INTERACTION OF LASER RADIATION WITH MATTER

Effect of stimulated emission on the distribution of CO molecules over vibrational levels

G.M. Grigorian, I.V. Kochetov

Abstract. **The vibrational distribution function (VDF) of CO molecules is measured in the presence and absence of generation in a CO laser pumped by a longitudinal dc discharge. Kinetic equations for the VDF and the Boltzmann equation for the energy distribution function of electrons are solved simultaneously in a theoretical model. A comparison of the experimental and calculated lasing spectra and VDF demonstrates their good agreement. By introducing an absorbing cell with different gases (NO, C_2H_4, CO_2, C_6H_6) into a resonator, the influence of selection of laser lines on the lasing spectrum and the VDF of CO molecules is studied. It is shown experimentally that the population of CO molecules at vibrational levels involved in lasing and at higher levels strongly decreases and the VDF at lower levels changes insignificantly. It is demonstrated that the VDF shape of CO molecules at high vibrational levels can be changed by introducing intracavity absorption.**

Keywords: *CO laser, vibrational distribution function, absorbing cell, line selection.*

1. Introduction

Experimental studies of a CO laser show that generation at vibrational–rotational transitions leads to the decrease in the population of excited electronic levels [1] and to the increase in the reduced electric field strength E/N (N is the density of neutral particles) at which a discharge is supported [2, 3]. This suggests that vibrationally excited CO molecules, whose concentration decreases during lasing, play a noticeable role in the balance of electronically excited and charged particles. Chemical reactions involving vibrationally excited CO molecules can occur [4]. In this connection the control of the vibrational distribution function (VDF) of CO molecules by varying the spectrum and intensity of laser emission is of certain interest. Such control of the VDF shape can be useful for studying elementary processes involving CO molecules in highly excited vibrational states.

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We know only three papers in which the VDF of CO molecules was measured during lasing. These are our papers [2, 3, 5], in which the VDF of CO molecules was measured in a longitudinal dc discharge cw CO laser upon cryogenic cooling of discharge tube walls. In addition, the VDF of CO molecules was measured in the supersonic jet of a gas mixture in the region located below the resonator down the gas stream. The size of the resonator was smaller than the height of the discharge chamber [6], and therefore the detected signal of the first overtone was the sum of signals from the gas parts that have and have not propagated through the lasing region, which introduced distortions to the shape of the VDF overtone signal reconstructed from the spectrum.

The selection of lines in the emission spectrum of a CO laser by the method of intracavity absorption was proposed in papers [7–12]. The authors of these papers studied the possibility of improving the propagation of radiation from a CO laser in the atmosphere by suppressing lasing at transition lines that are strongly absorbed by water vapour in the atmosphere. When a cell with water vapour was placed inside the resonator, lasing at lines absorbed by water vapour in the atmosphere was quenched, whereas the intensity of laser lines falling to transparency windows increased. The dependence of the cw CO laser efficiency on the rarefaction degree of the spectrum and spectral positions of the separated lines was theoretically studied in [13]. It was shown that the selection of individual lines weakly affected the output power of the CO laser. The output power in one line can achieve 50 % of the output power produced in the nonselective resonator. The dependence of the CO laser efficiency at overtones or fundamental transitions on the lasing spectrum width was theoretically studied in [14].

The control of the lasing spectrum in all previous experimental studies was performed by using a cell with water vapour only. It is of interest to investigate the control of the lasing spectrum by the method of intracavity absorption by using other gases.

The aim of this paper is to study experimentally and theoretically the VDF of CO molecules in the active medium and spectral emission characteristics of an electric-discharge CO laser with a gas mixture slowly circulating through the liquid nitrogen-cooled discharge tube or without circulation with cooling the discharge tube by flowing water in the free running regime, as well as with the use of intracavity absorption of radiation by NO, CO_2, C_2H_4, and C_6H_6 molecules.

2. Experimental

Measurements were performed in molybdenum glass discharge tubes, which were cooled by running water or liquid nitrogen. Tubes of diameter $D = 1.5$ or 2.0 cm and length $L = 50 - 60$ cm were used. Radiation was coupled out through calcium fluoride crystal windows oriented at the Brewster angle. Hollow cylindrical tantalum electrodes were located in side arms at a distance of 4.5 cm from the discharge tube axis. The active medium was excited by a dc glow discharge. A planospherical resonator was formed by a highly reflecting gold-plated mirror with the radius of curvature 5 m and a flat mirror with a hole of diameter 0.2 or 0.3 cm at the centre to couple out radiation. The resonator length was ~ 100 cm. As working gases, we used the $He - CO - O_2$ mixture (in the convection regime with liquid nitrogen cooling) and the $He - CO - Xe$ mixture (in the sealed off regime with running water cooling). The content of CO in working mixtures (He–CO) was varied from 5 % to 15 %. In the convection regime, oxygen was added to the mixture (the oxygen concentration was 1 % – 5 % of the CO concentration). In the sealed off regime, mixtures were used in which the concentration of Xe was close to that of CO. The discharge current I_d was varied from 20 to 80 mA and the gas pressure P was $5 - 20$ Torr.

The VDF of CO molecules was determined by measuring the spontaneous emission intensity at the fundamental $v \to v - 1$ transition (in the absence of lasing) and at the first $v \to v - 2$ and second $v \to v - 3$ overtones of vibrational transitions in the ground electronic state of CO molecules. Signals were recorded by using lock-in detection with the help of a liquid nitrogen-cooled HgCdTl photoresistor (with the sensitivity $\sim 10^5$ V W^{-1}) and InSb photoresistors without cooling.

The VDF of CO molecules was reconstructed from the intensity of overtones by the method similar to that described in [15–17]. The translational temperature of gas was measured by recording the Angström spectral bands of CO molecules with the unresolved rotational structure. Radiation was detected with a photomultiplier; the stray laser radiation was suppressed by means of a quartz filter mounted in front of the entrance slit of a monochromator.

The influence of intracavity absorption on the lasing spectrum was studied by placing an absorbing cell in the form of a glass tube of internal diameter 2.6 cm and length 35 cm into the resonator. The cell had a cooling jacket through which water circulated from a liquid thermostat. The temperature of the tube walls was controlled by the temperature of circulating water and measured with copper-constantan thermocouples. The CaF$_2$ windows at the cell ends were mounted at the Brewster angle. The gas parameters in the cell were changed by igniting a high-frequency induction discharge. In this case, the cell was placed inside a solenoid through which a high-frequency (~ 2 MHz) current was passed.

3. Theoretical model

We used a homogeneous model of the axial region of a discharge tube. The reduced electric field E/N and the translational gas temperature were set equal to their experimental values. The discharge power density on the axis was determined from the experimental discharge current by assuming that the electron concentration along the tube radius has the Bessel profile. The theoretical model used for calculating the VDF of CO molecules was described in our paper [18].

The intensity of vibrational–rotational emission transitions, at which the small-signal gain exceeded resonator losses, was calculated by using the condition of the equality of the gain to resonator losses at the wavelength of the corresponding transition [19]. In this case, lasing in calculations could occur only at one rotational transition corresponding to the maximum of the gain for the given vibrational transition.

4. Results

Figure 1 presents the VDFs of CO molecules measured in the amplification and lasing regimes for the $He - CO - Xe$ mixture in a sealed off CO laser with water-cooled discharge tube walls. One can see that the population of high vibrational levels ($v > 10$) noticeably decreases during lasing, whereas the population of lower vibrational levels does not change. After the appearance of lasing, the discharge current was maintained constant, the reduced electric field increased by 3 % – 5 %, and the gas temperature at the discharge tube axis slightly decreased (by ~ 10 K). The lasing efficiency was 13 %. The curves in Fig. 1 show calculated VDFs. One can see that experimental and theoretical VDFs are in good agreement in the amplification regime [curve (1)]. If lasing occurs at the fundamental mode and resonator losses are determined by the ratio of the area of the hole in the output mirror to the active medium area, the calculated populations of vibrational levels prove to be noticeably higher than experimental populations [curve (2)]. Such an estimate of resonator losses gives $\Gamma = 1.7 \times 10^{-4}$ cm^{-1}. Good agreement between calculations and experimental data was observed when losses were decreased by 4–5 times (down to 4×10^{-5} cm^{-1}) [curve (3)]. The size of the
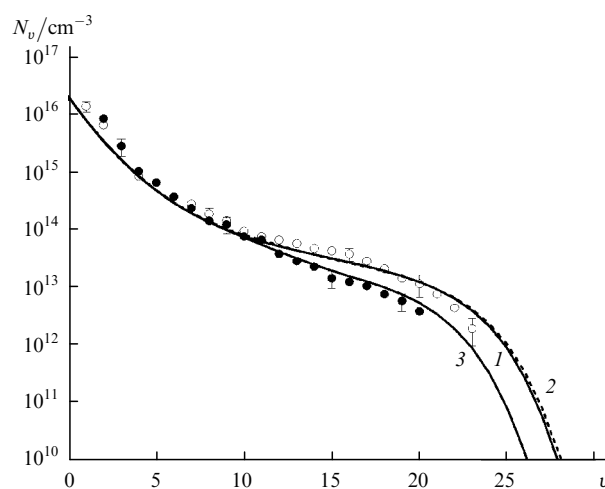

Figure 1. Vibrational distribution functions of CO molecules measured in the amplification (○) and lasing (●) regimes and calculated in the amplification regime (1) at $T = 450$ K and lasing regime at $T = 443$ K and resonator losses 1.7×10^{-4} (2) and 4×10^{-5} cm^{-1} (3); He : CO : Xe = 80 : 11 : 9, $P = 16$ Torr, $I_d = 30$ mA, $E/N = 27$ Td, $D = 1.5$ cm, $d = 0.2$ cm, and $L = 50$ cm.

fundamental mode on the mirror was estimated as 0.2 cm, which corresponds to the diameter of the output hole. Losses at the fundamental mode are large, and lasing will occur predominantly at high resonator modes. This is confirmed by the far-field radial intensity distribution of the laser beam observed in experiments. The estimate of losses in the resonator with a hole during lasing at overtones is complicated. Note that resonator losses caused by the misalignment of output windows are estimated as 2.4×10^{-5} cm^{-1} (the accuracy of alignment of resonator windows was assumed $\sim 1°$, which gives reflection from each of these windows of 0.03 %). This value does not contradict to total losses of 4×10^{-5} cm^{-1}.

Lasing occurred in experiments simultaneously at several $(3-5)$ rotational transitions of one vibrational transition. Therefore, the lasing intensity at an individual vibrational transition was determined as the sum of intensities for all rotational transitions corresponding to this vibrational transition. Figure 2 shows the experimental and calculated emission spectra of the laser under conditions corresponding to Fig. 1. One can see that, as the VDF, the experimental lasing spectrum well agrees with the spectrum calculated for resonator losses $\Gamma = 4 \times 10^{-5}$ cm^{-1}.
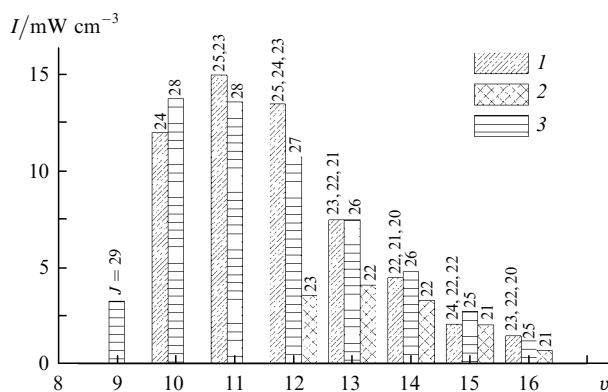

Figure 2. Experimental (*1*) and calculated emission spectra of the CO laser (the $v \rightarrow v-1$ transitions) for resonator losses 1.7×10^{-4} (*2*) and 4×10^{-5} cm^{-1} (*3*) (rotational quantum numbers J for the $J-1 \rightarrow J$ transitions on the P branch at which lasing was observed are indicated over the total intensity of the vibrational transition). Other parameters are as in Fig. 1.

When the left boundary of the lasing spectrum is shifted to higher vibrational levels, the intensity of lasing at higher vibrational transitions should increase. We studied the possibility of the influence of this process on the spectral parameters of the laser by using the intracavity suppression of lasing at the short-wavelength edge of the emission spectrum. Due to anharmonicity, the resonance between $CO(v \rightarrow v-1)$ and $NO(0 \rightarrow 1)$ is observed under these conditions in the beginning of a plateau of the VDF of CO molecules because the energy of the first vibrational quantum for NO molecules is somewhat smaller than for CO molecules.

The influence of intracavity absorption by NO molecules on the lasing spectrum was studied by placing a cell with NO or the He–NO mixture into the CO laser resonator (at a pressure of ~ 200 Torr).

Figure 3 shows the emission spectrum of the CO laser with an intracavity cell containing NO. Simulation was performed assuming that due to the presence of the absorbing cell, the small-signal gain for vibrational transitions with $v < 10$ did not achieve the threshold, and therefore the conditions for lasing were specified beginning from the 10th level. When the cell with NO was placed into the resonator, the lasing power decreased by no more than 10 % – 15 %. Our calculations predicted the decrease in the output power by 17 %.
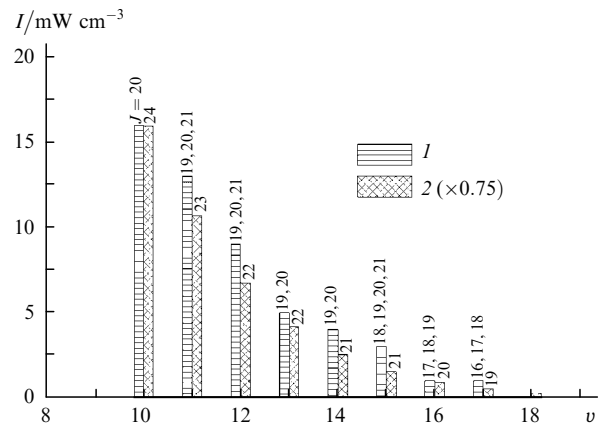

Figure 3. Experimental (*1*) and calculated (*2*) lasing spectra; the resonator contains an absorbing cell with NO; He : CO : O$_2$ = 92 : 8.92 : 0.08, $P = 12$ Torr, $I_d = 35$ mA, $E/N = 18.6$ Td, $D = 2.2$ cm, $d = 0.3$ cm, $L = 60$ cm, $T = 310$ K, $\Gamma = 4 \times 10^{-5}$ cm^{-1}.

Figure 4 shows VDFs of CO molecules in the amplification regime and in the cases of free running lasing and selection of laser lines with the help of the absorbing cell with NO.

The He–CO–O mixture was used and discharge tube walls were cooled with liquid nitrogen. One can see that experimental and theoretical results are in good agreement, as in the case of the water-cooled discharge tube. For the
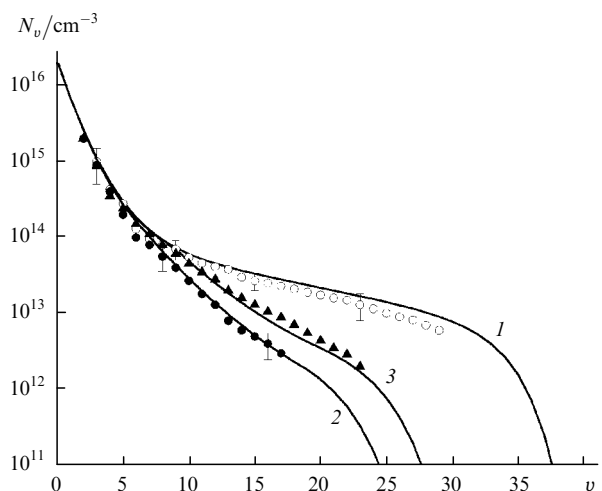

Figure 4. Vibrational distribution functions of CO molecules in the amplification ($\circ$) and free running ($\bullet$) regimes, and for lasing with an intracavity absorbing cell ($\blacktriangle$), and VDFs simulated in the amplification (*1*) and free running (*2*) regimes and for lasing with an intracavity absorbing cell (*3*). Other parameters are as in Fig. 3.

experimental conditions corresponding to Fig. 3, the lowest vibrational level involved in lasing is the level with $v = 7$. It is known [20] that the short-wavelength boundary of the emission spectrum of the CO laser lies near the vibrational level corresponding to the beginning of a plateau of the VDF of CO molecules. The plateau beginning is determined by the number of the vibrational level (v_{Tr}), which is called the Treanor minimum [21]. When lasing appears, the populations of vibrational levels above the Treanor minimum drastically decrease, the VDF slope above the Treanor minimum being dependent on the resonator Q factor [22].

The intracavity cell containing NO changed the VDF of CO molecules. Figure 4 shows that in this case the populations of high vibrational levels during lasing [curve (3)] noticeably exceed the populations of vibrational levels during free running lasing [curve (2)]. The NO intracavity cell leads to the red shift of the left boundary of the emission spectrum of the laser, which causes the observed increase in level populations. The heating of NO or the ignition of an electrodeless inductive high-frequency discharge in the NO cell (Figs 5, 6) resulted in a stronger red shift of the lasing spectrum, which can be explained by the increase in the population of vibrational levels of NO. It is also possible that more complex nitrogen oxides can be formed in the NO cell due to the dissociation of NO and subsequent chemical reactions, which can increase the absorption of radiation in the resonator. This absorption quenched lasing simultaneously at all rotational transitions of the vibrational level. This is probably explained by a small excess of the small-signal gain over the lasing threshold.

We studied the possibility of controlling the lasing spectrum by using other gases, in particular, CO_2, C_2H_4, and C_6H_6. Experiments have shown that the lasing spectrum can be considerably changed with a weak decrease in the output power. Figures 7 and 8 illustrate the modification of

Figure 6. Experimental emission spectra of the free running CO laser (a) and the CO laser with an intracavity NO cell in which a high-frequency discharge is ignited (b); He : NO = 7 : 3, $P = 80$ Torr.

lasing spectra with the help of intracavity absorption in the $CO_2 - C_2H_4$ and C_6H_6 mixtures. One can see that the emission range of the CO laser and radiation intensity distributions in spectral lines considerably change. The output power decreases by $\sim 30\%$. The optimisation of experimental conditions (the working mixture composition and discharge parameters, as well as the composition of gases used for intracavity absorption) can provide even more efficient modification of the emission spectrum.
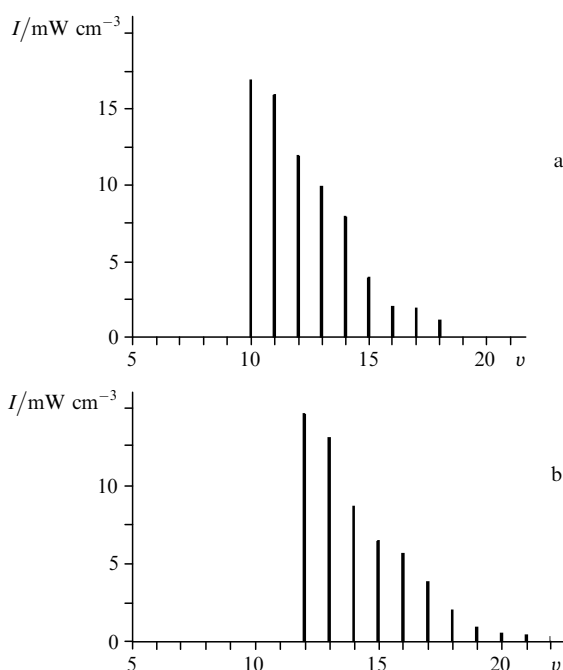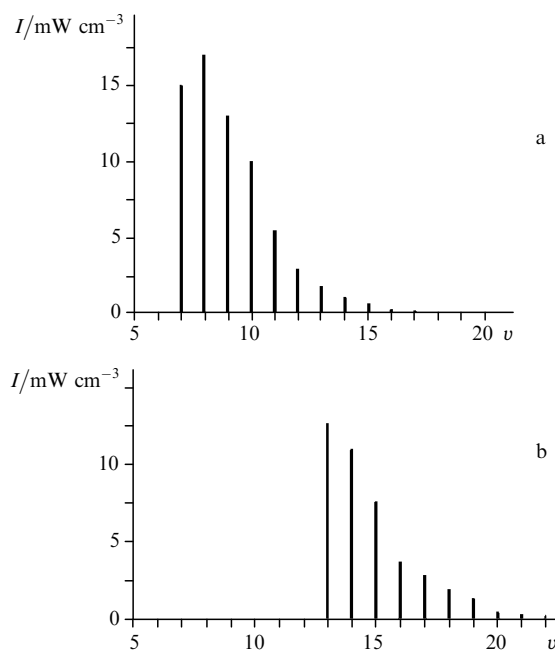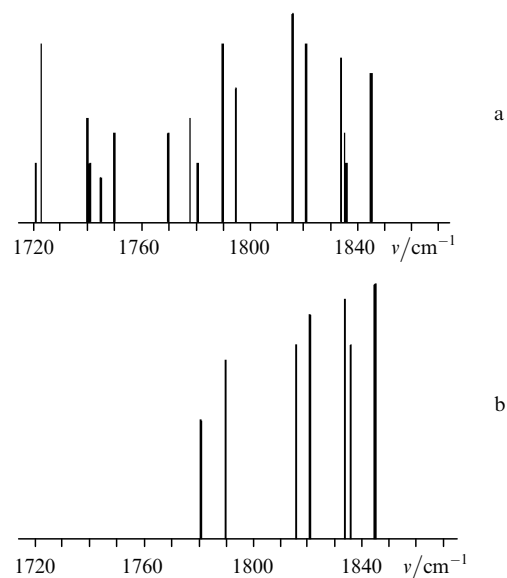

Figure 5. Experimental emission spectra of the CO laser with an unheated intracavity NO cell (a) and the cell heated to 360 K (b). Other parameters are as in Fig. 4.

Figure 7. Emission spectra of the free running CO laser (a) and the CO laser with an intracavity cell containing the $CO_2 : C_2H_4 = 7 : 3$ mixture ($P = 100$ Torr) (b); the selection efficiency is 74%, He : CO : N_2 : O_2 = 96 : 1 : 2 : 0.04, $P = 5$ Torr, $I_d = 30$ mA.
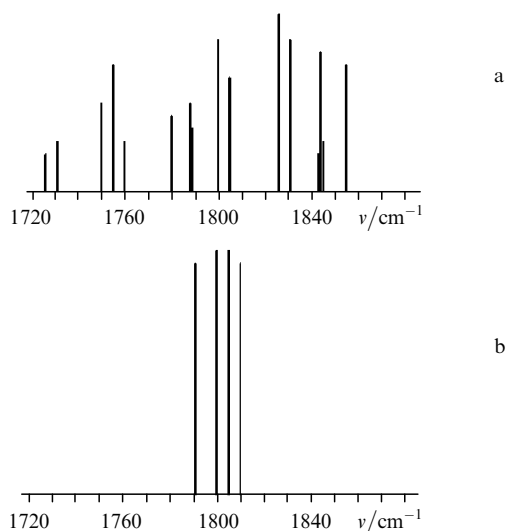
Figure 8. Emission spectra of the free running CO laser (a) and the CO laser with an intracavity cell containing the C_6H_6 ($P = 50$ Torr) (b); the selection efficiency is 67 %, He : CO : N_2 : O_2 = 96 : 2 : 2 : 0.02, $P = 13$ Torr, $I_d = 50$ mA.

5. Conclusions

The VDF of CO molecules has been measured in the presence and absence of generation in a CO laser excited by a longitudinal dc discharge. Experimental VDFs and lasing spectra are in good agreement with calculations. By using an intracavity absorbing cell with different gases (NO, CO_2, C_2H_4 and C_6H_6), it is possible to change considerably the lasing spectrum by decreasing the lasing efficiency only weakly. The use of an absorbing cell with NO causes the red shift of the lasing spectrum. This effect increases when the NO cell is heated or an inductive high-frequency discharge is ignited in the cell. The red shift of the lasing spectrum leads to the increase in the population of vibrational levels with large v.

By controlling the lasing spectrum in such a way, it is possible to form radiation from a CO laser which is weakly absorbed in a medium with impurities used in the absorbing gas cell.

The intracavity absorbing cell can be used to change the shape of the VDF of CO molecules in the region of high vibrational levels, which can be applied for studying elementary processes involving CO molecules in high excited vibrational states.

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