

## Diagnostics of Low Temperature Plasma by CO Laser Radiation (5.0-7.5 $\mu\text{m}$ )

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**Abstract.** A review of the work conducted in Gas Lasers Laboratory of P.N. Lebedev Physical Institute of the Russian Academy of Sciences and devoted to a research of the diagnostic of dynamics of small-signal gain, temperature and population of vibrational levels in the CO-containing gas mixtures excited by pulsed electron-beam sustained discharge is presented. With the help of laser probing techniques the nonlinear Zeeman splitting of the NO lines in a strong magnetic field was also investigated. The studies of gain of the train of nanosecond pulses allowed us to estimate the intensity of the gain saturation of the active medium of the pulsed electron-beam sustained discharge CO laser.

### 1. Introduction

A significant step forward in the study of kinetic processes occurring in the active medium of electric-discharge CO lasers is the transition from the comparison of the calculated and measured lasing properties of these lasers to a direct comparison of the gain of the active medium, and other parameters of low-temperature plasma. This considerably simplifies the theoretical modeling and reduces the number of parameters measured in the experiment, since it removes the need for modeling the processes of light generation. In addition, the characteristic time of the formation of the population inversion and, consequently, the small-signal gain (SSG), is very important for fast-flow CO lasers with both transverse and longitudinal circulation of the active medium because they have to be considered when developing the design of lasers. Thus, there is a need to study the temporal dynamics of the gain in the active medium of a CO laser under various experimental conditions, in particular, typical for the CO lasers with rapid circulation of the active medium. Also, most important among these conditions (specific input energy and composition of the gas mixture) can be simulated by a pulsed electron-beam sustained discharge (EBS) CO laser system with stationary cryogenic cooling of the active medium.

In the CO laser the dependence of the SSG of the temperature plays a particularly important role, however, it is very difficult to measure the temperature of the gas directly in the field of electrical discharge. In the paper [1] aimed at measuring the dynamics of the temperature of the active medium

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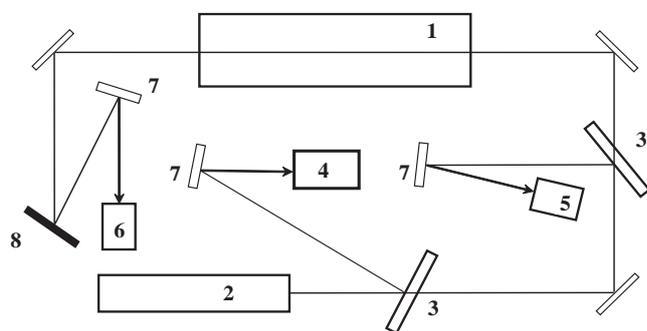
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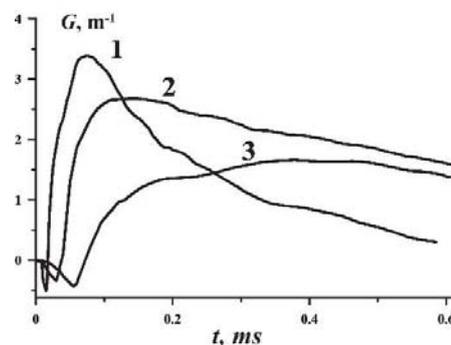
under conditions of electric discharge it was used the fact that the rotational temperature of excited states of the CO molecules became equal to the translational temperature during the time less than 1 ns. The dynamics of the rotational temperature of the gas was evaluated from the dynamics of the SSG for three rotational transitions within a single vibrational band, which was measured using the continuous frequency-selective CO laser as a probe laser. However, the implementation of such a technique with ten percent accuracy of measuring the SSG results in a high error ( $\sim 200$  K) in determining the gas temperature [2]. To increase the accuracy it is necessary, as it follows from [2], to increase the number of probed transitions by several times, i.e. develop a method for the multifrequency probing of active medium of the pulsed CO laser.

## 2. Multifrequency laser probing of the active medium of the CO laser

In [3, 4] a method of multifrequency laser probing was developed, which was used for the diagnosis of the SSG, temperature and population of vibrational levels in the CO-containing gas mixtures excited by a pulsed EBSD. Optical measuring scheme is shown in figure 1. The dynamics of the SSG at the vibrational-rotational transitions  $V+1 \rightarrow V P(J)$  of the CO molecule in various mixtures for various specific input energy  $Q_{in}$  was experimentally studied [3]. Figure 2 shows the dynamics of small-signal gain in the mixture  $\text{CO:N}_2 = 1:9$  for three transitions. It demonstrates that an increase in the vibrational number  $V$  the maximum gain  $G_{max}$  is reduced, and it is achieved at a later time. The maximum gain  $G_{max}$  is reduced threefold for  $V=31$  in comparison with value  $G_{max}$  for  $V=7$ . It was found that at a fixed value of  $V$  the rate of the rise of the SSG is increased with increasing the rotational number.



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

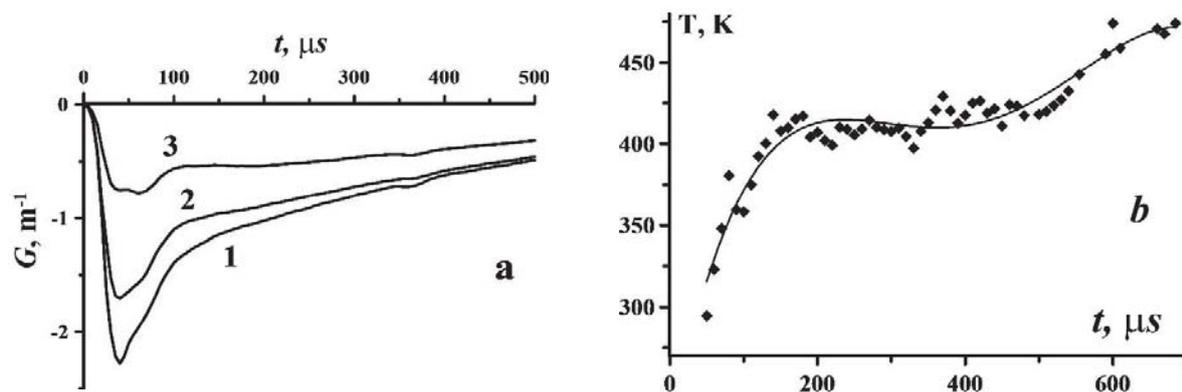


**Figure 2.** The SSG dynamics in the  $\text{CO:N}_2=1:9$  mixture at  $8 \rightarrow 7 P(10)$  (1),  $14 \rightarrow 13 P(10)$  (2) and  $21 \rightarrow 20 P(11)$  (3) transitions for  $Q_{in} = 800 \text{ J L}^{-1} \text{ Amagat}^{-1}$ .

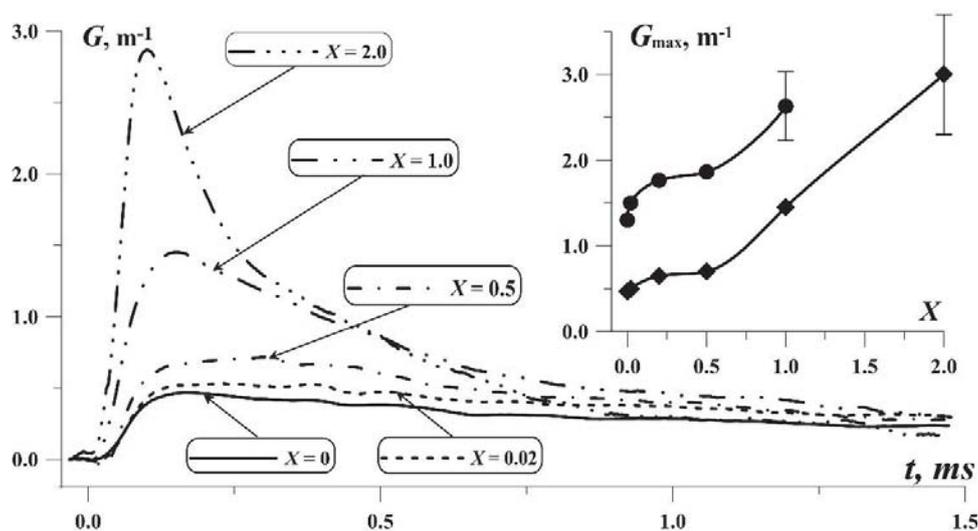
It was shown that for the oxygen-containing gas mixtures ( $\text{CO:O}_2 = 1:19$ ) the value  $G_{max}$  at lower vibrational transitions (for  $V < 13$ ) can be significantly greater than  $G_{max}$  for the mixture in which the nitrogen is used instead of oxygen ( $\text{CO:N}_2 = 1:19$ ). It has been found that the efficiency of the CO laser operating on a mixture with higher oxygen content is increased up to 47% in comparison with the efficiency of the CO laser operating on the nitrogen-containing gas mixture (where it equals to 30%).

It has been demonstrated that the method can reduce the error in the determination of the gas temperature up to 3% [4]. It was found that in the mixture of  $\text{CO:O}_2$  the population of lower vibrational

levels of the CO molecule can be several times higher than the population of respective levels in the laser mixtures CO:He and CO:N<sub>2</sub>. However, the temperature rise in the oxygen-containing mixtures is significantly higher than that in helium and nitrogen mixtures. Figure 3a shows the time dependence of the absorption for three transitions of the CO molecule, on the basis of which the temperature dynamics has been determined (figure 3b).



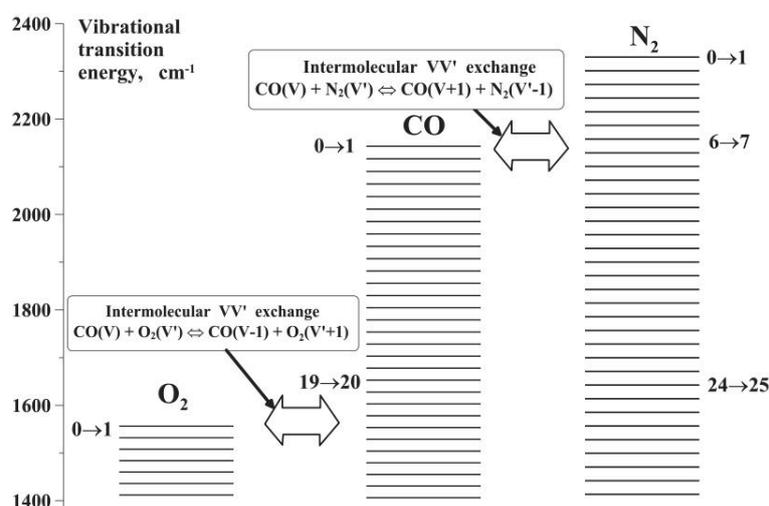
**Figure 3.** Absorption coefficient of the probe radiation (a) at the transitions in the 9→8 P band for  $J=9$  (1), 11 (2), and 15 (3), and the temperature dynamics (b) of the CO:O<sub>2</sub>=1:19 gas mixture for pressure 30 Torr, initial temperature 290 K, and  $Q_{\text{in}} = 250 \text{ J L}^{-1} \text{ Amagat}^{-1}$ .



**Figure 4.** The SSG dynamics in the CO:He:O<sub>2</sub>=1:4:X mixture at the 10→9 P(15) transition for five values of  $X$  ( $Q_{\text{in}} = 130 \text{ J L}^{-1} \text{ Amagat}^{-1}$ ). The inset shows the dependence of  $G_{\text{max}}$  on  $X$  for  $Q_{\text{in}} = 240$  (●) and  $130 \text{ J L}^{-1} \text{ Amagat}^{-1}$  (◆). The measurement error is within the marker size, except the shown error bars.

Further research showed [5] that the addition of molecular oxygen to the working mixture of a pulsed EBSD CO laser amplifier multiplies the SSG of the active medium in the vibrational transitions of the CO molecule for  $V$  from 6 to 13 (figure 4), which is associated with an increase of the population of the vibrational levels. However, the lifetime of the gain decreases for all the probed transitions.

Figure 5 shows the vibrational energies of transitions of  $O_2$ , CO and  $N_2$  molecules. This figure demonstrates the essential role of kinetic processes of intermolecular vibrational-vibrational (VV') exchange for the excitation of the CO molecules. It was found that the addition of oxygen increases the efficiency of the CO laser at the fundamental transitions, and the efficiency reaches its maximum value at lower values of specific input energy. It has been shown that the pulsed EBSD CO laser can operate on the air working mixture both at the fundamental transitions and the first overtone vibrational transitions  $V+2 \rightarrow V$ .



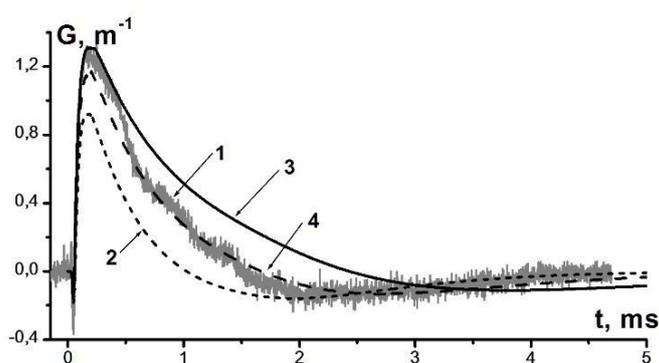
**Figure 5.** Vibrational transition energies of  $O_2$ , CO, and  $N_2$  molecules.

In the paper [6], we researched the influence of small additions of  $O_2$  on the dynamics of small-signal gain of the active medium of a pulsed EBSD CO laser at cryogenic temperature. Using a numerical model of the laser active medium has enabled us to find out how the VV'-exchange between vibrationally excited molecules at high levels of CO and oxygen molecules affects the dynamics of small-signal gain. Methodology based on a comparison of the measured and calculated dynamics of small-signal gain for a number of transitions (figure 6) enabled us to determine the parameters of the analytical approximation of the rate constants of the VV'-exchange for the pair of molecules CO- $O_2$  at cryogenic temperature of the active medium.

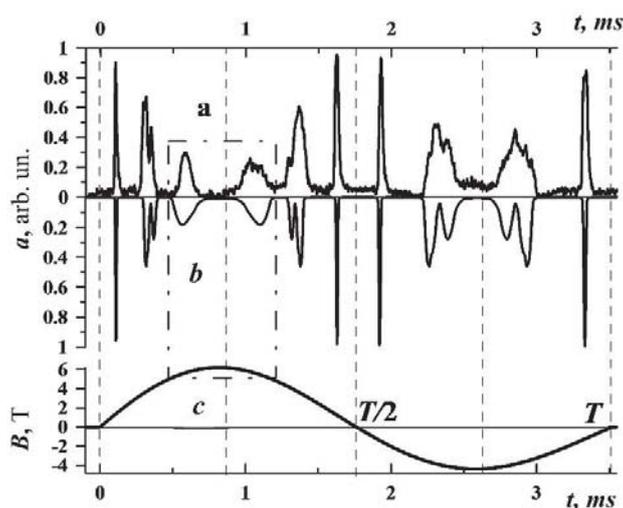
### 3. The research of the Zeeman splitting of the IR absorption lines of the NO molecule

In the paper [7], the dependence of the Zeeman splitting of the vibrational-rotational lines of the NO molecules on the value of a strong magnetic field with magnetic induction up to 6 Tesla was theoretically and experimentally studied. Measurement of absorption in a pulsed magnetic field was also carried out using a CW frequency-tunable CO laser operated in a single line selective mode. To analyze the dynamics of absorption, a numerical model of the nonlinear Zeeman splitting of the NO lines has been developed. The NO molecules absorption dynamics in a pulsed magnetic field was described by the calculation model

in a cubic approximation of the value of magnetic induction. A non-linearity of the Zeeman splitting lines of the Q- and P-branches of the vibrational-rotational transitions of the NO molecules in the  ${}^2\Pi_{3/2}$  state was found. Particularly, for the lines Q(2.5), P(5.5) and P(6.5) the non-linearity of the Zeeman components of the lines was identified for the magnetic induction of  $\sim 4$  T. Comparison of the calculated and measured absorption dynamics of the probe radiation showed that the calculation model reproduces well the position of the measured absorption peaks depending on the induction of a pulsed magnetic field (figure 7), that allowed, thus, to identify the Zeeman components of the absorption lines of the NO molecules.



**Figure 6.** The SSG dynamics at the  $18 \rightarrow 17$  P(15) transition for  $Q_{in} = 240 \text{ J L}^{-1} \text{ Amagat}^{-1}$  in the gas mixture  $\text{CO}:\text{He}:\text{O}_2=1:4:0.05$ . Experimental (1) and calculated values (2-4) are shown for various values of the  $VV'$  exchange rate constant.

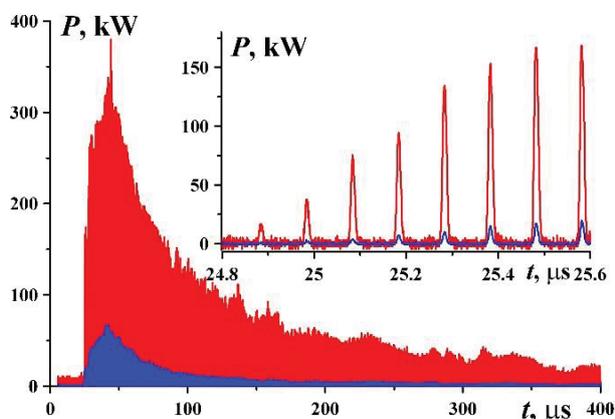


**Figure 7.** Experimental (a) and calculated (b) absorption dynamics  $a(t)$  for the NO molecules for the CO laser line  $9 \rightarrow 8$  P(15) in a damped oscillating magnetic field (c). Time instant corresponding to the value of  $B$  as high as 5 T is marked by the dash-dot lines.

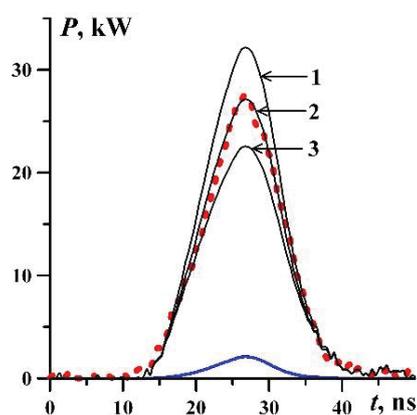
#### 4. The research of the CO laser nanosecond pulses amplification

The works [8, 9] were dedicated to a study of the active mode-locking of the EBSD CO laser, creating a master oscillator and laser amplifier system emitting a train of nanosecond pulses, as well as to a research of nanosecond pulse train amplification in the active medium of the CO laser amplifier. The duration of the spikes was  $\sim 10$  ns, with the minimum value of the duration, achieved in our experiments, equal to  $\sim 5$  ns. We got the dependence of the output laser energy of the amplifier from the input laser energy delivered to the amplifier for various values of the specific input energy delivered to the EBSD and of the relative density of the working gas mixture, which had a clearly non-linear character. This fact indicates the saturation of amplification of the active medium.

Maximum peak power  $P_{\text{peak}}$  of the radiation amplified in one pass for the CO : He = 1 : 4 mixture was 380 kW (figure 8) for the 3.2 J total energy of a train of nanosecond pulses at the output of the laser amplifier in the nonselective mode and 100 kW for, respectively, 1.1 J in the frequency-selective mode. At the same time the efficiency of the “master oscillator-laser amplifier” system compared to the pure laser mode of forming a train of nanosecond pulses increased by a factor of 2 in the selective mode, reaching 1.6%, and, respectively, by a factor of 1.5 for the non-selective mode, amounting to 5.3% [9]. To estimate the saturation parameter, the rotational relaxation time of the CO molecules was calculated for our conditions, and amounted to 1.7 ns that is significantly shorter than the duration of the radiation spikes. By comparing the experimental and calculated dynamics of the radiation spikes at the output of the laser amplifier the value of the saturation intensity of nanosecond pulses amplification was found in the active medium, amounting  $I_s = 14 \pm 4 \text{ kW/cm}^2$  for the vibrational-rotational transition of the CO molecule 9-8 P (11) at  $Q_{\text{in}} = 210 \text{ J/(L Amagat)}$ .



**Figure 8.** The power of a train of nanosecond pulses at the input (blue) and the output (red) of laser amplifier for nonselective mode ( $\sim 10$  spectral lines in the range from 5.1 to 5.4 microns). The inset shows the beginning of the train of nanosecond pulses, illustrating the shape and repetition frequency of the spikes.



**Figure 9.** Experimental data for dynamics of the radiation spike at the input (blue) and output (red) of the amplifier at  $t = 37 \mu\text{s}$ , and the calculation results (black curves) for the values of gain saturation intensity  $I_s = 20 \text{ kW cm}^{-2}$  (1),  $I_s = 15 \text{ kW cm}^{-2}$  (2), and  $I_s = 10 \text{ kW cm}^{-2}$  (3).

## 5. Conclusion

Developed and applied in the Gas Lasers Laboratory of P.N. Lebedev Physical Institute of the Russian Academy of Sciences, a method of multifrequency laser probing enabled us to hold a series of studies on the diagnosis of the dynamics of the small-signal gain, temperature and population of vibrational levels in the CO-containing gas mixtures excited by a pulsed electron-beam sustained discharge. With its help we also studied the non-linear Zeeman splitting of the NO lines in a strong magnetic field. The researches of amplification of a train of nanosecond pulses enabled us to estimate the intensity of the gain saturation in the active medium of a pulsed electron-beam sustained discharge CO laser.

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## References

- [1] Basov N G, Kazakevich V S, Kovsh I B and Mikryukov A N 1983 *Soviet Journal of Quantum Electronics* **13** 667
- [2] Vyazovetskii N M, Didyukov A I, Kirko V Yu, Kulagin Yu A and Shelepin L A 1990 *J. Appl. Spectr.* **52** 444
- [3] Vetoshkin S V, Ionin A A, Klimachev Yu M, Kozlov A Yu, Kotkov A A, Rulev O A, Seleznev L V and Sinitsyn D V 2007 *Quantum Electronics* **37** 111
- [4] Ionin A A, Klimachev Yu M, Kozlov A Yu, Kotkov A A, Rulev O A, Seleznev L V and Sinitsyn D V 2007 *Quantum Electronics* **37** 231
- [5] Ionin A A, Klimachev Yu M, Kozlov A Yu, Kotkov A A and Seleznev L V 2008 *Quantum Electronics* **38** 115
- [6] Ionin A A, Klimachev Yu M, Kozlov A Yu, Kotkov A A, Kurnosov A K, Napartovich A P and Shnyrev S L 2008 *Quantum Electronics* **38** 833
- [7] Ionin A A, Klimachev Yu M, Kozlov A Yu and Kotkov A A 2011 *J. Phys. B: At. Mol. Opt. Phys.* **44** 025403
- [8] Ionin A, Kinyaevskiy I, Klimachev Yu, Kotkov A and Kozlov A 2012 *SPIE Newsroom*, doi: 10.1117/2.1201112.004016
- [9] Ionin A A, Kinyaevskiy I O, Klimachev Yu M, Kozlov A Yu and Kotkov A A 2012 *Optics Communications* **285** 270