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Influence of CO on the population inversion in CO₂ lasers

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The population inversion in a CO₂-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 CO₂ (001) level. It is shown that the population inversion is negligible in the absence of CO. The importance of excitation and relaxation of CO₂ (001) by electrons is stressed.

The influence of CO₂, which is produced by dissociation in the discharge of an electrically excited CO₂ laser, is well recognized. Both experimental and theoretical work support the view that CO₂ plays a similar role to that of N₂ in supplying vibrational energy to the upper lasing level (001) of the CO₂ molecule.

Theoretical calculations by Gordietz et al., assuming that the excitation of the asymmetric stretch mode of the CO₂ molecule results entirely from vibrational energy transfer between excited CO₂ molecules and CO₂ molecules in the ground state, show the dependence of the population inversion on the CO₂ concentration in the lasing mixture. In those calculations, direct excitation by electrons of CO₂ 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 (001) 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 CO₂ molecules. (b) The symmetrical stretch mode (V00) and the bending mode (00V) have a common vibrational temperature T. (c) The vibrational temperature T, of the asymmetrical stretch of the CO₂ molecules and that of the CO₂ molecules are nearly equal.

Assumptions (a) and (b) are well established⁴, and assumption (c) is based on the relatively high relaxation constant for vibrational energy transfer from CO to CO₂ (00V).³

The vibrational energy balance expressed in terms of quanta, stored and exchanged in the electrical discharge for the CO₂ (00V) and CO (V) levels can be presented by

\[ \dot{Q}_{\text{V}}(T) = \dot{Q}_{0}(T) + \dot{Q}_{\text{V}}(T) + \dot{Q}_{\text{FP}}(T), \]

where \( \dot{Q}_{\text{V}} \) is the net rate of supply of vibrational quanta by electrons to the above-mentioned levels; \( \dot{Q}_{0} \) is the net relaxation rate from these levels to any of the other vibrational modes; \( \dot{Q}_{\text{V}} \) is the vibrational-translational relaxation rate and \( \dot{Q}_{\text{FP}} \) 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):

\[ \dot{Q}_{\text{V}}(T) = \dot{Q}_{0}(T) + \dot{Q}_{\text{FP}}(T) + \dot{Q}_{L}(T) + \dot{Q}_{\text{FP}}(T). \]

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 \( \dot{Q}_{\text{FP}} \) on the left-hand side includes contributions both from CO₂ and CO. Neglecting anharmonicity in both cases, one obtains expressions for \( \dot{Q}_{\text{FP}} \) and \( \dot{Q}_{\text{FP}} \) for the CO₂ and CO vibrational energy storage due to electron excitation, respectively:

\[ \dot{Q}_{\text{FP}} = N_{\text{CO}_2} L_1 T_1; \quad \dot{Q}_{\text{FP}} = N_{\text{CO}} L_2 T_2, \]

where \( N_{\text{CO}_2} \) and \( N_{\text{CO}} \) are the concentrations of CO₂ and CO, respectively, \( (Z_i)^{-1} \) 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 CO₂ and CO, respectively. Now

\[ Z_1 = (1-x_1)(1-x_2)(1-x_3), \quad Z_2 = (1-x_4), \]

where \( x_1 = \exp(-1388/kT); \quad x_2 = \exp(-667.4/kT); \quad x_3 = \exp(-2350/kT); \quad x_4 = \exp(-2180/kT). \]

1388, 667.4, 2350, and 2180 cm⁻¹ are the energies of the CO₂(100), CO₂(010), and CO₂(001) and CO (1) levels above the ground state, \( k \) is the Boltzmann constant;

\[ F_1 = \frac{1-x_3}{1-x_4}, \quad F_2 = \frac{1-x_4}{1-x_4} \]

\[ Q_{\text{FP}} = \frac{1}{1-x_4} \exp[2350/\bar{E}], \]

and \( \bar{E} \) is the mean electron energy.

The rate constants for excitation \( K_{\text{ex}} \) and \( K_{\text{em}} \) are

\[ K_{\text{ex}} = n_e \int_0^e f(E) \sigma_{\text{em}}(E)(2E/m)^{1/2} dE, \quad \text{for CO}_2 \]

\[ K_{\text{em}} = n_e \int_0^e f(E) \sigma_{\text{em}}(E)(2E/m)^{1/2} dE, \quad \text{for CO} \]

where \( \sigma_{\text{em}}(E) \) and \( \sigma_{\text{em}}(E) \) are the cross sections for excitation from the ground state by electrons to CO₂ (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 CO₂, the harmonic oscillator model was used, thus \( K_{\text{ex}} = (n+1)K_{\text{ex}} \). For CO, following Chen's model, excitation via a compound state was assumed, namely, \( K_{\text{ex}} = K_{\text{ex}} \).

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

\[ \dot{Q}_{\text{FP}} + \dot{Q}_{\text{FP}} + \dot{Q}_{\text{FP}} = \beta N_{\text{CO}_2} Z_1 x_3. \]

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FIG. 1. Population inversion as a function of discharge current; $y$ (degree of dissociation) = $[N_{CO}/(N_{CO} + N_{CO1})] \times 100$.

If the relevant constants in Eqs. 4–6 are known, Eq. (1) yields a vibrational temperature $T_v$ and the population inversion $\Delta N$ can be calculated for a given gas temperature. The population inversion is thus

$$\Delta N = N_{001} - N_{100} = N_{CO2} Z_1 (x_1 - x_1),$$

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 $\sigma_0$ and $\sigma_01$, the effective relaxation constant $\beta$, 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. Cross sections for CO$_2$ and CO given by Schultz and Boness and Schulz were used.

The effective relaxation constant $\beta$ was determined by measuring the spontaneous side light emission of the (001) level at 4.3 $\mu$m.

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 $\mu$s and the beam remained fully exposed for 850 $\mu$s.

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

$$\frac{d(N_{001} + N_{CO1})}{dt} = \beta N_{001} + P_{CO} - \gamma N_{CO1},$$

where $P_{CO}$ and $P_{CO}$ are the net excitation rates by electrons to the (001) and CO (1) level, respectively. The constant $\beta$ presents the effective rate constant for all depopulation processes of the (001) level, and $\gamma$ the rate constant for relaxation processes of CO (1) by collision with CO$_2$ molecules.

It follows that

$$\frac{dN_{001}}{dt} = \beta N_{001} - \left(\gamma N_{CO1} + \frac{dN_{CO1}}{dt}\right),$$

$$P = P_{001} + P_{CO2}.$$

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

$$\frac{dN_{001}}{dt} \approx \beta N_{001},$$

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

The relaxation constant $\beta$ was measured for different discharge currents and is in good agreement with values quoted by Flynn et al. 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 1. 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: $y = [N_{CO}/(N_{CO} + N_{CO1})] \times 100$; $\beta$ is the effective relaxation constant of the CO (001) level in the discharge; $n_0$ is the electron concentration; $T$ is the gas temperature; $I$ is the discharge current.

<table>
<thead>
<tr>
<th>Population inversion</th>
<th>$y = 20%$ (10$^{14}$ cm$^{-3}$)</th>
<th>$y = 10%$ (10$^{14}$ cm$^{-3}$)</th>
<th>$y = 0%$ (10$^{14}$ cm$^{-3}$)</th>
<th>$\beta$ (sec$^{-1}$)</th>
<th>$n_0$ (10$^9$ cm$^{-3}$)</th>
<th>$T$ (°K)</th>
<th>$I$ (mA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2, 1</td>
<td>1.5</td>
<td>1.2</td>
<td>1</td>
<td>1900</td>
<td>2</td>
<td>360</td>
<td>20</td>
</tr>
<tr>
<td>2, 55</td>
<td>1.8</td>
<td>1</td>
<td>1</td>
<td>2050</td>
<td>3</td>
<td>390</td>
<td>30</td>
</tr>
<tr>
<td>2, 8</td>
<td>1.85</td>
<td>0.97</td>
<td>0.97</td>
<td>2200</td>
<td>4</td>
<td>420</td>
<td>40</td>
</tr>
<tr>
<td>2, 65</td>
<td>1.75</td>
<td>0.75</td>
<td>0.75</td>
<td>2300</td>
<td>5</td>
<td>450</td>
<td>50</td>
</tr>
<tr>
<td>2, 75</td>
<td>1.65</td>
<td>0.55</td>
<td>0.55</td>
<td>2500</td>
<td>6</td>
<td>450</td>
<td>60</td>
</tr>
<tr>
<td>2, 6</td>
<td>1.45</td>
<td>0.25</td>
<td>0.25</td>
<td>2650</td>
<td>7</td>
<td>510</td>
<td>70</td>
</tr>
<tr>
<td>2, 4</td>
<td>1.2</td>
<td>negative</td>
<td>2800</td>
<td>8</td>
<td>540</td>
<td>80</td>
<td></td>
</tr>
<tr>
<td>2, 2</td>
<td>0.92</td>
<td>negative</td>
<td>2950</td>
<td>9</td>
<td>570</td>
<td>90</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>0.7</td>
<td>negative</td>
<td>3100</td>
<td>10</td>
<td>600</td>
<td>100</td>
<td></td>
</tr>
</tbody>
</table>

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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-CO₂ 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 CO₂-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 CO₂ (001) molecule to the population inversion. This contribution has been neglected in previous investigations.

Finally, it should be noticed that the population inversions presented here are in full agreement with gain measurements 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-CO₂ mixtures.