PULSED LASER ACTION ON CO ELECTRONIC TRANSITIONS WITH COOLING OF THE GAS

M. D. BARANOV, V. M. KASLIN, and G. G. PETRASH

P. N. Lebedev Physics Institute, USSR Academy of Sciences

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The properties of pulsed laser action on the 0-5, 0-4, 0-3, and 0-2 bands of the Angstrom system of the CO molecule with cooling of the working gas are investigated experimentally. Cooling of the gas from room temperature to 125°K results in a redistribution with respect to the rotational structure and in a sharp increase of laser output (by about 50 times for the line corresponding to the maximum of the distribution over J). The spectrum of the laser radiation is studied. The dependence of generated power and its duration on capacitor voltage V and gas density N is studied over a wide temperature range. It is found that at all temperatures the most power is attained at the same gas density $N_{opt} = 1.2 \times 10^{17}$ cm^{-3}. At any temperature all properties of the laser pulsed are determined by the parameter $\gamma = (V - V_0)/N$. Generation occurs in a limited interval of the parameters V and N. The pulse arises at the beginning of the strongly pointed phase of the discharge. The pulse length decreases with decreasing pressure and increasing voltage. In these experiments it varies from 300 to 30 ns. Under optimal conditions the pulse length is about 100 ns. The optimal conditions for different bands are attained at different V and N, which permits tuning from band to band, thereby obtaining laser radiation in all principal portions of the visible spectrum, by changing pressure or voltage. The inversion mechanism for the CO Angstrom bands is discussed. In particular, it is shown that laser action in this system has a quasicontinuous character, which suggests the possibility of CW operation.

INTRODUCTION

PULSED laser action on electronic transitions of the CO molecule was first observed in 1963.[1] However, it has not been studied much since then, possibly because of the relatively low power of the laser and the associated experimental difficulties. In[2,3] the temporal characteristics of the pulse and the inversion establishment time were studied. In[4] a further investigation of the radiated spectrum was carried out. Use of pure flowing gas and internal mirrors permitted the observation of many new lines. At the present time laser action is obtained on six bands of the Angstrom system, $B^1\Sigma^+ \to A^1\pi$, which are spread over almost the entire visible spectrum from red to blue:

Band $v' \to v''$:	0—5	0—4	0—3	0—2	0—1	0—0
Edge of the band of spontaneous emission (in air), Å:	6620	6080	5610	5198	4835	4511

All six bands originate from a single upper vibrational level $v' = 0$. The total number of laser lines is about 90. Action was observed during the current pulse. Different authors give different pulse lengths and different optimal conditions. In[2] the opinion is expressed that inversion is produced by direct excitation by electrons and that laser action stops as a result of a radiative cascade from a C state. Experimental data, however, are insufficient to make any reliable conclusions about the creation and annihilation of the inversion.

There is interest in the further investigation of laser action on the Angstrom bands in CO because of the following peculiarities of this system. The CO molecule is the only asymmetrical molecule for which laser action on electron transitions has been obtained so far.[1)] In contrast to other molecules which give action on electronic transitions (N_2, H_2, D_2), vibrational transitions are allowed in CO. There is even known laser action on the vibrational transitions of the ground state of CO[6-8] The possible interaction between electronic and vibrational laser action is of interest,[9] particularly cascade action. In addition, the presence of vibrational transitions can in principle lead to peculiarities in the mechanism of creation and annihilation of inversion.

Laser action in the visible region of the spectrum can be of great practical interest, if one can succeed in significantly increasing its power. A significant increase in gain and power of electronic laser action in molecules, as was shown in[10,11], can be obtained by cooling the working gas. Preliminary results[11] have shown that for CO the power increase can be extremely large. It was difficult, however, to predict beforehand what the effect of temperature on laser action in CO would be, and this also is due to the asymmetry of the molecule. The point is that other molecules are efficiently produced in the CO discharge (CO_2, C_2, and O_2, in particular), so that the composition of the gas undergoes marked changes. With cooling, the chemical equilibrium can be shifted; hence it is difficult to predict the results of cooling. More experiments were required to clarify the situation. The investigation of temperature effects is also of independent interest, for it casts additional light on the generation mechanism.

In the present paper we set ourselves the task of carrying out a thorough experimental investigation of the properties of laser action on the Angstrom bands of the CO molecule as a function of discharge conditions and, especially, of temperature. Such an investigation is essential to elucidate the optimal conditions for generation and gives valuable information about the

[1)]Except for one weak, uninteresting laser line in HD.[5]

mechanisms of creating and destroying inversion. It was necessary to give particular attention to a study of the temporal characteristics of generation, since the available data on this question are contradictory,[1-4] and the data on the lifetimes of the working levels[12-14] suggests the possibility of obtaining CW action.

EXPERIMENTAL ARRANGEMENT

In the experiments a laser of typical construction with a longitudinal electric field and external mirrors was employed. The arrangement and construction of the laser tube are diagrammed in Fig. 1. The discharge tube, 8 mm in internal diameter, had two internal cold electrodes of thoriated tungsten, located in side branches. The discharge span was 111 cm in length. Excitation of the working gas was accomplished by discharging a 0.01 μF capacitor through a controllable three-electrode air discharger CD. The capacitor voltage V was established within the limits of 8 to 40 kV. The peak value of the current was 0.1 to 1.8 kA, depending on the experimental conditions. The repetition rate of the current pulses was several hertz; most of the measurements were made at a frequency of 2 Hz.

The discharge tube was enclosed by two jackets with diameters 16 and 30 mm over almost the entire active length (100 mm). The outer vacuum jacket VJ, pumped out by a diffusion pump, served as thermal insulation, and the internal one was for cooling the discharge tube. This was done by introducing cold nitrogen vapors from a dewar containing liquid nitrogen into the internal jacket. By varying the rate of evaporation of the nitrogen, it was possible to vary the temperature of the wall of the discharge tube from room temperature to about 100°K. The temperature was monitored by a copper-constantan thermocouple TC attached to the outside wall of the discharge tube. In doing this, it was assumed that the temperature of the working gas did not differ very much from the temperature of the outside wall of the tube at the low pulse repetition rate used in the experiment. Experiments with nitrogen[10,11] done under similar conditions demonstrated the validity of this assumption. On the other hand, calculations made on the basis of the observed intensity distribution of the rotational lines showed that the temperature of the gas in the tube was approximately 25°K higher than the temperature of the outside wall. Since such a control was not applied at all investigated temperatures, we shall henceforth always quote the temperature of the tube wall.

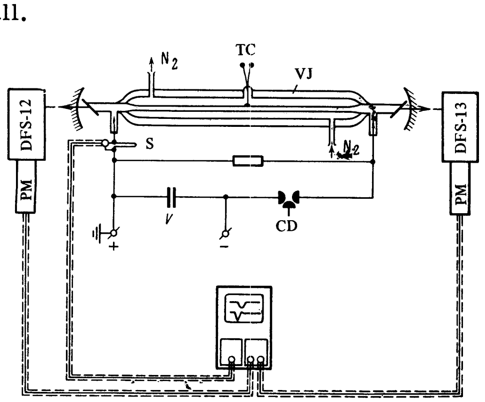

FIG. 1. Setup and construction of laser tube.

The laser resonator was formed by two spherical mirrors in almost confocal configuration. The radius of curvature of the mirrors was 2 m. For work with different bands, we could choose mirrors with dielectric coatings possessing high reflectivity over a given band. In special cases, aluminum mirrors were used.

The study of the spectra of generation and spontaneous emission were carried out photographically and oscillographically. A DFS-13 spectrograph with 1200 and 600 line/mm gratings was used in the photographic method. With the first grating we worked in first order at a dispersion of 2 Å/mm, with the second, in third order at a dispersion of 1.35 Å/mm. For accurate measurement of wavelength and identification of the spontaneous and generated lines, the spectra were photographed on photographic plates. The apparatus allowed positive resolution of almost all the lines of the rotational structure. According to our estimates, the accuracy of wavelength determination was ±0.03 Å. An arc spectrum of Fe was used as a standard. Since the precision of measurement of the relative position of the lines was ~0.005 Å, a graphical identification of the spontaneous emission lines of the corresponding bands was made, according to the data of[15,16]; thereafter, the lines participating in laser action were already identified. The correctness of the identification was verified by the construction of Forter graphs.

To study the temporal characteristics of the radiation, a DFS-12 spectrometer with a nominal dispersion of 5 Å/mm was used. The total intensity of the two or three strongest rotational lines of a given band was registered. In the visible, photomultipliers FÉU-28 and FÉU-36 were used to detect the induced and spontaneous emission, in the ultraviolet, an FÉU-39 with a uviol window. (To isolate the 0-5 band, which lies outside the working range of the DFS-12, a UM-2 universal monochromator was used.) The signal from the photomultiplier was fed through a carefully shielded coaxial cable to an S1-11 fast oscilloscope. The oscillograms were photographed and then measured. The time resolution of the apparatus, including the photomultipliers, was about 10 ns. This was determined by investigating the superradiance of the 6143-Å Ne line, the observed duration of which was found to be 10 ns. The duration of both the generation and the spontaneous emission is considerably longer, so that we can say that the apparatus did not introduce any distortion in the temporal characteristics. The photomultipliers were operated in the linear region.

The duration, magnitude, and shape of the discharge current pulse was measured from an oscillogram of the voltage pulse from a non-inductive coaxial shunt S of resistance 0.227 Ω connected in series with the discharge tube. The voltage from the shunt was fed through a voltage divider to the S1-11 oscilloscope. Analogous measurements of the current pulse were made with a Rogovskiĭ bridge working in the current pulse transformer regime.[17] Both methods gave the same result.

Oscillograms of the current pulses and radiation pulses were also obtained on a dual-beam oscilloscope S1-17 with a time resolution of 40 ns. The total average power over all lines was measured with a calibrated thermopile and an M-17/13 galvanometer. The peak generated power could be estimated from the known duration and repetition rate of the pulses. Under optimal conditions it was about 20 W, most of it being concentrated in the 0-4 band.

In the experiments we used carbon monoxide prepared in the chemical group of the Optical Laboratory at FIAN (Physics Institute, Academy of Sciences) by the decomposition of formic acid, as well as commercial bottled gas. The gas was not further purified. The carbon monoxide obtained from formic acid usually contained a trace of nitrogen. Before the tube was filled with the working gas, it was pumped out to a pressure of $\sim 10^{-5}$ Tor. All the measurements were made without flow-through of the working gas.

EXPERIMENTAL RESULTS

Using the above apparatus, we observed four of the six known laser bands of CO (see above): 0-5, 0-4, 0-3, and 0-2. The 0-1 and 0-0 bands were not observed in generation. The table gives the identification and measured wavelengths of the rotational components of these bands. It is to be understood that the table is a summary, and the conditions under which one of the lines shown in it is obtained differ from those under which the others are obtained. Most of these laser lines were observed previously in[1,4]. Two new lines are indicated in the table.

The distribution of power over the bands depends to a marked extent on the choice of mirrors and on the parameters of the gas discharge. By using mirrors with specific reflective properties, one can isolate a specific band from the laser output. The use of aluminum mirrors, which have approximately the same reflectivity for all bands, showed that the highest laser gain was in the 0-4 band. The gain in the 0-5 and 0-3 bands was much less and roughly the same. It is still less in the 0-2 band. However, irrespective of the choice of mirrors, the power distribution over the rotational structure is of the same form in all bands. For example, laser action is observed only in the Q and P branches, with the principal fraction of the power (just as for spontaneous emission) in the Q branch, the P branch lines being considerably weaker. The R-branch was not observed in generation. This relation among the branches is explained by the linear dependence of the gain coefficient on the rotational strength of the line (the Hanie-London factor), which for transitions with a change of orbital momentum $\Delta\Lambda = \pm 1$ is greatest for the Q branch and least for the R branch. An exception in our case is in the 0-2 band, for which the line λ = 5198 Å, belonging to the P branch, is much more intense than all the other lines in this band.[2)] The explanation of this anomaly is evidently provided[4] by the coincidence within the Doppler width of the P4 and P5 lines, the total gain coefficient of which greatly exceeds the generation threshold. A similar phenomenon, although not so clearly expressed, is observed in the lines P6, 2 and P7, 1 in the 0-4 band.

Rotational structure of the laser bands in the Angstrom system of the CO molecule

$v' \longrightarrow v''$ band	λ meas, Å (in air)	Rotational transition	
0—5	6620.04		P5
	6619.29		P6 *
	6618.18		P7
	6616.33	Q2	
	6615.12	Q3	
	6613.53	Q4	
	6611.51	Q5	
	6609.10	Q6	
	6606.30	Q7	
	6603.06	Q8	
	6599.46	Q9	
0—4	6080.07		P4
	6079.95		P3 *
	6079.88		P5
	6079.35		P6,2
	6078.50		P7,1
	6077.34	Q1	P8
	6076.66	Q2	
	6075.66	Q3	
	6074.35	Q4	
	6072.72	Q5	
	6070.75	Q6	
	6068.48	Q7	
	6065.86	Q8	
	6062.93	Q9	
0—3	5606.17	Q3	
	5605.15	Q4	
	5603.85	Q5	
	5602.27	Q6	
	5600.44	Q7	
	5598.35	Q8	
0—2	5198,06		P4,5

*New lines.

Another property of the rotational spectrum common to all laser bands is the presence of a sharp maximum in the distribution of radiated power over the rotational structure within the branches. The shape of this distribution is somewhat like the analogous distribution in the spontaneous spectrum. Figure 2 shows the distribution of generated power in the Q branch of the 0-4 band for different temperatures of the discharge tube walls. It is seen from the figure that a change in gas temperature has a marked influence on the laser action in CO: When the gas is cooled, J_{max} (the rotational quantum number corresponding to the maximum of the distribution) shifts toward smaller values, and there is a very strong increase in power. Cooling of the working gas from room temperature to 125°K led to a 50-fold increase in average power at J_{max}.

An analysis of the data in Fig. 2 gives the following empirical relation between the average generated

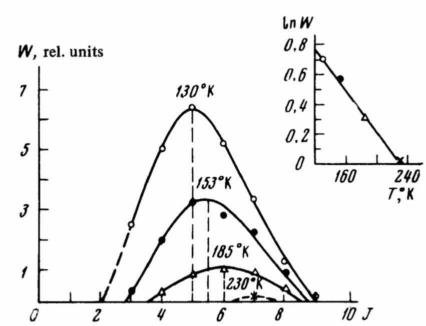

FIG. 2. Distribution of energy of generation in the Q branch of the 0-4 band for $N_{opt} = 1.2 \times 10^{17}$ cm^{-3} at various temperatures of the walls of the discharge tube. The graph of ln W = f(T) is constructed for J_{max}.

2)The table gives only this line, since the other lines that happened to be located on both sides of it were very weak and their wavelengths were difficult to measure accurately. The P4,5 line is the last line at the edge of the 0-2 band.

power and the gas temperature for optimum density:

$$W(J_{\max}) = Ce^{-\alpha T};$$

here C and α are empirical constants. The linear approximation of the function $\ln W = f(T)$ is good to within 6%.

It must be mentioned that the marked increase of gain and generated power upon cooling of the gas was of great value in the conduct of our experiment. The fact is that at room temperature the gain in the system is so low that one has to work near the generation threshold, and this makes all the measurements very difficult. More cooling of the gas permitted a careful study of the properties of this interesting laser.

The character of the change in properties of the CO laser with cooling was found to be generally similar to that observed earlier in nitrogen.[10,11] However, unlike the nitrogen case, where it was possible to cool to the temperature of liquid nitrogen, the approach to such low temperatures for CO encountered certain difficulties, hence most of the measurements were made for $T \geq 125°K$. We note, however, that when the temperature is lowered from 125°K to nitrogen temperatures (78°K), there is a further increase in gain and generated power.

The decrease in CO concentration in the discharge at low temperatures may be due to the following cause. Molecules of O_2 and CO_2 are formed in the discharge from the dissociation of CO. If the partial pressure of any product of the reaction in the discharge exceeds the vapor pressure of this product at a given temperature, this product will be deposited on the walls. As a result, there will be a reduction of CO until equilibrium is reestablished, but this will now be at some other gas density. The lower the temperature, the lower the equilibrium density. Under our operating densities, the CO_2 formed in the discharge evidently begins to freeze out at about 125°K (the vapor pressure of CO_2 at 125°K is somewhat less than 1 Tor). At the temperature of liquid nitrogen the deposition of CO_2 is so significant that the gas pressure in the discharge falls to about 10^{-2} Tor in just a few minutes.

Since the character of the change in all the investigated laser bands as a function of voltage, current, and gas pressure is the same in its general features, we shall limit ourselves below mainly to a detailed examination of the properties of only one, the most easily obtained generation in the 0-4 band.

To determine the optimal conditions for reaching the highest average power, we used the method of photographic photometry to measure the dependence of average power (or pulse energy) at $J_{\max}$ on the applied voltage at various working gas pressures. These measurements were made at 296, 265, 232, 195, 160, and 125°K. Figure 3a shows the dependence for T = 232°K. The pattern is analogous at the other temperatures. On one hand, it is seen from the figure that the pulse energy does not rise monotonically with voltage V, but reaches a certain maximum value at V_{opt}, after which it quickly falls to zero. On the other hand, it is seen that both V_{opt} and the general region in which laser action exists depend on the gas pressure: the higher the pressure, the more are they shifted toward higher voltages. Figure 3c illustrates the de-

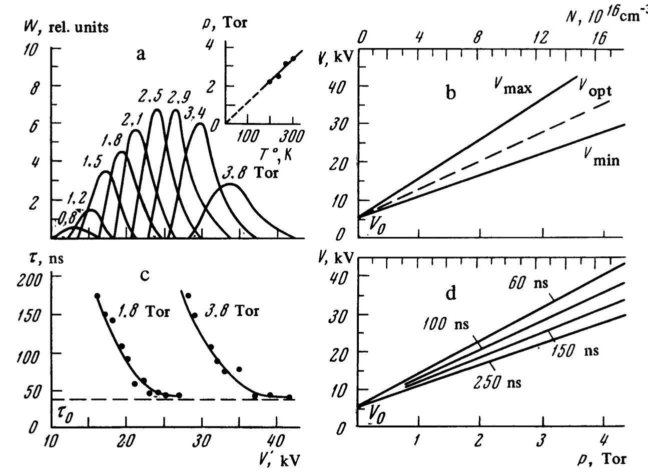

FIG. 3. Power and temporal characteristics of generation in the 0-4 band at T = 232°K: a – dependence of generated pulse energy on applied voltage at various gas pressures; b – region of existence of generation as a function of the parameters V and p (or N); c – dependence of duration of generation on applied voltage for different gas pressures (τ_0 is the radiation lifetime of the upper laser level); d – isotimes of generation in coordinates (V, p (or N)).

pendence of $V_{\min}$, V_{opt}, and $V_{\max}$ on gas pressure. Interestingly, when these functions, which are straight lines, are extrapolated to lower pressures, they intersect in a single point; the point is on the voltage axis at $V = V_0$. Similar results are obtained at the other temperatures. In all cases the value of V_0 is the same; only the slopes of the lines change. The slope is higher, the higher the gas temperature.

In investigating the temporal characteristics of the generation, we found that the laser pulse length was not constant, as previously assumed, but depended strongly on the applied voltage V and the gas pressure p. It was found that within the limits of those values of these parameters for which laser action is observed, the generation length τ at a constant temperature is a monotonic function of them. Figure 3c illustrates the reduction of τ with increasing V and decreasing p at T = 232°K. The pulse length varied from ~300 to ~30 ns. (Note that the radiative lifetime of the upper laser level is τ_0 = 25 ns.[14]) Thus an explicit dependence of the pulse length on the operating conditions of the laser can evidently explain the contradictions in the data given for it by different authors.[1-4] It is interesting that if one constructs the lines of equal pulse length (isotimes) in the coordinate system (V, p), these lines are straight, with different slope, but come together in one point on the ordinate axis at $V = V_0$ (Fig. 3d). It is remarkable that the value of the parameter V_0 obtained by extrapolating the temporal characteristics is exactly the same as in the case of the power characteristics, within the limits of experimental error. The slope of the isotimes is greater, the smaller τ is. Similar measurements made at different gas temperatures gave analogous results.

It can also be seen in Fig. 3a that at a given temperature there is a certain optimal gas pressure (here it is ~2.7 Tor) for which the highest pulse energy is obtained at V_{opt}. The experiment showed that the dependence of the optimal pressure on temperature (Fig.

3a, upper part) is approximated well by a straight line passing through the origin. Since in these experiments there was a large volume of uncooled gas connected to the tube, the pressure of the gas in the working tube hardly changes upon cooling. The pressure, density, and temperature of the gas are related by the formula $p = NkT$. It follows from this formula and Fig. 3a that over the entire investigated interval of temperature the optimal conditions for generation are attained at the very same gas density $N_{opt} = 1.2 \times 10^{17}$ cm^{-3}.

If in Figs. 3b and 3d the gas pressure is recalculated into density, then it turns out that at all temperatures the functions for V_{max}, V_{opt}, V_{min} and τ = const in the coordinates (V, N) almost completely coincide. A small difference is observable in that lowering the temperature slightly diminishes the slope of all the lines and increases V_0. This change bears a systematic character, but it is scarcely outside the limits of experimental error. As an example we show in Fig. 4 the function V(N) for τ = const. The points indicated by different symbols correspond to different temperatures. It can be seen that the systematic shift with temperature is of the order of the scatter of the points. Thus, it may be stated that the region of existence of generation in the plane (V, N) and the pulse length for given V and N do not depend on the gas temperature.

Since the lines in Figs. 3b and 3d intersect in a single point V_0, an attempt was made to describe the generation properties as a function of the parameter $\gamma = (V - V_0)/N$. In Fig. 5 are shown the dependence of the pulse energy with respect to the energy at γ_{opt} (a) and of the pulse length (b) on the parameter γ. It was found that these functions are universal, within experimental error, and satisfy all our experimental data.

As we mentioned earlier, all the above considered functions pertain to the strongest generation in the 0-4 band. Qualitatively the same behavior is observed also in the other bands of the Angstrom system. However, the optimal conditions for generation in the different bands differ markedly. As an example, we show in Fig. 6 the dependence of pulse energy in the 0-2, 0-3, and 0-4 bands on V for a single density and temperature of the gas. It is seen from the figure that the highest energy for these three bands is attained at

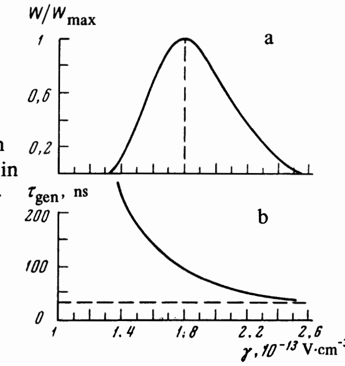

FIG. 5. Shape of the distribution of pulse energy (a) and duration (b) in the 0-4 band as a function of the parameter $\gamma = (V - V_0)/N$.

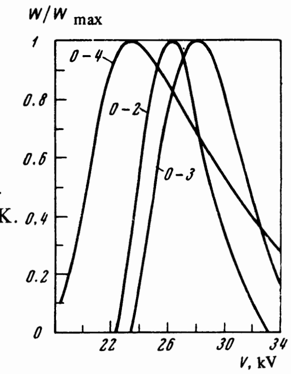

FIG. 6. Dependence of pulse energy in the 0-2, 0-3, and 0-4 bands on the voltage for $N = 1 \times 10^{17}$ cm^{-3} and T = 180°K.

different V_{opt}. The differences are so marked that one could visually observe how the color of the laser light changed, e.g., from red to green, with a change in applied voltage (or gas pressure). This feature of laser action in the Angstrom band of CO makes it possible to tune the laser over bands whose wavelengths are spread over almost all parts of the visual spectrum, by simply changing the voltage on the working condenser or the gas pressure.

Figure 7 shows oscillograms of the generated pulses (a) and of the current (b). The oscillograms were obtained on a dual beam oscilloscope. It is seen from Fig. 7 that generation is observed strictly at the leading edge of the current pulse in its sharply pointed phase, the beginning of generation being delayed relative to the beginning of the sharply pointed phase by about 150 ns. In this, we have taken into account the delay between the light and electrical pulses associated with the different times of passage of the signals from

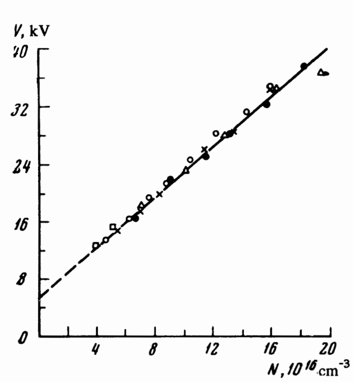

FIG. 4. Illustration of scatter of experimental points in measurements of pulse duration in the 0-4 band at different gas temperatures (K): □ – 265, ○ – 232, × – 195, ● – 160, △ – 125. $\tau \approx 100$ ns.

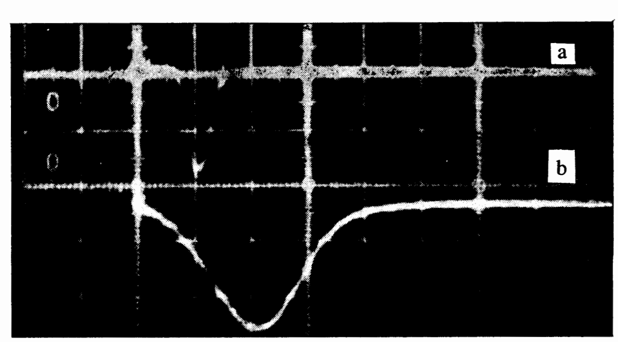

FIG. 7. Oscillograms of the pulses of current (a) and generation (b). Scale, 0.5 μs per division.

the respective detectors to the oscilloscope. In addition, an oscillographic study of the spontaneous emission (observed from the end of the gas-discharge tube) showed that the spontaneous emission from the laser levels is much shorter than the current pulse, attaining its maximum in the upper level somewhat earlier than in the lower. The generation, in turn, is slightly ahead of the luminescence from the upper level.

Furthermore, it is important to note that with an increase in applied voltage and a decrease in gas pressure, as is the case with the generation, the duration of current and of spontaneous emission also decreases rapidly in the lasing region (Fig. 8). The relation between the durations of generation, current, and spontaneous emission from the upper laser level is shown in Fig. 8b.

An investigation of the visual spectrum of the discharge disclosed the presence of C_2 molecules. The maximum in the emission of the electronic bands of C_2 lies on the back edge of the current pulse.

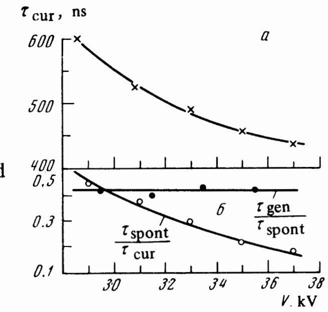

FIG. 8. Dependence of current duration (a) and relations among the durations of current, generation, and spontaneous emission in the 0-4 band (b) on voltage.

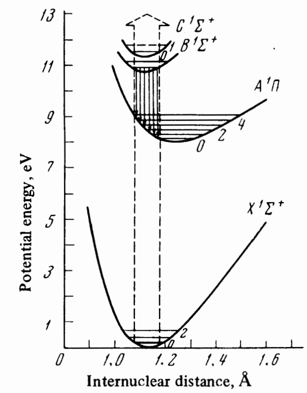

FIG. 9. Level diagram of the CO molecule.

DISCUSSION OF RESULTS

The potential curves for the CO molecule are sketched out in Fig. 9. The investigated generation is observed in the electronic transition between the singlet levels $B^1\Sigma^+$ and $A^1\pi$. Both levels are coupled by strong optical transitions to the ground state of the molecule $X^1\Sigma^+$. The upper level $B^1\Sigma^+$ radiatively decays into the ground and lower laser levels. According to Hesser,[14] its lifetime is 25 ns, and the excitation cross section by direct electron collision is 2.4×10^{-18} cm^2, according to the data of Zapesochnyĭ and Skubenich.[18,19] Cascade transitions to it are unknown. The lower laser level $A^1\pi$ decays radiatively only to the ground state. Its lifetime is ~10 ns,[14] and the cross sections for excitation by direct electron collision taking cascades from above into account are as follows[18,19]: $\sigma(v'' = 5) = 1.38 \times 10^{-18}$, $\sigma(v'' = 4) = 2.06 \times 10^{-18}$, $\sigma(v'' = 3) = 2.74 \times 10^{-18}$, $\sigma(v'' = 2) = 3.02 \times 10^{-18}$, $\sigma(v'' = 1) = 2.66 \times 10^{-18}$, and $\sigma(v'' = 0) = 1.70 \times 10^{-18}$ cm^2. Besides the Angstrom system, the system of Herzberg bands ends at the lower laser level, beginning at the level $C^1\Sigma^+$ with radiative lifetime ~1 ns[14] and cross section for direct electronic excitation of 0.48×10^{-18} cm^2.[18,19] The level $C^1\Sigma^+$ decays into the level $A^1\pi$ and also by two other paths: a strong optical transition to the ground state and a weak intercombination transition to the state $a'\,{}^3\Sigma^+$. Levels B and C predissociate, beginning respectively with vibrational levels $v' = 2$ and $v' = 1$. The vertical dashed lines indicate the region of excitation of the molecule by direct electron collision with fulfillment of the Franck-Condon principle. The arrows indicate the bands of the Angstrom system observed in the laser action.

Our experiments have shown that at all investigated gas temperatures the fundamental properties of the generation are determined by the parameter γ $\gamma = (V - V_0)/N$. Since the generation occurs in the strongly pointed phase of the discharge, when a more or less uniform field is established in the discharge tube,[20,21] this parameter, which is analogous to the usually employed parameter E/p, determines the properties of the electron gas in the discharge. This amounts to saying that the processes of creation and annihilation of inversion in the electronic transitions of the CO molecule are principally determined by interaction with electrons.

The results of the temperature investigations lead to a similar conclusion. It was shown in[10,11] that with electronic excitation of the working levels, cooling of the working gas evokes a strong growth in gain and generated power by electronic transitions in molecules. This effect arises as a consequence of a redistribution of the population over the rotational levels and a decrease in the Doppler width of the lines. The behavior of generation in CO upon cooling (in particular, the sharp increase in generated power) is in complete accord with the ideas about electronic excitation developed in[10,11].

With respect to the direct mechanism of creation of inversion, all the known experimental data on this laser action in CO,[1-4] as well as the results of our work, agree well with the assumption of direct excitation of the upper working state of the molecule by electrons from its ground state. Particular evidence for this is given by the observed generation spectrum, the appearance of generation at the leading edge of the current pulse, and other characteristics of the generation. A calculation of the gain coefficient[3)] with the assumption of direct excitation of both working levels by electrons from the ground state and using the effective cross sections from[18,19] has shown qualitative agreement with the observed facts. For example, according to the calculation the strongest gain is in the 0-4 band; somewhat less, but comparable, gain is found in 0-5

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3)To be published elsewhere.

and 0-3, and then, in descending order, in 0-2, 0-1, and 0-0. Just such a sequence was observed in the experiment. The estimated rate of excitation was completely adequate to explain the observed power.

The reasons for disruption of generation are not completely clear at the moment. The investigation of the spontaneous emission of the discharge does throw some light on this problem. In particular, it showed that the spontaneous emission from the working levels is shorter than the current pulse, and its maximum lies on the leading edge of the pulse. This says that the excitation of the laser levels ceases before the discharge current ends. An obvious reason for this limitation in the duration of excitation of the working levels under our experimental conditions is the decrease in voltage on the tube as the condenser discharges and the concomitant decrease in the energy of the electrons in the discharge. In addition, as the discharge develops, the working gas dissociates into products which could have high effective cross sections and low excitation and ionization potentials, which would lead to a sharp increase in the losses sustained by the electrons and, consequently, to a cooling off of the electrons. Moreover, if the decay of CO is intense, then some shortening of the duration of spontaneous emission can also occur as the result of the reduced amount of CO.

A change in the conditions of excitation of the upper level during development of the discharge cannot, however, explain all the observed features of the generation. If the formation of inversion were determined only by the excitation of the upper level, i.e., if the population of the lower level were neglected, then the behavior of the generation in all bands (e.g., attainment of optimal conditions) would be identical, since all bands originate from one level. But in the experiment we observed a marked difference in the optimal conditions for different bands, as well as some difference in their temporal behavior. This means that an important role is played by the lower laser levels, which are different for different bands, i.e., their populations must be close to the population of the upper level. A process that so effectively populates the lower laser levels may be excitation by direct electron collision from the ground state. This is corroborated by the data on the respective cross sections and the calculation based on them: the populations of the vibrational levels of the upper and lower states are close, and the ratio between them $N_{V'}/N_{V''}$ varies from 1 to 5 for the different bands. However, consideration of this process alone shows that there should be gain in the working transition for practically any electron energy, i.e., the duration of generation should in fact be the same as the duration of spontaneous emission from the upper level.

Thus, in order to explain the shortening of generation compared to emission, as well as the fact that generation ceases for high V and N, it is necessary to include additional processes in the consideration of the population of the lower levels. These processes do not have to be so effective as the basic one, since not much needs to be added to disrupt the inversion in the working transition. Such processes may be: radiative cascade from the state $C^1\Sigma^+$ (Herzberg bands)[2] and superelastic collisions with electrons, i.e., nonradiative transitions over the working path with transfer of excess energy to electrons. An estimate of the effect of the cascade from the C state made on the basis of the excitation cross section data shows that the contribution of this cascade does not eliminate the problem. Moreover, the excitation functions for the B and C levels are approximately the same in form, so that the role of the cascade will change little with changing discharge conditions. Superelastic collisions could play a particularly large role at high electron densities. It is known that they significantly limit the attainable power in lasers based on electronic transitions in the nitrogen molecule.[21,22] The possibility that the rapid cessation of generation in CO with time and at high V and N is partially due to such a process cannot be excluded.

Obviously, one must also consider possible processes by which energy is exchanged between the lower working levels and the levels of other electronic states, since some close resonances exist for some of them: e.g., between the levels $A^1\pi(v = 4)$ and $a'^3\Sigma^+$ $(v = 14)$ the energy defect amounts to only 23 cm^{-1}. Finally, it is not excluded that in the process of decomposition of CO in the discharge there are formed molecules and radicals that absorb in the region of the working transitions.

At the present time there is insufficient data to evaluate the roles of all these factors in the process of cessation of generation. More experiments are required.

The circumstance that the excitation cross sections of a number of the lower levels (see beginning of this section) exceed the excitation cross section of the upper level means that inversion is possible only as the result of a sufficiently rapid decay of the lower level. This fact, as well as the fact that the duration of generation varies as a function of changing discharge conditions (it follows changes in current duration) and can significantly exceed the lifetime of the upper level, amounts to saying that generation in the bands of the Angstrom system of the CO molecule has a quasicontinuous character. This means that with the proper choice of conditions it may be possible to obtain continuous generation in these bands.

Finally,,the present investigations have shown that application of the method of cooling[10,11] to increase the gain in gas-discharge lasers based on electronic transitions in asymmetrical molecules has certain limitations. In particular, the lower limit of temperature is determined by the vapor pressures of the products of decomposition of the working substance. The higher the vapor pressures, the lower the temperature that can be employed. In addition, the new substances formed in the discharge, as mentioned above, can markedly affect the properties of the electron gas or, for example, absorb energy at the working wavelength. Continuous pumping on the working gas may be a way to eliminate these decay products.

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