Effect of Proton Beam on the Generation of a CO<sub>2</sub> Gas Laser

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The effect of passing a beam of protons of several MeV energy through the laser medium on the generation power of a  $CO_2$  gas laser is investigated. Introduction of the beam may either increase or decrease the generation power, depending on the composition of the gas mixture and the laser operating conditions. The results may be explained on the basis of an influence of the proton beam on the volt-ampere characteristics of the discharge and also by assuming the existence of several channels for population of the (001) vibrational level of the  $CO_2$  molecule.

 $\mathbf{I} \mathbf{N}^{[1]}$  we reported an increase of the generation power of a laser based on  $\mathbf{CO}_2$  by introducing a proton beam into the working mixture. The present paper is devoted to an explanation of the mechanism whereby the beam influences the generation, for the purpose of ascertaining the possibility of directly exciting a  $\mathbf{CO}_2$  laser by bombarding the gas mixtures with charged particles, and also to aid in the analysis of the physical phenomena occurring in such a laser under ordinary electricpumping conditions.

1. The experimental setup remained in principle the same as in<sup>[1]</sup>. A resonator of 1.7 m length was made up of an internal metallic flat mirror with eccentric hole of 10 mm diameter for the entry of the beam, and with an external spherical glass mirror (R = 3 m) with a central opening of 5 mm diameter for the emergence of the radiation. The working surface of the mirror was covered with gold by vacuum sputtering. The discharge gap was 85 cm. Gases of technical purity were used. The system operated in the flow-through regime with a gas flow rate of  $\sim 1 \text{ m/sec}$ . The laser generation power was registered with an evacuated thermopile designed by B. P. Kozyrev and with a M-95 instrument. A beam of protons of energy 2.0-2.5 MeV and current  $10-30 \ \mu A$  was introduced into the resonator. The power scattered by the beam in the working volume of the tube did not exceed 10 W.

All the experiments were performed with  $CO_2$  gas and with  $CO_2-N_2$  mixtures at different partial pressures of the component and at total pressures ranging from 1 to 10 mm Hg. In each case we measured the generation power as a function of the discharge current, and also the corresponding current-voltage characteristic of the discharge.

The experimental results, some of which are shown in Figs. 1–3, point to the following conclusion. In all the electric-current and pressure ranges investigated by us, in the case of  $CO_2$  gas, passage of a beam of protons through the discharge increases the generation power (see Fig. 1). On the other hand, for mixtures with an appreciable admixture of N<sub>2</sub>, the generation power is increased by the proton beam only at those gas-discharge laser regimes in which the current greatly exceeds a certain optimal value corresponding to the maximum generation power and depending on the concrete gas mixture (see Figs. 2 and 3). 2. To explain the experimental results, we assume that the main influence of the proton beam on the generation mechanism reduces to the following. The proton beam introduced into the discharge is an additional source of electron ionization and therefore changes the current-voltage characteristic of the discharge in such a way that the previous values of the current correspond to smaller values of the electric field (see Figs. 1, 2, and 3).

The decrease of the electric field, in turn, changes the electron energy distribution function and consequently changes the kinetics of the population of the working levels of the  $CO_2$  molecule. The direct influence of the proton beam on the electron distribution function can be neglected since the relatively small change of the electric field offers evidence of comparable production rates of the electrons in the gas discharge itself and by the external ionizing source. (Direct population of the molecular vibrational levels by the protons is obviously negligible, for otherwise the proton beam would never lead to enhancement of generation, since it does not have, in contrast to the electrons of the gas discharge, the ability to populate selectively the working levels.)

The mechanism of the physical processes in the gas discharge plasma of a laser based on CO2 is quite complicated and has not yet been fully explained to date. The main difficulties are connected with the very approximate knowledge of the electron energy distribution function and with the dissociation of the CO<sub>2</sub> molecules in the discharge, a dissociation that can either improve the operation of the laser or make it worse, depending on the pressure of the working mixture, on its composition, on the electric characteristics of the discharge, and on the rate of flow of gas through the resonator (see, for example, [2,3]). The reason for this is that the dissociation of the CO<sub>2</sub> molecules, by decreasing the number of actually working molecules, at the same time produces an additional channel for populating the upper laser level, by resonant transfer of the energy of the vibrationally excited CO molecules to the CO<sub>2</sub> molecules (either directly or via  $N_2$ ).

Let us therefore analyze our experimental results by considering three possible mechanisms of populating the upper laser level (001): (i) direct electron impact, (ii) resonant energy transfer from the vibrationally ex-



FIG. 1. Dependence of the generation power and of the electric field intensity on the discharge current.  $P_{CO_2} = 1.5 \text{ mm Hg}$ ,  $\bullet$ -discharge without beam,  $\bigcirc$ -discharge with beam.



FIG. 2. Dependence of generation power and electric field intensity on the discharge current.  $P_{CO_2} = 1 \text{ mm Hg}$ ,  $P_{N_2} = 1.7 \text{ mm Hg}$ ,  $\bullet$ -discharge without beam, O-discharge with beam.



FIG. 3. Dependence of the generation power and of the electric field intensity on the discharge current.  $P_{CO_2} = 1.3 \text{ mm Hg}$ ,  $P_{N_2} = 2.6 \text{ mm Hg}$ ,  $\bullet$ -discharge without beam, O-discharge with beam.

cited  $N_2$  molecules to the  $CO_2$  molecules, and (iii) resonant energy transfer of the vibrationally excited CO molecules to the  $CO_2$  molecules.

The rate of population of the upper laser level by the electrons directly is determined by the following expression:

$$W_{e \operatorname{CO}_2} = \left(\frac{2}{m}\right)^{\frac{1}{2}} \int_{0}^{\infty} \varepsilon f(\varepsilon) \, \sigma_{e \operatorname{CO}_2}(\varepsilon) \, d\varepsilon \, n_e n_{\operatorname{CO}_2} \equiv K_{e \operatorname{CO}_2} n_e n_{\operatorname{CO}_2}. \tag{1}$$

Here  $n_e$  and  $n_{CO_2}$  are the densities of the electrons and of the unexcited  $CO_2$  molecules;  $\sigma_{eCO_2}$  is the cross section for the excitation of the (001) level of the  $CO_2$  molecules by electron impact; m is the electron mass;  $f(\epsilon)$ is the electron energy distribution function normalized as follows:

$$\int_{0}^{\infty} f(\varepsilon) \, \varepsilon^{1/2} \, d\varepsilon = 1.$$

The rate of population of the upper laser level by the excited  $N_2$  and CO molecules depends on the rate of excitation of the resonant levels of these molecules by the electrons:

$$W_{eN_2} = \left(\frac{2}{m}\right)^{1/2} \int_{0}^{\infty} \varepsilon f(\varepsilon) \sigma_{eN_2}(\varepsilon) d\varepsilon \, n_e n_{N_2} \equiv K_{eN_2} n_e n_{N_2}, \qquad (2)$$

$$W_{eCO} = \left(\frac{2}{m}\right)^{\eta_{e}} \int_{0}^{\varepsilon} ef(\varepsilon) \sigma_{eCO}(\varepsilon) d\varepsilon \, n_{e} n_{CO} \equiv K_{eCO} \, n_{e} n_{CO}. \tag{3}$$

Here  $\sigma_{eN_2}$  and  $\sigma_{eCO}$  are the cross sections for the excitation of the first eight vibrational levels of the  $N_2$  and CO molecules by electron impact, and  $n_{N_2}$  and  $n_{CO}$  are the concentrations of the unexcited  $N_2$  and CO molecules.

It is seen from (1)-(3) that the rate of population of the upper laser level of the CO<sub>2</sub> molecule by direct electron impact and the rates of population of the vibrational levels of the CO and N<sub>2</sub> molecules are determined both by the total electron density and by the form of the electron energy distribution function.

The passage of the proton beam through the gas discharge leads, as already noted, to a decrease of the electric field at a fixed value of the gas-discharge current. The decrease of the electric field, naturally, shifts the electron distribution function towards lower energies. In addition, the indicated change of the electric field leads to a change of the electron concentration that is established in the weakly-ionized gas-discharge. Indeed, at a constant value of the gas-discharge current the electron concentration in the positive column of a glowing gas discharge is inversely proportional to the electron drift velocity  $v_{dr}$ , which at a specified partial composition of the gas mixture is a function of the parameter E/N (E is the electric field intensity and N is the total molecular concentration).

The determination of the electron distribution function in  $CO_2$  lasers has been the subject of many investigations (see, for example,  $[4^{-6}]$ ), from which it follows that in the investigated discharges the relative weight of the low-energy electrons is larger than in either a Maxwellian or in a Druyvesteyn distribution function.

To interpret our experimental data, we can use the results of Nighan<sup>[6]</sup>, who solved numerically the Boltzmann integro-differential equation and calculated with the aid of the obtained electron distribution function the rates of excitation of the molecular levels, and also the average electron energy. The results were obtained for a large number of laser gas mixtures based on  $CO_2$  as functions of the parameter E/N. With the aid of these data, and also with the aid of the measured electron mobilities<sup>[7]</sup>, it is possible to plot the functions  $K_{eCO_2}$ ,  $N_2$ ,  $CO/v_{dr}$  against E/N, which determine the change of the rate of population of the molecular levels by the electrons at a constant value of the gas-discharge current.

Figure 4 shows the plot of  $K_{eCO_2}/v_{dr}$  against E/N for pure CO<sub>2</sub>. Figure 5 shows the analogous plot for pure N<sub>2</sub>. The solid line of Fig. 6 is the plot of  $K_{eCO}/v_{dr}$ against E/N for the gas mixture CO<sub>2</sub>: CO = 1:1, with account taken of the calculated data on the average electron energy established in the CO<sub>2</sub>-CO gas mixture<sup>[6]</sup>, and also of the fact that the electron mobility in this mixture is determined mainly by collisions with the CO molecules.

3. As already noted, a character of the change in the generation power when a proton beam passes through  $CO_2$  gas is entirely different from the case of  $CO_2$ -N<sub>2</sub> mixtures. We shall show that this difference is connected with the fact that in gas mixtures with an appreciable

 $10^{16} \text{ K}_{eCO_2}/\text{v}_{dr}, \text{ cm}^2$ 



FIG. 5. Plot of  $K_{eN_2}/v_{dr}$  against E/N for pure N<sub>2</sub>.

amount of  $N_2$  the main channel for populating the (001) level of the  $CO_2$  molecule is resonant energy transfer from the vibrationally-excited  $N_2$  molecules.

Let us consider a simplified scheme for producing inverted population of the vibrational  $CO_2$  molecule levels in  $CO_2$ - $N_2$  and  $CO_2$ -CO gas mixtures. Following the authors of<sup>[8,9]</sup>, we introduce the vibrational temperatures for each type of normal oscillations of the  $CO_2$ ,  $N_2$ , and CO molecules, namely,  $T_1$ ,  $T_2$ , and  $T_3$  are respectively the temperatures of the symmetrical, deformation and asymmetrical oscillation modes of the  $CO_2$  molecules, and  $T_4$  is the vibrational temperature of the  $N_2$  or CO molecules. Taking into account the strong energy coupling between the deformation and symmetrical modes<sup>[9]</sup>, we assume that  $T_1 = T_2$ .

Let us consider the kinetics of populating the working levels of the  $CO_2$  molecules in the  $CO_2$ -N<sub>2</sub> gas mixture. Assuming that the main channel for populating the (001) level of the  $CO_2$  molecule is the resonant energy transfer from the vibrationally-excited N<sub>2</sub> molecules to the asymmetrical oscillations of the  $CO_2$  molecules, we write down the energy-balance equations for the different vibrational modes in the following simplified form:

$$dE_{4}/dt = hv_{4}n_{S_{2}}\{an_{c}K_{e}n_{3} + n_{CO_{2}}x_{3}K_{34} - n_{CO_{2}}x_{3}K_{34}\} = 0,$$
  
$$dE_{3}/dt = hv_{3}n_{CO_{2}}\{n_{S_{2}}x_{3}S_{34}K_{34} + n_{CO_{2}}x_{2}^{3}S_{32}K_{32} - n_{S_{2}}x_{3}K_{34} - n_{CO_{2}}x_{3}K_{32}\} = 0,$$
  
$$dE_{3}/dt = hv_{3}n_{CO_{2}}\{n_{S_{2}}x_{3}S_{34}K_{34} + n_{CO_{2}}x_{2}^{3}S_{32}K_{32} - n_{S_{2}}x_{3}K_{34} - n_{CO_{2}}x_{3}K_{32}\} = 0,$$

$$dE_2/dt = hv_2 n_{\rm CO_2} \{ 3n_{\rm CO_2} x_3 K_{32} - 3n_{\rm CO_2} x_2^3 \mathscr{C}_{32} K_{32} - N x_2 K_{20} \} = 0; \quad (4)$$

where  $\mathscr{E}_{ij} = \exp\{-\Delta \epsilon_{ij}/T\}$ ;  $x_i = \exp\{-h\nu_i/T_i\}$ ;  $\nu_i$  are the fundamental frequencies of the different modes; T is the gas temperature; and  $\alpha$  is the effective number of vibrational quanta excited in the electron-N<sub>2</sub> collisions.

The following molecular processes were taken into account in (4): (a) relaxation of the upper laser level of the CO<sub>2</sub> molecules:  $h\nu_3 \rightarrow 3h\nu_2 + \Delta\epsilon_{32}$  with probability  $K_{32}$ ; (b)  $h\nu_2 \rightarrow$  kinetic energy of the CO<sub>2</sub> and N<sub>2</sub> molecules, with probability  $K_{20}$ ; (c) resonant energy transfer from the vibrationally-excited N<sub>2</sub> molecule to the asymmetrical oscillations of the CO<sub>2</sub> molecules:  $h\nu_4 + \Delta\epsilon_{34}$  $\rightarrow h\nu_3$  with probability  $K_{34}$ .

It should be borne in mind that besides the process

(a) there occur also other processes that lead to energy transfer from the asymmetrical modes to the deformation and symmetrical modes of the  $CO_2$  molecule. For example,

$$hv_3 \rightarrow hv_1 + hv_2 + 272 \text{ cm}^{-1}, \quad hv_3 \rightarrow 4hv_2 - 204 \text{ cm}^{-1}.$$
 (5)

At the present time, there are no data on the rates of all these individual processes, theoretically these rates cannot differ strongly, and in the experiment the corresponding relaxation losses (including the processes (5)) make comparable contributions to  $K_{32}$ . Since we have used in all the estimates the experimental values of  $K_{32}^{[0]}$ , it follows, generally speaking, that effective account is taken of all the processes that lead to vibrational relaxation of the upper laser level.

After solving (4) for the quantities  $x_i$ , the inverted population of the laser levels can be estimated as follows:

$$n_{\rm CO_2}(x_3 - x_2^2) = \alpha n_e K_e N_2 n_{\rm N_2} / n_{\rm CO_2} K_{32}.$$
(6)

We consider now the kinetics of populating the working levels of the  $CO_2$  molecule in a gas mixture containing no N<sub>2</sub>. Assuming that the main channel of populating the (001) level of the  $CO_2$  molecule is direct electron excitation, we write down the energy-balance equation for the deformational and asymmetric modes in the following form:

$$dE_{2}/dt = hv_{3}n_{\text{cO}_{2}}\{n_{c}K_{s}c_{0} - n_{c}c_{2}x_{3}K_{32} + n_{c}c_{2}x_{3}^{2}\mathscr{F}_{32}K_{32}\} = 0,$$
  

$$dE_{2}/dt = hv_{2}n_{c}c_{0}\{3n_{c}c_{2}x_{3}K_{32} - 3n_{c}c_{2}x_{2}^{3}\mathscr{F}_{32}K_{32} - Nx_{2}K_{20}\} = 0.$$
(7)

By solving these equations, we can obtain the following estimate for the inverted population:

$$n_{\rm CO_2}(x_3 - x_2^2) = n_e K_{e \, \rm CO_2} / K_{32}. \tag{8}$$

If we assume, on the other hand, that in the absence of  $N_2$  the main channel of populating the (001) level is resonant energy transfer from the vibrationally-excited CO molecules produced in the discharge as a result of the dissociation of the  $CO_2$  molecules, then the estimate for the inverted population can be obtained in analogy with (6):

$$n_{\rm CO_2}(x_3 - x_2^2) = \beta n_e K_e \operatorname{co} n_{\rm CO} / n_{\rm CO_2} K_{32}; \tag{9}$$

here  $\beta$  is the effective number of the vibrational quanta excited in electron-CO collisions.

With the aid of (6), (8), and (9) we can obtain a semiquantitative interpretation of our experimental results.

We assume that in the absence of  $N_2$  the dissociation of  $CO_2$  is low and the main channel for populating the (001) level is direct electronic excitation. It can then be seen from (8) and from Fig. 4 that by turning on a proton beam, which lowers the electric field in the discharge at each specified value of the current, more effective conditions are created for generation at the experimentally realized values of the parameter E/N. This agrees with the experimentally observed increase of the generation power, shown in Fig. 1.

We now make another assumption, namely that dissociation of  $CO_2$  in the absence of  $N_2$  is high (about 50%) and that the main channel for populating the (001) level is resonant energy transfer from the vibrational excited CO molecules to the  $CO_2$  molecules. It is then seen from (9) and from Fig. 6 that the decrease of the electric field at a specified value of the gas-discharge cur-



FIG. 6. Plot of  $K_{eCO_2}/v_{dr}$  against E/N. Solid line-mixture composition  $CO_2$ : CO = 1:1, dashed-CO<sub>2</sub>-CO mixture with CO<sub>2</sub> predominating.

rent produces conditions that are less effective for the generation at the experimentally realized values of the parameter E/N, i.e., the passage of the proton beam through the gas mixture should decrease the generation power.

Let us obtain, finally, one more evaluation of the experimental results. We assume that the dissociation of the CO<sub>2</sub> is low, but the inverted population is produced mainly via the CO molecules. In this case it can be assumed that both the distribution function and the mobility of the electrons are determined by the collisions with the CO<sub>2</sub> molecules. Then, using the calculated distribution function of the electrons in pure  $CO_2$  gas<sup>[6]</sup> and the measured values<sup>[10]</sup> of the cross section for electronic excitation of the first eight vibrational levels of the CO molecule, we can plot  $K_{eCO}/v_{dr}$  against E/N for  $CO_2$  gas with a low degree of dissociation. The results of such a calculation are shown by the dashed line of Fig. 6. As seen from this figure, the population of the (001) level via the CO molecules produced as a result of weak dissociation of CO2 also explain the favorable influence of the proton beam on the generation power (Fig. 1).

For a qualitative explanation of the influence of the proton beam on the generation power of  $CO_2$ - $N_2$  gas mixtures with a predominant amount of  $N_2$  molecules, we can use expression (6) and the  $K_{eN_2}n_e \sim K_{eN_2}/v_{dr}$  plot of Fig. 5. It is seen from this figure that passage of the proton beam, which lowers the electric field intensity in the discharge at each specified value of the current, creates conditions that are less favorable for the generation at the experimentally realized values of the parameter E/N (see Figs. 2 and 3). Therefore the negative influence of the proton beam on the generation power in the  $CN_2$ - $N_2$  mixtures in the region of low gas-discharge currents can be attributed to the shift of the electron distribution function.

On the other hand, the enhancement of the generation by the proton beam at large gas-discharge current may be due to a number of causes. Principal among them is apparently the lowering of the gas temperature when the beam is turned on. Indeed, at a fixed value of the gasdischarge current, the Joule energy released in the discharge decreases with decreasing electric field. On the other hand, the direct heating of the gas where the proton beam can be neglected since the energy lost by the beam in the gas-discharge volume is much lower than the Joule energy released there.

The lowering of the gas temperature at low gasdischarge currents (lower than the optimal value for the concrete gas mixture) does not exert a noticeable influence on the generation power, for in this range of currents the gas temperature is still low and its small variations have little influence on the inverted population of the working levels. On the other hand, at currents that are larger than the optimal value, it is precisely the rise of the gas temperature which determines the decrease in the generation power with increasing gas-discharge current.

It can thus be concluded that the change observed  $in^{[1]}$  of the generation of a gas laser based on  $CN_2$  when a proton beam passes through the discharge is due to the influence of the photon beam only on the current-voltage characteristic of the discharge. It can be concluded from this that direct generation of standard gas mixtures based on  $CO_2$  by irradiation with high-energy charged particles is hardly realizable, since the conditions for producing inverted population of the laser levels are extremely sensitive to the electron distribution function. In the absence of an electric field, the maximum of the electron distribution function is shifted towards lower energies, which leads to a strong decrease of  $K_{eN_2}$ ,  $K_{eCO_2}$ , and  $K_{eCO}$ .

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