Gain of a CO_2–N_2–He atmospheric-pressure gas mixture excited by a quasistationary non-self-sustaining discharge

E. P. Velikhov, Yu. K. Zemtsov, A. S. Kovalev, I. G. Persantsev, V. D. Pis'mennyĭ, and A. T. Rakhimov

Nuclear Physics Institute, Moscow State University

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The time dependence of the gain of CO_2–N_2–He atmospheric-pressure gas mixtures excited by a quasistationary non-self-sustaining discharge is investigated. The stability limits imposed on the non-self-sustaining discharge by the dissociative capture of electrons by CO_2 molecules are also investigated.

An earlier paper[1] reported on the quasistationary generation observed in a CO_2—N_2—He gas mixture at atmospheric pressure, in which a low-current electron beam maintained a quasistationary non-self-sustaining discharge. In the present paper we report an investigation of the temporal and spatial characteristics of the gain realized in such discharges. We also investigate some aspects of the stability of non-self-sustaining discharges in gas mixtures containing CO_2.

1. The maximum duration of generation of a high-pressure CO_2 laser is theoretically limited by a single factor: gas heating during the generation pulse that is totally uncompensated for by heat transfer to the walls of the discharge chamber. Gas heating by the generation pulse changes the relaxation times of the laser levels and the population of the lower laser level in such a way that the medium ceases to be optically active after a certain period of time. Furthermore, gas heating causes the development of overheat instability limiting the homogeneous stage of the non-self-sustaining discharge[2].

The characteristic time of development of overheat instability

$$\tau = \frac{\gamma}{\gamma-1}\frac{P}{W}$$

(γ is the gas adiabatic exponent, P is gas pressure, and W is the electric power input) is in good agreement with experiments involving non-self-sustaining gas discharges that are free of ionization instabilities[3-5]. At the same time, it is difficult to make even simple estimates of the generation time as a function of the electric power input and of the various plasma parameters because of the strong temperature dependence of the relaxation times of the laser levels of the CO_2 molecule[6]. Therefore, numerical computations of the time behavior of gain in CO_2—N_2—He mixtures at atmospheric pressure, taking into account the rise in gas temperature during the pulse, were performed to obtain the theoretical characteristics determining the generation pulse length.

The equations determining the population of molecular levels are written in the usual approximation of three-mode temperatures[7]:

$$\frac{d\varepsilon_4}{dt} = \frac{W}{h\nu_4 n_{N_2}} - K_{34}(\varepsilon_4-\varepsilon_3)n_{CO_2},$$

$$\frac{d\varepsilon_3}{dt} = K_{34}(\varepsilon_4-\varepsilon_3)n_{N_2} - \frac{1}{8}(K_{32CO_2}n_{CO_2}+K_{32N_2}n_{N_2} + K_{32He}n_{He})[\varepsilon_3(2+\varepsilon_2)^3 - e^{-\tilde{\varepsilon}/T}\varepsilon_2^3(1+\varepsilon_3)],$$

$$\frac{d}{dt}\left[\frac{2\varepsilon_2+3\varepsilon_2^2}{2(1+\varepsilon_2)}\right] = \frac{3}{8}(K_{32CO_2}n_{CO_2}+K_{32N_2}n_{N_2} + K_{32He}n_{He})[\varepsilon_3(2+\varepsilon_2)^3 - e^{-\tilde{\varepsilon}/T}\varepsilon_2^3(1+\varepsilon_3)] - (K_{20CO_2}n_{CO_2}+K_{20N_2}n_{N_2}+K_{20He}n_{He})(\varepsilon_2-\varepsilon_2^0);$$

$$\varepsilon_i = c_i\exp(-h\nu_i/T_i)/[1-\exp(-h\nu_i/T_i)]. \quad (1)$$

Here ϵ_i is the average number of vibrational quanta per molecule in the corresponding vibrational modes and c_i is the degree of degeneracy of the corresponding mode. The indices i = 4, 3, 2 refer to the vibrational levels of N_2, and to the antisymmetric and deformational modes of CO_2, respectively; n_{CO_2}, n_{N_2}, and n_{He} are the concentrations of CO_2, N_2, and He molecules, $\epsilon_2 \equiv \epsilon_2(T)$, T is the gas temperature, $\tilde{\epsilon} = 500°$, K_{34} is the probability of vibrational exchange between N_2 and the antisymmetric mode of CO_2, K_{32} is the probability of vibrational-vibrational relaxation for the antisymmetric mode of CO_2, and K_{20} is the probability of vibrational-translational relaxation for the deformation mode of CO_2. We note that (1) is based on the assumption that the entire Joule energy liberated in the discharge is transformed into vibrational energy of the N_2 molecules.

The probabilities of intermode transitions depend strongly on the gas temperature, whose rise during a long quasistationary pulse determines the time behavior of the gain.

The gas energy balance equation determining the variation of gas temperature has the form

$$\frac{d}{dt}\left(\frac{n_{CO_2}}{\gamma_{CO_2}-1}+\frac{n_{N_2}}{\gamma_{N_2}-1}+\frac{n_{He}}{\gamma_{He}-1}\right) = {}^1/_8\tilde{\varepsilon}n_{CO_2}(K_{32CO_2}n_{CO_2}+K_{32N_2}n_{N_2}+K_{32He}n_{He})[\varepsilon_3(2+\varepsilon_2)^3 - e^{-\tilde{\varepsilon}/T}\varepsilon_2^3(1+\varepsilon_3)] + n_{CO_2}(K_{20CO_2}n_{CO_2}+K_{20N_2}n_{N_2}+K_{20He}n_{He})(\varepsilon_2-\varepsilon_2^0)h\nu_2. \quad (2)$$

Numerical solution of (1) and (2) was used to find the time behavior of the population of the vibrational levels of the CO_2 molecules and of the gas temperature. The absolute values of the transition probabilities and their temperature dependences in the range 300°K < T < 600°K were obtained by processing experimental data[6-9] and were used in the following form (all values are given in cm^3/sec):

$$K_{34} = 5\cdot 10^{-13}; \quad K_{32CO_2} = 1.31\cdot 10^{-10}\exp(-9.37/\theta^{1/3}),$$
$$K_{32N_2} = 2.42\cdot 10^{-10}\exp(-11.3/\theta^{1/3}), \quad K_{32He} = 1.9\cdot 10^{-11}\theta^{1/2}\exp(-8.79/\theta^{1/3}),$$
$$K_{20CO_2} = 4.28\cdot 10^{-11}\exp(-8.85/\theta^{1/3}),$$
$$K_{20N_2} = 3.18\cdot 10^{-11}\exp(-10.32/\theta^{1/3}),$$
$$K_{20He} = 4.8\cdot 10^{-11}\exp(-6.07/\theta^{1/3}).$$

In these expressions, $\theta = T/300°K$ is the normalized gas temperature.

Solution of (1) and (2) yields the time behavior of gain of the medium with the aid of the expression

$$K = \left(\frac{\lambda}{2\pi}\right)^2\frac{A}{\Delta\nu}\frac{2hcB}{T}(2j+1)\exp\left(-\frac{hcBj(j+1)}{T}\right) \times 16n_{CO_2}\frac{(1+\varepsilon_2)(4\varepsilon_3+4\varepsilon_2\varepsilon_3-\varepsilon_2^2)}{(2+\varepsilon_2)^6(1+\varepsilon_3)^2}. \quad (3)$$

Here λ is the wavelength of the coherent radiation, A is

the Einstein coefficient, B is the rotational constant, j = 20 is angular momentum, and $\Delta\nu$ is the impact width of the working transition.

Figure 1 shows some results of numerical computation of the gain corresponding to various values of Joule input power and various compositions of the CO_2—N_2—He gas mixture at atmospheric pressure. These diagrams show that optical activity of the medium lasting on the order of 100 μsec can be reached by maintaining power input at the level of a few kW/cm^3 in gas mixtures with a low concentration of CO_2. This results in an energy input of approximately 0.4 J/cm^3.

2. In order to verify the above results, experiments were performed to study the time behavior of gain during quasistationary excitation of the CO_2—N_2—He gas mixture. Figure 2 shows the experimental setup.

An electron beam 1 × 30 cm in cross section, serving as the external ionizer, was extracted from the vacuum diode into the discharge chamber through an aluminum-foil window. The electron energy was 120—160 keV, and the electron current pulse length was 1—3 msec. The distance between the anode and cathode of the discharge chamber was 2 cm. When the peak value of the electron beam current was reached, the anode of the discharge chamber was connected to a 37 μF capacitor charged to 5–15 kV.

The gain was measured from the oscilloscopic traces of the signal from the Ge-Au detector intercepting radiation from the driving CO_2 laser after amplification in a single pass through the discharge chamber. The driving-laser beam, 3 mm in diameter at the entrance to the active medium, passed through the discharge chamber at a distance of 0.5 cm from the anode. The selection of the anode region of the gas discharge as the site for measuring the time behavior of gain was based on the following considerations. An earlier report[5] indicated that in the investigated gas discharges there is significant propagation of heat from the cathode region, setting up a gas-density gradient and consequently a refractive-index gradient near the cathode and perpendicular to the direction of the amplified beam. This gradient causes refraction of the beam and is thus naturally capable of distorting the experimental results in gain measurements.

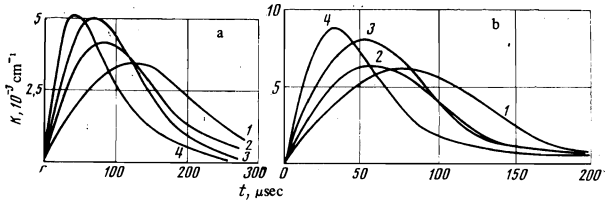

FIG. 1. Computed time behavior of gain. a—At a pump power level of 1 kW/cm^3; composition of N_2–CO_2–He mixture: 1–85:5:10; 2–8:1:1; 3–6:1:3; 4–5:2:3; b—at a pump power level of 2 kW/cm^3; composition of N_2–CO_2–He mixture: 1–65:5:30; 2–8:1:1; 3–6:1:3; 4–5:2:3.

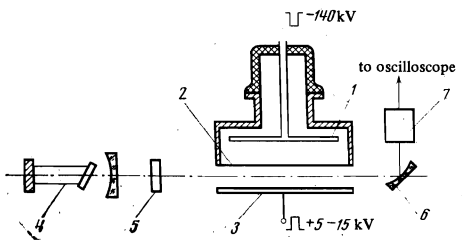

FIG. 2. Experimental setup. 1–Cathode assembly of electron gun; 2–aluminum foil window admitting electron beam to discharge chamber; 3–anode of discharge chamber; 4–driving laser; 5–shutter; 6–mirror; 7–Ge-Au photodetector.

To eliminate the effect of refraction in the gain measurements we performed experiments to study the deviation of the propagating beam as a function of its orientation in the discharge gap. The measurement method was based on the fact that the indices of refraction were practically identical in the investigated gases for visible and infrared radiation. Therefore a He-Ne laser beam 2 mm in diameter was allowed to pass for the duration of a current pulse through the discharge chamber parallel to the electrodes. The beam emerging from the discharge chamber was photographed with a streak camera. The streak photographs of the beam were synchronized with the oscilloscopic traces of the discharge current and voltage by means of two photoemitters in front of the film inside the streak camera. Figure 3 shows a typical streak photograph of an He-Ne beam propagating 3 mm from the cathode and the corresponding traces of the discharge current and voltage. We see the significant effect of light refraction in the near-cathode region during the periods in which the gain was studied. The broad horizontal band in the streak photograph corresponds to a strongly overexposed section of the film by multiple passes of the undeflected beam during shutter release.

These experiments showed that for all mixtures used and for pump powers up to 2 kW/cm^3, beam deflection was not observed if the beam passed at a distance greater than 14 mm from the cathode. At shorter distances we observed deflection of the beam during the non-self-sustaining pulse, and the magnitude of the deflection depended on both the pump power and the distance from the cathode. For this reason the beam of the driving CO_2 laser was passed through the anode region in all experiments investigating the time behavior of the gain.

Figure 4 shows the experimental results from study of the time behavior of gain for various gas mixtures at pump powers of 1 and 2 kW/cm^3. The current density of the electron beam was 110 μA/cm^2 and the desired pump power level was reached by selecting an appropriate electric field intensity of the non-self-sustaining discharge.

The obtained experimental and theoretical results allow us to draw a number of conclusions as to the magnitude and time behavior of gain in CO_2—N_2—Ne mixtures at atmospheric pressure excited by quasistationary non-self-sustaining discharge. We see that in all mixtures excited by the investigated discharges the peak gain is at the level of 10^{-2} cm^{-1}, while the duration of population inversion at laser levels is of the order of 100 μsec. Therefore we can say that changes in gas composition within fairly wide limits have no pronounced effect on the optical characteristics of the investigated quasistationary discharges. On the other hand, a change in the composition of the gas mixture does have a significant effect on the current density of the non-self-sustaining discharge maintained by definite levels of the electron-beam power and the applied electric field. An increase in the proportion of CO_2 reduces considerably the conductivity of the plasma in the non-self-sustaining discharge.

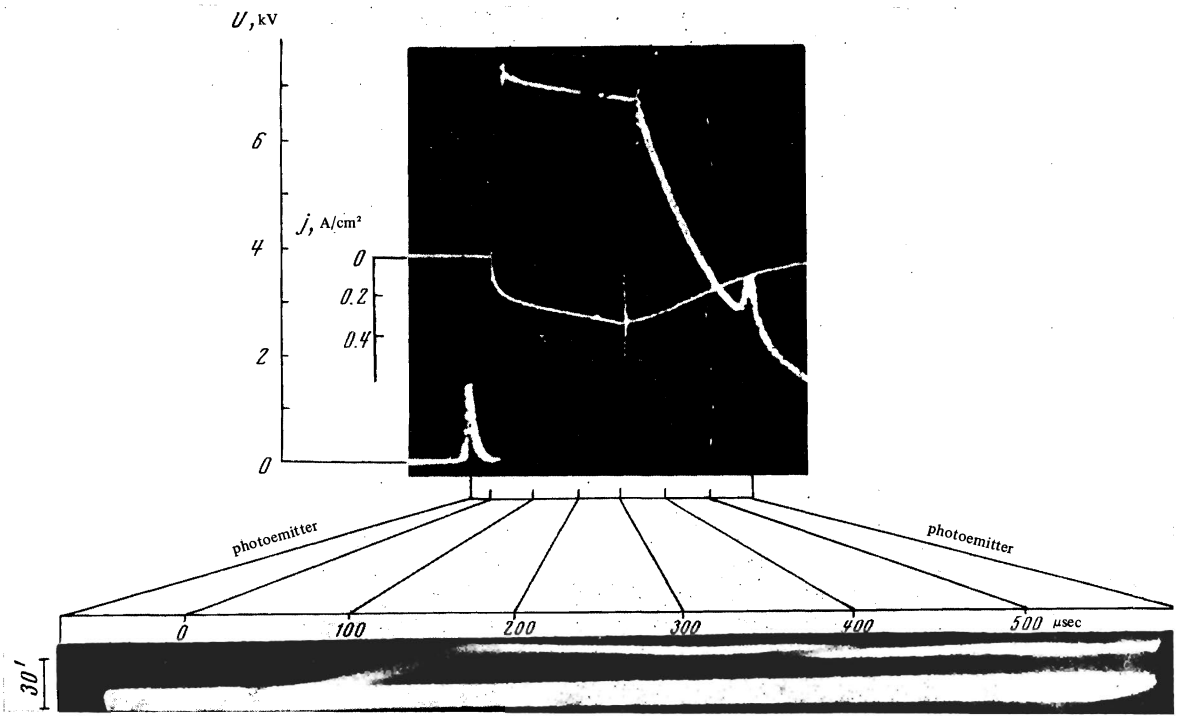

FIG. 3. Oscilloscopic traces of discharge current and voltage and streak photograph of He-Ne laser beam deflection due to refraction 3 mm from the cathode.

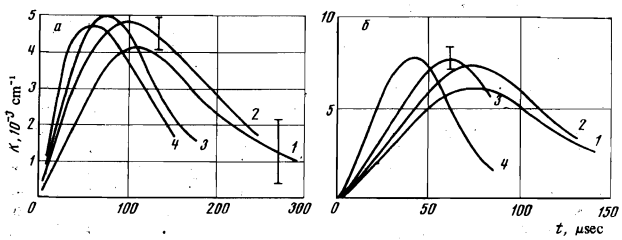

FIG. 4. Time behavior of gain. a–For pump power level of 1 kW/cm³; N_2–CO_2–He mixture composition: 1–8:1:1; 2–6:1:3; 3–4:1:5; 4–5:2:3; b–for pump power level of 2 kW/cm³; N_2–CO_2–He mixture composition: 1–8:1:1; 2–6:1:3; 3–4:1:5; 4–5:2:3.

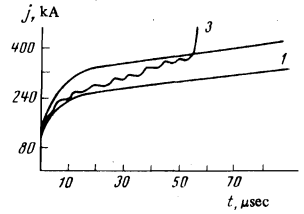

FIG. 5. Oscillocopic traces of discharge current. N_2–CO_2–He mixture composition: 5:2:3. 1–U = 8 kV; 2–U = 10 kV; 3–U = 12 kV.

The cause of the effect of CO_2 on plasma conductivity is the fact that the concentration of charged particles is sufficiently small ($n_e < 10^{12}$ cm^{-3}) in the investigated discharges and therefore dissociative capture of electrons by CO_2 molecules may play a significant role in the balance of charged particles. A direct experimental proof that dissociative capture of electrons is responsible for reducing plasma conductivity is the fact that above a certain value of E/P the discharge current in gaseous mixtures enriched with CO_2 decreases with increasing applied electric field. Such a relationship can be explained only if dissociative capture is the main mechanism of electron loss, since we know that the rate of dissociative capture of electrons by CO_2 molecules, in contrast to the electron recombination rate, increases strongly with increasing electron temperature within the range of the investigated values of E/P. Therefore in a non-self-sustaining discharge in which the electron generation rate in no way depends on electron temperature, the electron concentration can decrease with increasing applied E/P. This can lead in turn to a drop in the discharge current.

Figure 5 shows three oscilloscopic traces of the discharge current in a gas mixture containing 20% CO_2, corresponding to three different values of the electric field intensity. These traces clearly show the nonmonotonic nature of the volt-ampere characteristic of the non-self-sustaining discharge in a gas mixture enriched with CO_2. We can also see that the drop in peak current in a non-self-sustaining discharge is accompanied by small-scale oscillations on the current trace (trace 3 in Fig. 5). In our experiments, these oscillations always accompanied the above current drop; most likely they indicate the appearance of an instability of the non-self-sustaining discharge to striations formed perpendicular to the discharge current. In fact, this instability[10] derives from the predominant role of dissociative capture of electrons by CO_2 molecules in the charged-particle balance of a non-self-sustaining discharge plasma. The fluctuating growth of electron temperature here (as distinct from the usual situation) results in a reduced concentration of electrons which, in turn, causes a drop in plasma conductivity and further heating of the electrons at a fixed discharge-current density.

The result thus indicates the need to limit the partial pressure of the CO_2 and the electric field intensity in the excitation of CO_2–N_2–He gas mixtures by quasi-stationary non-self-sustaining discharges.

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