{11} = 49 \pm 15$ pm/V.

5 Conclusions

The sum-frequency generation (SFG) of CO laser radiation is studied with the $BaGa_2GeSe_6$ crystal of 4.7 mm thickness. Several series of measurements are performed using different lenses with the focal length $F = 20$, 11.5 and 6 cm. The maximum SFG external efficiency is observed at phase-matching (PM) angle of 22°. At F = 11.5 cm external efficiency reaches 0.7%, which corresponded to the SFG internal efficiency of 1% taking into account optical losses under reflection from uncoated facets of the crystal. The PM angle width at 0.5-level is 1.1°. At $F = 20$ cm, the SFG internal efficiency is 0.7%, and the PM angle width is 0.8°. When using lens with $F = 6$ cm, the maximum internal efficiency is 0.46%. The SFG spectrum lay in the wavelength range from 2.48 to 2.80 μm. It consisted of 110 lines at $F = 20$ cm, and 209 lines at $F = 11.5$ cm. Comparison of the results obtained with different lenses shows that tighter focusing leads not only to an increase of the conversion efficiency, but also to an increase of the angular and spectral width of phase-matching. However, if focusing is too sharp, the conversion efficiency begins to decrease because of the short effective length of the crystal.

The SFG of CO laser radiation in GaSe crystal of 6 mm length is studied too. The CO laser radiation is focused by a lens with $F = 20$ cm. The maximum external SFG efficiency is observed at PM angle of 10.8° and reaches 0.15%, i.e. internal efficiency reaches 0.31%.

Table 1 Parameters used for the estimation of the BGGSe crystal nonlinear coefficient

Crystal	ω_0 (μm)	L_f (mm)	ρ (degr./rad.)	L_a (mm)	L_c (mm)	L_{eff} (mm)	K (%)
GaSe	90	42	3.4/0.06	2.7	6.1	4.06	0.31
BGGSe	51	14	1.3/0.02	4.5	4.8	4.65	1.02

The SFG spectrum consists of more than 140 lines in the wavelength range from 2.48 to 2.8 μm with maximum at 2.6 μm.

Effective nonlinearity of BGGSe crystal is determined through the ratio between measured efficiency in BGGSe and GaSe crystals because we obtain close spectral characteristics of SFG emission in both cases: wavelength range from 2.48 to 2.8 μm with maximum at 2.6 μm and full width at a half of the maximum of a spectra envelope is ~ 0.15 μm. Effective nonlinearity of SFG of the CO laser emission in the BGGSe crystal is $d_{eff} = 42 \pm 14$ pm/V, and nonlinear coefficient of this crystal was $d_{11} = 49 \pm 15$ pm/V.

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