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Influence of CO on the population inversion in CO₂ lasers

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The population inversion in a CO_2-He glow discharge has been calculated, taking into account CO produced by dissociation in the discharge. The calculation was based on experimentally determined electron energy distribution functions and the measured effective relaxation constant of the CO_2 (001) level. It is shown that the population inversion is negligible in the absence of CO. The importance of excitation and relaxation of CO_2 (001) by electrons is stressed.

The influence of CO, which is produced by dissociation in the discharge of an electrically excited CO_2 laser, is well recognized.[1,2] Both experimental and theoretical work support the view that CO plays a similar role to that of N_2 in supplying vibrational energy to the upper lasing level (001) of the CO_2 molecule.

Theoretical calculations by Gordietz *et al.*,[3] assuming that the excitation of the asymmetric stretch mode of the CO_2 molecule results entirely from vibrational energy transfer between excited CO molecules and CO_2 molecules in the ground state, show the dependence of the population inversion on the CO concentration in the lasing mixture. In those calculations, direct excitation by electrons of CO_2 molecules was neglected, a Maxwellian electron energy distribution was assumed, and both theoretically and experimentally obtained relaxation constants were used.

It is the purpose of this paper to show that by using experimentally obtained rate constants and measured electron energy distribution functions (non-Maxwellian), and by taking into account direct electron excitation of the $(00V)$ mode, the population inversion would indeed be marginal if it were not for the presence of the CO produced by dissociation in the discharge.

The following simple assumptions are made: (a) A defined vibrational temperature exists for each of the different vibrational modes of the CO_2 molecules. (b) The symmetrical stretch mode $(V00)$ and the bending mode $(0V0)$ have a common vibrational temperature T. (c) The vibrational temperature T_v of the asymmetrical stretch of the CO_2 molecules and that of the CO molecules are nearly equal.

Assumptions (a) and (b) are well established[4,5] and assumption (c) is based on the relatively high relaxation constant for vibrational energy transfer from CO to CO_2 $(00V)$.[3]

The vibrational energy balance expressed in terms of quanta, stored and exchanged in the electrical discharge for the CO_2 $(00V)$ and CO (V) levels can be presented by

$$\dot{Q}_{ev}(T_v) = \dot{Q}_{vv}(T_v) + \dot{Q}_{VT}(T_v) + \dot{Q}_{VD}(T_v), \tag{1}$$

where $\dot{Q}_{ev}$ is the net rate of supply of vibrational quanta by electrons to the above-mentioned levels; $\dot{Q}_{vv}$ is the net relaxation rate from these levels to any of the other vibrational modes; $\dot{Q}_{VT}$ is the vibration-translation relaxation rate and $\dot{Q}_{VD}$ is the rate of loss of vibrational energy quanta due to diffusion to the walls.

When lasing ensues, an additional term $\dot{Q}_L$ the rate of radiation energy emitted has to be added to Eq. (1):

$$\dot{Q}_{ev}(T_v) = \dot{Q}_{vv}(T_v) + \dot{Q}_{VD}(T_v) + \dot{Q}_L(T_v) + \dot{Q}_{VT}(T_v). \tag{2}$$

In order to calculate the population inversion and its dependence on the CO concentration, both Eqs. (1) and (2) have been solved using only experimentally measured data and constants.

The term $\dot{Q}_{ev}$ on the left-hand side includes contributions both from CO_2 and CO. Neglecting anharmonicity in both cases, one obtains expressions for $\dot{Q}_{ev_1}$ and $\dot{Q}_{ev_2}$ for the CO_2 and CO vibrational energy storage due to electron excitation, respectively:

$$\dot{Q}_{ev_1} = N_{CO_2} Z_1 F_1; \quad \dot{Q}_{ev_2} = N_{CO} Z_2 F_2, \tag{3}$$

where N_{CO_2} and N_{CO} are the concentrations, $(Z_1)^{-1}$ and $(Z_2)^{-1}$ are the partition functions, and F_1 and F_2 are the average number of inelastic collisions per unit time and unit volume, which cause excitations to the relevant vibrational levels of CO_2 and CO, respectively.

Now

$$Z_1 = (1 - x_1)(1 - x_2)^2(1 - x_3), \quad Z_2 = (1 - x_4),$$

where

$$x_1 = \exp(-1388/kT); \quad x_2 = \exp(-667.4/kT);$$
$$x_3 = \exp(-2350/kT_v); \quad x_4 = \exp(-2180/kT_v);$$

1388, 667.4, 2350, and 2180 cm^{-1} are the energies of the CO_2(100), CO_2(010), and CO_2(001) and CO (1) levels above the ground state, k is the Boltzmann constant;

$$F_1 = K'_{01} \frac{1 - x_3 \exp(2350/\overline{E})}{(1 - x_3)^2},$$

$$F_2 = \sum_{n=1}^{8} K_{0n} \, n \frac{[1 - x_4^n \exp(n2180/\overline{E})]}{1 - x_4}, \tag{4}$$

and $\overline{E}$ is the mean electron energy.

The rate constants for excitation K_{01} and K_{0n} are

$$K'_{01} = n_e \int_0^\infty f(E) \sigma'_{01}(E)(2E/m)^{1/2} dE, \quad \text{for } CO_2$$

$$K_{0n} = n_e \int_0^\infty f(E) \sigma_{0n}(E)(2E/m)^{1/2} dE, \quad \text{for } CO \tag{5}$$

where $\sigma'_{01}(E)$ and $\sigma_{0n}(E)$ are the cross sections for excitation from the ground state by electrons to CO_2 (001) and CO (V), respectively, $f(E)$ is the electron energy distribution function, E the electron energy, and n_e the electron concentration.

In the case of CO_2 the harmonic oscillator model was used, thus $K'_{n,n+1} = (n+1)K'_{01}$. For CO, following Chen's[6] model, excitation via a compound state was assumed, namely, $K_{0V} = K_{n,n+V}$.

Now let us turn to the relaxation terms of Eq. (1). If we define an effective constant β which includes all the processes that cause the depopulation of the (001) level, then

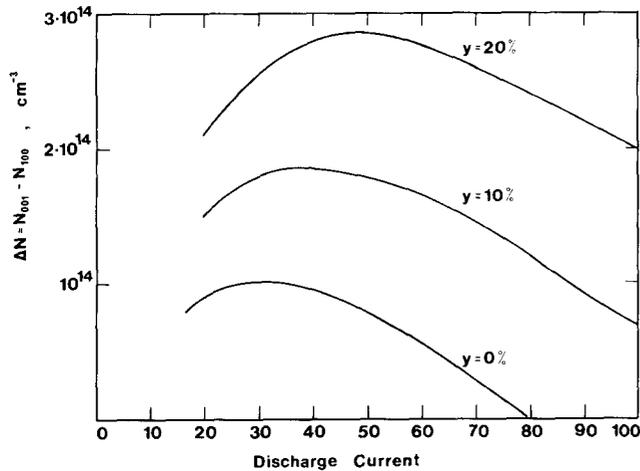$$\dot{Q}_{VV} + \dot{Q}_{VT} + \dot{Q}_{VD} = \beta N_{CO_2} Z_1 x_3. \tag{6}$$

FIG. 1. Population inversion as a function of discharge current; y (degree of dissociation) = $[N_{CO}/(N_{CO} + N_{CO_2})] \times 100$.

If the relevant constants in Eqs. 4—6 are known, Eq. (1) yields a vibrational temperature T_V and the population inversion ΔN can be calculated for a given gas temperature. The population inversion is thus

$$\Delta N = N_{001} - N_{100} = N_{CO_2} Z_1 (x_3 - x_1), \tag{7}$$

where N_{001} and N_{100} are the populations per cm^3 of the upper and lower lasing levels, respectively.

In order to solve Eq. (1) for T_V, one has to know the electron density n_e, the electron energy distribution function $f(E)$, the cross sections for excitations σ'_{01} and σ_{0n}, the effective relaxation constant β, and the concentration of the gas-mixture constituents.

These constants have been found experimentally in a conventional water-cooled He-CO_2 gas laser with a central cathode and two anodes placed at the ends of the tube. The diameter of the tube was 26 mm, the distance of anode to cathode in each arm 43 cm, and the gas pressures CO_2—0.8 Torr and He—3.4 Torr. The discharge was run at 20—100 mA and a 15% transmission germanium mirror was used. The gas was pumped at a speed of 15 liter/min.

The distribution function $f(E)$ and the electronic density were determined from probe measurements as described elsewhere.[7] Cross sections for CO_2 and CO given by Schultz[8] and Boness and Schulz[9] were used.

The effective relaxation constant β was determined by measuring the spontaneous side light emission of the (001) level at 4.3 μ.

This was done as follows: A mechanical chopper within the cavity switched the discharge periodically from a lasing to a nonlasing condition. The chopper exposed the laser beam within 60 μsec and the beam remained fully exposed for 650 μsec.

The 4.3-μ wavelength spontaneously emitted radiation was detected and from the temporal variation of the emission β can be deduced as follows: The temporal variation of the populations of the CO (1) and CO (001) is given by

$$\frac{d(N_{001} + N_{CO(1)})}{dt} = P_{001} - \beta N_{001} + P_{CO} - \gamma N_{CO(1)}, \tag{8}$$

where P_{001} and P_{CO} are the net excitation rates by electrons to the (001) and CO (1) level, respectively. The constant β presents the effective rate constant for all depopulation processes of the (001) level, and γ the rate constant for relaxation processes of CO (1) by collision with CO_2 molecules.

It follows that

$$\frac{dN_{001}}{dt} = P - \beta N_{001} - \left(\gamma N_{CO(1)} + \frac{dN_{CO(1)}}{dt} \right), \tag{9}$$

$$P = P_{001} + P_{CO_2}.$$

It can be shown that for CO concentrations in He-CO_2 lasing mixtures not exceeding 25% the term in the parentheses can be neglected; thus

$$\frac{dN_{001}}{dt} \approx P - \beta N_{001}, \tag{10}$$

namely, the temporal variation of the spontaneous emission measured at 4.3 μ is indeed proportional to the population of the upper lasing level N_{001}.

The relaxation constant β was measured for different discharge currents and is in good agreement with values quoted by Flynn *et al.*[10] for similar discharges.

Equation (7) has been solved for different discharge currents which involved separate solutions of Eq. (1) for three different concentrations of CO. The results are presented in Table I. The population inversion for a given concentration of CO is shown in Fig. 1.

TABLE I. Population inversion as a function of the degree of dissociation: $y = [N_{CO}/(N_{CO} + N_{CO_2})] \times 100$; β is the effective relaxation constant of the CO_2 (001) level in the discharge; n is the electron concentration; T is the gas temperature; I is the discharge current.

| Population inversion | | | | | | |
$y = 20\%$ (10^{14} cm^{-3})	$y = 10\%$ (10^{14} cm^{-3})	$y = 0\%$ (10^{14} cm^{-3})	β (sec^{-1})	n_e (10^9 cm^{-3})	T ($0°K$)	I (mA)
2.1	1.5	0.9	1900	2	360	20
2.55	1.8	1	2050	3	390	30
2.8	1.85	0.97	2200	4	420	40
2.85	1.75	0.75	2350	5	450	50
2.75	1.65	0.55	2500	6	480	60
2.6	1.45	0.25	2650	7	510	70
2.4	1.2	negative	2800	8	540	80
2.2	0.92	negative	2950	9	570	90
2	0.7	negative	3100	10	600	100

The influence of CO on the population inversion is striking. It is seen that were it not for CO, the population inversion at usual operating gas temperatures (450–500 °K) would not suffice to allow lasing in He-CO$_2$ mixtures. Moreover, even at a temperature as low as 360 °K, negligible gain should be detected in the absence of CO. It should be noticed that if the CO$_2$-CO (1) relaxation term in Eq. (9) were not neglected, even lower population inversions would result.

These results demonstrate, however, the contribution of direct excitation and relaxation by electrons of the CO$_2$ (001) molecule to the population inversion. This contribution has been neglected in previous investigations.[3]

Finally, it should be noticed that the population inversions presented here are in full agreement with gain measurements[11] which were performed in similar mixtures. Moreover they are also in agreement with the experimentally measured influence of CO on the lasing properties of He-CO$_2$ mixtures.[1]

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