THE ROLE OF CO IN CO_2 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 distributions and the measured effective relaxation constant of CO_2 (001). It is shown that the population inversion is negligible in the absence of CO.

CO is produced by dissociation in the glow discharge of a CO_2-laser. Its influence on the laser performance is well recognized [1, 2]; similarly to nitrogen, it receives energy from electrons and supplies it to the CO_2(001) level. Calculations by Gordietz [5], assuming that the excitation of the CO_2 (oov) mode 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 laser gas. In those calculations direct excitation of CO_2 molecules by electrons was neglected and a Maxwellian electron energy distribution assumed.

We want to show here 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 CO_2 (oov) levels, 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 assumptions are made:

1. A defined vibrational temperature exists for each of the different vibrational modes of the CO_2 molecule.

2. The symmetrical stretch mode (voo) and the bending mode (ovo) have a common vibrational temperature T.

3. The vibrational temperature T_v of the asymmetrical stretch of the CO_2 molecules and that of the CO molecules are nearly equal.

Assumptions (1) and (2) are well established [3, 4], and assumption (3) is based on the relatively high relaxation constant for vibrational energy transfer from CO to CO_2 (oov) [5].

The vibrational energy balance for the CO_2 (oov) and CO (v) levels is

$$\dot{Q}_{ev}(T_v) = \dot{Q}_{vv}(T_v) + \dot{Q}_{vt}(T_v) + \dot{Q}_{vd}(T_v) \quad (1)$$

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 by diffusion.

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

$$\dot{Q}_{ev}(T_v) = \dot{Q}_{vv}(T_v) + \dot{Q}_{vd}(T_v) + \dot{Q}_L(T_v) + \dot{Q}_{vt}(T_v). \quad (2)$$

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 obtained data.

The term $\dot{Q}_{ev}$ on the left hand side of (2) 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 transfer due to electron excitation, respectively:

$$\dot{Q}_{ev_1} = N_{CO_2}(Z_1)F_1, \quad \dot{Q}_{ev_2} = N_{CO}(Z_2)F_2$$

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

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

where

$$x_1 = \exp\left(-\frac{1388}{kT_v}\right); \quad x_2 = \exp\left(-\frac{667.4}{kT}\right);$$

$$x_3 = \exp\left(-\frac{2350}{kT_v}\right); \quad x_4 = \exp\left(-\frac{2180}{kT_v}\right).$$

1388 cm^{-1}, 667.4 cm^{-1}, 2350 cm^{-1} 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,

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

$$F_2 = \sum_{n=1}^{8} K_{0n}\, n\left[1 - x_4^n \exp\left(n\,\frac{2180}{\bar{E}}\right)\right]/(1-x_4); \quad (4)$$

$\bar{E}$ is the mean electron energy.

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

$$\left.\begin{aligned} K'_{01} &= n_e \int_0^\infty f(E)\,\sigma'_{01}(E)\sqrt{\frac{2E}{m}}\,dE \quad \text{for } CO_2 \\ K_{0n} &= n_e \int_0^\infty f(E)\,\sigma_{0n}(E)\sqrt{\frac{2E}{m}}\,dE \quad \text{for CO} \end{aligned}\right\} \quad (5)$$

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, $f(E)$ the electron energy, and n_e the electron concentration.

In the case of CO_2 the harmonic oscillator model was used, thus $K'_{m,n+1} = (n+1)K'_{01}$. For CO, following Chen's model [6], 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

$$\dot{Q}_{vv} + \dot{Q}_{vt} + \dot{Q}_{vd} = \beta N_{CO_2} Z_1 x_3 . \quad (6)$$

If the relevant constants in eqs. (4), (5) and (6) are known, eq. (1) yields a vibrational temperature T_v, and the population inversion N can be calculated for a given gas temperature. The population inversion is thus

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

where N_{001} and N_{100} are the populations per cm^3 of the upper and lower 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 He-CO_2 gas laser with a central cathode and

Table 1

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(100) level in the discharge; n_e is the electron concentration; T is the gas temperature; I is the discharge current

Population inversion			β	n_e	T	I
y = 20%	y = 10%	y = 0%				
($10^{14}/cm^{-3}$)	($10^{14}/cm^{-3}$)	($10^{14}/cm^{-3}$)	(sec^{-1})	($10^9/cm^{-3}$)	(°K)	(mA)
2.1	1.5	0.9	1 900	2	360	20
2.55	1.8	1.0	2 050	3	390	30
2.8	1.85	0.97	2 200	4	420	40
2.85	1.75	0.75	2 350	5	450	50
2.75	1.65	0.55	2 500	6	480	60
2.6	1.45	0.25	2 650	7	510	70
2.4	1.2	negative	2 800	8	540	80
2.2	0.92	negative	2 950	9	570	90
2.0	0.7	negative	3 100	10	600	100

two anodes placed at the ends of the tube. The diameter of the tube was 26 mm, the distances anode-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 30 ltr/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 measured by Schulz [8, 9] were used. The effective relaxation constant β was determined by measuring the spontaneous light emission of the (001) level at 4.3 μm, while switching the discharge to a non-lasing condition. The method was similar to that used by Flynn et al. [10].

Eq. (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 1. 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. The population inversions presented here are in agreement with gain measurements [11] performed in similar mixtures. They are also in agreement with the measured influence of added (non-dissociative) CO on the lasing properties of He-CO_2 mixtures [1].

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