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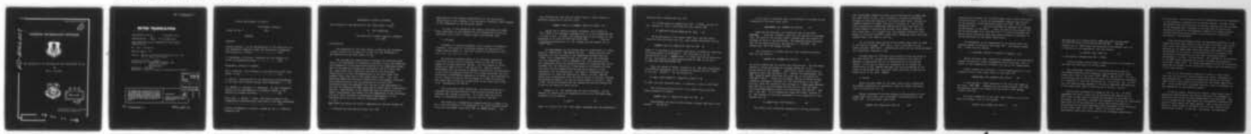

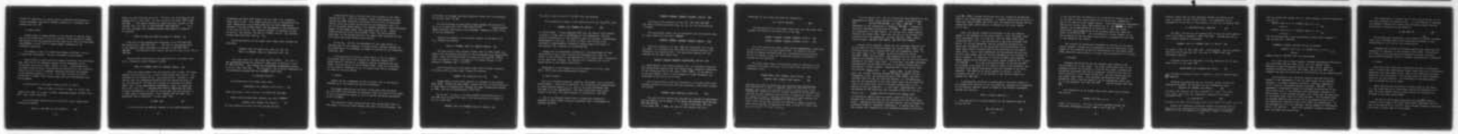

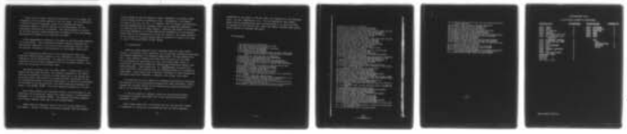

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THE INFLUENCE OF GAS ADDITIVES ON THE LASER EFFECT IN CO_2

By

Dan C. Dumitras

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Monographic studies of assembly

The influence of gas additives on the laser effect in CO_2*

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1.Introduction

The appearance of the laser effect in pure CO_2 was discussed in [1], the aim of this paper is to deal with the influence of gas additives on the laser effect in CO_2.

The gas additives significantly influence the processes which lead to the achievement of population inversion in CO_2, and especially the processes of collisional de-excitation of the vibrational energy. Most of the additives disactivate efficiently the 01^10 level from CO_2, where there was a choking on the way of the vibrational de-excitation, so that the depopulation of the lower laser level becomes faster than that of the higher laser level. Also, the additives change the excitation conditions in the plasma, increasing the production rate of the CO_2 molecules on the level 00^01, optimizing either the energetic distribution of the electrones, or the rate of producing Nitrogen molecules or carbon oxide, vibrationally excited. Nevertheless, it was shown [2] that gas additives do not modify significantly the density and the temperature of the electrones and thus, their influence is manifasted preponderantly in the modification of the life times of vibrational levels.

Some additives reduce the kinetic temperature of the gas through the

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modification of the thermic conductibility of the gas mixture, contributing to the increase in the population inversion, which depends on the temperature in the molecular lasers.

At last, in the sealed-off gas lasers, some geses are added, which contribute to the reduction in the dissociation of the CO_2 molecules, with a view to obtain longer functionning times.

2. Nitrogen

Nitrogen is a biatomic molecule, with a degree of freedom of vibration. The levels of vibrational energy are depicted by a single quantum number v; the first levels are situated at 2,331 cm^{-1} (v=1) and 4631 cm^{-1} (v=2).

The electric discharge leads to the efficient electronic excitation on the first 8 vibration levels of over 50% of the number of nitrogen molecules [3], [4], and this number is little affected by laser action in the CO_2-N_2 mix. Schultz [5], [6] established that the section of maximum collision for the excitation of the first 8 vibration levels of the nitrogen molecules with the electrones is 3×10^{-6} cm^2 and appears at an energy of the electrones of 2.3 eV, around the peak of the discharge of a laser with CO_2-N_2. The higher $N_2(v)$ levels are de-excitated on the lower $N_2(1)$ level in an approximate 1 μs time [7].

The N_2 molecule being homonuclear, its dipole radiation is forbidden and the deactivation of the vibration levels can be achieved only through collisions. It is said that the $N_2(v=1)$ level is a metastable state. Its life time is 1.36s (8).

The situation is dramatically changed when CO_2 is added to N_2. Due to the coincidence of the energies of the first vibration level of N_2 with the higher laser level 00^01 of $CO_2 (\Delta E = 18\ cm^{-1})$, the

CO_2 molecules can take over the energy from N_2, which leads to a selective population of the 00^01 level.

$$CO_2(000) + {}^{14}N_2(v = 1) \rightarrow CO_2(00^01) + {}^{14}N_2(v = 0) - 18\ \mathrm{cm}^{-1}. \quad (1)$$

Among the $V-V$ energy exchange processes, this process is very close to the energetic resonance and it is to be expected that its rate is big. At 300K, the transfer rate of the process (1) was determined to be $(1{,}6 \pm 0{,}1) \times 10^4\ s^{-1}\ torr^{-1}$ [7], [9]–[12]. The rate calculated by Gordietz et al. [13] through the SSH method [14] is $1 \times 10^4\ s^{-1}\ torr^{-1}$.

The measurements of the process rate (1) effectuated in a large range of temperatures (300–2500 K) [15]–[17], showed that the rate decreases with the increase in the temperature between 300K and 1,200K, with a dependence $T^{-3/2}$, and over 1,200K the rate increases with the temperature, with a dependence $T^{3/2}$. This variation with the temperature cannot be explained through the formalism SSH, which presumes that the interaction is dominated by the repulsive forces with short action between molecules and which predicts a rate increasing with the temperature. The negative dependence on the temperature to 1,200K was explained by Sharma and Brau [18], [19], taking into consideration only the action forces at distance, between the quadrupole moment of N_2 and the dipole moment of $CO_2(v_3)$. Above 1,200K, the rate increases with the temperature and the interaction is due to the forces of short action.

Rosser et al. [11] showed that the rate $k\ (s^{-1}\ torr^{-1})$ of the process (1) varies with the temperature T(K) in the 300-1000K range, according to the relation

$$k = DT^{-3/2} \quad (2)$$

where $D = 8{,}6 \times 10^7\ s^{-1}\ torr^{-1}\ K^{3/2}$. This result coincides with the theoretical analyses done by Sharma and Brau [19].

In a wider range of temperatures (300 - 3,000K), the rate of the process (1) can be calculated with the formula [20]

$$k = 4{,}055 \times 10^{5}\, T^{-1/2} \exp\left[10^{-3}\, T(8{,}84 \times 10^{-4}\, T - 2{,}07)\right]. \qquad (3)$$

The de-excitation of the higher laser level can take place, along with the collisions with CO_2, also through collisions with N_2.

$$CO_2(00^01) + N_2(v = 0) \rightarrow CO_2(n\,, m^1,\, 0) + N_2(v = 0) + \Delta E, \qquad (4)$$

where it is obtained a CO_2 molecule excited on the modes of symetrical stretching and/or of deformation. The efficiency N_2 in the de-excitation of the $CO_2(00^01)$ level, is lower than the de-excitation of CO_2, the process (4) being characterized at 300K by the rate $k = (110 \pm 10)\, s^{-1}\, torr^{-1}$ [11], [12], [21]—[24].

Using the method of phase, Biriukov et al. [25] have established the following dependence on the temperature between 300 and 1,000K of the rate $k(s^{-1} torr^{-1})$ for the process (4)

$$k = \exp(-\,11{,}484 + 507{,}83\, T^{-1/3} - 4574{,}9\, T^{-2/3} + 12\,724\, T^{-1}). \qquad (5)$$

At 300K, the rate indicated above, is obtained, a rate of $110\, s^{-1}\, torr^{-1}$.

The vibrational de-excitation in the system CO_2-N_2 can also happen through the process

$$CO_2(000) + N_2(v = 1 \rightarrow CO_2(n,\, m^1,\, 0) + N_2(v = 0) + \Delta E. \qquad (6)$$

Nevertheless, the rate of this process is lower than that of the process (4) [11], [12].

Little data is available about the influence of nitrogen on the relaxation of the lower laser level.

$$CO_2(10^00,02^00)_I + N_2 \rightarrow CO_2(01^10) + N_2 + 721\ cm^{-1}. \qquad (7)$$

Cheo [21] noticed that the relaxation time of the level $(10^00, 02^00)_I$ does not increase linear with the pressure of nitrogen. Thus, the value of the process rate(7) depends upon the pressure of nitrogen at which it was determined. The measured values are lower than in the case of pure CO_2 $26\ s^{-1}\ torr^{-1}(p_{N_2} = 1 - 7\ torr)$ [24] and respectively $370\ s^{-1}\ torr^{-1}\ (p_{N_2}/p_{CO_2} = 1/3)$ [26].

The influence of nitrogen upon the final relaxation process from the CO_2 molecule

$$CO_2(01^10) + N_2 \rightarrow CO_2(000) + N_2 + 667\ cm^{-1}, \qquad (8)$$

was little studied and the results are contradictory. The SSH theory predicts a triple efficiency of nitrogen versus CO_2 in the de-excitation of the $CO_2(01^10)$ $(650\ s^{-1}\ torr^{-1})$ level, value which is adopted by Cheo [24] and Moore [12]. Another two measurements [27], [28], found an equal efficiency of N_2 and CO_2 in the process (8) $(180-200\ s^{-1}\ torr^{-1})$. Taylor and Bitterman [29] who reviewed the main relaxation processes, important for the functionning of the laser with CO_2, draw the conclusion that nitrogen is 5 times less efficient than the CO_2 in the collisional de-excitation of the 01^10 level. Two recent measurements confirm this conclusion: $38\ s^{-1}\ torr^{-1}$ [7] and $60\ s^{-1}\ torr^{-1}$ [30]. In the temperature range 300-3,000K, Gordietz et al. [20] propose the following dependence on the temperature of the process rate (8)

$$k = 4{,}538\ T^{-1} \exp(-72\ T^{-1/3})\ (s^{-1}\ torr^{-1}). \qquad (9)$$

The table of the vibrational relaxation in the CO_2-N_2 system, thus established, permits to clear up the influence of nitrogen upon the laser effect in CO_2. This influence is primarily manifasted through the efficient pumping of the higher laser level, through the resonant transfer of energy from $N_2(v=1)$ (process 1). Due to the great efficiency of vibrational excitation of N_2 on the level v=1, a large number of molecules are formed, which remain in the state for a long time. In a mix with CO_2, the resonant transfer of this energy on the level $CO_2(00^01)$ takes place, achieving a more efficient pumping than in the case of pure CO_2.

The collisional deactivation of the higher laser level is lower in the case of nitrogen ($110\ s^{-1}\ torr^{-1}$), than in the case of pure carbon dioxide ($330\ s^{-1}\ torr^{-1}$), contributing positively to the maintainence of a population inversion in CO_2.

Nitrogen does not influence significantly the de-excitation of the level $CO_2(01^10)$, so that the choking formed on this level in pure CO_2 is maintained, in the way of de-excitation of vibrational energy. In the CO_2-N_2 system, the appearance of CO through the dissociation of CO_2, remains important for the functionning of the laser of continuous wave, since the carbon oxide has a high rate of deactivation of the level $CO_2(01^10)$.

3. Helium

Among the gases added to the laser, next to CO_2, helium has the highest potenssial of ionization, since the lowest electronic level is situated at 19.7 eV.

Helium influences little the process of de-excitation of the higher laser level and a lot the process of de-excitation of the level 01^10. For the higher laser level

$$CO_2(00^01) + He \rightarrow CO_2(n, m^l, 0) + He + \Delta E, \qquad (10)$$

helium and in general, all the noble gases, have a small efficiency of de-excitation. A series of measurements indicate for the rate of this process a value comprised between 64 s^{-1} $torr^{-1}$ 105 s^{-1} $torr^{-1}$ [7], [12], [31] — [36]. The rate calculated by Gordietz [13] through the SSH method, is 62 s^{-1} $torr^{-1}$. Thus, the rate of the process (10) can be established to be (85 ± 20) s^{-1} $torr^{-1}$. It can be concluded that helium has an efficiency lower than carbon dioxide or nitrogen, in the de-excitation of the higher laser level.

The dependence on the temperature of the process rate (10), studied through the method of the phase [25], was placed between 300 - 1,000K with the formula

$$k = \exp(24{,}538 - 265{,}48\, T^{-1/3} + 763{,}32\, T^{-2/3} + 712{,}83\, T^{-1}). \quad (11)$$

Rosser and Gerry [32] , studied the dependence on the temperature through the method of laser fluorescence and noticed that, this is linear at high temperatures ($T > 500K$), but not at low temperatures.

Helium presents an efficiency a little higher than CO_2 in the relaxation of the lower laser level, on the level 01^10

$$CO_2(10^00, 02^00)_I + He \rightarrow CO_2(01^10) + He + 721\ cm^{-1}. \quad (12)$$

It was noticed a linear variation of the life time invers of the state $(10^00, 02^00)_I$ depending on the helium pressure [37], [21]. The measured rates of the process (12) are 4×10^3 s^{-1} $torr^{-1}$ [37] and $4{,}7 \times 10^3$ s^{-1} $torr^{-1}$ [21].

The final relaxation of the lower laser levels is, as in the case of nitrogen, given by the process

$$CO_2(01^10) + He \rightarrow CO_2(000) + He + 667\ cm^{-1}. \quad (13)$$

The data for this process rate at 300K tally well on the value $(3{,}8 \pm 0{,}5) \times 10^3\ s^{-1}\ torr^{-1}$ [7], [12], [15], [21], [30], [34], [35], [38], [39]. Gordietz et al. [20] placed the variation of the process rate (13) in function of the temperature, with the formula

$$k = 4{,}635\, T^{-1} \times \exp(-40{,}63\, T^{-1/3}), \qquad (14)$$

in the range of temperatures 300 - 1,000K.

Unlike nitrogen, helium plays a complex role in the increase in the power and efficiency of the CO_2 laser.

The collisional processes of vibrational de-excitation are influenced especially with regards to the relaxation in the lower laser level, both of the state $(10^00, 02^00)_I$, and of 01^10. The CO_2-He collisions are 20 times more efficient in the process of relaxation V-T (process 13), than the collisions CO_2 - CO_2 or CO_2 - N_2. In this way, the choking which appeared in the way of emptying the population of the lower laser level in pure CO_2 or in the CO_2 - N_2 mix.

Helium also influences the plasma of the CO_2 - N_2 - He laser, increasing the rate at which the CO_2 molecules are excited on the 00^01 level, either directly through the processes of collision with electrones, or indirectly, through the increase in the rate of vibrational excitation of the N_2 molecules, in other words helium leads to a distribution of the electrone energy more favourable to the excitation of the higher laser level [10]., Patel [27] brought another experimental proof that helium modifies the discharge properties of the CO_2 laser. The nitrogen was excited separately and then introduced in the resonator, together with the carbon dioxide. The exit power was greater when the helium was introduced together with the nitrogen in the discharge zone, compared to the case when the helium was introduced directly in the resonator together with carbon dioxide, proving that helium participates in the process of vibrational excitation of the nitrogen.

Weber and Deutsch [40] studied the spontaneous emission of the higher and lower laser levels in the presence of helium, and noticed that, while the emission on the higher laser level increases, the emission on the lower laser level decreases, when helium is added. The decrease of the latter is associated with the increase in the non-radiative de-excitation on the lower vibrational levels, originating in the CO_2 - He collisions. The increase in the emission of the 00^01 level indicates that helium increases the pumping of the higher laser level. All these experiments confirm the conclusions drawn now as regards to the iinfluence of helium upon the populations of the laser levels.

Due to its high thermic conductivity (about 6 times higher than that of N_2 or CO_2), helium reduces the kinetic temperature of the discharge gas. A lower gas temperature implies on one hand, lower rates of collisional de-excitation of the higher laser level, and , on the other hand, increases the gain, which is dependent on the temperature of the molecular lasers.

In the CO_2 lasers, excited with impulse, there takes place a negligible cooling during the impulse, and thus, the presence of helium will not affect the peak temperature of the gas [41]. Also, it is improbable that there would be a considerable relaxation of the 01^10 level during the laser impulse. That is why the role of helium in these lasers is different from the role played by the conventional lasers. In the TEA - CO_2 lasers, the favourable dielectric properties of helium helps to establish and maintain a steady discharge, and helium becomes essential at a pressure that exceeds 250 torr [42].

4. Carbon Oxide

Carbon oxide is always present in the gas mix of the CO_2 laser, due to the dissociation of the CO_2 molecule, under electronic impact. The degree of the CO_2 dissociation can reach up to 70% in a stationary regime [43], or even 100% in impulse [44], depending on the discharge conditions.

CO presents a level of vibrational energy at $2143\ \mathrm{cm}^{-1}$, active in infra-red, metastable. CO has a permanent dipole moment.

Carbon oxide is easily excited through collisions with electrones. More than 15% of the electrone energy can be transfered to the vibrational levels of CO [3]. Schultz [5], [6], established that the added cross sections, calculating the excitation of the vibration levels up to v=8, give a resonant character and reach the maximum at an electrone energy of 1.7 eV. This cross section $\sigma\ (\bar{e},CO) = 8\times10^{-16}\ \mathrm{cm}^2$, is almost three times bigger than the corresponding one for nitrogen.

De-excitation of the higher levels on the v=1 level

$$CO(v=2) + CO(v=0) \rightarrow CO(v=1) + CO(v=1) - 27\ \mathrm{cm}^{-1} \quad (15)$$

takes place fast, at a rate, at 295K, comprised between $6{,}2\times10^{4}\ \mathrm{s}^{-1}\ \mathrm{torr}^{-1}$ [45] and $1{,}1\times10^{5}\ \mathrm{s}^{-1}\ \mathrm{torr}^{-1}$ [46].

The collisional deactivation of the CO(v=1) level takes place through the process

$$CO(v=1) + M \rightarrow CO(v=0) + M + 2143\ \mathrm{cm}^{-1}, \quad (16)$$

where M is the collision particle. For M=CO, the efficiency of the process (16) is very small, the rate being of the order $10^{-3}\ s^{-1}\ torr^{-1}$ [47]–[49]. The carbon dioxide is a more efficient partner in the process (16); the rate is $(2{,}8 \pm 0{,}5) \times 10^{3}\ s^{-1} torr^{-1}$ [50], [51]. In a mix with nitrogen, the transfer process from CO(v=1) to $N_2(v = 1)$ is possible.

$$CO(v = 1) + N_2(v = 0) \rightarrow CO(v = 0) + N_2(v = 1) - 188\ cm^{-1}. \quad (17)$$

The rate of this process was determined to be $177\ s^{-1}\ torr^{-1}$ [46] and respectively $130\ s^{-1}\ torr^{-1}$ [52]. The rate of the process (17) in the opposite sense (exothermic direction) is $440-460\ s^{-1}\ torr^{-1}$ [46], [53]. The radiative life time of the CO(v=1) state is 30 -33 ms [54], [55].

Carbon oxide contributes to the population of the higher laser level, through an almost resonant process

$$CO(v = 1) + CO_2(000) \rightarrow CO(v = 0) + CO_2(001) - 206\ cm^{-1}. \quad (18)$$

Since the energy defect for the reaction (18) is about ten times greater for CO than for N_2, it can be anticipated that the CO - CO_2 transfer will be slower for N_2 - CO_2 (reaction (1)). Further, it was noticed that the spontaneous emission and the induced emission in CO_2 are reduced 30 -35% when the nitrogen is replaced with carbon oxide [56]. Recent measurements registered a rate, for which the data tally very well $(2{,}2 \pm 0{,}1) \times 10^{3}\ s^{-1}\ torr^{-1}$ at 300 K [51], [57], [58]. A linear dependence of the rate of the process (18) was noticed in a large range of temperatures. Thus, between 300 and 900K, this dependence can be described " of the right" [51]

$$k = 9{,}9T - 840. \quad (19)$$

By fitting with the smallest squares of the values measured by

Stephenson and Moore [57] results for the right (19) a slightly lower slope (9.25). This linear variation can be extended both for lower temperatures (up to 180K) [58] and for high temperatures (between 1,500 and 2,500K) [59]. The dependence on the temperature of the rate of the CO - CO_2 transfer process, is opposite to that of the N_2 - CO_2 process, so that at $T > 500K$, the CO - CO_2 transfer becomes more efficient than N_2 - CO_2.

The de-excitation of the higher level takes place through the processes:

$$CO_2(00^01) + CO(v=0) \rightarrow CO_2(n, m', 0) + CO(v=0) + \Delta E, \quad (20)$$

$$CO(v=1) + CO_2(000) \rightarrow CO(v=0) + CO_2(n, m', 0) + \Delta E, \quad (21)$$

the final state for (21) being $CO_2(11^10,03^10)_I$. For (20) it was determined through the method of laser gain in impulse, a rate of $193\ s^{-1}\ torr^{-1}$ [21], and through the vibrational fluorescence method, a rate of $230\ s^{-1}\ torr^{-1}$ [51]. For this rate, the following dependence on the temperature between 500 and 900K [51] was established

$$k = \exp(15{,}82 - 72{,}76\ T^{-1/3}). \quad (22)$$

The relaxation of the lower laser level

$$CO_2(10^00,02^00)_I + CO \rightarrow CO_2(01^10) + CO + 721\ cm^{-1}, \quad (23)$$

takes place fast, with a rate of $(4-5)\times10^4\ s^{-1}\ torr^{-1}$ [21], [60].

Carbon oxide de-excitates strongly the level $CO_2(01^10)$

$$CO_2(01^10) + CO \rightarrow CO_2(000) + CO + 667\ cm^{-1}, \quad (24)$$

the rate being $(2{,}5-5)\times10^4\ s^{-1}\ torr^{-1}$ [21], [27], [38], [61], [62].

Carbon oxide plays a similar role with nitrogen in the laser system with CO_2. Like the nitrogen, carbon oxide presents a cross section of great electronic excitation for the first 8 vibrational levels, which are quickly de-excited on the metastable level v=1. Due to the great energy defect, the transfer process of the vibrational energy to the higher laser level is slower than in N_2. Carbon oxide de-excites little the $CO_2(00^01)$ level, but has a greater efficiency in the de-excitation of the lower laser level and of the 01^10 level (one order greater than helium).

In the CO_2 - He mix, the appearance of the laser effect is possible only in the presence of carbon oxide, produced through the CO_2 dissociation in discharge, as shown by calculus of the population inversion [63].

It was possible to obtain a laser effect in CO_2 by pumping the higher laser level only through the resonant process (18). Carbon oxide is excited in a separate electric discharge and then mixed with CO_2 and He in the resonator [64]. When CO is replaced in the electric discharge by N_2, the power level can be increased at least by two size orders.

5. Oxygen

Oxygen can be a component of the CO_2 laser, due to the partial breaking up of CO_2 during the electric discharge.

The oxygen molecule has the first vibrational level situated at 1,556 cm^{-1}, which presents a relatively long life time (2.4 ms) [65]. It has a dissociation energy of 5.12 eV and an ionization potenssial of 12.2 eV [66].

The excitation cross section of the first vibrational level of the oxygen is one size order smaller than that of the electrones - CO, collisions, at average electrone energies typical for the discharge of the CO_2 laser [3], [65].

The de-excitation of the $O_2(v=1)$ level through collisions with O_2 or N_2 is slow $(10^{-2} - 10^{-1}\,s^{-1}\,torr^{-1})$ [29], [47], [67]. Carbon dioxide shows a greater efficiency in this process, compared to that of CO_2 in the N_2 relaxation.

Oxygen can transfer its vibrational energy to the mode v_2 of CO_2, through the process

$$O_2(v=1) + CO_2(000) \to O_2(v=0) + CO_2(01^10) + 889\ cm^{-1}. \quad (25)$$

The high rate of this process $(3\times10^3\,s^{-1}\,torr^{-1}$ at 300K) [29], can explain the low oxygen efficiency in the de-population of the $CO_2(01^10)$ level. Also, the de-excitation of oxygen on the $CO_2(10^00,\ 02^00)_{II}$ level is possible, since it presents a V-V process closer to the energetic resonance ($\Delta E = 270\ cm^{-1}$).

The de-excitation of the higher laser level through inellastic collisions with O_2 takes place through the process

$$CO_2(00^01) + O_2 \to CO_2(n, m^l, 0) + O_2 + \Delta E. \quad (26)$$

Oxygen shows an efficiency similar to nitrogen in the process [26], [22]. Rosser and Gerry [32], studied the rate of this process in a large range of temperatures, through the laser fluorescence method. At 300K, the rate is approximately $125\,s^{-1}\,torr^{-1}$.

In the CO_2 - O_2 mix, it is also possible the de-excitation of the 00^01 level on the 01^10 level, with the excitation of oxygen on the v=1 level (68).

$$CO_2(00^01) + O_2(v=0) \to CO_2(01^10) + O_2(v=1) + 127\ cm^{-1}, \quad (27)$$

for which a rate of $\sim 270\ s^{-1}\ torr^{-1}$ at 293K was measured.

The influence of oxygen in the deactivation of the $CO_2(01^10)$ level

$$CO_2(01^10) + O_2 \rightarrow CO_2(000) + O_2 + 667\ cm^{-1} \qquad (28)$$

is little known. It was suggested [29] that the rate of this process is equal, in size, and in dependence to the temperature, with the rate of de-excitation of the $CO_2(01^10)$ level by nitrogen. Nevertheless, measurements in the CO_2 - O_2 level at 300K, with the aid of an ultrasonic interferometer, indicate a double oxygen efficiency in the process (28), compared to nitrogen [28].

Oxygen has an unfavourable influence in the achievement of laser oscillation in CO_2, since, through the processes (25), and (27) leads to the increase in the population of the lower level, introducing at the same time, through the process (27) a parallel way in the deactivation of the higher laser level.

Experiments to add oxygen in an active medium of the CO_2 laser [69] - [71], have given contradictory results.

6. Water vapours

H_2O is a triatomic molecule, simetric, non-linear, with a non-null dipole moment. Like the CO_2 molecule, the vibrations are characterized by three quantum numbers v_1, v_2, and v_3, with the difference that the mode v_2 is not degenerated. The only vibrational level which occurs in the CO_2, 010 laser process, is situated at 1,595 cm^{-1}.

Due to its extremly rapid time of vibrational relaxation, compared to other simple triatomic molecules, H_2O is considered an abnormal molecule

$$H_2O(010) + H_2O(000) \rightarrow H_2O(000) + H_2O(000) + 1595\ \mathrm{cm}^{-1}. \qquad (29)$$

The rate of this process is very big: $10^5 - 10^6\ \mathrm{s}^{-1}\ \mathrm{torr}^{-1}$ [29]. For other collision partners (CO_2, N_2, O_2, He) the de-excitation is slower.

The vibrational energy can be transfered from the excited state of water vapours on the level $CO_2(01^10)$

$$H_2O(010) + CO_2(000) \rightarrow H_2O(000) + CO_2(01^10) + 928\ \mathrm{cm}^{-1}. \qquad (30)$$

For this process a big rate [29] was calculated, but in the CO_2 laser, it is probable to take place the reverse reaction (30), followed by the reaction (29). The same thing happens with the transfer on the lower laser level

$$H_2O(010) + CO_2(000) \rightarrow H_2O(000) + CO_2(10^00, 02^00)_I + 207\ \mathrm{cm}^{-1}. \qquad (31)$$

Nevertheless, the reverse process (31), is excluded by Bulthuis and Ponsen [72], arguing that this would imply a quadratic dependence of the relaxation rate of the level $(10^00, 02^00)_I$, on the density of the H_2O vapours, while it was measured a linear dependence for pressures of H_2O for up to 0.4 torr.

The de-excitation of the CO_2 molecules on the higher laser level through inellastic collisions with H_2O molecules, takes place through the process

$$CO_2(00^01) + H_2O \rightarrow CO_2(n, m^l, 0) + H_2O + \Delta E. \qquad (32)$$

The rate of this process was measured in a series of experiments, and the results can be placed in the range $(3{,}3 \pm 0{,}9) \times 10^4\ \mathrm{s}^{-1}\ \mathrm{torr}^{-1}$ [12], [20], [21], [32], [73], [74]. It was established [32] that in the range 300 - 1,000K, the rate of the process (32) has a dependence on the linear and negative temperature

$$k = -15{,}7\ T + 3{,}37 \times 10^4. \qquad (33)$$

The de-excitation of the higher laser level can take place also through collisions of the order two, of the type

$$CO_2(00^01) + H_2O(000) \rightarrow CO_2(000) + H_2O(010) + 754\ cm^{-1}, \qquad (34)$$

$$CO_2(00^01) + H_2O(000) \rightarrow CO_2(01^10) + H_2O(010) + 87\ cm^{-1}. \qquad (35)$$

The first process can lead, under certain experimental conditions, to the reduction in the laser power [75]. The process (35) was called by Yardley and Moore [76] "the intermolecular distribution of the V-V energy", since the vibrational energy is distributed between the two molecules.

The lower laser level is de-excited under the influence of the inellastic collisions with H_2O, on the level $01^1 0$, and this in the fundamental state

$$CO_2(10^00,\ 02^00)_I + H_2O \rightarrow CO_2(01^10) + H_2O + 721\ cm^{-1}, \qquad (36)$$

$$CO_2(01^10) + H_2O \rightarrow CO_2(000) + H_2O + 667\ cm^{-1}. \qquad (37)$$

The data for these two processes are uncertain and contradictory. From experiments in grain in impulse, Chel [21] decucted for the process (36) a rate of $1.15 \times 10^6\ s^{-1}\ torr^{-1}$. Rosser et al. [26], assuming the addivity of the CO_2 and H_2O effects in the process (36), obtained through averaging some results of fluorescent laser, a rate of $1.2 \times 10^5\ s^{-1}\ torr^{-1}$. Bulthuis and Ponsen [77] have measured the rate of the vibrational relaxation of the level CO_2 $(10^00,\ 02^00)$ by H_2) and D_2O from the decrease in power, after the laser short circuit, and obtained at 500K, $5.79 \times 10^4\ s^{-1}\ torr^{-1}$ for H_2O and $3.86 \times 10^4\ s^{-1}\ torr^{-1}$ for D_2O.

For the process (37), the rate determined experimentally at the temperature of 300K, can be placed in the range $(1-5)\times 10^{5}$ s^{-1} $torr^{-1}$ [12], [21], [27], [29], [38], [62], (at 990K), [78], [79]. The dependence on the temperature is negative [12], [29], assuming a variation inversely proportional to T^{3} [20]; this dependence is not explained by the SSH theory. In contrast with these results, Bulthuis and Ponsen [72], found that the rate of the process (37) increases with the temperature, between 500 and 1,000K, taking values between $3,86\times 10^{4}$ s^{-1} $torr^{-1}$ (500 K) and 10^{5} s^{-1} $torr^{-1}$ (1 000 K).

Bulthuis [17] proposes a model for the processes (36) and (37) on the basis of the coupling between the levels $(10^{0}0, 02^{0}0)_I$ and $01^{1}0$ and respectively, between $01^{1}0$ and the fundamental level. In the first case, of the "strong" coupling between the levels $CO_2(10^{0}0, 02^{0}0)_I$ and $CO_2(01^{1}0)$, the relaxation of the level $01^{1}0$ towards the fundamental state, is the determinant rate of the deactivation process of the lower laser level. In this case, the power decreases exponentially and the reduction time is inversely proportional to the density of water vapours. Experimentally these things were noticed when $p_{H_2O}/p_{CO_2} < 0,12$ or $p_{D_2O}/P_{CO_2} < 0,20$. In the second case, the "strong" coupling between the level $CO_2(01^{1}0)$ and the fundamental state, the coupling between $(10^{0}0, 02^{0}0)_I$ and $01^{1}0$ (process (36) with CO_2 as collision partner), is the determinant rate for the relaxation of the lower laser level and the reduction time of the laser power will be inversely proportional to the density of the CO_2 molecules. It was found, experimentally, that the reverse of the reduction time increases linear with the CO_2 density for $p_{H_2O}/p_{CO_2} > 0,12$ and respectively $p_{D_2O}/p_{CO_2} > 0,20$.

In conclusion, adding relatively small quantities of H_2O, the process (42) is determinant for the relaxation of the lower laser level. Increasing the density of water vapours, the two relaxation processes become of comparable importance. By adding also H_2O, the process (36) together with the relaxation process of the state

$CO_2(10^00, 02^00)_I$ through collisions with CO_2, become the determinant step of the vibrational relaxation. A similar explanation was given by Gower and Carswell [62], who found that the transition between the two relaxation steps takes place at a pressure of the water vapours of 0.4 torr.

Thus, the fastest relaxation processes in the CO_2 molecule appear in the presence of the water vapours. Both the higher laser level (processes (32), (34) and (35), and the lower laser level (processes (36), (37)), are de-excited by H_2O with extremely high rates. At low H_2O pressures, the CO_2 relaxation processes are positively influenced, in the sense that the de-excitation of the lower laser level is faster than the de-excitation of the higher laser level. This is the explanation for the maximum noticed both for the laser power, and for the gain depending on the pressure of water vapours. In comparison to the mix without water vapours, double powers can be obtained at a water vapour pressure of 0.2 torr [17], [75], [80]. At a gas circulation laser, it was noticed [80] an increase in the power of 2-3 times at small total pressures, the increase being smaller and smaller with the increase in the total pressure. The addition of water vapours to the CO_2 - N_2 mix, increases the gain, as it was noticed in the case of lasers with dynamic gas [81]—[84].

A considerable influence is excercized by the water vapours on the dissociation reaction of the CO_2 molecule, through the intermediary of the hydroxil radical, which is formed in the electric discharge, through the reaction

$$H_2O + \bar{e} + 4{,}98\ \text{eV} \rightarrow OH + O + \bar{e}. \tag{38}$$

The radical OH is a good catalyst for the formation reaction of CO_2 from CO

$$OH + CO \rightarrow CO_2 + H. \tag{39}$$

It was noticed that the degree of CO_2 dissociation reaches only 20% in the case of water vapour addition to the gas mix, compared to 60% in the case of water vapour absence. Mass spectroscopic measurements [86] and of lateral light [87] proved that in the presence of water vapours, the concentration of CO molecules is very low, being significant the concentration of OH radicals, which can reach the value $(2-4)\times10^{12}$ cm^{-3} [88]. The introduction of water vapours in the active medium of the CO_2 laser significantly increases the life time of the sealed-off gas lasers.

The calculus of population inverssion in function of the water vapour pressure, taking into consideration only the reduction in the degree of CO_2 dissociation and the de-excitation of the higher level [88], shows the same maximum at a water vapour pressure of 0.1-0.2torr.

7. Hydrogen

Hydrogen and deuterium are the lightest gases added to the active medium of the CO_2 laser. The first vibrational level of the hydrogen molecule is situated at 4,159 cm^{-1} and cannot influence the relaxation processes from the CO_2 molecule. Deuterium has the v=1 level situated a little lower (2,990 cm^{-1}) and can interaction with the higher laser level. Both hydrogen and deuterium have small relaxation rates, from the level v=1 on the fundamental level; for H_2, it was measured 4,4 s^{-1} torr^{-1} [47], [89], and for D_2 0,83 s^{-1} torr^{-1} [47], [90].

The relaxation of the higher laser level takes place through the process

$$CO_2(00^01) + M \rightarrow CO_{2(n,\, m^l,\, 0)} + M + \Delta E \qquad (40)$$

where M is H_2 or D_2. For $M=H_2$, the rate measured through the vibrational fluorescent method (3 890 s^{-1} torr^{-1}) [12], [91] is slightly lower than the rate measured through the method of the gain in impulse (4,500 s^{-1} $torr^{-1}$) [21]. The dependence on the temperature of the rate of this process is given by the formula [20]

$$k = 1{,}738\times10^{6}\, T^{-1/2} \exp\left(-22{,}22\, T^{-1/3}\right). \qquad (41)$$

For M=D_2, the rate of the process (40) is one size order smaller than in the case of hydrogen : 350 s^{-1} $torr^{-1}$ [33]. Between CO_2 and D_2 it is always possible the process

$$CO_2(00^01) + D_2(v = 0) \rightarrow CO_2(000) + D_2(v = 1) - 641\ cm^{-1} \qquad (42)$$

for which a rate of 467 s^{-1} $torr^{-1}$ [57]. was measured. For the exotherm direction of the process (42), the rate of 10 500 s^{-1} $torr^{-1}$ was determined [12], [57].

Hydrogen de-excites efficiently the CO_2 molecules on the lower laser level, on the level 01^10

$$CO_2(10^00,\ 02^00)_I + H_2 \rightarrow CO_2(01^10) + H_2 + 721\ cm^{-1}. \qquad (43)$$

[21] Through the method of gain in impulse, a rate of $3{,}3\times10^{4}$ s^{-1} $torr^{-1}$ was measured.

Hydrogen and deuterium have an efficient action also on the process of CO_2 (01^10) de-excitation on the fundamental state

$$CO_2(01^10) + M \rightarrow CO_2(000) + M + 667\ cm^{-1}. \qquad (44)$$

For M=H_2, was measured the rate $(7\pm3)\times10^{4}$ s^{-1} $torr^{-1}$ [7], [12], [21], [92], [93], with the temperature dependence the simple subtrahend [20]

$$k = 5{,}118\times10^{8}\, T^{-3/2}. \qquad (45)$$

For M=D_2, the rate of the process (44) is $(1\text{-}2)\times10^{4}$ s^{-1} $torr^{-1}$ [12, 94].

Sharma [94] proposed a mechanism whereby the process (44) is depicted by the coupling of the dipole mement of the $01^10 \leftrightarrow 000$ trasition, with the permanent H_2 quadrupole moment, converting the CO_2 vibrational energy into H_2, which suffers a rational transition with $\Delta J = 2$:

$$CO_2(01^10) + H_2(v=0,\ J=1) \rightarrow CO_2(000) + H_2(v=0,\ J=3) + \\ + 80{,}5\ \mathrm{cm}^{-1}. \tag{46}$$

$$CO_2(01^10) + H_2(v=0,\ J=2) \rightarrow CO_2(000) + H_2(v=0,\ J=4) - \\ - 146{,}3\ \mathrm{cm}^{-1}. \tag{47}$$

For the process (46) it was established a double rate compared to the process (44), [92], [93].

Similarly, Sharma proposed for D_2 the process

$$CO_2(01^10) + D_2(v=0,\ J=4) \rightarrow CO_2(000) + D_2(v=0,\ J=6) - \\ - 25{,}2\ \mathrm{cm}^{-1} \tag{48}$$

where a rate of $2{,}9 \times 10^4\ \mathrm{s}^{-1}\,\mathrm{torr}^{-1}$ was established.

As in the case of water vapours, hydrogen depopulates efficiently both the higher and the lower level, but with smaller rates. Deuterium is more favourable to the CO_2 laser effect, since it strongly de-excites only the 01^10 level, and for the 00^01 level presents the same low efficiency of de-excitation as the carbon dioxide.

The similarity with the water vapours continues also with respect to the dependence of the CO_2 laser power to the hydrogen pressure. The power increases initially, reaching a maximum at 0.1-0.2 torr H_2, afterwards rapidly decreases [95]—[97]. This dependence is explained through the stronger de-excitation of the lower laser level at the hydrogen lower pressures; together with the increase in the hydrogen pressure, the vibrational de-excitation of the higher laser level will stop the gain. The increased power at hydrogen addition can be also due to the modifications in the gas composition, especially due to the lower CO_2 dissociation. In the presence of a 0.2 torr hydrogen, the CO_2 degree of dissociation halves [71], [96]—[98]. Above this pressure, hydrogen influences less the degree of dissociation.

The dissociation is reduced due to the catalyst effect of the OH radical through the reaction (39). In the presence of hydrogen, the hydroxil radicals are formed from the atomic oxygen from the dissociation, according to the reaction

$$O + H_2 \rightarrow OH + H. \tag{49}$$

The introduction of hydrogen in the sealed off gas lasers increases their life time [87], [97], being absolutely necessary to the maintainance of the initial CO_2 concentration.

Hydrogen has a positive influence upon the CO_2 lasers excited cross-wise in impulse, at high pressure (TEA) [99]. The addition of 3-10% H_2 to the CO_2-N_2-He mix removes the spring, giving a more uniform discharge, where the impulse repeatability is improved.

8. Xenon

Unlike the other additives, xenon does not have a vibrational level to excite the prefferential higher laser level or to efficiently depopulate the lower laser level. Due to its relatively big mass, xenon has a low efficiency in the de-excitation of the vibrational levels of the CO_2 molecule. Its influence is manifasted mainly upon the plasma of the CO_2 laser.

Xenon improves the excitation of carbon oxide on the state v=1, the cross section of vibrational excitation reaching 0.5 eV, about 30 times greater than the cross section of excitation of nitrogen at the same energy [100].

The rate at which xenon de-excites the $CO_2(00^01)$ level is situated in the range 0-30 $s^{-1}torr^{-1}$ [12], [21], [33]-[35], and the rate of de-excitation of the 01^10 level through inellastic collisions with xenon is lower than 44 $s^{-1}torr^{-1}$ [34].

Xenon has the lowest ionization potensial (12.1 eV) among the components of the active medium of the CO_2 laser. In its presence, a smaller enrgy is necessary, which is to produce the given ionization density, and the portion of the high energy of the electronic distribution is reduced. The spectroscopia in visible of the CO_2-N_2-He-Xe laser proves that the number of the high energy electrones is reduced by the addition of xenon.

Probe measurements indicate an increase in the electrone density of 40-45% [101]. This increased electrone density produces more efficiently molecules of N_2 or CO on the vibrational levels, with an ulterior increase in the population of the higher laser level.

The functioning conditions of the CO_2-N_2-He-Xe laser are modified towards larger currents, with a decrease in the discharge tension of 20-25% [100]-[102]. The addition of xenon reduces the variations of the plasma impedance with the discharge current, thus, also the variations in the laser power.

The xenon influence upon the laser power is significant in the sealed off gas laser systems. The highest increases in the power were noticed [100] in the case of xenon addition to the CO_2-He and CO_2-CO-He mixes, where there is a more efficient excitation of the CO originating either from the dissociation, or specially introduced. In the case of the CO_2-N_2-He mix, the xenon addition increases the power by 25% [101], [103]. The xenon optimum pressure is 1-2 torr.

Since xenon reduces the discharge tension, the efficiency of the conversion of the electric energy into laser energy, is considerably increased, by 12-25% [101], [102]. The maximum efficiency appears at a current smaller than that for the maximum power.

Xenon plays an important role in the life of CO_2 sealed off gas lasers. Having a minimum ionization potensial and the largest cross ionization section among the major components of plasma, xenon is the primar source of discharge ions. Consequently, the number of ions of CO_2, O_2, CO, etc., is reduced, and is limited the oxidation and the spreading at the cathode, which is an important mechanism in limiting the life of these lasers. Reducing the number of electrones with high energies, it is also reduced the probability of collisions, which lead to the dissociation of the CO_2 molecule. The xenon addition to the CO_2-He or CO_2-N_2-He mixes made it possible to reach life times of over 1,000 hours [101], [103], [104].

9. Conclusions

Additive gases manifest their influence upon the laser effect in CO_2, in different ways, modifying the vibrational relaxation processes, the plasma parameters, the chemical composition, the gas temperature, etc., which affects finally the power and efficiency of the laser. In the presence of some gases N_2, CO), the higher laser level is efficiently populated through the transfer of vibrational energy from these molecules. Other gases (He, CO, H_2O, H_2) strongly de-excite the level $CO_2(01^1 0)$, thus contributing to the maintainance of a population inverssion. Noble gases have a smaller efficiency in these relaxation processes and their influence is smaller, the larger their mass.

The addition of water vapours or hydrogen contributes to minimize the reaction of the CO_2 molecule dissociation under electronic impact, through the catalyst effect of the dissociation reaction between CO and O, by the hydroxil radical.

At the CO_2 lasers in impulse, there can be obtained uniform discharges by adding some gases with a low ionization potensial (hydrogen, xilen).

Under these conditions, the optimum mix for the gas flow lasers is comprised of CO_2-N_2-He, the proportion and the total pressure depending on the speed of the gas flow, the diameter of the discharge pipe, the gas temperature, etc. For the sealed-off gas lasers, where the CO_2 concentration must be maintained at a value as close as possible to the initial one, there are added, besides these gases, water vapours or hydrogen and xenon.

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