Quantum Electronics **39** (3) 229 – 234 (2009)

LASERS

PACS numbers: 42.55.Lt; 42.60.By; 42.60.Jf
DOI: 10.1070/QE2009v039n03ABEH013811

A cryogenic slab CO laser

A.A. Ionin, A.Yu. Kozlov, L.V. Seleznev, D.V. Sinitsyn

Abstract. **A compact capacitive transverse RF-discharge-pumped slab CO laser with cryogenically cooled electrodes, which operates both in the cw and repetitively pulsed regimes, is fabricated. The laser operation is studied in the free running multifrequency regime at the vibrational – rotational transitions of the fundamental ($V + 1 \rightarrow V$) vibrational bands of the CO molecule in the spectral region from 5.1 to 5.4 μm. Optimal operation conditions (gas mixture composition and pressure, RF pump parameters) are determined. It is shown that only gas mixtures with a high content of oxygen (up to 20 % with respect to the concentration of CO molecules) can be used as an active medium of this laser. It is demonstrated that repetitively pulsed pumping is more efficient compared to cw pumping. In this case, quasi-cw lasing regime can be obtained. The maximum average output power of $\sim$12 W was obtained for this laser operating on fundamental bands and its efficiency achieved $\sim$14 %. The frequency-selective operation regime of the slab RF-discharge-pumped CO laser was realised at $\sim$ 100 laser lines in the spectral region from 5.0 to 6.5 μm with the average output power of up to several tens of milliwatts in each line. Lasing at the transitions of the first vibrational overtone ($V + 2 \rightarrow V$) of the CO molecule is obtained in the spectral region from 2.5 to 3.9 μm. The average output power of the overtone laser achieved 0.3 W. All the results were obtained without the forced gas mixture exchange in the discharge chamber. Under fixed experimental conditions, repetitively pulsed lasing (with fluctuations of the output characteristics no more than $\pm$10 %) was stable for more than an hour.**

Keywords: *CO laser, RF discharge, slab (planar) geometry, vibrational – rotational transitions, fundamental band, overtone band, repetitively pulsed regime.*

1. Introduction

A CO laser operating at the vibrational – rotational transitions of the fundamental ($V + 1 \rightarrow V$) vibrational bands of the CO molecule ($\lambda \sim 4.9 - 7.5$ μm) is a source of mid-IR laser radiation, which is convenient for solving various fundamental and applied problems. Under special conditions, this laser can also operate at the overtone ($V + 2 \rightarrow V$) transitions of the CO molecule ($\lambda \sim 2.5 - 4.2$ μm) (see, for example, reviews [1, 2]). The emission spectrum of the CO laser (at fundamental and overtone transitions) can consist of more than 1000 spectral lines [3, 4] covering the spectral regions containing the absorption lines of not only usual substances (H_2O, CO_2, CH_4, NO_2, NO, acetone, benzene, methanol, etc.) but also atmosphere pollutants, toxins, explosives and drugs, whose reliable detection is an urgent problem at present. In addition, overtone CO-laser radiation overlaps a rather broad transparency window of atmosphere (3.3 – 4.0 μm) [5]. Estimates presented in [6] show that a CO laser is one of the best candidates for solving problems of qualitative and quantitative laser spectroscopic analysis of multicomponent gas mixtures containing small impurities of the above substances.

Significant advance has been achieved recently in the development of capillary and slab diffusively cooled gas lasers (including gas-flow and sealed-off CO_2 and CO lasers) excited by a capacitive transverse radio-frequency (RF) discharge (see, for example, [7 – 16]). In these lasers, heat is removed through cooled electrodes, to which exciting RF voltage is applied. A RF discharge has a number of advantages compared to the most simple and widely used dc discharge. They, first of all, include lower voltage of the power supply, higher energy efficiency and simplicity of the laser output power modulation and control. In addition, a RF discharge allows excitation of rather large volumes of the active medium (in the planar geometry) at pressures of several tens of torr without using external ionisation sources. Compared to the dc discharge, a RF discharge is more stable.

It is known that a CO laser has the best output characteristics at cryogenic temperatures of the active gas mixture. The low temperature of the active medium is especially important for the efficient operation of an overtone CO laser. However, till now RF-pumped CO lasers have been studied mainly at room temperature or at slightly lower (down to $-20 \div -30\,^{\circ}$C) temperature [7 – 9, 11, 17] of the active medium in the lasing regime at fundamental band transitions. The exception is some papers where overtone lasing in the RF-pumped CO lasers was achieved by using cryogenic diffusive cooling of the active medium of a slab CO laser at fundamental transitions (see, for example, [13]) or cooling in a supersonic gas flow (see, for example, [18, 19]). Therefore, the problem of fabrication of a compact

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A.A. Ionin, A.Yu. Kozlov, L.V. Seleznev, D.V. Sinitsyn P.N. Lebedev Physics Institute, Russian Academy of Sciences, Leninsky prosp. 53, 119991 Moscow, Russia; e-mail: aion@sci.lebedev.ru

Received 5 February 2008; revision received 9 February 2009
Kvantovaya Elektronika **39** (3) 229 – 234 (2009)
Translated by I.A. Ulitkin

RF-pumped slab CO laser with diffusive cooling of its active medium down to low temperatures is of practical importance. Such a laser combining a relative simplicity and compactness inherent in RF-excited slab lasers as well as a high efficiency and a broad emission spectrum typical of electric-discharge CO lasers can become a unique spectroscopic instrument for detecting different explosives, toxic and other dangerous substances and materials. This paper is devoted to the development of a RF-excited slab CO laser with cryogenic diffusive cooling of the active medium operating at the fundamental ($V+1 \rightarrow V$) transitions and studies its lasing parameters for obtaining lasing at overtone ($V+2 \rightarrow V$) transitions.

2. Experimental setup

A specially developed and fabricated setup described in detail in [20] was used to study experimentally the parameters of a cryogenic RF-excited CO laser. The discharge chamber was made of stainless steel and had an internal volume of ~ 8 L. The electrode system of the setup consisted of two hollow brass electrodes through which liquid nitrogen was continuously pumped, which enabled electrodes to be cooled down to ~ 120 K. In the experiments, we used two electrode systems (A and B) of different configurations. In both systems the length of electrodes along the laser resonator axis was 250 mm and the height of the vertically located discharge gap was 16 mm (system A) and 30 mm (system B). The interelectrode distance in both systems was 3 mm.

The discharge was excited using a radio-frequency RFPS-500AM generator with the maximum output power $P_{\mathrm{RF}}^{\max}$ of 620 W in the cw regime, which was connected to the discharge chamber via a controlled T-like matching LC system. The working frequency F of the RF generator was 81.36 MHz. The generator could also operate in the repetitively pulsed regime with a low-frequency amplitude modulation of the output RF power (the modulation frequency is $F_{\mathrm{mod}} = 0.1 - 25$ kHz). The average RF power $\langle P_{\mathrm{RF}} \rangle$ applied to the discharge gap could be changed by varying both the duration t of modulating pulses in the range $t = (0.01 - 1)T_{\mathrm{mod}}$ ($T_{\mathrm{mod}} = 1/F_{\mathrm{mod}}$ is the modulation period) and instantaneous RF power $P_{\mathrm{RF}} = (0.25\text{–}1.0)P_{\mathrm{RF}}^{\max}$ in each pulse. Information on the pump parameters and the ways of their variation will be presented below for each series of experiments.

Note that the operation of electric-discharge CO lasers at cryogenic temperatures of the oxygen-containing plasma of the active medium (oxygen is usually added in the active gas mixture to compensate for the CO dissociation in the discharge), as a rule, is accompanied by the production of ozone in it, which condenses and percipitates as a liquid on cryogenic elements of the laser. Liquid ozone represents a high explosive, which can sometimes lead to destruction of experimental cryogenic electric-discharge setups on the basis of gas mixtures containing carbon oxide and/or oxygen [21–23]. To prevent such a situation, the liquid ozone should be continuously removed from the electric-discharge chamber. For this reason, cryogenic CO lasers, as a rule, should operate at a continuous gas flow, which needs to be maintained for a long time even after the discharge is switched off. The design of the cryogenic RF-pumped slab CO laser used in this paper allows one, due to the vertical location of the discharge gap, to remove liquid ozone from the RF discharge region by the gravity action and provides its heating and gasification in the buffer volume of the chamber having the room temperature.

3. Experimental results

3.1 Fundamental vibrational transitions of CO molecule

We performed a series of experiments to determine optimal conditions for the operation of a cryogenic RF-pumped slab CO laser at which the maximum output power and/or efficiency are achieved. In these experiments, we varied the parameters of the laser active medium (the gas mixture composition, its pressure) or the pump conditions (power and the excitation regime, modulation frequency). The 270-mm-long laser resonator was formed by a highly reflecting (aluminium-coated quartz) spherical ($R_{\mathrm{crv}} = 1500$ mm) mirror and a plane output mirror (a $\mathrm{CaF_2}$ plate with a dielectric coating) having the reflection coefficient $85\,\% \pm 5\,\%$ in the wavelength range from 4.9 to 7.1 μm.

The output parameters of electric-discharge CO lasers depend on the composition of the gas mixture and its pressure. Nitrogen and oxygen in different proportions are usually present in gas mixtures. In our experiments, we used air (the ratio of the concentrations of nitrogen and oxygen was 4:1) as an addition to the CO–He gas mixture. In sealed-off CO lasers operating at room [17] or slightly lower [8, 9] temperatures, gas mixtures with a low content of oxygen are usually used, i.e. $\mathrm{CO}:\mathrm{O_2}:\mathrm{M} = 1:(0.03-0.05):\mathrm{Y}$, where M are other components of the mixture and Y is their total content. However, the use of such mixtures in our experiments lead to an intense production of a carbon film on working surfaces of cooling electrodes, which substantially changed the RF discharge structure (the uniform distribution of the discharge current across the area of the working surfaces of electrodes was violated). As a result, the output power of the slab CO laser significantly decreased (by four–five times) after a short-term (within several minutes) operation. For this reason, only mixtures with a high content of oxygen (up to 20 % with respect to the concentration of CO molecules) proved suitable as an active medium of a RF-pumped slab CO laser with cryogenically cooled electrodes. We also optimised laser parameters concerning the content of helium in the three-component gas mixture, $\mathrm{CO}:\mathrm{air}:\mathrm{He} = 1:1:\mathrm{X}$. Experiments showed that it is more preferable to use gas mixtures with a relatively high content (70 %–90 %) of He as an active medium in a RF-excited slab CO laser.

The optimal pressure of the gas mixture was found experimentally (Fig. 1). The maximum efficiency of the RF-excited slab CO laser achieved in the experiments with the electrode system A was obtained at the gas mixture pressure of 37.5 Torr and was equal to $\sim 11\,\%$ (the mixture $\mathrm{CO}:\mathrm{air}:\mathrm{He} = 1:1:10$). In these experiments the pulse value of the supplied RF power was constant and the average supplied power was varied by varying the duration t of modulation pulses.

For the further parametric study of the RF-excited CO laser, it was necessary to determine the optimal RF modulation frequency of the pump power. In this series of experiments (Fig. 2) the gas mixture composition and its pressure were fixed ($\mathrm{CO}:\mathrm{air}:\mathrm{He} = 1:1:10$; 37 Torr). With the constant average power of RF excitation $\langle P_{\mathrm{RF}} \rangle = 62$ W (the duration of modulating pulses was 10 % of the

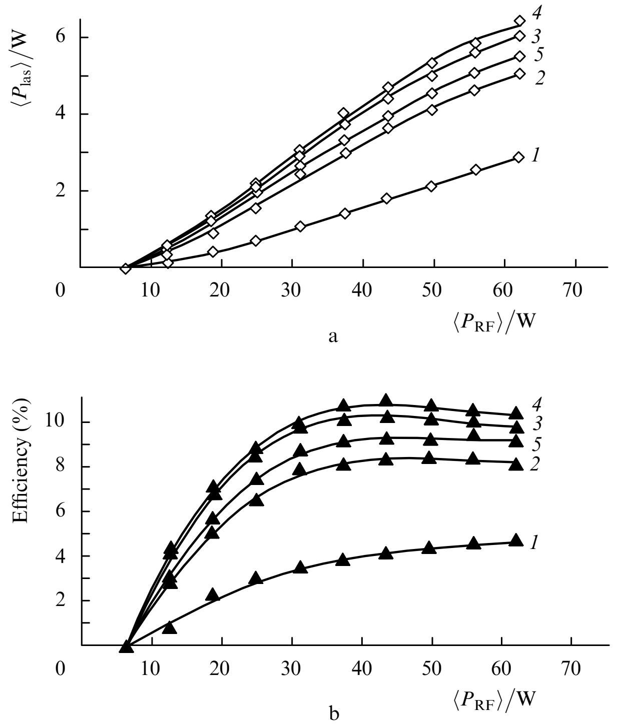

Figure 1. Dependences of the average output power of the RF-pumped slab CO laser (a) and its efficiency (b) on the average RF-excitation power (electrode system A) for the mixture CO : air : He = 1 : 1 : 10 at $F_{\rm mod} = 500$ Hz, $P_{\rm RF} = 620$ W and the pressure of the active medium 11 (*1*), 18.5 (*2*), 30 (*3*), 37.5 (*4*) and 45 Torr (*5*).

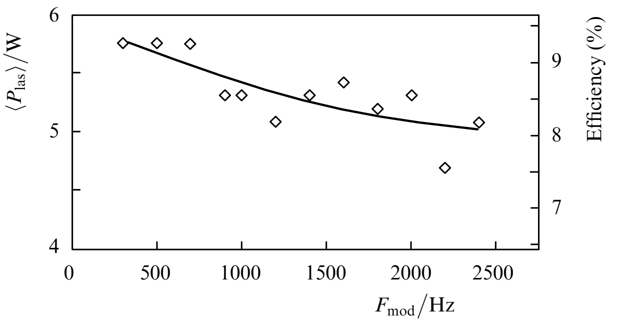

Figure 2. Dependences of the average output power $\langle P_{\rm las}\rangle$ and efficiency of the slab CO laser on the RF modulation frequency of the excitation power (electrode system A) for the gas mixture CO : air : He = 1 : 1 : 10 at the pressure 37 Torr and $\langle P_{\rm RF}\rangle = 62$ W.

modulation period) and with increasing $F_{\rm mod}$, we observed a slight decrease in the average laser power $\langle P_{\rm las}\rangle$ and the efficiency of the CO laser. In further experiments, the modulation frequency was 100 – 1000 Hz, which corresponded to the highest values of the laser power and efficiency of the CO laser (Fig. 2).

The average RF-excitation power in the next series of experiments was increased by increasing the duration t of the RF-excitation pulse (at constant $F_{\rm mod}$) with a transition, as a result, to the cw pump regime. We determined the upper limit of the average RF-excitation power, whose excess resulted in a significant decrease in the output power of the CO laser and its efficiency (Fig. 3) due to the gas mixture heating in the discharge region. In these experiments, RF excitation was performed at $F_{\rm mod} = 1000$ Hz and the peak RF pulse power $P_{\rm RF} = 400$ W. The average RF excitation power $\langle P_{\rm RF}\rangle$ was changed from 40 to 280 W. The extreme right point in Fig. 3 corresponds to the cw operation regime of the RF generator.

With increasing the average RF-excitation power from ~40 to ~150 W, the average output power of the CO laser increased and achieved its maximum value (~12 W). In this range of $\langle P_{\rm RF}\rangle$, the efficiency of the RF-excited slab CO laser remained approximately at the same level and was 8 % – 9 % (Fig. 3). A further increase in $\langle P_{\rm RF}\rangle$ lead to a drastic decrease both in the output power of the CO laser and its efficiency. Under these conditions, liquid nitrogen probably boiled up inside the electrodes and a vapour layer with a low thermal conductivity, which prevented the heat removal from the RF discharge region, appeared between the internal surface of the electrode and liquid nitrogen. To make the heat removal more efficient, it was necessary to increase the flow rate of liquid nitrogen through electrodes, which was taken into account while designing the electrode system B.

A series of experiments similar to those described above were performed with the electrode system B in which the height of the discharge gap was enlarged to 30 mm and the flow rate of liquid nitrogen (under pressure) inside the electrodes was increased. Figure 4 presents the energy parameters of the slab CO laser equipped with different

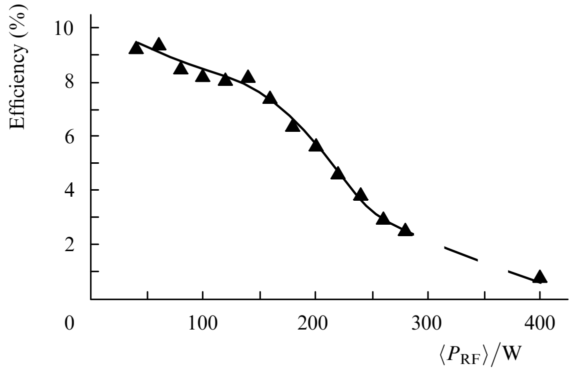

Figure 3. Dependence of the efficiency of the slab CO laser on average RF-excitation power (electrode system A) for the gas mixture CO : air : He = 1 : 1 : 5 at the pressure 26 Torr, $F_{\rm mod} = 1000$ Hz and $P_{\rm RF} = 400$ W.

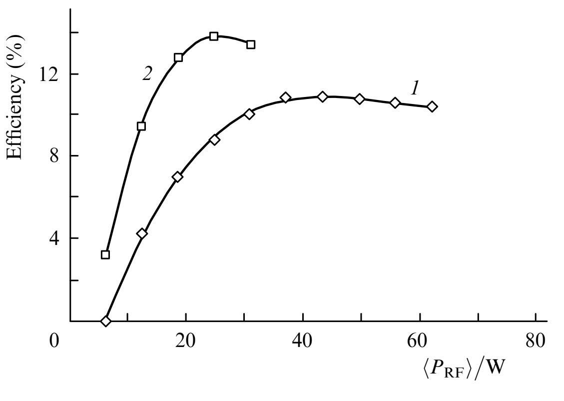

Figure 4. Dependences of the efficiency of the slab CO laser on average RF-excitation power for the electrode systems A [the height of electrodes is 16 mm (*1*)] and B [30 mm (*2*)]. The gas mixture is CO : air : He = 1 : 1 : 10.

electrode systems under optimal pump conditions for each system. The maximum efficiency of 14 % was obtained for the electrode system B. For this reason, all further experiments were performed using this electrode system.

The emission spectrum of the RF-excited slab CO laser lied in the wavelength range from 5.1 to 5.4 μm and strongly depended on the experimental conditions. As an example, Fig. 5 presents one of the obtained emission spectra. Note that the spectrum in these experiments was detected without the proper sensitivity. In this connection, the number of laser lines at which lasing occurred could be much greater than those presented in Fig. 5.

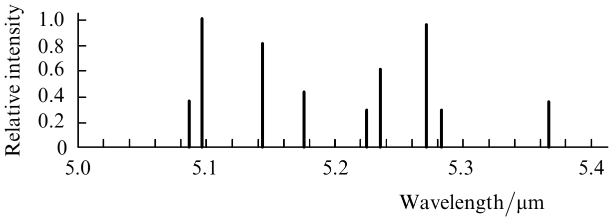

Figure 5. Typical emission spectrum of the RF-excited slab CO laser. The gas mixture is CO : air : He = 1 : 1 : 10, the pressure is 30 Torr and $F_{\mathrm{mod}} = 500$ Hz.

In the experiments we also realised the frequency-selective operation regime of the cryogenic RF-excited slab CO laser. The laser resonator consisted of a highly reflecting (aluminium-coated quartz substrate) concave mirror with a radius of curvature 1 m and a 210-grooves mm^{-1} diffraction grating with the laser output through the zero diffraction order. The experimental conditions (laser mixture CO : air : He = 1 : 1 : 10, pressure of 22 Torr, pump regime – 0.1-ms, 500-Hz RF pulses with the peak RF power of 620 W) being fixed, single-frequency lasing was obtained at ~100 laser lines in the spectral region from 4.98 to 6.26 μm. The average output power was from several milliwatts to several tens of milliwatts at each line. A small increase in the average RF-excitation power by increasing the pump pulse duration to 0.14 ms lead to the expansion of the laser emission spectrum to the red till ~6.5 μm.

Figure 6 presents typical intensity oscillograms of output radiation of a repetitively pulsed RF-excited slab CO laser operating in the free-running multifrequency regime. The active medium was excited by pulses of different durations t at a repetition rate of 100 Hz. Experiments showed that under our conditions, already at the pump pulse duration $t \sim 400$ μs (Fig. 6b), the radiation intensity starts decreasing even before the end of the pump pulse due to the gas mixture overheating. Note in whole that depending on the RF-excitation regime (modulation frequency, off-duty ratio and pulse durations), the CO laser can operate both in the repetitively-pulsed regime (short pulses with a large off-duty ratio) and in the quasi-cw regime, when each next RF-pump pulse overlaps the previous laser pulse that has not terminated yet.

We performed a series of control experiments on measuring the average output power of the slab CO laser during a long time interval at invariable pump conditions (the laser pulse repetition rate was 100 Hz, the average output power of the laser was ~1.5 W). When the laser operated for more than an hour without the renewal of the working mixture, fluctuations of the average output power did not exceed ~10 %. Note that the total number of RF-pumping cycles (and, correspondingly, laser pulses), at which no significant degradation of the laser gas mixture as well as no decrease in the output laser parameters were observed, achieves $(3 - 5) \times 10^5$. In this case, depending on the specific task which can be solved using the CO laser, the laser pulse repetition rate can be varied in a broad range – from several hertz to several kilohertz and the time of stable continuous operation (at minimum frequencies) can be manifold increased.

3.2 Overtone vibrational transitions of CO molecule

Based on the obtained results and on the operational experience with other types of electric-discharge CO lasers [3, 4, 24], we performed experiments to obtain lasing at overtone ($\mathrm{V} + 2 \rightarrow \mathrm{V}$) transitions of the CO molecule in the RF-excited slab laser.

Taking into account a relatively small length of the active medium in the slab CO laser (25 cm) and a significantly lower expected gain at overtone transitions compared to the fundamental transitions [2], we used in the experiments a stable laser resonator with interference highly reflecting mirrors. Note that due to the high reflection coefficient (more than 99 %) in the region from 2.5 to 4.0 μm and the small coefficient in the spectral region $\lambda > 5$ μm, lasing at fundamental transition was suppressed, which is important for ensuring efficient 'overtone' lasing [2, 3].

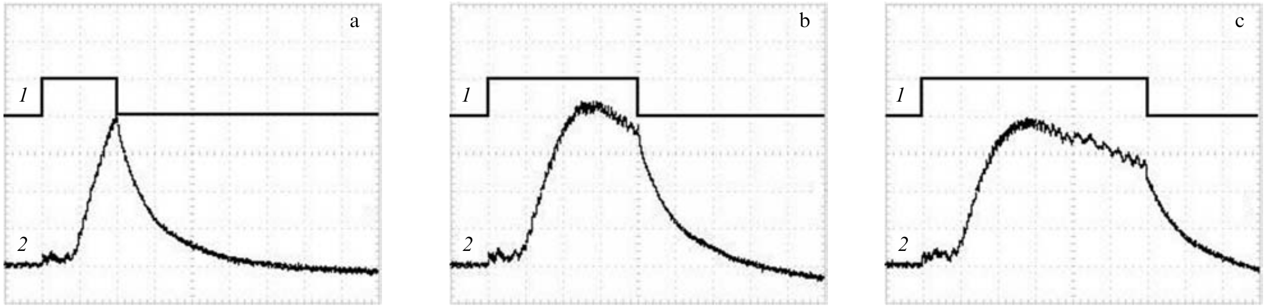

Figure 6. Oscillograms of the RF pump pulse envelope (*1*) and laser pulses (*2*) in the case of the fixed modulation frequency ($F_{\mathrm{mod}} = 100$ Hz) for the mixture CO : air : He = 1 : 1 : 10 at the pressure of 26 Torr and the pulse duration of RF pumping $t = 200$ (a), 400 (b) and 600 μs (c); the sweep is 100 μs div^{-1}.

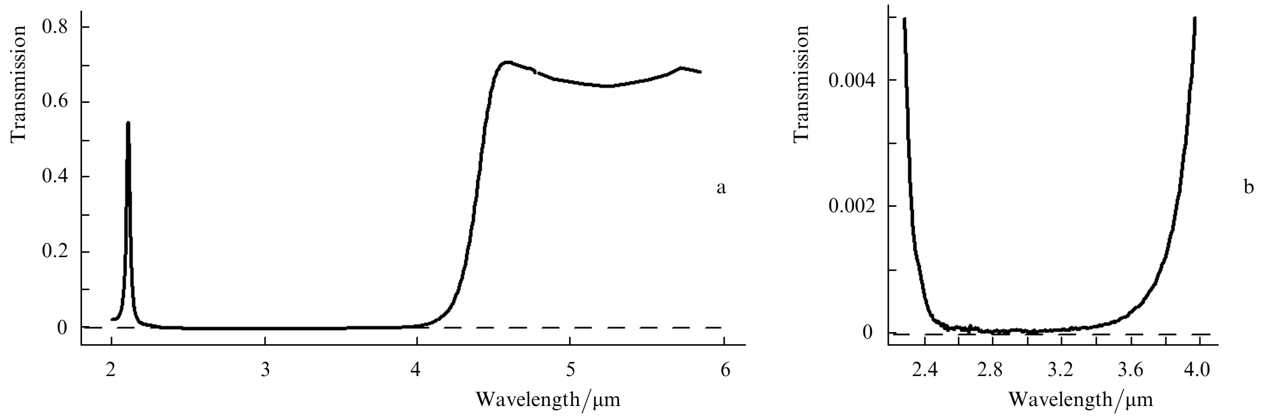

Figure 7. Total spectral characteristic (a) and its part near the transmission minimum (b) of the back spherical mirror with the curvature radius of 2000 mm (a silicon substrate with a dielectric coating).

We used the following optical scheme to detect spectral and energy parameters of the laser. Laser radiation propagated through a plane plate made of 20-mm-thick IR quartz, which totally absorbed radiation with the wavelengths exceeding 4 μm, to cut off luminescence and lasing (if it appeared) at the fundamental transitions of the CO molecule. Then, after the propagation through the CaF_2 beamsplitter, radiation was directed to the diffraction monochromator. The lasing power concentrated in some lines was measured at the monochromator output. Absolute calibration of the spectral and energy distribution of the laser power was performed by using a second power meter, which measured the energy reflected by the beamsplitter.

We used two sets of resonator mirrors with different spectral parameters. At the invariable rear mirror, whose spectral parameters are presented in Fig. 7, the emission spectrum of the laser was determined by the spectral properties of the output mirrors. Figure 8 shows the transmission spectra of output mirrors and typical lasing power distributions corresponding to them, which were measured relative to the lines of the overtone slab CO laser with repetitively-pulse RF excitation in the nonselective free-running regime. The total (by two sets of resonator mirrors) output spectrum of the laser consisted of ~90 lines of vibrational transitions $V + 2 \rightarrow V$ (from $8 \rightarrow 6$ to $36 \rightarrow 34$) and covered the spectral region from 2.55 to 3.9 μm. The

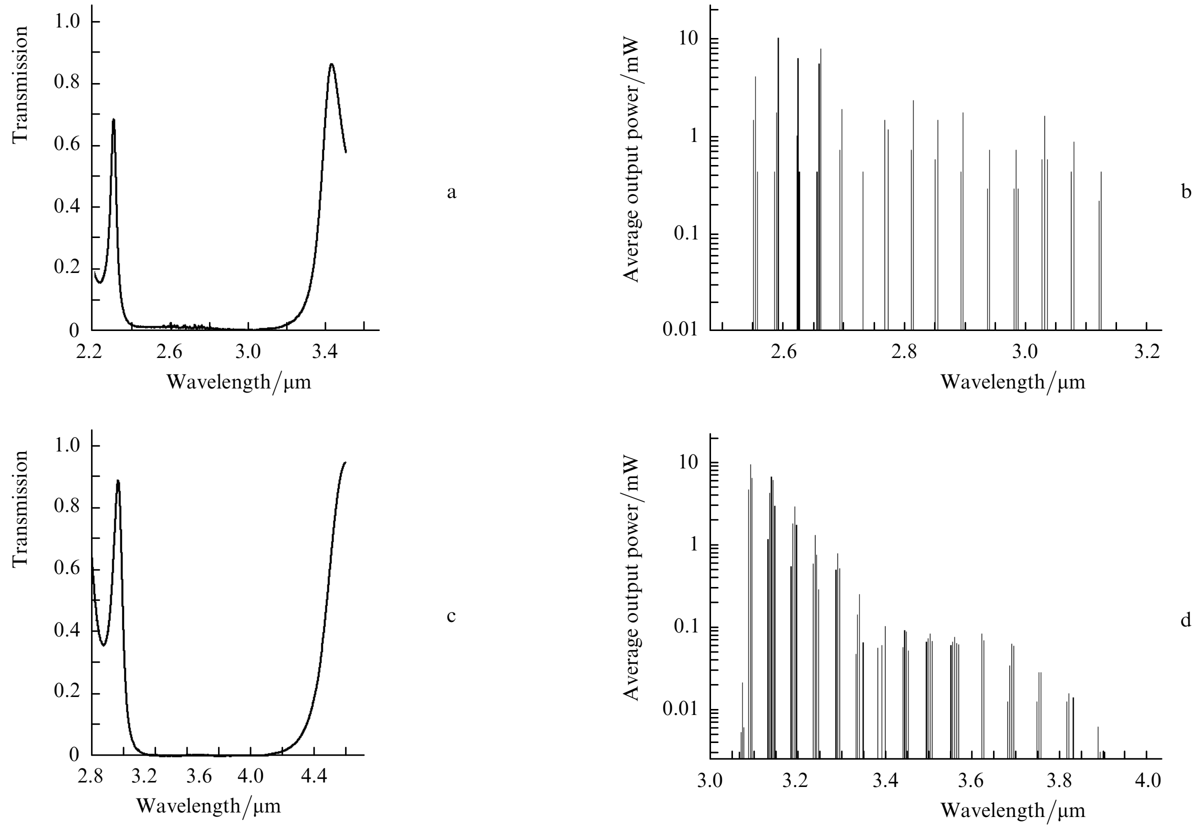

Figure 8. Transmission spectra of output plane mirrors (a, c) and spectra of free-running multifrequency lasing of the repetitively pulsed RF-pumped overtone slab CO laser (b, d) corresponding to them.

average output power of the overtone CO laser with the output mirror, whose transmission spectrum is presented in Fig. 8a, achieved ~0.3 W for the efficiency of about 0.5 % (in total over all the lasing lines), the maximum power in a separate line being ~12 mW. The maximum average output power of the overtone CO laser with the other output mirror (Fig. 8c), which was achieved in the experiments, was somewhat lower (~90 mW in total over all the lasing lines). The decrease in the output power is obviously caused by the less optimal parameters of the resonator, which strongly affects the operation efficiency of the laser taking into account small values of the gain at the overtone transitions of the CO molecule.

4. Conclusions

We have developed a compact slab CO laser pumped by a repetitively pulsed and cw capacitive RF discharge and having cryogenically cooled electrodes. It has been shown that only mixtures with a high content of oxygen (up to 20% with respect to the concentration of CO molecules) are suitable as the active medium of this laser. We have performed the parametric study of this laser operating at vibrational–rotational transitions of the fundamental ($V + 1 \rightarrow V$) vibrational bands of the CO molecule. The optimal conditions of its operation (the composition and the pressure of the gas mixture, RF-pumping parameters) have been determined in the regime of free-running multifrequency lasing. It has been demonstrated that repetitively-pulsed pumping is more efficient than the cw pumping, lasing proceeding in the quasi-cw regime in this case. The emission spectrum lied in the region from 5.1 to 5.4 μm. The maximum average power of the laser obtained in the experiments was ~12 W and its efficiency achieved ~14 %.

The frequency-selective lasing regime of the RF-excited slab CO laser has been realised at ~100 laser lines in the spectral region from 5.0 to 6.5 μm. The average output power of the CO laser in this regime is from several milliwatts to several tens of milliwatts in each line.

All the results have been obtained without the forced gas mixture exchange. Experimental conditions being fixed, stable lasing (with the fluctuations of the output parameters of no more than ±10 %) has been realised for more than an hour, which corresponds to the number of laser pulses $(3-5) \times 10^5$.

The RF CO laser has operated for the first time at overtone vibrational–rotational transitions ($V + 2 \rightarrow V$) of the CO molecule in the spectral region 2.5–3.9 μm with the average output power up to 300 mW.

This laser combining relative simplicity and compactness inherent in RF-excited slab lasers as well as a high efficiency and a broad emission spectrum typical of electric-discharge CO lasers can become a unique spectroscopic instrument for detecting different explosives, toxic and other dangerous substances and materials.

Acknowledgements. This work was supported by the Russian Foundation for Basic Research (Grant Nos 06-02-08041 and 07-05-00765) and the Program for the Youth Support at the Educational and Scientific Complex of P.N. Lebedev Physics Institute, RAS. The authors thank Yu.V. Terekhov and A.V. Shelestovich for their help in the experiments, A.A. Kotkov, Yu.M. Klimachev and O.A. Rulev for their help in preparing the experiments and discussion of the results as well as A.P. Mineev for useful advice.

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