

Multifrequency laser probing of CO-containing gas mixtures excited in a pulsed discharge

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Abstract. The method of multifrequency laser probing is developed which can be used for diagnostics of the temperature and population of vibrational levels in gas mixtures containing CO molecules in excited vibrational states. The method is tested by studying the dynamics of the gas temperature and population of vibrational levels of the CO molecule in gas mixtures excited by a pulsed discharge. It is shown that the method provides the reduction of the gas temperature measurement error down to 3%. It is found that the population of lower vibrational levels in the CO – O₂ mixture can exceed the population of levels in CO – He and CO – N₂ laser mixtures by several times.

Keywords: vibrational exchange, vibrational distribution function, nonself-sustained discharge, active medium, carbon oxide, CO laser.

1. Introduction

The vibrational levels v of N₂, O₂, CO and other molecules are populated in gas-discharge plasmas. These processes are used to pump active media of gas-discharge CO, CO₂ and other molecular lasers. The energy exchange between molecules results in the formation of the plateau-like vibrational distribution function (VDF) [1–3], the gas temperature considerably affecting the rate of nonequilibrium kinetic processes proceeding in the medium. Therefore, to study transient kinetic processes, it is necessary to develop methods for measuring the gas temperature and population of vibrational levels. In [4–6], the method of multifrequency laser probing of the active medium of a cw CO laser amplifier was developed for this purpose. The method is based on the fact that the small-signal gain (SSG) at the vibrational–rotational transition of molecules is related to the gas temperature T and populations of the upper (N_{v+1}) and lower (N_v) vibrational levels [7]. These quantities can be calculated (recovered) from the measurements of absorption or amplification of radiation at several ($m > 2$) vibrational–

rotational molecular transitions. The recovery procedure involves the search for a set of parameters T , N_v , and N_{v+1} for which the calculated values of the SSG $g_{v,J}$ describe in the best way the gain (absorption) $G_{v,J}$ measured for the rotational components J of the $v + 1 \rightarrow v$ vibrational transition. Therefore, the more accurately the values of the SSG $G_{v,J}$ are measured, the more accurately the required parameters will be recovered. Note that in this case the rotational temperature is recovered, but the equality of the rotational and translational temperatures in molecular gases is established during several collisions [2] (the RT relaxation time under normal conditions is $\tau_{RT} \sim 10^{-9}$ s).

In [8, 9], the attempt was made to determine the temperature of the active medium of the pulsed CO laser amplifier by measuring the SSG at three ($m = 3$) rotational transitions of one vibrational band. The relative error of measuring the SSG with respect to the maximal measured value was 9%–15% [9]. However, it was shown in [6] that the typical error of measuring the SSG for $m = 3$ ($\delta G = 10\%$) leads to a large relative error ($\delta T > 60\%$) of the gas temperature recovery (for example, for $T = 330$ K the absolute error is $\Delta T > 200$ K [6]). To reduce the relative error (for example, down to $\delta T \sim 8\%$ for $m = 3$), the SSG values should be measured more accurately ($\delta G < 1\%$ [6]).

In [4–6], the accuracy of temperature recovery was increased by increasing the number m of spectral lines being probed because analysis performed in [4, 5] has shown that for $m = 10$ and $\delta G = 10\%$, the temperature recovery error ΔT can be reduced down to 1 K. In [5, 6], the active medium of a low-pressure cw CO laser amplifier pumped by a dc discharge was probed at a few tens ($m \approx 50$) of lines, however, the temperature recovery error was large ($\Delta T = 23$ K). The authors themselves note that ‘the gain measurement accuracy was probably insufficient’ [6]. In addition, to calculate the SSG, it is necessary to identify accurately the $v + 1 \rightarrow vP(J)$ vibrational–rotational transitions being probed. The problem appears because several spectral lines corresponding to different vibrational–rotational molecular transitions can lie within the spectral line of probe radiation. To solve this problem, the authors of paper [6] varied the quantum numbers v и J , and then used a rather uncertain criterion for estimates based on the fact whether ‘reasonable’ or ‘non-physical’ results were obtained after data processing. However, even in the case of a correct identification of the spectral lines, the insufficient measurement accuracy of the SSG can lead to incorrect results (for example, to the negative population and temperature). Therefore, the development of the method of multifrequency laser probing requires an increase in the SSG

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measurement accuracy and a reliable assignment of the transitions being probed.

Both these problems were solved in this paper for the development of the methods for diagnostics of the temperature and population of vibrational levels in molecular gases. In addition, the application of the method of multi-frequency laser probing upon pulsed pumping required the modification of the data processing procedure. The method developed in the paper was tested by studying the dynamics of the gas temperature and population of vibrational levels in the CO–He, CO–N₂, and CO–O₂ mixtures pumped by a pulsed discharge.

2. Experimental

The dynamics of the SSG $G_{vJ}(t)$ was measured at the vibrational–rotational transitions of the carbon oxide molecule. The optical scheme of experiments is presented in Fig. 1. A gas mixture containing CO molecules was excited by a pulsed e-beam sustained discharge (EBSD) in a discharge chamber of laser amplifier (1) with cryogenic cooling [10]. A low-pressure tunable cw CO laser pumped by a dc discharge was used as probe laser (2) [10]. A part of the laser beam was directed by beamsplitter (3) to IKS-31 spectrometer (4) for measuring the spectral parameters of probe radiation. The temporal parameters of radiation incident on and transmitted through the medium under study were measured with detectors (5) and (6) [Hg–Cd–Zn–Te photodetectors (PEM-L-3)]. Spherical mirrors (7) focused radiation on the entrance slit of the spectrometer and photodetectors. The output signals of detectors were recorded with a Tektronix TDS1012 oscilloscope. The probe radiation was separated from the spontaneous emission spectrum with the help of 150 lines mm^{-1} diffraction grating (8).

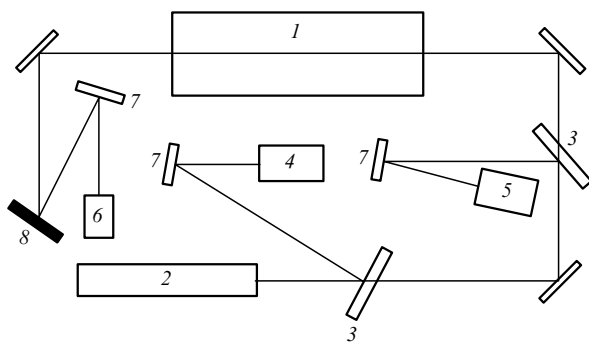


Figure 1. Optical scheme of experiments: (1) laser amplifier; (2) probe laser; (3) beamsplitter; (4) IR spectrometer; (5, 6) photodetectors; (7) spherical mirror; (8) diffraction grating.

We carefully selected the lines for probing in order to provide their reliable identification. Each of the selected lines should not overlap with the lines corresponding to other transitions (including isotopes and gases contained in the gas mixture) at which noticeable absorption or amplification of the probe radiation occurs. The line selection criterion was the condition that the frequency detuning $\Delta\nu_{vJ}$ of the probed line from the centre of the nearest line exceeds by many times the widths of the probe and probed lines; in addition, the accuracy of measuring the radiation frequency

was taken into account. The accuracy of measuring the radiation frequency of a probe CO laser in [6] was ‘insufficient’ ($\sim 2 \text{ cm}^{-1}$) because the spectral lines of CO are located so close to each other that neither of the hundred of the $\nu + 1 \rightarrow \nu P(J)$ lines of the $^{12}\text{C}^{16}\text{O}$ molecule satisfies the criterion $\Delta\nu_{vJ} > 2 \text{ cm}^{-1}$.

The full width at half-maximum of the lines probed under our experimental conditions did not exceed 0.02 cm^{-1} , while the error of measuring the laser radiation frequency was $\sim 0.2 \text{ cm}^{-1}$ ($\Delta\lambda = 0.8 \text{ nm}$). The lines for probing were selected by analysing the vibrational–rotational spectrum [11] of the natural isotopic mixture of carbon oxide (the most abundant isotope $^{12}\text{C}^{16}\text{O}$ has the relative concentration $y_1 \approx 0.987$, while the concentration of the $^{13}\text{C}^{16}\text{O}$ isotope is $y_2 \approx 0.011$). The lines were selected according to the criterion $\Delta\nu_{vJ} > 0.2(y_n/y_1) \text{ cm}^{-1}$ taking into account the relative concentration y_n ($n = 1, 2, \dots$) of the isotope to which the interfering line belongs. Of 700 lines corresponding to the $\nu + 1 \rightarrow \nu P(J)$ laser transitions of the $^{12}\text{C}^{16}\text{O}$ molecule, where $\nu = 2 - 36$ and $J = 4 - 23$, 250 suitable lines were selected (7 ± 3 lines in each vibrational band). For a stricter selection criterion $\Delta\nu_{vJ} > 0.3(y_n/y_1) \text{ cm}^{-1}$, the number of suitable lines was reduced almost by half (down to 130 lines).

Then, the spectral parameters of a set of 250 lines were compared with those of a probe CO laser (~ 200 spectral lines, $\nu = 5 - 31$). This comparison has shown that the number of lines within one vibrational band that can be used to recover the gas temperature and level populations rarely exceeds three. However, these parameters can be recovered by using spectral lines belonging to different vibrational bands. As a result, we found several sets containing ten spectral lines each, which belong to three successive vibrational bands.

To measure the SSG with good accuracy ($\delta G < 10\%$), the probe radiation intensity should be high enough ($10^{-1} - 1 \text{ W cm}^{-2}$ in our experiments), but lower than the gain saturation intensity I_s in the medium under study ($I_s \sim 10^2 \text{ W cm}^{-2}$ for transitions in the CO molecule [12]). The tuning range of the probe laser within each vibrational band should cover the transitions having the maximum absolute values of the SSG in the medium. To match the tuning range of the probe CO laser to the absorption spectrum of the medium under study, the probe laser cell was cooled with liquid nitrogen. In this case, the tuning range of probe radiation covers transitions with J from 6 to 18, the transitions with maximum SSG values ($\sim 1 \text{ m}^{-1}$) in the medium under study corresponding to $J = 6 - 10$. However, when amplification in the active medium is high, it is necessary to separate probe radiation from a broad spontaneous emission spectrum. This is achieved with the help of grating monochromator (8) (Fig. 1).

The procedure of selecting the lines for probing and the measurement of the emission spectrum of the probe laser provided the accurate assignment of each of the lines. The SSG $G_{vJ}(t)$ for each of the lines was measured several times (three measurements in each of three or four series) to eliminate random errors and find the absolute measurement error $\sigma_{vJ}(t)$. The absolute measurement error σ_{vJ} for the CO–N₂ and CO–He laser mixtures was $\sim 0.03 \text{ m}^{-1}$, while the relative measurement error δG was $\sim 2\%$.

The gas temperature $T(t)$ and population $N(t)$ of vibrational levels were recovered by minimising the target function

$$F(T, N) = \sum_{v,J} \left[\frac{g_{vJ}(T, N_v, N_{v+1}) - G_{vJ}(t)}{\sigma_{vJ}(t)} \right]^2, \quad (1)$$

where N is the two-row matrix (N_v, N_{v+1}) of the population of vibrational levels; g_{vJ} is the SSG value to be calculated; and $G_{vJ}(t)$ is the SSG value measured at the instant t ($t = 0$ is the instant of the discharge pulse onset). Note that the factor $\sigma_{vJ}(t)$ in the target function was neglected in papers [4–6], thereby assuming that SSG measurement errors are the same, whereas they differ by several times for different lines being probed (see [6]).

The procedure for minimisation of function (1) proposed in [4, 5] involved the determination of the minimum of the target function from the population $N_0(T_1)$ for some start value of the gas temperature T_1 . Then, the minimum of the function $F[T, N_0(T_1)]$ was determined from the gas temperature. However, such a simplified procedure for searching the minimum of function (1) can give incorrect results because the population $N_0(T_1)$ can strongly change even upon small variations in the gas temperature. For this reason, the result of the temperature recovery depends on the choice of the start value of T_1 . The procedure described above can be applied only when the gas temperature is measured by other methods and almost does not change [$T(t) \approx T_1$], for example, upon pumping of the gas by a dc discharge. However, in the case of pulsed pumping, when the gas temperature changes insignificantly, it was necessary to modify the procedure of gas-temperature recovery.

Because the gain g_{vJ} depends linearly on the populations N_v and N_{v+1} of vibrational levels, the matrix of the population $N_0(T)$ providing the minimum of function (1) is calculated by solving the system of linear equations (for different values of v):

$$\left. \frac{\partial F}{\partial N_v} \right|_T = 0, \quad \left. \frac{\partial F}{\partial N_{v+1}} \right|_T = 0. \quad (2)$$

The computer-aided solution of the system of equations (2) can be represented in the form of the subprogram function, and the search for the minimum of function (1) is reduced in this case to the determination of the minimum of the function of one variable $F[T, N_0(T_1)]$. To solve this problem, various descent methods were developed (we used the Levenberg–Marcar method [13]). The method of singular expansion, which was used in [4–6] to find $N_0(T_1)$, has a better stability to the perturbation of initial data but at the possible sacrifice of the large calculation error.

To eliminate the problems appearing upon minimisation of function (1), such as the absence or excess of solutions and also solutions with the negative values of temperature and population, it was proposed in [5] to limit the region of values of the required parameters (for example, $T > 0$, $N_v > N_{v+1} > 0$) and to find the constrained minimum of function (1). We used a different approach for processing our experimental data. Incorrect solutions appeared when the data were processed with a large relative error (for example, for small values of the SSG at the beginning and end of the amplification time interval). To find such solutions, we calculated several (from 15 to 25) values of temperature for random SSG values being simulated, which had the normal distribution near the average measured value $G_{vJ}(t)$ with the standard deviation equal to $\sigma_{vJ}(t)$. The

statistical analysis of the results of calculations with simulated initial data gave the average value of the required parameters and standard deviation, which characterised the sensitivity of the results of calculations to the perturbation of the initial data. The results of calculations for which the standard deviation exceeded the average deviation during the amplification time by a factor of five and more were discarded.

Depending on the parameters of a medium under study (pressure, density, and composition of the gas mixture), the method of multifrequency laser probing allows the recovery of either the absolute population N_v of vibrational levels or the relative population $n_v = N_v/N$ (N is the concentration of active molecules). When the Doppler broadening of spectral lines dominates in the active medium, the absolute population N_v is recovered. In the case of collision broadening, the linewidth changes almost proportionally to the concentration of gas particles, and therefore the relative population n_v is recovered, which is less sensitive than N_v to variations in the gas density, for example, upon gas-dynamic perturbations of the medium. Under our experimental conditions, the collision broadening of lines dominated, and therefore the relative population n_v of levels was recovered.

3. Experimental results

Figure 2 presents the dynamics of the gas temperature $T(t)$ for the CO : He = 1 : 4 and CO : N₂ = 1 : 9 laser mixtures for the average gas concentration in the laser amplifier $N_g = 0.12$ amagat. Here, amagat is the unit of the gas concentration equal numerically to the number of gas moles in the molar volume (22.4 L). The initial gas temperature T_g

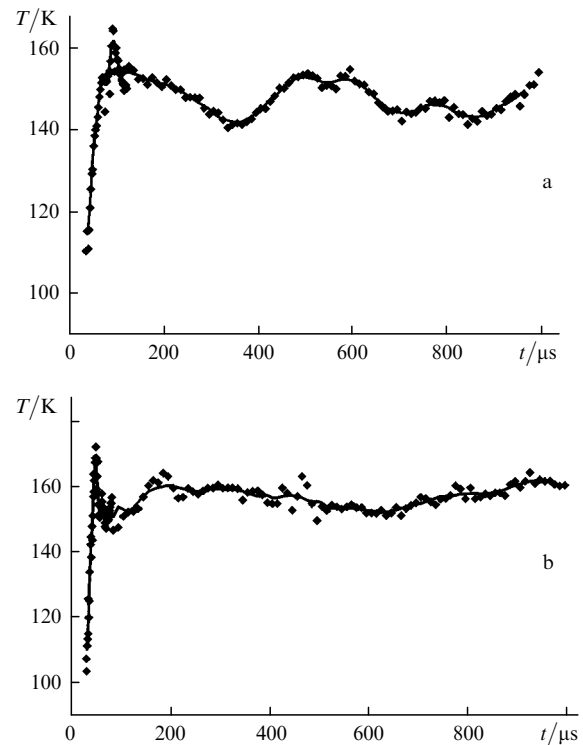


Figure 2. Temperature dynamics for the CO : He = 1 : 4 (a) and CO : N₂ = 1 : 9 (b) gas mixtures for the specific energy input $Q = 180 \text{ J L}^{-1} \text{ amagat}^{-1}$ and the average gas concentration $N_g = 0.12$ amagat.

measured with thermocouples in the region under study (without a discharge) was $\sim 108 \pm 3$ K for the helium mixture and $\sim 117 \pm 4$ K for the nitrogen mixture, the error of these two measurements being determined by the inhomogeneity of the gas temperature distribution in a cooled discharge chamber. The use of the method of multifrequency [$m = 10$, the $13 \rightarrow 12P$ ($J = 9, 13, 15$), $14 \rightarrow 13P$ ($J = 8, 10, 13, 14$), $15 \rightarrow 14P$ ($J = 10, 14, 16$) lines] laser probing provided the accuracy of measuring the laser medium temperature $\Delta T = 5$ K (root-mean-square error), which is comparable to the inhomogeneity of the distribution of the initial gas temperature T_g .

After the pump pulse of duration $40 \mu\text{s}$, the gas temperature increased during the first $100 \mu\text{s}$, but then it weakly changed for a long time (up to $1000 \mu\text{s}$). The gas temperature increased up to 148 ± 5 K for the helium mixture (Fig. 2a) and up to 157 ± 5 K for the nitrogen mixture (Fig. 2b), i.e. it increased by the same value 40 ± 9 K after the electroionisation discharge. The specific heat at a constant volume was $0.64 \text{ J L}^{-1} \text{ amagat}^{-1} \text{ K}^{-1}$ for the $\text{CO} : \text{He} = 1 : 4$ mixture and $0.94 \text{ J L}^{-1} \text{ amagat}^{-1} \text{ K}^{-1}$ for the $\text{CO} : \text{N}_2 = 1 : 9$ mixture. The local specific energy input (in the probed region) determined in [14] by comparing the experimental and calculated gain dynamics was $Q \approx 180 \text{ J L}^{-1} \text{ amagat}^{-1}$. For this specific energy input, the fraction of the discharge energy spent for the ‘fast’ heating of the gas was $14\% \pm 4\%$ for the $\text{CO} : \text{He} = 1 : 4$ mixture and $20\% \pm 5\%$ for the $\text{CO} : \text{N}_2 = 1 : 9$ mixture.

Figure 3a presents the dynamics of the relative population $n_v(t)$ of the vibrational level $v = 12$ for the $\text{CO}-\text{He}$ and $\text{CO}-\text{N}_2$ mixtures. The accuracy of recovering n_v during the amplification time was $8\% - 15\%$. For the same pump energy, the value of n_v in the nitrogen mixture exceeded on average n_v in the helium mixture by a factor of 2.4. Recall that we are dealing here only with the population of vibrational levels, while the amplification of radiation occurs in the case of a partial inversion at the vibrational-

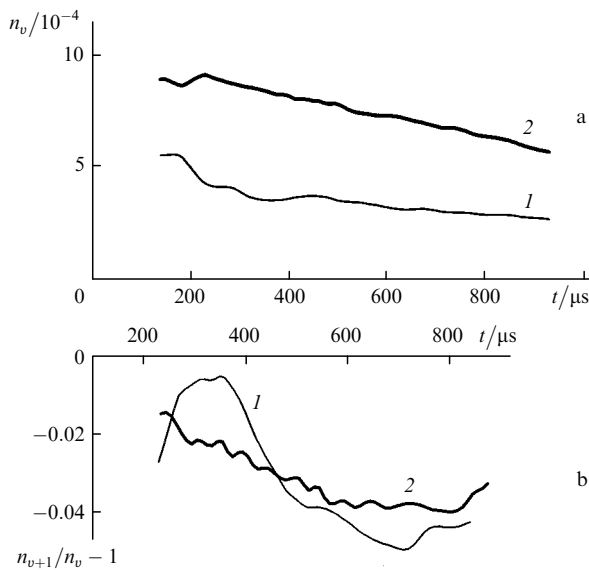


Figure 3. Dynamics of the relative population $n_v(t)$ of the $v = 12$ vibrational level (a) and the quantity $n_{v+1}/n_v - 1$ for $v = 12$ (b) in the $\text{CO} : \text{He} = 1 : 4$ (1) and $\text{CO} : \text{N}_2 = 1 : 9$ (2) gas mixtures. The rest of the parameters are as in Fig. 2.

rotational transition. The slope of the distribution function f_v , which can be estimated from the relation $n_{v+1}/n_v - 1 \approx (f_v)^{-1} df_v/dv$, plays an important role in this case. Variations in $n_{v+1}/n_v - 1$ with time are presented in Fig. 3b for both mixtures. Note that the VDF slope in the nitrogen mixture changes slower than in the helium mixture because a part of the vibrational energy stored in N_2 molecules is gradually transferred to CO molecules.

It was shown in [15, 16] that the addition of carbon oxide stabilises a discharge in gas mixtures containing great amounts of oxygen and provided high energy inputs. The increase of the energy input in such mixtures provides the conditions for excitation of singlet oxygen (O_2 in the $a^1\Delta_g$ state) with a high yield. In addition, lasing was obtained at the transitions of the CO molecule in such mixtures [16, 17]. The diagnostics of the gas temperature and population of vibrational levels of the molecule was performed by measuring the SSG dynamics in the $\text{CO} : \text{O}_2 = 1 : 19$ mixture for spectral lines within the $10 \rightarrow 9P$ band ($J = 7, 8, 10-16$). These lines ($m = 9$) were selected by using the weaker selection criterion [$\Delta v_{vJ} > 0.1(v_n/y_1) \text{ cm}^{-1}$] and neglecting the influence of two weak interfering absorption lines. The measurements were performed for a small local energy input $Q = 90 \text{ J L}^{-1} \text{ amagat}^{-1}$ at which gas-dynamic perturbations in the $\text{CO}-\text{O}_2$ mixture were weak. The gas temperature in the oxygen mixture (Fig. 4) increased by 45 ± 12 K (the initial gas temperature was $T_g \approx 120$ K). For comparison, Fig. 4 demonstrated the dynamics of the gas temperature in the $\text{CO}-\text{He} = 1 : 4$ mixture, in which temperature increased approximately by 10 K ($T_g \approx 110$ K). Thus, the EBSD in the oxygen mixture is characterised by a stronger heating of the active medium than in the helium mixture. A part of the energy spent to direct heating of the gas in the EBSD was $35\% \pm 10\%$ for the oxygen mixture with the 5% addition of carbon monoxide, which is close enough to the results obtained in [18] for pure oxygen ($40\% \pm 10\%$). A change in the gas concentration and temperature during its expansion to a ballast volume after the discharge was manifested in the decrease in the gas temperature for the oxygen mixture (Fig. 4) in the interval from 200 to $500 \mu\text{s}$.

It is known (see, for example, [19–21]) that the addition of a small amount of oxygen to the active medium of a self-sustained discharge-pumped low-pressure CO laser improves the output parameters of a flowing-gas CO laser

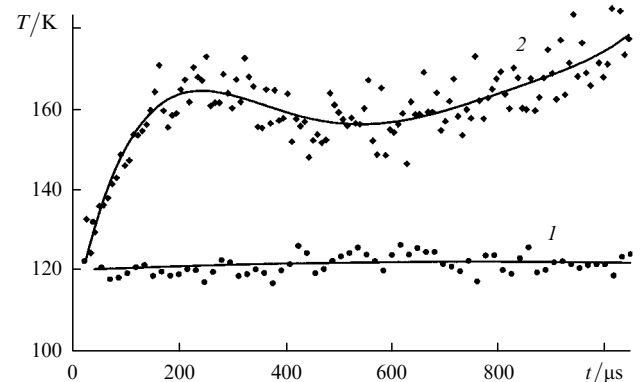


Figure 4. Temperature dynamics for the $\text{CO} : \text{He} = 1 : 4$ ($N_g = 0.12 \text{ amagat}$) (1) and $\text{CO} : \text{O}_2 = 1 : 19$ ($N_g = 0.06 \text{ amagat}$) (2) mixtures and $Q = 90 \text{ J L}^{-1} \text{ amagat}^{-1}$.

(unlike a sealed laser operating without the gas-mixture replacement; see [22]). It was demonstrated in [23] that the population of vibrational levels of the CO molecule in the case of a self-sustained discharge can increase by 5–10 times after the addition of oxygen to the CO : He = 1 : 4 gas mixture in the amount $[O_2] = 0.25[CO]$ determined by the ratio of concentrations of O_2 and CO molecules. The analysis of our experimental results obtained in a pulsed nonself-sustained discharge showed that the relative population $n_v(t)$ of vibrational levels (Fig. 5a) in the CO : $O_2 = 1 : 19$ mixture also was higher than in the CO : He = 1 : 4 mixture. In the average the relative population of the $v = 9$ vibrational level in the oxygen mixture during amplification exceeded the value of n_v in the gas mixture by a factor of fifteen. The weakening of the line selection criterion did not increase the recovery error of the population of vibrational levels. Moreover, some increase in the accuracy ($\delta n_v = 7\% - 12\%$) is caused by a decrease in the number of unknown quantities because probed lines belong to one vibrational transition rather than to three. Figure 5b shows the dynamics of the ratio $n_{v+1}/n_v - 1$ for two mixtures. The slope of the VDF in the oxygen mixture is twice as large as that in the helium mixture, which is probably explained by vibrational energy transfer from CO to O_2 molecules during the intermolecular vibrational energy exchange $CO(v) + O_2(0) \rightarrow CO(v-1) + O_2(1)$. As a result, CO molecules undergo the $v \rightarrow v-1$ transition and are accumulated on the vibrational levels located below the levels involved in the intermolecular exchange {in [17], radiation was not amplified but absorbed for $v > 14$, $G_{vJ}(t) \leq 0$ }.

The increase in the initial temperature from 110–120 K up to room temperature ($T_g \approx 290$ K) resulted in the decrease in the amplification of laser radiation in the active medium, which then changed to absorption (Fig. 6a). Figure 6b shows the dynamics of the gas temperature in the CO : $O_2 = 1 : 19$ mixture recovered from the measurements

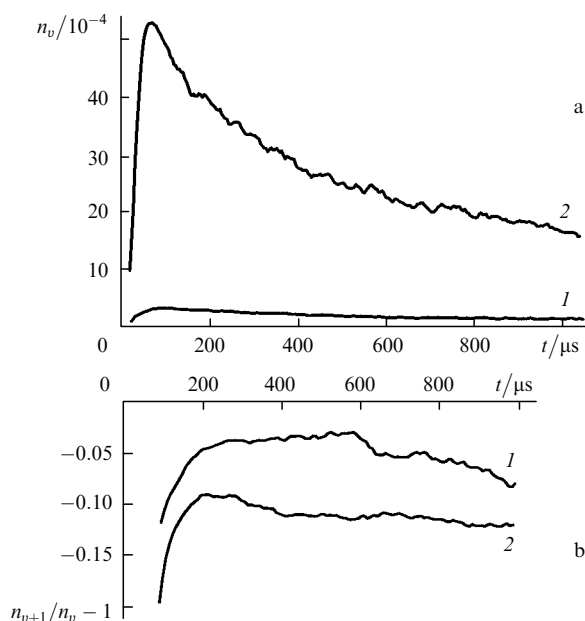


Figure 5. Dynamics of the relative population $n_v(t)$ of the $v = 9$ vibrational level (a) and of the quantity $n_{v+1}/n_v - 1$ for $v = 9$ (b) in the CO : He = 1 : 4 (1) and CO : $O_2 = 1 : 19$ (2) gas mixtures. The rest of the parameters are as in Fig. 4.

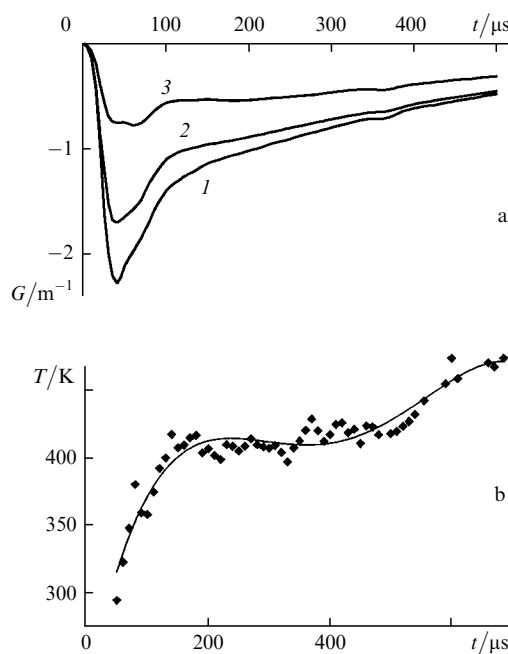


Figure 6. Absorption of the probe radiation at the transitions in the $9 \rightarrow 8P$ band for $J = 9$ (1), 11 (2), and 15 (3) and the temperature dynamics of the CO : $O_2 = 1 : 19$ gas mixture for $P_0 = 30$ Torr ($N_g \approx 0.04$ amagat), $T_0 \approx 290$ K, and $Q = 250$ J L $^{-1}$ amagat $^{-1}$ (b).

of absorption of radiation from a probe CO laser at ten spectral lines $9 \rightarrow 8P$ ($J = 9, 11, 15$); $10 \rightarrow 9P$ ($J = 10, 11, 13, 15$) and $11 \rightarrow 10P$ ($J = 10, 12, 16$). For the energy input $Q = 250$ J L $^{-1}$ amagat $^{-1}$, the gas temperature increased from the initial value $T_g \approx 290$ K up to 415 ± 20 K for the first 200 μs . Then, the gas temperature changed weakly during $\sim 500 \mu s$ probably due to the gas expansion upon heating. Thereafter the gas temperature continued to increase due to relaxation of the vibrational energy stored in molecules.

Absorption measurements revealed the systematic error caused by the oxidation of carbon oxide molecules ($O_2 + e \rightarrow O + O^-$, $CO + O^- \rightarrow CO_2 + e$). These reactions resulted in the increase in the amount of carbon dioxide molecules in the gas mixture. The concentration of CO_2 molecules was measured with an AF-3 Fourier spectrometer (Scientific and Technical Centre of Unique Instrument Making, RAS, Moscow). A gas cell was filled with the gas mixture under study, and absorption coefficients were measured in the wavelength range from 4.2 to 4.4 μm . After thirty discharge pulses, the partial pressure of CO_2 achieved 0.2 Torr for the initial pressure of carbon oxide equal to 1.5 Torr. The increase in the amount of CO_2 molecules in the CO – O_2 gas mixture resulted in a change in the absorption dynamics $G_{vJ}(t)$ due to the increase in the relaxation rate of vibrational energy. To eliminate this systematic error, we analysed variations in $G_{vJ}(t)$ depending on the discharge pulse number and found the correction function. Then, the results of measurements were corrected to reduce all absorption data to the first (after the gas admission) discharge pulse. The statistical processing and recovery of the gas temperature were performed by using these corrected data (Fig. 6a).

4. Conclusions

Our study has shown that the method of multifrequency laser probing can be used for measuring the dynamics of the gas temperature and population of vibrational levels in molecular gases containing carbon oxide. The spectral lines corresponding to the vibrational–rotational transitions of the CO molecule within overlapped vibrational bands are located so closely to each other that the problem of the choice and identification of probed transitions appears. This problem was solved by analysing the mutual location of the spectral lines of the CO molecule. The lines were selected by using a strict selection criterion taking into account the spectral resolution of a spectrometer used for measuring the probe radiation frequency. As a result, each of the selected lines was reliably identified. In addition, the data processing procedure was modified for the conditions of pulse pumping of the gas. The method developed in the paper was tested by studying the dynamics of the gas temperature and population of vibrational levels in CO – He, CO – N₂, and CO – O₂ gas mixtures pumped in the plasma of a pulsed EBSD. The relative error of the gas temperature recovery was 3 % – 5 %. The analysis of the results of this study has shown that the relative population of the $\nu = 9$ vibrational level of the CO molecule in the CO : O₂ = 1 : 19 gas mixture is higher approximately by a factor of fifteen than in the CO : He = 1 : 4 mixture probably due to the intermolecular vibrational exchange between carbon monoxide and oxygen molecules. The method developed in the paper can be used for studying thermodynamic and kinetic processes in gas mixtures containing CO molecules in excited vibrational states.

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