INVESTIGATION OF THE LASER EFFECT IN CO_2 DURING PULSED EXCITATION

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Processes occurring in a pure CO_2 laser during pulsed excitation were investigated. A complex spectral and time dependence of the generation of two lines at 9.5 and 10.6 μ was observed. A study of the dependences of the pulse amplitude and emission lag on the electron energy showed that electron impact was the decisive mechanism. Maximum power was obtained for the 9.5 μ line: it was 5 kW for a pulse of 1–2 μsec duration.

MANY papers[1,2] have been published on CO_2 lasers operating continuously or under Q-switching conditions. The operation of these lasers under pulse excitation conditions has not yet been investigated sufficiently thoroughly. The present paper describes an investigation of the pulse operation of a pure CO_2 laser: our purpose was to obtain sufficiently powerful short light pulses which could be very useful in many applications such as nonlinear infrared optics, the generation of nonequilibrium carriers in semiconductors, etc.

1. EXPERIMENTAL METHOD

We used a laser in which a gas was pumped along a discharge gap 200 cm long. The radiation was emitted through a window made of an NaCl plate, 12 mm in diameter. Spherical (R = 10 m) gold-coated mirrors were placed within the laser cavity. The discharge tube walls were cooled by circulating tap water at a temperature of 13–15°C. The discharges in the gas were produced by pulses of 300 A maximum amplitude. A capacitor was discharged via a thyratron directly through the laser without any additional ballast resistance. The radiation was detected with Ge + Hg photoresistors operating at 77°K and having a time constant of 10^{-7} sec. Spectroscopic measurements were carried out using an IKS-12 monochromator with an NaCl prism. The apparatus is shown schematically in Fig. 1.

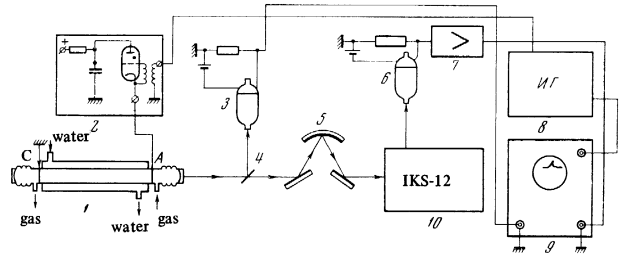

FIG. 1. Schematic diagram of the apparatus. 1) Laser; 2) generator of power pulses; 3), 6) Ge + Hg photoresistors; 4) plane-parallel NaCl plate; 5) focusing system of mirrors; 7) amplifier; 8) generator of triggering and synchronization pulses; 9) S1-17 oscillograph; 10) IKS-12 monochromator.

2. RESULTS OF EXPERIMENTS

A study of the dependence of the nature of the radiation on the gas pressure p and on the amplitude of the voltage pulse E applied to the laser showed that the radiation pulse had a complex form due to the superposition of various spectral components shifted relative to one another along the time axis. Initially, a large-amplitude short-duration component appeared at 9.5 μ. Next, after several microseconds, a second component of much lower amplitude and longer duration appeared at 10.6 μ. The time lag of the second component depended appreciably on the CO_2 pressure and on the applied voltage pulse.[1)]

Figure 2 shows a series of oscillograms illustrating the nature of the observed radiation and its dependence on the CO pressure. Qualitatively similar dependences were obtained when the voltage was varied: an increase of the pressure produced the same effects as a decrease of the voltage applied to the laser electrodes. These oscillograms and the dependences of the principal parameters of the pulses on the pressure (Fig. 3) indicated that the amplitude of the 9.5 μ line had a well-defined maximum, which disappeared at pressures higher than 3 torr. When the CO_2 pressure was increased, the time lag before the emission of a light pulse (measured from the beginning of the current pulse) decreased. This effect was particularly strong for λ = 10.6 μ, so that at pressures above 2.5 torr the 9.5 and 10.6 μ lines were generated practically simultaneously. A very important property was the power of the generated radiation. Direct calorimetric measurements

[1)] The spectral components observed at 9.5 and 10.6 μ each consisted of separate lines representing various vibration–rotation transitions which occurred at different times. Because of the poor resolution of our apparatus, it was difficult to resolve in detail these lines. However, the fine structure of the laser radiation appeared particularly clearly in the region of the first spectral component and this structure increased the duration of the first component at high values of E/p. At low values of E/p, the number of additional rotational transitions decreased. As the energy of the transition decreased, the time lag of the emission increased.

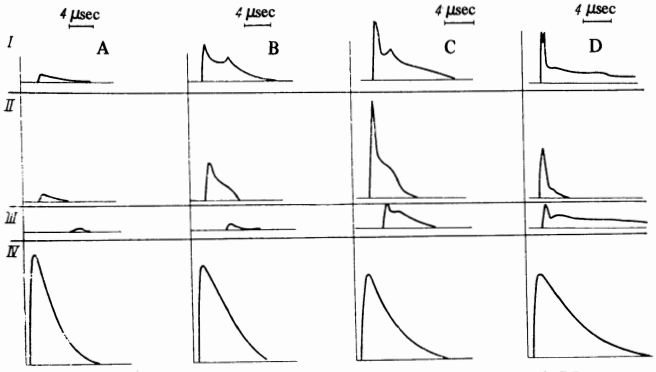

FIG. 2. Oscillograms of laser radiation pulses; A) p = 0.75 torr; B) p = 1 torr; C) p = 1.5 torr; D) p = 2.5 torr. Electric field E = 50 V/cm. I) Unresolved radiation-pulse spectrum; II) $\lambda = 9.5\ \mu$ radiation pulse; III) $\lambda = 10.6\ \mu$ radiation pulse; IV) discharge current pulse (the ordinate scales are relative).

showed that when the pulse duration was 8–10 μsec, the power emitted in the form of laser radiation was 3–5 kW, which was comparable with the emission powers of $CO_2 + N_2 +$ He lasers working under the Q-switching conditions and producing pulses lasting hundreds of nanoseconds. The most interesting result was the generation of a strong $\lambda = 9.5\ \mu$ line and of time-shifted spectral components whose time lag was altered by variation of the value of E/p.

3. DISCUSSION OF RESULTS

In considering the mechanism of the operation of CO_2 lasers, the most interesting point is the nature of the excitation producing population inversion and the competition between the various transitions responsible for the characteristic features of the laser radiation spectrum. The reported experimental data show that the phenomena which take place during the pulse excitation of CO_2 lasers are quite complex, and at present only some qualitative conclusions can be drawn about the processes taking place in these lasers. The very short time lag of the first spectral component indicates that the population inversion necessary for the generation of this component is due to inelastic collisions of electrons with CO_2 molecules. In this case, $A_1 \propto (N_2 - N_1)$, where N_1, N_2 are the populations of the lower and upper active levels, which, in this case, are the 02^00 and 00^01 levels. On the other hand, $N_1 \propto n_e'\langle\sigma_{01}v_{e1}\rangle N_0$ and $N_2 \propto n_e''\langle\sigma_{02}v_{e2}\rangle N_0$, where σ_{01} and σ_{02} are the cross sections for the excitation of CO_2 molecules, by electron impact, to the 02^00 and 00^01 levels, respectively. Here, N_0 is the concentration of CO_2 molecules in the discharge; n_e' and n_e'' are the densities of electrons having suitable energies for the excitation of the molecules to the active levels; v_{e1}, v_{e2} are the velocities of such electrons. Assuming, in the first approximation, that $n_e' = n_e'' = n_e$, we can write A_1 in the form

$$A_1 \sim n_e\langle\sigma_{02}v_{e2} - \sigma_{01}v_{e1}\rangle N_0 = n_e\gamma N_0. \qquad (1)$$

Thus, if the population inversion, necessary for the $00^01 - 02^00$ transition, does indeed take place due to electron impact, the amplitude of the radiation pulse should be governed by the density of electrons and CO_2 molecules in the discharge and it should also depend linearly on the value of γ. On the other hand, n_e, γ, and

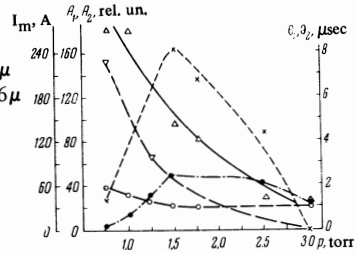

FIG. 3. Dependences of the experimental values of the laser radiation parameters on the CO_2 pressure: X) amplitude of the 9.5 μ line (A_1); ●) amplitude of the 10.6μ line (A_2); O) time lag between the beginning of a discharge and the laser emission (θ_1); ▽) time lag between the emission of the 9.5 and 10.6 μ lines (θ_2); Δ) amplitude of the discharge current pulse (I_m). E = 50 kV/cm.

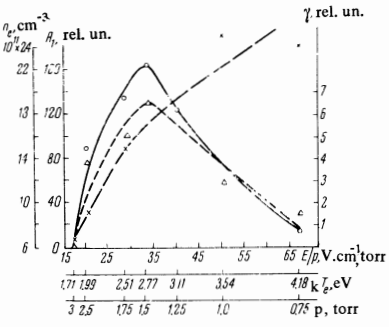

FIG. 4. Dependences of the laser radiation parameters on the ratio E/p for E = 50 kV/cm: O) amplitude of the 9.5 μ line (A_1); X) electron density in the discharge (n_e); Δ) quantity γ representing the difference between the populations of levels.

N_0 are single-valued functions of the electric field E and of the pressure p, as well as of the ratio E/p. Thus, we can find the dependence of γ on the energy of electrons taking part in the excitation by investigating the dependence $A_1 = f(E/p)$ and by measuring independently the value of n_e. The dependence of γ on the electron energy has not yet been investigated for CO_2 molecules but, by analogy with other molecular gases such as N_2 or CO, it should be of the resonance type with a maximum at energies of the order of 2–3 eV. The value of E/p can be changed in two ways: either by varying the electric field or by altering the pressure. We used both methods.

Figure 4 shows the dependences $\gamma = f(E/p)$, $n_e = f(E/p)$, plotted using the results given in Fig. 3; these dependences were obtained by varying the gas pressure in the system and keeping the electric field intensity constant.[2)] The dependence $A_1 = f(E/p)$ is included for convenience in Fig. 4. When E/p was increased, the value of n_e increased and N_0 decreased. The changes in these two quantities approximately compensated each other and the dependence of γ followed the dependence of the amplitude of the first component A_1. The dependences of A_1 and γ on E/p and, consequently, on the energy of electrons causing the excitation, were of the resonant type. Assuming that the electron distribution was Maxwellian, the energy corresponding to the maximum was 2.7 eV.[3)]

Figure 5 shows curves similar to those in Fig. 4 but in this case the amplitude of the voltage pulse was varied while the pressure was kept constant. It was found,

[2)] The electron density n_e was deduced from the experimentally determined discharge current and drift velocity, calculated from the value of E/p using the results given in [3]. The values of n_e obtained in this way could have been overestimated somewhat (up to 15%) because the cathode voltage drop was ignored in the calculation of these values.

[3)] This energy should be regarded as an estimate because the cathode voltage drop was ignored in the calculation of its value.

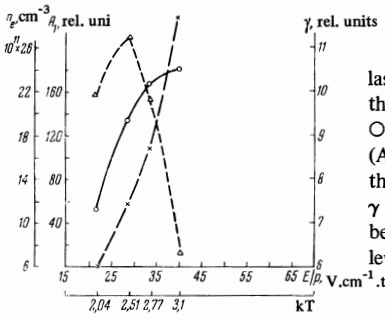

FIG. 5. Dependences of the laser radiation parameters on the ratio E/p for p = 1.5 torr: O) amplitude of the 9.5 μ line (A_1); X) electron density in the discharge (n_e); Δ) quantity γ representing the difference between the populations of levels.

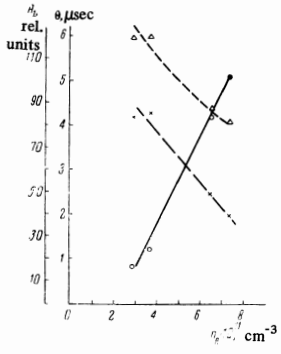

FIG. 6. Dependences of the amplitude of the 9.5 μ line (A_1) and of the time lags of the 9.5 μ (θ_1) and 10.6 μ (θ_2) lines on the electron density in the discharge (n_e). O) A_1; X) θ_1; Δ) θ_2. kT_e = 2.27 eV; p = 1.15 torr; E/p = 25.4 V.cm^{-1}.torr^{-1}.

as in the preceding case of constant electric field and varying pressure, that the value of γ exhibited a resonance dependence with a maximum corresponding to an energy of 2.5 eV, which could be regarded as being in good agreement with the value found in the preceding case, bearing in mind the experimental error. Since the dependence $v_e(E/p)$ was fairly weak and had no inflection points in the region of maximum of $\gamma = f(E/p)$ and since $\sigma_{02} > \sigma_{01}$, we could assume (bearing in mind the approximations made) that the resonance value was 2.5 eV or less if the cathode voltage drop was considerable. This estimate was reasonable when compared with the positions of the maximum values of the excitation cross sections of such molecular gases as nitrogen and carbon monoxide, for which ϵ_{max} was found to be 2.3 and 1.85 eV, respectively.

Figure 6 shows curves recorded keeping E/p constant and varying the current pulses by connecting ballast resistances. This altered the electron density. The electron energy was 2.2 eV. When the electron density n_e was increased the time lag of the first spectral component, θ_1, decreased and its amplitude A_1 increased linearly in accordance with Eq. (1). All these results, and particularly the resonance nature of the dependence of the excitation cross sections of the laser levels on the field-pressure ratio, convincingly support the mechanism of direct electron excitation of the vibrational levels responsible for the 9.5 μ laser emission. These results made it possible to estimate the lower limit of the cross section for the excitation of the upper laser level 00^01.

Since the energy q recorded in a single pulse (per unit volume of the active gas) was governed by the difference between the populations at the moment when the emission began, i.e.,

$$q = h\nu(N_2 - N_1) = n_e v_e(\sigma_{02} - \sigma_{01})N_0 t_p h\nu,$$

where t_p was the effective duration of the 9.5 μ pulses, and bearing in mind that $\sigma_{02} > \sigma_{01}$, we found that $\sigma_{02} \geq 1.4 \times 10^{-16}$ cm^2. This calculation was carried out using the following values of the parameters:

$$n_e = 1.7 \cdot 10^{12}\,\text{cm}^3; \quad v_e = 9.9 \cdot 10^7\,\text{cm/sec} \quad N_0 = 4.5 \cdot 10^{16}\,\text{cm}^{-3}; \quad t_n = 2 \cdot 10^{-6}\,\text{sec}.$$

There are no published values of the cross sections of the excitation of CO molecules by inelastic collisions with electrons. The only result with which we could compare our cross section was that given in[3], where the total cross section of elastic and inelastic conditions was given as 8.5×10^{-16} cm^2 for similar conditions and which naturally was larger than our value of σ_{02}.

We shall now consider the problem of the time lag between the first and second spectral components. At high values of E/p, Figs. 2 and 3 show that the 00^01-02^00 transition takes place first and the 00^01-10^00 line begins to be emitted after about 8 μsec. However, at low values of E/p, the line at $\lambda = 9.5\ \mu$ disappears and the time lag before the emission of the line at $\lambda = 10.6\ \mu$ reaches its lowest value of about 1 μsec. This indicates that at high values of E/p the gain for the 00^01-02^00 transition is initially higher than the corresponding gain for the 00^01-10^00 line, but at low values of E/p the position is reversed. The main cause of the difference between these gains is the inequality of the corresponding inverted populations. At low signal levels, the amplification in a molecular system can be written in the form

$$K = e^{\alpha l_1}. \tag{2}$$

For the P branch,

$$\alpha = \frac{8\pi^3 c^4 K_{21}}{3kT_r}\left(\frac{2\pi kT}{M}\right)^{1/2}(J+1)\left\{N_2 B_1 \exp\left[-F_2(J)\frac{hc}{kT_r}\right] - N_1 B_2 \exp\left[-F_1(J+1)\frac{hc}{kT_r}\right]\right\}, \quad F(J) = BJ(J+1), \tag{3}$$

where M is the molecular weight; J is the rotational quantum number; $B_{1,2}$ are the rotational constants; K_{21} is the matrix element of the transition; T_g is the temperature of the gas.

Since, in our case, the time lag θ_2 before the appearance of the second spectral component depends strongly on the electric field and the gas pressure and since, in accordance with[1,5], the matrix elements of the transitions 00^01-10^00 and 00^01-02^00 differ slightly, we may assume that all the quantities occurring in Eq. (3)—with the exception of the populations N_1 and N_2—cannot alter greatly the value of α for the observed transitions. On the other hand, the difference between the populations of the 02^00 and 10^00 levels may be due to the difference between the degree of occupancy of these levels and the nature of their relaxation. In particular, the observed time lags may be explained by assuming that the maxima of the electron-impact excitation cross sections of the CO_2 molecule are shifted along the electron energy scale so that $\epsilon^{max}_{02^00} > \epsilon^{max}_{00^01} > \epsilon^{max}_{10^00}$. Then, at high values of E/p, the 10^00 level is filled to a considerable degree and we observe first the line corresponding to the 00^01-02^00 transition. The appearance of the line at $\lambda = 10.6\ \mu$ after a time lag may be due to the pumping of the upper level by the displacement (after the beginning of the discharge) of the electron energy distribution maximum in the direction of lower energies and a consequent increase of the rate of filling of the 00^01 level. Some role in the observed time lags may also be

played by the processes of suppression of the lower laser levels, particularly by collisions with CO molecules, which are formed in the discharge by dissociation.[6,7] At low values of E/p, the 10^00 level is not strongly populated with molecules excited by electron impact and, therefore, the delay in the appearance of the $\lambda = 10.6\ \mu$ line is short.

However, we must mention that, since such important characteristics of the laser discharge in CO_2 as the excitation cross sections of the levels, the electron energy distribution functions, etc., are not yet known, our conclusions cannot be regarded as final and additional investigations are necessary to obtain a complete understanding of the observed relationships.

In conclusion, we shall consider the problem of the structure and duration of the 10.6 μ pulses. We can see from Fig. 2 that when the CO_2 pressure is increased the time lag θ_2 decreases and the duration of the $\lambda = 10.6\ \mu$ pulses increases. Moreover, a definite structure appears in these pulses; first, we observe a relatively short peak, then a dip, and later a plateau-like region of up to 40 μsec duration. The first peak is due to the excitation of molecules to the 00^01 level by electron impact, while the plateau of the radiation pulse is due to the excitation of CO_2 molecules to the same level by resonance interactions with CO molecules formed during the discharge. This interpretation is supported also by the fact that a reduction in the rate of pumping of the gas through the discharge gap increases the duration of the radiation pulse. Moreover, to obtain information on this point, we compared the radiation pulses produced in pure CO_2 and in CO_2 containing a small admixture of nitrogen; the pressure in pure CO_2 was such that the current pulses were the same in both cases. The form of the initial part of the radiation pulse and the time lag was practically unaffected by the admixture of nitrogen but the "tail" of the 10.6 μ pulse became considerably longer when nitrogen was added to the carbon dioxide. Bearing in mind that the role of N_2 in the excitation of the upper laser level was considerably greater than the role of CO, we can conclude that when the CO_2 pressure is increased, the duration of the radiation pulse increases because of a rise in the number of CO molecules in the discharge.

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