

# Broadband carbon monoxide laser system operating in the wavelength range of 2.5–8.3 $\mu\text{m}$

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**Abstract.** A two-cascade frequency conversion of CO-laser radiation is demonstrated in a single sample of a nonlinear  $\text{ZnGeP}_2$  crystal. The crystal is pumped by a repetitively pulsed cryogenic low-pressure CO laser operating on  $\sim 150$  vibration–rotational transitions in the wavelength range 5.0–7.5  $\mu\text{m}$ , which corresponds to the frequency range of a half octave. In the first conversion cascade, generation of second harmonic and sum frequencies of various pairs of CO-laser radiation give  $\sim 350$  emission lines in the wavelength range 2.5–3.7  $\mu\text{m}$ . In the second cascade, by mixing the radiation converted in the first cascade with the residual radiation of the CO laser we have obtained  $\sim 90$  lines in the range 4.3–5.0  $\mu\text{m}$  and more than 80 lines in the range 7.5–8.3  $\mu\text{m}$ . Thus, using a single sample of the nonlinear  $\text{ZnGeP}_2$  crystal pumped by the radiation of a single CO laser we have produced a source of broadband (more than one and a half octaves) laser radiation, simultaneously operating at  $\sim 670$  lines in the wavelength range 2.5–8.3  $\mu\text{m}$ .

**Keywords:** repetitively pulsed cryogenic low-pressure CO laser, second harmonic generation, sum-frequency and difference-frequency generation.

## 1. Introduction

Development of laser radiation sources of mid- and far-IR ranges is of particular interest in spectroscopy, photochemistry, isotope separation, and other applications. One promising approach to get into new spectral ranges is parametric frequency conversion of the radiation of existing lasers by methods of nonlinear optics, in particular, sum-frequency and difference-frequency generation.

A  $\text{ZnGeP}_2$  crystal (ZGP) is a most promising nonlinear optical crystal used for laser frequency conversion in the mid-IR spectral range. It is characterised by high mechanical (hardness is 5.5 by Mohs scale) and thermal (the heat conductivity is  $0.36 \text{ W cm}^{-1} \text{ K}^{-1}$ ) properties, high damage threshold [1], and low optical loss (the absorption coefficient is at most

$0.01 \text{ cm}^{-1}$ ) in the wavelength range 3–8  $\mu\text{m}$  [2]. Various three-wave interactions were realised with ZGP crystals. For example, it was reported [3] about obtaining the internal efficiency of 80% for second harmonic generation (SHG) of a  $\text{CO}_2$  laser. The authors of paper [4] developed a sum-frequency generator (SFG) of CO- and  $\text{CO}_2$ -laser radiations.

A carbon monoxide laser can operate over a wide spectral range at approximately 1000 vibration–rotational lines both on the transition of the fundamental vibration band (at the radiation wavelengths of 4.7–8.2  $\mu\text{m}$  [5]) and of the first vibration overtone band (at the wavelengths of 2.5–4.2  $\mu\text{m}$  [6–8]). However, a number of applications require radiation wavelengths between the lines of a CO laser or beyond the ranges mentioned above.

The spectral range of 4.2–4.7  $\mu\text{m}$  between the generation bands of the CO laser is interesting for realising remote sensing methods, in particular, because there is a main transparency window of the atmosphere. New laser sources aimed at adapting to this range are intensively developed (see, e.g., [9–11]). The problem of creating a new radiation source in the range 4.2–4.7  $\mu\text{m}$  can be solved as well by means of efficient parametric frequency conversion of CO-laser radiation in nonlinear crystals. In particular, one possible solution suggested in [12] is the difference-frequency generator (DFG) of the fundamental and first overtone bands of CO-laser radiation lines by using ZGP crystals. Later [13], it was suggested to substitute the overtone band radiation for a second harmonic of the fundamental band of CO-laser radiation. Note that concurrent SHG and DFG in a single sample of a nonlinear crystal reduces losses and saves the nonlinear optical material. A two-cascade frequency conversion in a single crystal sample was earlier realised in near-IR nonlinear crystals pumped by a neodymium glass laser [14–16]. The wavelength of the radiation generated at a difference frequency was within the limits of near-IR range below 2.5  $\mu\text{m}$ . We also realised [17] the two-cascade frequency conversion of CO-laser radiation in a single sample ZGP crystal, where the DFG covered the wavelength range of 4.3–4.9  $\mu\text{m}$ .

Calculations of the phase matching in a ZGP crystal performed in the present work show that the method of two-cascade frequency conversion in a single crystal provides obtaining radiation beyond a long-wavelength limit of the fundamental band of the CO laser. Mid-IR radiation at wavelengths longer than 8  $\mu\text{m}$  is actual because there is one more transparency window of the atmosphere in the wavelength range of 8–12  $\mu\text{m}$  [18]. Here we investigate the two-cascade frequency conversion of CO-laser radiation in a single nonlinear ZGP crystal in order to expand the radiation spectrum to both short- and long-wavelength parts of mid-IR range.

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Received 25 July 2012; revision received 20 November 2012

Kvantovaya Elektronika 43 (2) 139–143 (2013)

Translated by N.A. Raspopov

## 2. Phase matching conditions

Two-cascade frequency conversion of CO-laser radiation lines can be realised in a single sample of a nonlinear crystal if the internal phase matching (PM) angles coincide for the two parametrical processes of frequency conversion: SHG and SFG on the one hand (the first conversion cascade) and DFG of this radiation and the residual CO-laser radiation (the second cascade) on the other hand. In the present work we only consider PM conditions for collinear interaction in both the processes. In view of the fact that SHG is a degenerated case of SFG, in what follows we will consider both the processes as SFG for simplicity.

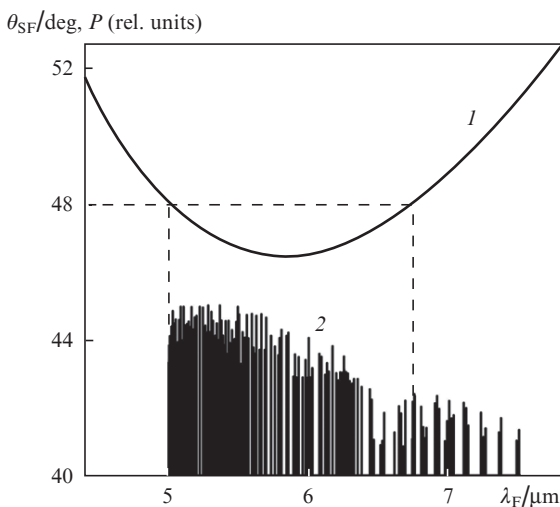
In calculating the PM angles for the ZGP crystal in the case of three-wave interactions  $e + e \rightarrow o$ ,  $o + e \rightarrow o$ , and  $e + o \rightarrow o$  for SFG and  $o - e \rightarrow o$ ,  $o - o \rightarrow e$  and  $o - e \rightarrow e$  for DFG by the formulae [1]:

$$\frac{n_{F1}^{o,e}}{\lambda_{F1}^{o,e}} + \frac{n_{F2}^{o,e}}{\lambda_{F2}^{o,e}} = \frac{n_{SF}^o}{\lambda_{SF}^o}, \quad (1)$$

$$\frac{n_{SF}^o}{\lambda_{SF}^o} - \frac{n_{F1(F2)}^{o,e}}{\lambda_{F1(F2)}^{o,e}} = \frac{n_{DF}^{o,e}}{\lambda_{DF}^{o,e}} \quad (2)$$

we employed the dispersion equations from [19]. In (1) and (2),  $\lambda_{F1}^{o,e}$  and  $\lambda_{F2}^{o,e}$  are the wavelengths of two interacting lines (F1 and F2) of the fundamental band of CO-laser radiation with ordinary (o) and extraordinary (e) polarisations;  $\lambda_{SF}^o$  and  $\lambda_{DF}^{o,e}$  are the radiation wavelengths of the sum frequency (SF) and difference frequency (DF);  $n_{F1,F2}^{o,e}$ ,  $n_{SF}^o$  and  $n_{DF}^{o,e}$  are the refractive indices of the ZGP crystal at the corresponding wavelengths.

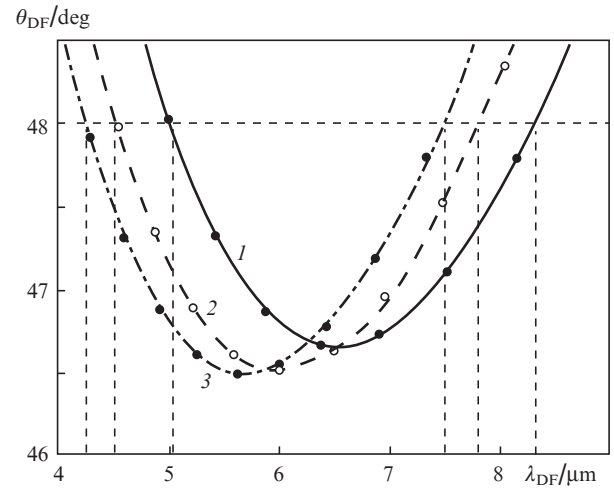
A dispersion dependence of the internal PM angle  $\theta_{SF}$  corresponding to the  $e + e \rightarrow o$  type SHG of CO-laser radiation in a ZGP crystal is shown in Fig. 1. Vertical dashed lines denote the wavelengths of 5.0 and 6.7  $\mu\text{m}$ , which correspond to  $\theta_{SF} = 48^\circ$ . In the same figure, a measured spectrum of cryogenic CO-laser radiation is shown, where these lines are pres-



**Figure 1.** Internal angle of PM for the  $e + e \rightarrow o$  type SHG in a ZGP crystal (1) and spectrum of CO-laser radiation (2).

ent. Since the CO-laser spectrum ( $\lambda_F = 5.0 - 7.5 \mu\text{m}$ ) resides in the range of the minimal dispersion dependence of the PM angle (the minimum corresponds to  $\lambda_F \approx 5.8 \mu\text{m}$ ), taking into account a large spectral width of PM [20] one may assume that actually all the spectrum of laser radiation may be involved in the SFG process.

As an example, Fig. 2 shows the dispersion dependence of internal PM angle  $\theta_{DF}$  calculated for DFG of the sum frequency o-wave and CO-laser residual e-wave emission of the  $o - e \rightarrow e$  type. The three PM curves for DFG shown in Fig. 2 correspond to the CO-laser radiation wavelengths  $\lambda_F = 5.0$ , 5.6, and 6.0  $\mu\text{m}$ . The sum-frequency radiation wavelength  $\lambda_{SF}$  increases along the PM curves from left to right; its increase from 2.5 to 3.4  $\mu\text{m}$  is marked (for spectral reference) in the curves by dots in 0.1  $\mu\text{m}$  increments. The vertical dashed lines denote the radiation wavelengths  $\lambda_{DF}$ , which correspond to the same angle  $\theta_{DF} = 48^\circ$  as for SFG.



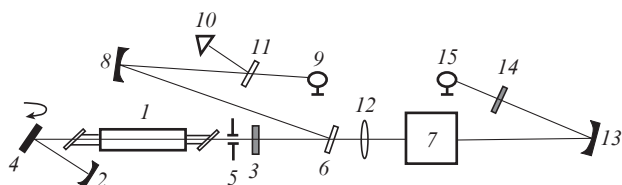
**Figure 2.** Internal PM angle for DFG of the  $o - e \rightarrow e$  type in a ZGP crystal calculated for the CO-laser radiation wavelengths  $\lambda_F = 5.0$  (1), 5.6 (2), 6.0  $\mu\text{m}$  (3)

From Figs 1 and 2 it follows that if there is radiation at the wavelengths  $\lambda_F = 5.0$  and 5.6  $\mu\text{m}$  in the spectrum of CO-laser and the angle inside ZGP crystal between the laser beam propagation direction and crystal optical axis is  $48^\circ$  then SHG will occur of the  $e + e \rightarrow o$  type at the wavelength  $\lambda_{SF} = 2.5 \mu\text{m}$  (Fig. 1). In this case, the interaction of the  $o - e \rightarrow e$  type between the radiation at  $\lambda_{SF} = 2.5 \mu\text{m}$  and the laser radiation at  $\lambda_F = 5.6 \mu\text{m}$  results in the DF radiation arising at the wavelength  $\lambda_{DF} = 4.5 \mu\text{m}$  (Fig. 2).

Similarly, if there is radiation at the wavelengths  $\lambda_F = 6.0$  and 6.7  $\mu\text{m}$  in the spectrum of the CO laser and if the angle inside the ZGP crystal between the laser beam propagation direction and crystal optical axis is  $48^\circ$  then SHG will occur at the wavelength  $\lambda_{SF} = 3.35 \mu\text{m}$  according to the  $e - e \rightarrow o$  type (Fig. 1). Here, the interaction of the  $o - e \rightarrow e$  type between the radiation at  $\lambda_{SF} = 3.35 \mu\text{m}$  laser radiation at  $\lambda_F = 6.0 \mu\text{m}$  leads to the DF radiation at the wavelength  $\lambda_{DF} \approx 7.6 \mu\text{m}$  (Fig. 2). Due to a large width of PM in ZGP the frequency conversion of a broadband CO-laser radiation may cover a wide spectral range, in particular, the intervals designated in Fig. 2 by vertical dashed lines.

### 3. Experimental setup

An optical schematic diagram of an experimental frequency conversion of CO-laser radiation in the ZGP crystal is shown in Fig. 3. A repetitively pulsed low-pressure cryogenic CO laser (1) with a DC discharge pumping was used. The repetitively pulsed operation regime was realised by modulating the  $Q$ -switched laser cavity. The cavity was formed by a highly reflecting spherical mirror (2) (with the curvature radius of 9 m) and a plane outcoupling mirror (3). In contrast to [17], in the present work in the CO laser we employed an outcoupling mirror with the higher reflection coefficient of above 80% in a wider spectral range (4.5–7.5  $\mu\text{m}$ ). In addition, we used the working gas mixture with much higher contents of carbon monoxide molecules (CO:N<sub>2</sub>:He = 1:1:12 at the total pressure of 8.2 Torr with a small amount of air). As later measurements showed, this resulted in a wider spectrum of CO-laser radiation in the wavelength range of 5.0–7.5  $\mu\text{m}$ . The spectrum obtained comprised  $\sim 150$  lines, i.e., approximately twice more than reported in [17]. The  $Q$ -factor was modulated by a rotating mirror (4) placed inside the laser cavity with the rotation frequency varying from 30 to 130 Hz. The diaphragm (5) with a diameter of 8 mm provided laser operation on the fundamental transversal mode. The laser radiation was a train of pulses with the duration varying from 0.3 to 1  $\mu\text{s}$  depending on the mirror rotation frequency. The laser radiation peak power reached 4 kW. The laser radiation was directed to an uncoated ZGP crystal (7) of length 17 mm. The pump radiation power was controlled by means of a plane-parallel plate (6) made of CaF<sub>2</sub>, which directed a part of the radiation ( $\sim 5\%$ ) to a spherical mirror (8) and then to a power meter (9) (Ophir 3A-SH) and photodetector (10) (PEM-L-3) for recording the time shape of the pulses using one more plane-parallel plate (11) made of CaF<sub>2</sub>.



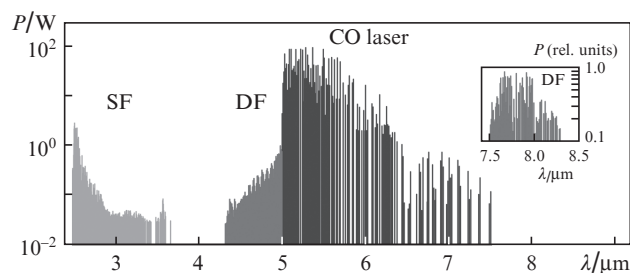
**Figure 3.** Optical schematic diagram of experiment: (1) CO laser; (2) spherical mirror with the radius of curvature 9 m; (3) partially reflecting mirror; (4) rotating mirror; (5) diaphragm; (6, 11) plates made of CaF<sub>2</sub>; (7) ZGP crystal; (8, 13) spherical mirrors; (9, 15) power meters; (10) photodetector; (12) lens made of CaF<sub>2</sub>; (14) plate made of IR quartz.

The radiation of the laser was focused onto a ZGP crystal (7) by a lens (12) made of CaF<sub>2</sub> with a focal length of 115 mm. A spherical mirror (13) collects radiation from the crystal which then passes through a spectral filter (14) to a power meter (15) (Ophir 3A-SH). The spectral filter (14) made of quartz separates the converted radiation (with the wavelength of 2.5–3.7  $\mu\text{m}$ ) from the pump radiation (5.0–7.5  $\mu\text{m}$ ). For measuring spectral characteristics the converted radiation passed (without spectral filtering) to an IKS-31 spectrometer (not shown in Fig. 3) with the spectral resolution of 0.4 nm.

### 4. Spectral-energy characteristics of SFG and DFG

A maximal average radiation power of SFG was obtained at the crystal position corresponding to the internal PM angle of 47°. The external conversion efficiency was calculated by dividing the average SF radiation power at an exit from the crystal by the average pump radiation power passing to the crystal. A maximal external efficiency of SFG conversion was 3.4%, which, taking into account the losses to Fresnel reflection from uncoated crystal edges, corresponds to the internal efficiency of 6.5.

The crystal was installed in such a way that the internal PM angle was 48°, which corresponds to the SHG PM angle for the wavelength of 2.5  $\mu\text{m}$ . Radiation spectra of the CO-laser and converted radiation are presented in Fig. 4. The pump radiation spectrum measured without the crystal was in the wavelength range 5.0–7.5  $\mu\text{m}$  and comprised approximately 150 spectral lines. The total peak power was 4 kW. The spectrum of SF radiation in the ZGP crystal was in the range 2.5–3.7  $\mu\text{m}$  and comprised approximately 350 emission lines. The external efficiency of conversion was  $\sim 1\%$  and the total peak power was 40 W.



**Figure 4.** Measured spectra of pump CO-laser radiation, sum, and difference frequencies.

The spectrum of DF radiation generated in the ZGP crystal included almost 90 lines with the wavelengths from 4.3 to 5.0  $\mu\text{m}$ . The radiation power of these lines was measured in the same way and conditions as the radiation power of CO-laser pumping. The external efficiency of DFG was 0.23%, which corresponds to the internal efficiency of 0.45%. Lack of DF radiation wavelengths shorter than 4.3  $\mu\text{m}$  may be explained by a lower radiation power for lines with the wavelength longer than 6.0  $\mu\text{m}$  and by atmospheric carbon dioxide absorption in the wavelength range 4.2–4.3  $\mu\text{m}$ . Energy and spectral characteristics of DFG obtained in the range 4.3–5.0  $\mu\text{m}$  are close to the results of [17].

Unlike [17], due to a wider radiation spectrum of the CO laser and enhanced sensitivity of measurements though with a lower resolution power of a spectrometer we succeeded to detect  $\sim 80$  lines in the wavelength range 7.5–8.3  $\mu\text{m}$  (see inset in Fig. 4). Note that the wavelength of 8.3  $\mu\text{m}$  is a limiting operation band wavelength of the diffraction grating of the spectrometer.

### 5. Comparison of measured and calculated radiation spectra

In calculating the SF and DF radiation spectra we used the identified experimental lines of the CO laser with measured

powers (Figs 4 and 5). The internal angle between the laser pump beam (with a radius of 0.5 mm) and crystal optical axis was taken 48°.

The expression for the SF radiation power can be presented in the form [21]

$$P_{\text{SF}} = \frac{8\pi^2 d_{\text{eff}}^2 L^2 P_{\text{F1}} P_{\text{F2}}}{\varepsilon_0 c n_{\text{F1}} n_{\text{F2}} n_{\text{SF}} \lambda_{\text{SF}}^2 A} \text{sinc}^2\left(\frac{|\Delta k| L}{2}\right), \quad (3)$$

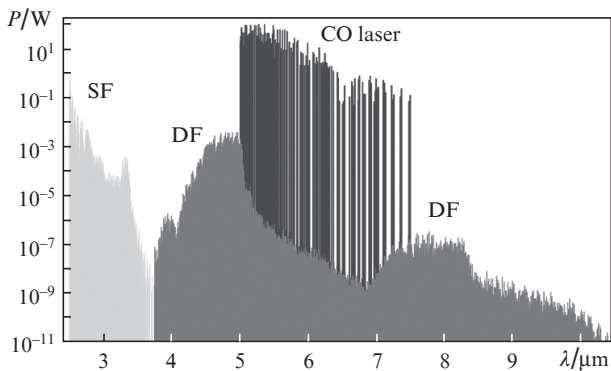
where  $d_{\text{eff}}$  is the effective nonlinear coefficient (in the ZGP crystal  $d_{36} = 75 \text{ pm V}^{-1}$ );  $L = 17 \text{ mm}$  is the crystal length;  $P_{\text{F1}}$  and  $P_{\text{F2}}$  are the pump laser radiation power for two lines;  $\varepsilon_0$  is the dielectric constant;  $c$  is the speed of light in vacuum;  $A$  is the cross-section of the laser beam; and  $\Delta k$  is the wave mismatch.

Calculation results for SF radiation spectrum are presented in Fig. 5. Both the calculated and measured spectra have a local maximum of the SF radiation power near the wavelength of 3.5  $\mu\text{m}$ , which is explained by an exact phase matching for SFG in this spectral range. A small distinction ( $\sim 0.2 \mu\text{m}$ ) in the calculated and experimental positions of the maximum may be related to the error of the crystal angle adjustment, which is  $\pm 0.3^\circ$ . An analysis of the calculated spectrum revealed that a higher pump radiation power and sensitivity of measurements would result in a greater number of detected SF radiation lines in the range 2.5–3.7  $\mu\text{m}$ .

In order to calculate the spectrum of DF radiation (Fig. 5) in the conditions of the realised two-cascade conversion of CO-laser radiation in a single ZGP crystal we used the calculated spectrum of SF emission. The expression for the DF radiation power can be presented in the form [21]

$$P_{\text{DF}} = \frac{8\pi^2 d_{\text{eff}}^2 L^2 P_{\text{SF}} P_{\text{F}}}{\varepsilon_0 c n_{\text{F}} n_{\text{SF}} n_{\text{DF}} \lambda_{\text{DF}}^2 A} \text{sinc}^2\left(\frac{|\Delta k| L}{2}\right). \quad (4)$$

The processes of SFG and DFG occur concurrently and it is necessary to solve the corresponding system of combined differential equations. However, the efficiency of SFG was small and equal to  $\sim 1\%$  and in a model analysis we employed the preset field approximation so that the power of SF radiation lines  $P_{\text{SF}}$  and the pump wave power  $P_{\text{F}}$  were taken constant and equal to the corresponding power at the crystal exit.



**Figure 5.** Calculated spectra of sum and difference frequency radiation for the measured spectrum of pump CO-laser radiation.

Since the absorption of radiation by carbon dioxide was not taken into account and the general structure of the calculated DF spectrum in the range 4.3–5.0  $\mu\text{m}$  is similar to the spectrum structure of measured frequency-converted radiation (compare Figs 4 and 5) it is likely that the lower DF radiation power at shorter wavelengths is mainly related to a lower pump radiation power.

Lack of DF emission lines at wavelengths longer than 8.3  $\mu\text{m}$  in the spectrum measured (Fig. 4) is related to a spectral operation limit of the diffraction grating employed in the spectrometer (4.0–8.3  $\mu\text{m}$ ). The calculations confirm that based on a single sample of the nonlinear ZGP crystal pumped by a single CO laser one can design a broadband (more than two octaves) source of laser radiation operating, at least, in a wavelength range 2.5–10.3  $\mu\text{m}$ .

## 6. Conclusions

In the present work, a concurrent two-cascade frequency conversion is realised for CO-laser radiation wavelengths of 5.0–7.5  $\mu\text{m}$  in a single sample of the nonlinear ZGP crystal. Emission in the three wavelength ranges was obtained: 2.5–3.7  $\mu\text{m}$ , 4.3–5.0  $\mu\text{m}$ , and 7.5–8.3  $\mu\text{m}$ . The pump radiation comprised  $\sim 150$  spectral lines in the wavelength range 5.0–7.5  $\mu\text{m}$  at the peak power of up to 4 kW. In performing SHG and SFG with the CO laser (the first frequency conversion cascade), the wavelength range of 2.5–3.7  $\mu\text{m}$  was covered, within which approximately 350 emission lines were detected. In the same ZGP crystal sample at the same internal angle of pumping (48°), DFG was observed with the radiation obtained in the first cascade and the pump CO-laser radiation (the second cascade of frequency conversion). In the wavelength range 4.3–5.0  $\mu\text{m}$ , approximately 90 emission lines were detected and more than 80 lines were detected in the range 7.5–8.3  $\mu\text{m}$ .

Thus, on a basis of a single nonlinear crystal sample pumped by a single gas CO laser, the broadband (more than one and a half octaves) carbon monoxide laser system was developed operating at approximately 670 lines in the wavelength range 2.5–8.3  $\mu\text{m}$ . Calculations show that such a laser source can operate in a wider wavelength range of at least 2.5–10.3  $\mu\text{m}$ .

**Acknowledgements.** The authors are grateful to A.V. Soluyanov, a student of the National Research Nuclear University MEPhI, for help in carrying out the experiments.

The work was partially supported by the Russian Foundation for Basic Research (Grant Nos 12-02-01085-a, 12-02-31121-mol-a), the Presidential Grant No. NS-512.2012.2, Federal Target Program ‘Scientists and Science Educators of Innovative Russia’ (Agreement No. 8883) and Educational Scientific Complex of P.N. Lebedev Physics Institute.

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