

Pulsed laser operating on the first vibrational overtone of the CO molecule in the 2.5–4.2- μm range:

1. Multifrequency lasing

N G Basov, A A Ionin, A A Kotkov, A K Kurnosov, J E McCord,
A P Napartovich, L V Seleznev, N G Turkin, G D Hager

Abstract. Characteristics of a pulsed, multifrequency electroionisation CO laser operating on the first vibrational overtone of the CO molecule are studied experimentally and theoretically. It is shown experimentally that a pulsed, multifrequency overtone CO laser is an efficient source of coherent radiation in the 2.5–4.1- μm region whose specific output reaches $50 \text{ J l}^{-1} \text{ amagat}^{-1}$ and the electro-optical efficiency amounts to 11%. A theoretical analysis based on experimental data shows the feasibility of obtaining overtone emission with efficiency as high as $\sim 20\%$. The alternation of the intensity of vibrational bands in the overtone CO laser is discussed. This effect is caused by the cascade mechanism of formation of the emission spectrum. The comparison of experimental data with calculations showed that they agree well with respect to spectral and energy characteristics of the overtone CO laser. Moreover, it revealed the necessity of refinement of the kinetic model of the active medium of the CO laser for obtaining an adequate description of time parameters of a laser pulse.

1. Introduction

Owing to the kinetics of vibrational-vibrational (VV) exchange [1, 2], a free-running laser operating on the first overtone ($\Delta v = 2$) of the CO molecule [3, 4] can use vibrational transitions lying considerably higher than the transitions used in a CO laser operating on the fundamental band ($\Delta v = 1$). The spectral range of emission of an overtone CO laser overlaps the range of HF and DF lasers, and the density of its filling with vibrational-rotational lines is considerably higher. Many of these lines coincide with absorption lines of various organic and inorganic compounds. An overtone CO laser emitting at these vibrational-rotational lines can be used for the resonant action on various media in nonlinear spectroscopy, laser monitoring of the atmosphere, laser chemistry, etc. Note that the long-wave-

length part of the emission spectrum of an overtone CO laser falls into the atmospheric transparency window, whereas the short-wavelength emission is efficiently absorbed by water molecules and can be used, for instance, in laser medicine.

By the early 90s, the emission spectrum of a pulsed, multifrequency overtone laser, which was observed in experiments [3–7], lay in a rather narrow spectral range from 2.7 to 3.3 μm , which corresponds to vibrational transitions $v + 2 \rightarrow v$ from $12 \rightarrow 10$ to $23 \rightarrow 21$. As shown in Refs [5–7], the electrooptical efficiency of a pulsed overtone multifrequency CO laser (the ratio of the output energy to the pump energy deposited into an active medium before the end of the laser pulse, hereafter referred to as the laser efficiency) reaches 5%, with the specific output of up to $10 \text{ J l}^{-1} \text{ amagat}^{-1}$ and the total output of up to 50 J. In a cw low-pressure laser [8], overtone emission at a separate vibrational-rotational transition of the CO molecule was obtained, and in [8, 9], lasing was observed on 330 separate spectral lines in the 2.62–4.07- μm range (from the $10 \rightarrow 8$ transition to the higher $37 \rightarrow 35$ transition), the maximum emission power reached $\sim 0.5 \text{ W}$ (the laser efficiency was not reported there), and the spectral line width was $\sim 110 \text{ kHz}$ [10].

Because of the lack of experimental data on the rates of VV exchange involving higher vibrational levels of the CO molecule, it is common to use the extrapolation of kinetic constants in the kinetic model of the active medium of a CO laser [11–17]. For instance, in Ref. [17], based on the agreement between the calculated and the experimental data, a set of kinetic constants for a laser operating on fundamental transitions of the CO molecule from $5 \rightarrow 4$ to $16 \rightarrow 15$ was used for the calculation of energetic potentialities of a supersonic overtone electroionisation CO laser operating on overtone transitions up to $44 \rightarrow 42$. However, the applicability of extrapolation of kinetic constants for such high vibrational transitions requires justification based on comparison with experimental data.

This comparison was made in Ref. [16], where lasing properties of a multifrequency electroionisation CO laser with a short $\sim 1.5\text{-}\mu\text{s}$ pump pulse were studied. In Ref. [16], a good agreement between the experimental and calculated data was found, but the emission spectrum was rather narrow (11 lines corresponding to transitions from $14 \rightarrow 12$ to $19 \rightarrow 17$), the laser efficiency did not exceed 1.2%, and the maximum specific output was $12 \text{ J l}^{-1} \text{ amagat}^{-1}$.

Here, we report results of the experimental and theoretical study of a pulsed electroionisation laser operating on the first vibrational overtone of the CO molecule in the multifrequency regime (see also [18–24]). Results of our study of

N G Basov, A A Ionin, A A Kotkov, L V Seleznev P N Lebedev Physics Institute, Russian Academy of Sciences, Leninskii prosp. 53, 117924 Moscow, Russia; e-mail: aion@sci.lebedev.ru

A K Kurnosov, A P Napartovich, N G Turkin Troitsk Institute for Innovation and Thermonuclear Research, 142190 Troitsk, Moscow oblast, Russia; e-mail: apn@triniti.ru

J E McCord, G D Hager AFRL, Albuquerque, New Mexico, USA

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frequency-selective operation of an overtone CO laser on separate vibrational-rotational transitions will be reported elsewhere [25].

2. Laser setup and methods used for the measurements of parameters

Experiments were carried out in Levedev Physics Institute of the Russian Academy of Sciences at a pulsed cryogenic electroionisation laser setup with an active medium 1.2 m long. The maximum current density in an electron beam was 20 mA cm^{-2} , and the electron energy was about 150 keV. The electroionisation discharge current pulse was nearly triangular, with a maximum on the leading edge. Its duration could be varied from 25 to 1500 μs (at a level of 0.1). The specific deposited energy was varied from 50 to $1000 \text{ J l}^{-1} \text{ amagat}^{-1}$. The deviation of the deposited energy from these values at the ends of an electric-discharge gap did not exceed 10%. In most of the experiments, the ratio of the electric field strength to the gas mixture density E/N was $\sim 4 \text{ kV cm}^{-1} \text{ amagat}^{-1}$.

The experimental system was able to measure the energy and the dynamics of a laser pulse, radiation intensity distributions in the near- and far-field regions, and the laser emission spectrum. Emission at the overtone or fundamental transitions was separated out by spectral filters. They represented thin quartz plates totally absorbing radiation with wavelength larger than $5 \mu\text{m}$ or broad-band interference mirrors with reflectivity in the $2.5\text{--}4.0\text{-}\mu\text{m}$ range higher than 90% and transmission in the $5\text{--}6\text{-}\mu\text{m}$ range higher than 90%.

The vibrational-rotational structure of the laser emission spectrum was analysed using a normal-incidence slit spectrograph with a $150 \text{ lines mm}^{-1}$ diffraction grating blazed at $4.0 \mu\text{m}$. The emission spectrum in the focal plane of the spectrograph was recorded on a diffusely scattering screen using an infrared imager and subsequent digital processing. In the experiments, we used laser cavities with dielectric mirrors and an intracavity spectral filter.

3. Laser with an intracavity filter

Lasing characteristics of the overtone CO laser were studied using an intracavity broad-band spectral filter, which totally suppressed lasing in the fundamental band. The filter represented a thin $\sim 0.5\text{-mm}$ plane-parallel plate made of fused quartz, which was 50 mm in diameter. It was positioned in the laser cavity near the output window of a discharge chamber at a certain angle to the optical axis of the cavity. The laser cavity was formed by two copper mirrors. One of them was plane, and the other was spherical and had a $\sim 5\text{-m}$ radius of curvature. The spherical mirror was mounted directly on the discharge chamber. The lasing region was bounded by an intracavity aperture 36 mm in diameter, and its volume was $\sim 1.2 \text{ l}$. Radiation was outcoupled from the cavity through Fresnel reflection from both faces of the filter. Fig. 1 presents the spectral transmission of the filter, its absorption, and a typical spectrum of laser emission.

3.1. Spectral characteristics

Multifrequency overtone laser emission was observed in the $2.7\text{--}3.6\text{-}\mu\text{m}$ range (Fig. 1), which corresponds to vibrational transitions from $13 \rightarrow 11$ to $30 \rightarrow 28$ in the absence of lasing on the fundamental transitions. The spectral range of laser emission was limited because of the intracavity absorp-

tion of atmospheric water vapour at wavelengths below $2.7 \mu\text{m}$ and absorption of the filter material above $3.6 \mu\text{m}$. Using conditions in which lasing on the fundamental transitions of CO was totally suppressed, we carried out a multipartametric study of characteristics of a pulsed multifrequency electron-beam-controlled discharge laser operating on the first vibrational overtone of the CO molecule.

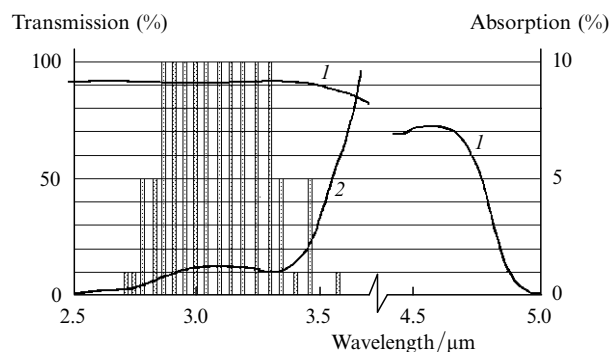


Figure 1. Spectral transmission (1) and absorption (2) of the filter, and a typical histogram of laser emission.

3.2. Energy characteristics

When studying energy characteristics of the overtone CO laser, we analysed the effect of its parameters on the specific output and the laser efficiency and compared experimental and calculation data (see also [18–20]). In the experiments, we varied the following parameters of a laser mixture and a pulse of the electroionisation discharge: the density (from 0.02 to 0.5 amagat), the initial temperature (from ~ 100 to 300 K), the composition of the laser mixtures $\text{CO} : \text{N}_2 = 1 : x$ (x was varied from 1.5 to 39) and $\text{CO} : \text{N}_2 : \text{He} = 1 : 9 : y$ (y was varied from zero to 20); the specific deposited energy (from 50 to $1000 \text{ J l}^{-1} \text{ amagat}^{-1}$), the pump pulse duration (from 25 to 1500 μs), the capacity of a capacitor bank (from 12 to 67 μF). We also varied the outcoupling coefficient of a laser cavity from 0.5 to 10% by tilting the filter with respect to the optical axis and varied its Q factor by introducing additional optical loss inside the cavity (up to 30%).

In this study, we determined the optimum conditions providing the highest efficiency in the pulsed, multifrequency overtone electroionisation CO laser: the gas density from 0.1 to 0.3 amagat, a gas temperature of $T \sim 100 \text{ K}$, the composition of the laser mixture $\text{CO} : \text{N}_2 : \text{He} = 1 : 9 : 10$, the specific deposited energy from 200 to $300 \text{ J l}^{-1} \text{ amagat}^{-1}$, with a $\sim 25\text{-}\mu\text{s}$ pump pulse, and an outcoupling coefficient of 4–8%. However, the conditions required for obtaining the maximum specific output were somewhat different. For this purpose, we used the $\text{CO} : \text{N}_2 = 1 : 9$ mixture, and the deposited energy was $500\text{--}600 \text{ J l}^{-1} \text{ amagat}^{-1}$. Fig. 2 presents the dependences of the specific output on the specific deposited energy for the $\text{CO} : \text{N}_2 = 1 : 9$ mixture with density $N = 0.06\text{--}0.5 \text{ amagat}$. The maximum specific output above $20 \text{ J l}^{-1} \text{ amagat}^{-1}$ was obtained for a gas density of 0.2 amagat and a deposited energy of $\sim 600 \text{ J l}^{-1} \text{ amagat}^{-1}$.

The comparison of the experimental data with theoretical calculations performed using the kinetic model of the active medium of the CO laser taking into account the experimental conditions showed [18–20] that the energy and spectral char-

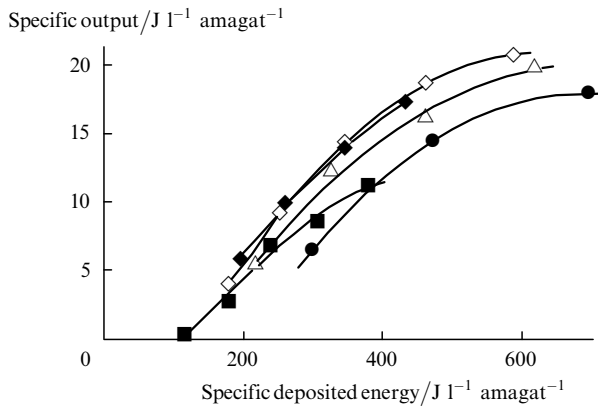


Figure 2. Dependences of the specific laser output on the specific deposited energy for the CO : N₂ = 1 : 9 gas mixture with density $N = 0.5$ (■), 0.3 (◆), 0.2 (◇), 0.12 (△), and 0.06 amagat (●).

acteristics of the overtone CO laser were well described by this model.

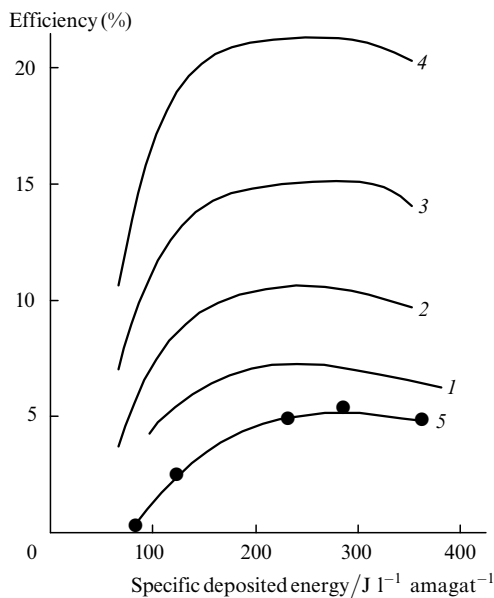


Figure 3. Calculated (1–4) and experimental (5) dependences of the efficiency of the overtone CO laser on the specific deposited energy for different values of optical loss through absorption and scattering in optical elements for a round trip of the laser cavity for the CO : N₂ : He = 1 : 9 : 10 mixture with $N = 0.3$ amagat at $T \sim 100$ K. (1) 6, (2) 4, and (3, 4) 2% (curve 4 was obtained under the assumption that the laser cavity contained no atmospheric water vapour).

Fig. 3 presents the experimental and theoretical dependences of the efficiency of the overtone CO laser on the specific deposited energy for the CO : N₂ : He = 1 : 9 : 10 mixture with a density of 0.3 amagat and temperature of ~ 100 K. In these experiments, the efficiency of the overtone CO laser reached a maximum value of $\sim 5.5\%$ at $\sim 300 \text{ J l}^{-1} \times \text{amagat}^{-1}$. Theoretical curves 1–4 in Fig. 3 correspond to different loss through absorption and scattering in optical elements during a round trip over the cavity. Curve 1 (6% loss) well agrees with the experimental data. For optical loss as low as 2% (curve 3), which can be really achieved in this laser cavity, the calculated efficiency increased up to $\sim 15\%$. The calculation performed for the optical cavity con-

taining no atmospheric water vapour, which corresponds to the extension of the emission spectrum to the short-wavelength region, predicts for the overtone CO laser the efficiency as high as $\sim 20\%$.

3.3. Time characteristics

Under conditions giving the maximum laser efficiency, we observed a nearly-triangular pulse of overtone emission 0.5–2 ms long, which almost always started after the end of a pump pulse (see below an exception from this rule). As the gas mixture density was decreased from 0.5 to 0.02 amagat, the emission pulse lengthened from 0.3 to 3 ms, and the delay of its onset with respect to the onset of a pump pulse increased from 80 to 500 μs [18–20]. An increase in deposited energy above the optimum values ($\sim 300 \text{ J l}^{-1} \text{ amagat}^{-1}$) shortened the emission pulse.

We also showed in these experiments that shortening of a pulse of an electroionisation discharge down to 25 μs , with deposited energy being fixed, caused a monotonic increase in the laser efficiency and specific output. A similar increase in energy characteristics of the overtone CO laser with decreasing pump pulse duration, but under other experimental conditions is illustrated in Fig. 4.

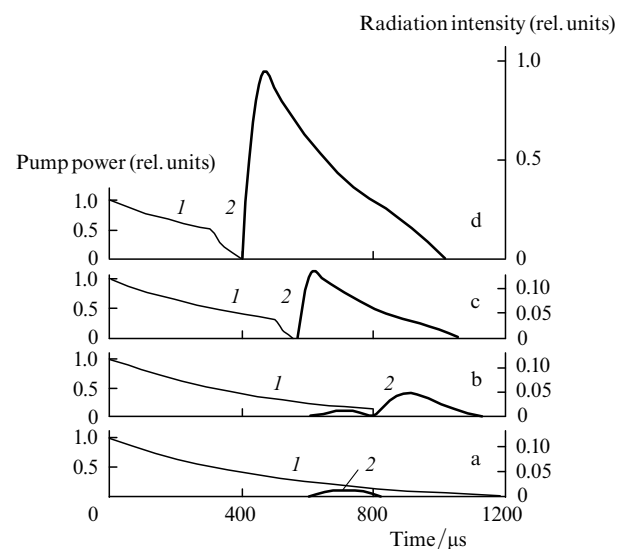


Figure 4. Time dependences of the pump power (1) and the overtone emission intensity (2) for a specific deposited energy of $300 \text{ J l}^{-1} \text{ amagat}^{-1}$ in the CO : N₂ : He = 1 : 9 : 10 mixture with $N = 0.3$ amagat.

In this experiment, the time dependences of the pump power were identical up to the end (interruption) of an electroionisation discharge pulse. The pump pulse duration was varied from 400 to 1200 μs . Fig. 4a shows a complete pump pulse (curve 1) with the duration $t_p \sim 1200 \mu\text{s}$ and specific deposited energy of $300 \text{ J l}^{-1} \text{ amagat}^{-1}$ in the CO : N₂ : He = 1 : 9 : 10 laser mixture with a density of 0.3 amagat. Under these conditions, the overtone CO laser (curve 2) emitted a very weak pulse upon pumping an active medium by an electroionisation discharge. This pulse was ~ 0.2 ms long, and its onset was delayed with respect to the pump pulse by $\sim 600 \mu\text{s}$.

The interruption of an electroionisation discharge pulse after the formation of a laser pulse at the moment $t = t_p \approx 800 \mu\text{s}$ (Fig. 4b) caused a sharp increase in the overtone emission intensity and an increase in the laser pulse duration up to

0.5 ms. A decrease in the pump pulse duration from ~ 600 (Fig. 4c) to ~ 400 μs (Fig. 4d) resulted in an increase in the intensity and energy of the overtone emission almost by an order of magnitude.

The results of this experiment show that a discharge in a gas substantially affects kinetic processes taking place in the active medium of the overtone CO laser. The calculated delay time of the laser pulse and its duration were considerably smaller than their typical values observed in the experiment. This disagreement calls for the refinement of mechanisms responsible for energy transfer to relatively high vibrational levels of the CO molecule (see, e.g., [19]).

Using the intracavity filter, we suppressed lasing on the fundamental transitions of CO and expanded the spectral range of multifrequency laser emission to the long-wavelength region up to 3.6 μm . In the multiparametric study of characteristics of the laser with this filter, we determined optimum conditions for increasing efficiency and specific output of the overtone CO laser.

4. Laser with interference mirrors

Using results of our study of the effect of cavity parameters on characteristics of the overtone CO laser with an intracavity filter, we formulated requirements imposed on laser mirrors. The coefficient of outcoupling from the cavity for a round-trip for the 2.5–4.2- μm radiation should be 4–8%, and the reflection coefficient in the 4.8–6.5- μm range should be lower than 10% (for a symmetric cavity). In the experiments, we used different pairs of dielectric interference laser mirrors with different spectral characteristics, which were deposited on CaF_2 substrates.

4.1. Spectral range of laser emission

Fig. 5 presents reflectivities of three pairs of laser mirrors of an equivalent symmetric cavity: $R(\lambda) = (R_i R_j)^{1/2}$, $i, j = 1, 2$,

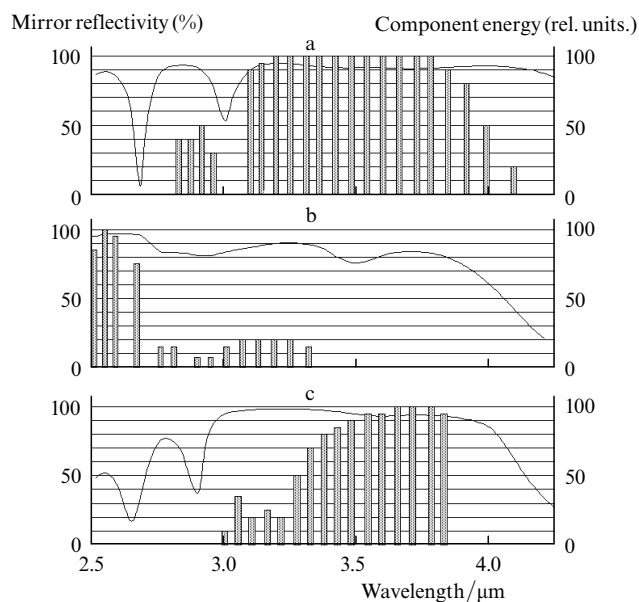


Figure 5. Vibrational spectra of the overtone emission (histograms) and the corresponding reflectivities of mirrors of an equivalent symmetric cavity (curves) for three pairs (a–c) of laser mirrors for the CO : N_2 : He = 1 : 9 : 10 mixture with $N = 0.12$ amagat and specific deposited energy of $300 \text{ J l}^{-1} \text{ amagat}^{-1}$.

where R_i and R_j are the mirror reflectivities. The vibrational spectra of overtone emission are shown in the form of histograms. In each vibrational band, we observed lasing on 2–3 vibrational-rotational lines. Using in these experiments different pairs of cavity mirrors, we expanded the spectrum of multifrequency overtone laser emission both to the long-wavelength region up to 4.1 μm (the $37 \rightarrow 35$ vibrational transition, Fig. 5a) and to the short-wavelength region up to 2.5 μm (the $6 \rightarrow 4$ transition, Fig. 5b). In spite of a low reflectivity of the mirrors in the 5–6- μm range ($R < 10\%$, i.e., the outcoupling coefficient for a round trip was higher than 99%), lasing on the fundamental transitions of CO was excited in the cavity.

4.2. Energy characteristics

Fig. 6 presents the experimental and theoretical dependences of the laser efficiency on the specific deposited energy for the cavity whose spectral characteristic is shown in Fig. 5c. The major portion of the energy emitted by this laser corresponded to higher-lying transitions (with wavelengths above 3.5 μm). It is likely that this fact is caused by a too high (nonoptimal) Q factor of the cavity in the 3.1–3.5- μm range.

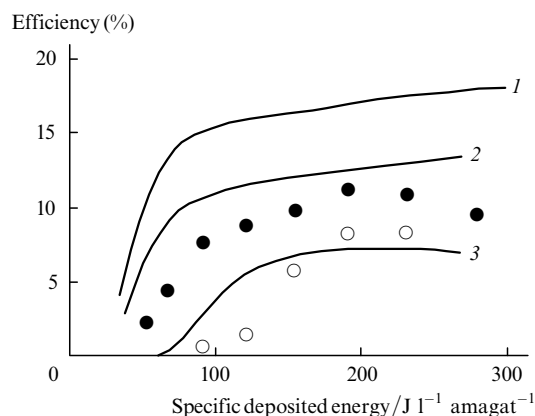


Figure 6. Experimental (points) and theoretical (solid curves) dependences of the electro-optical laser efficiency on the specific deposited energy for the experimental conditions corresponding to Fig. 5c: (●, 1, 2) overtone transitions; (○, 3) fundamental transitions (curve 2 was calculated for 0.5% additional loss for a round trip of the laser cavity).

The maximum efficiency of the overtone laser was $\sim 11\%$, and it was obtained for the specific deposited energy of $\sim 200 \text{ J l}^{-1} \text{ amagat}^{-1}$. Curves 1 and 3 (Fig. 6) calculated for the experimental conditions (the CO : N_2 : He = 1 : 9 : 10 mixture with a density of 0.12 amagat) represent the efficiency of the laser operating on overtone and fundamental transitions, respectively, as a function of the specific deposited energy. Theoretical curve 1 predicts the laser efficiency exceeding the experimental value by a factor of ~ 1.5 (up to 17%).

However, curve 2, which was calculated for a cavity with additional 0.5% loss for a round trip, agrees with the experimental data better. This suggests that a decrease in the optical loss in a laser cavity, even by a value as small as 0.5%, can substantially increase the overtone laser efficiency. A decrease in the efficiency of lasing on overtone transitions with increasing specific deposited energy (above $200 \text{ J l}^{-1} \times \text{amagat}^{-1}$) was caused by gas mixture heating and an increase in the energy emitted in the fundamental band.

The efficiency for lasing in the fundamental band reached 9%, i.e., the total efficiency of the CO laser was 20%. For the laser cavity whose spectral characteristic is presented in Fig. 7, the fundamental transitions emitted a considerably lower laser energy than the overtone transitions. As the specific deposited energy was increased up to $\sim 650 \text{ J l}^{-1} \text{ amagat}^{-1}$, the specific output energy of the overtone laser monotonically increased up to $\sim 50 \text{ J l}^{-1} \text{ amagat}^{-1}$ (Fig. 8).

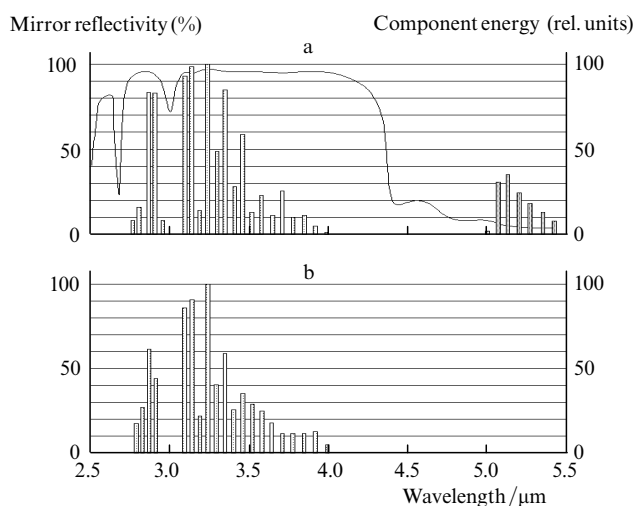


Figure 7. Reflectivities of mirrors of the equivalent symmetric cavity (curve) and the distribution of laser emission energy over vibrational bands for a specific deposited energy of $300 \text{ J l}^{-1} \text{ amagat}^{-1}$ in the CO : N₂ : He = 1 : 9 : 10 mixture with $N = 0.12 \text{ amagat}$ (a – experiment, b – theory).

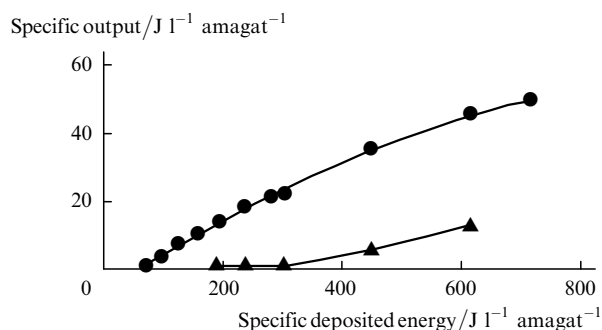


Figure 8. Dependences of the specific output power on the specific deposited energy for the CO laser with the cavity whose spectral characteristic is presented in Fig. 7. The results correspond to the CO : N₂ = 1 : 9 mixture with $N = 0.12 \text{ amagat}$ for overtone (●) and fundamental (▲) transitions.

4.3. Narrow-band lasing

Using laser mirrors with a high reflectivity in a relatively narrow 200–300- cm^{-1} spectral range, one can obtain a high efficiency of the overtone CO laser. The emission spectrum of a laser with a cavity formed by a pair of such mirrors consisted only of five-six vibrational bands (similarly to the spectrum in Fig. 5b), but the laser efficiency exceeded 5%. In this case, no lasing on the fundamental transitions was observed [21–23].

To find out potentialities of the narrow-band oscillation in the overtone CO laser, we calculated its efficiency for the specific deposited energy of $220 \text{ J l}^{-1} \text{ amagat}^{-1}$ for the CO : N₂ : He = 1 : 9 : 10 mixture (Fig. 9). In the calcula-

tions, the laser mirrors were assumed to have reflectivity of 97.5%, transmission of 1.5%, and passive loss of 1% in the spectral range containing five (the range 200–250 cm^{-1} wide, Fig. 9a) or three (100–150 cm^{-1} , Fig. 9b) neighbouring overtone vibrational bands. The numbers of vibrational components of the *P*-branch were determined from the condition of competition of vibrational-rotational transitions. The maximum efficiency calculated for the overtone CO laser operating on five or three overtone bands exceeded 6 and 4%, respectively.

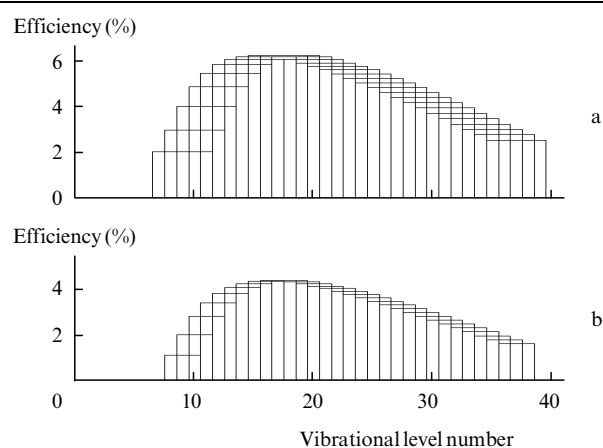


Figure 9. Summary (over all lines) efficiency of the overtone CO laser operating on five (a) and three (b) neighbouring transitions $v + 2 \rightarrow v$ as a function of the number of the vibrational level v for a deposited energy of $220 \text{ J l}^{-1} \text{ amagat}^{-1}$ in the CO : N₂ : He = 1 : 9 : 10 mixture with $N = 0.12 \text{ amagat}$. The corresponding transitions are connected by a horizontal line.

Note that the laser output in the cases considered above was distributed among neighbouring transitions rather non-uniformly. For $v > 16$, more than 50% of the emission energy corresponded to the lowest (short-wavelength) transition $v + 2 \rightarrow v$ (see also [25]).

4.4. Alternation of the intensity in the vibrational spectrum

Fig. 7a presents the distribution of the laser output over vibrational bands in the form of a histogram. The measurements were performed with a calorimeter in the focal plane of the spectrograph. This distribution shows an alternation of strong and weak vibrational bands in the 3–4- μm range. This alternation cannot be attributed to a change in the *Q* factor of the laser cavity, whose spectral characteristic in this range is virtually constant (the solid curve in Fig. 7a).

This effect has been theoretically predicted in Ref. [12]. It is caused by lasing on two vibrational cascades including only even or only odd vibrational levels. The calculated spectrum of the overtone emission is presented in Fig. 7b, and it agrees well with the measured spectrum, including the alternation effect. Note that the laser output in the fundamental band in the experiments was very low; in this case, the calculations gave no lasing on these transitions.

The alternation of the intensity was also observed for other laser mixtures (CO : N₂, CO : He, and pure CO) for different deposited energies down to threshold values and gas densities from 0.02 to 0.3 amagat. The observation of the alternation effect in the experiments gives evidence of

the cascade mechanism of oscillation in the CO laser and of the excess of the rate of radiative transitions over the rate of the VV exchange.

4.5. Spectral and time characteristics

Time characteristics of laser emission for each vibrational band were measured under the experimental conditions corresponding to Fig. 7. Emission pulses were nearly triangular, and their peak was found near the pulse onset. The duration of an emission pulse and its time delay as functions of the number of the lower vibrational level ν are presented in Fig. 10 in the form of a histogram. Their values for overtone vibrational transitions were 1–4 ms and 130–500 μs , respectively.

The pulses emitted at vibrational transitions of the fundamental band were considerably shorter, and their duration was 0.05–0.5 ms. Typical delay times for emission pulses of the fundamental band ranged from 30 to 60 μs . The only exception from this rule (see Fig. 10) are two vibrational bands with $\nu = 9$ and 10 whose delay times (150–180 μs) are comparable to the delay times for the neighbouring overtone transitions with $\nu = 11$ and 12. This is probably related to the cascade mechanism of inverse-population formation and the mutual effect of lasing on the overtone and the fundamental band. This mutual effect has been also observed in Ref. [5]. Note that the intensity alternation observed in the emission spectrum of the overtone CO laser (Fig. 7) for $\nu > 19$ was almost absent in the spectrum of time characteristics.

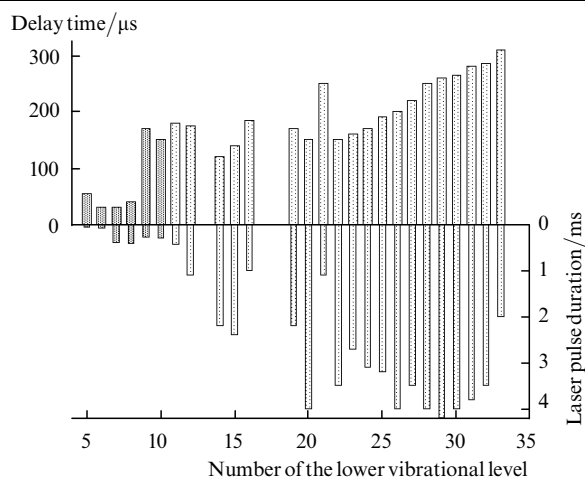


Figure 10. Dependences of the duration and the delay time of an emission pulse on the number of the lower vibrational number ν for the experimental conditions corresponding to Fig. 7 (the fundamental transitions are specified by dark grey, and the overtone transitions are light grey).

5. Conclusions

Based on the multiparametric study of characteristics of the overtone electroionisation CO laser with an intracavity filter with totally suppressed lasing on the fundamental band, we found the optimum experimental conditions required for increasing the laser efficiency, namely, the pumping conditions, the Q factor of the laser cavity, the temperature of a gas medium, its density, and composition. The experimental efficiency of the overtone laser reached $\sim 5.5\%$. The theoretical analysis based on the comparison of experimental and theoretical data predicts the efficiency of the overtone electroionisation laser as high as 20%.

The use of broad-band dielectric mirrors with a high reflectivity and a low optical loss provided a considerable expansion of the range of overtone laser emission up to 2.5–4.1 μm , which corresponds to vibrational transitions from $6 \rightarrow 4$ to $37 \rightarrow 35$. We experimentally observed the alternation of intensities of strong and weak vibrational bands, which has been predicted theoretically. The experimental and theoretical data were found to be in good agreement. The maximum efficiency of the overtone laser was 11%, and the specific output reached $50 \text{ J l}^{-1} \text{ amagat}^{-1}$. It follows from the theoretical calculations that a decrease in the optical loss for a round trip by a value as small as 0.5% can increase the overtone CO laser efficiency up to 17%. A combination of spectral and energy characteristics of a pulsed CO laser makes it competitive with chemical HF(DF) lasers.

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