Population mechanisms in visible carbon monoxide pulsed lasers

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The population mechanism in the Ångstrom system of a CO pulsed laser are studied through analysis of the rotational intensities distribution of the emitted bands. The observed spectra were simulated by using a simple excitation model. The results suggest that, apart from electron impact, there is a strong radiative contribution of the VUV 4^+ system to the gain of the visible emission through a selective depletion of the lower laser level.

I. Introduction

Carbon monoxide laser action can be obtained in three different spectral regions: the IR (~5 μm) involving transitions between vibrational levels of the ground electronic state; the visible ($B^1\Sigma^+ \rightarrow A^1\Pi$, Ångstrom system) covering nearly the whole visible spectrum in seven bands, and the VUV emission ($A^1\Pi \rightarrow X^1\Sigma^+$ fourth positive system) in the 1800–2000-Å region.

While the IR emission has been extensively studied,[1] mainly due to its potential applications in fields such as medical sciences, material processing, and atmospheric studies, there are few papers dealing with the visible[2–5] and VUV[6] emission characteristics.

Although CO can easily be obtained in a pure chemical way, and can be made to lase using a simple excitation circuit, its main drawback is the poor stability of the gas under high power electric discharges.[1–5] To overcome this problem for the IR laser, special mixtures including O_2, N_2, He, and Xe have been used.[1] With pulsed excitation and at low repetition frequencies, the difficulty can be avoided by removing the gas in the tube between successive excitation pulses and working under a fairly slow flow regime.

It has been previously shown[7,8] that the population mechanisms of the N_2 laser can be studied by analyzing the rotational structure of the bands emitted. In this work we use this method for studying the processes by means of which population inversion in the Ångstrom system of the CO molecule is achieved.

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II. Experimental setup

We used a 10-mm bore discharge tube of 85-cm active length, with a cladding for liquid air to increase the gain of the stimulated emission by working at low temperature.[5] The pressure of CO in the tube varied from 0.5 to 1.5 Torr. The gas was used in a flow regime to avoid decomposition of the CO molecule due to electric discharge phenomena.

Excitation was accomplished by discharging a 5-nF capacitor bank charged to 20 kV. The spectra were recorded on 103 a–F Kodak spectroscopic plates using an Ebert-mount spectrograph of 3.4-m focal length with a 600 grooves/mm plane grating. Dispersion in the first diffraction order was 4.8 Å/mm.

The microdensitograms corresponding to the spectra obtained were reduced to intensities through the characteristic curve of the plate.

III. Experimental results

The spectra of the 0–5, 0–4, 0–3, and 0–2 visible CO laser bands at pressures ranging from 0.6 to 1.3 Torr were recorded with the same exposure time. Figure 1 shows four typical microdensitograms of these bands for certain pressure values. We observed that the rotational structure of the different bands consisted of Q and P branches. No R-branch lines were observed in any case.

All the spectra shown in Fig. 1 correspond to optimum pressure. For lower pressure values, Q-branch lines are the only ones observed, except for the 0–2 band [Fig. 1(a)] which showed P-branch lines for all pressure values.

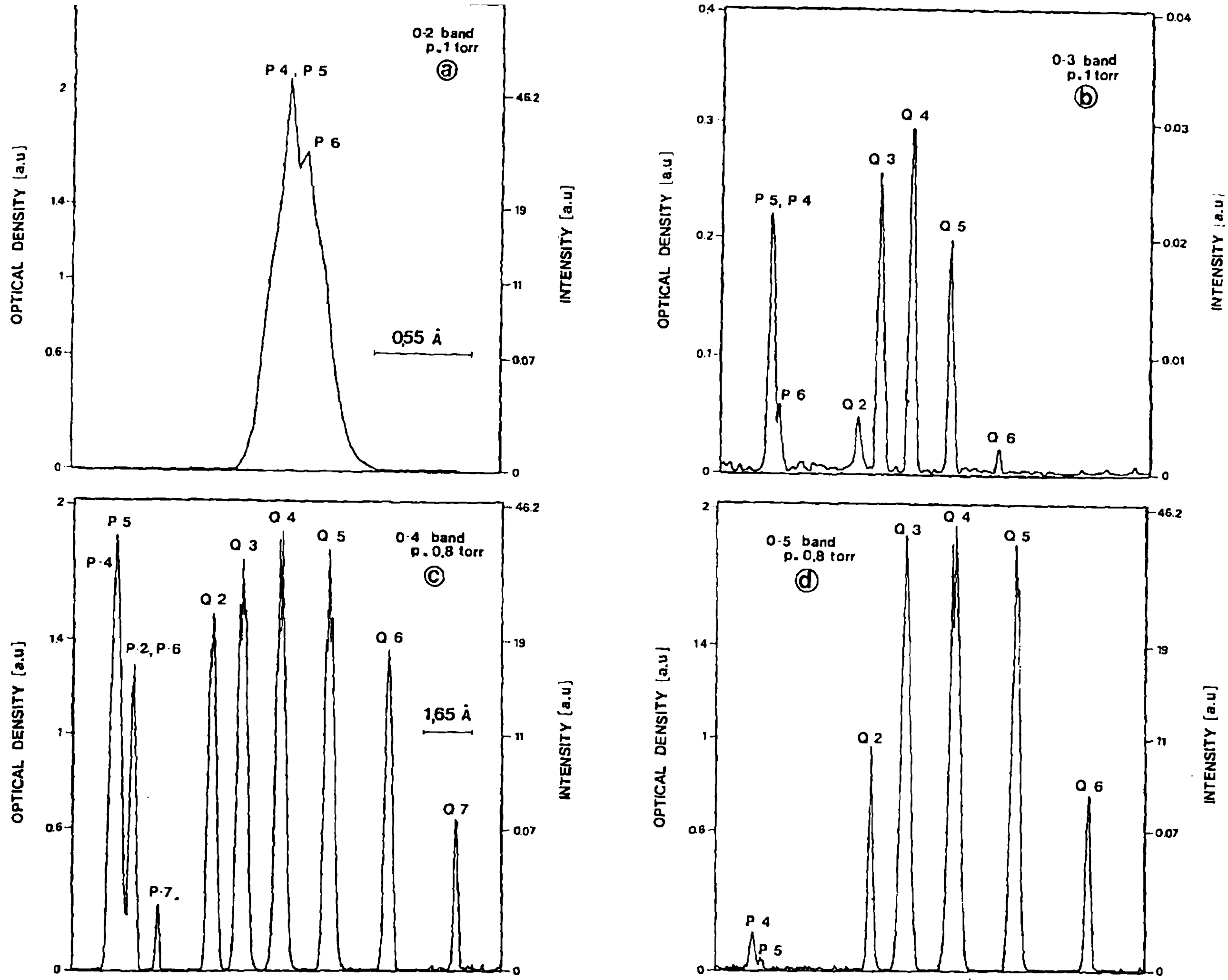


Fig. 1. Microdensitograms of the CO laser bands: (a) 0–2, (b) 0–3, (c) 0–4, and (d) 0–5 Typical pressure values are shown. Dispersion of (b), (c), and (d) is that shown in (c).

IV. Theoretical Calculations and Discussion

The population mechanisms of the upper and lower laser levels can be studied by simulating the spectra obtained. Such simulation is carried out by calculating the gain of the electronic—vibro—rotational transitions of interest, based on a simple excitation model of both laser levels (Fig. 2).

We assume that

(a) The $B^1\Sigma^+$ state belongs to Hund's case (a).[9] This state is populated by direct electron impact (DEI) from the $X^1\Sigma^+$ ground state.[3] Due to its short lifetime (25 ns),[10] the collision frequency in the range of pressures employed is not high enough[11] to thermalize the population during the time the visible laser emission occurs (100 ns). Thus, the rotational population distribution reproduces that of the ground state, so the population of a rotational level of the $B^1\Sigma^+$ state can be written as

$$N_J^B = N_v^B(2J+1)\exp\left[-B_x\frac{hc}{kT}J(J+1)\right], \qquad (1)$$

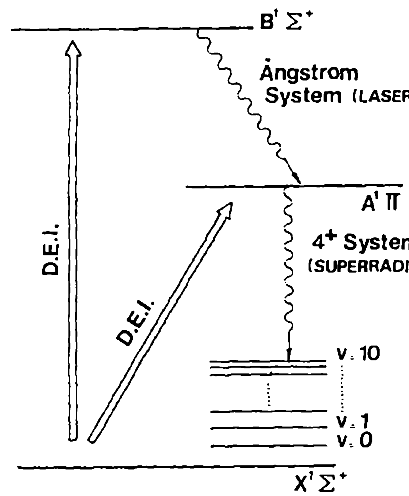


Fig. 2. Simplified energy level diagram of the CO molecule. The excitation and decay processes of the laser levels are shown.

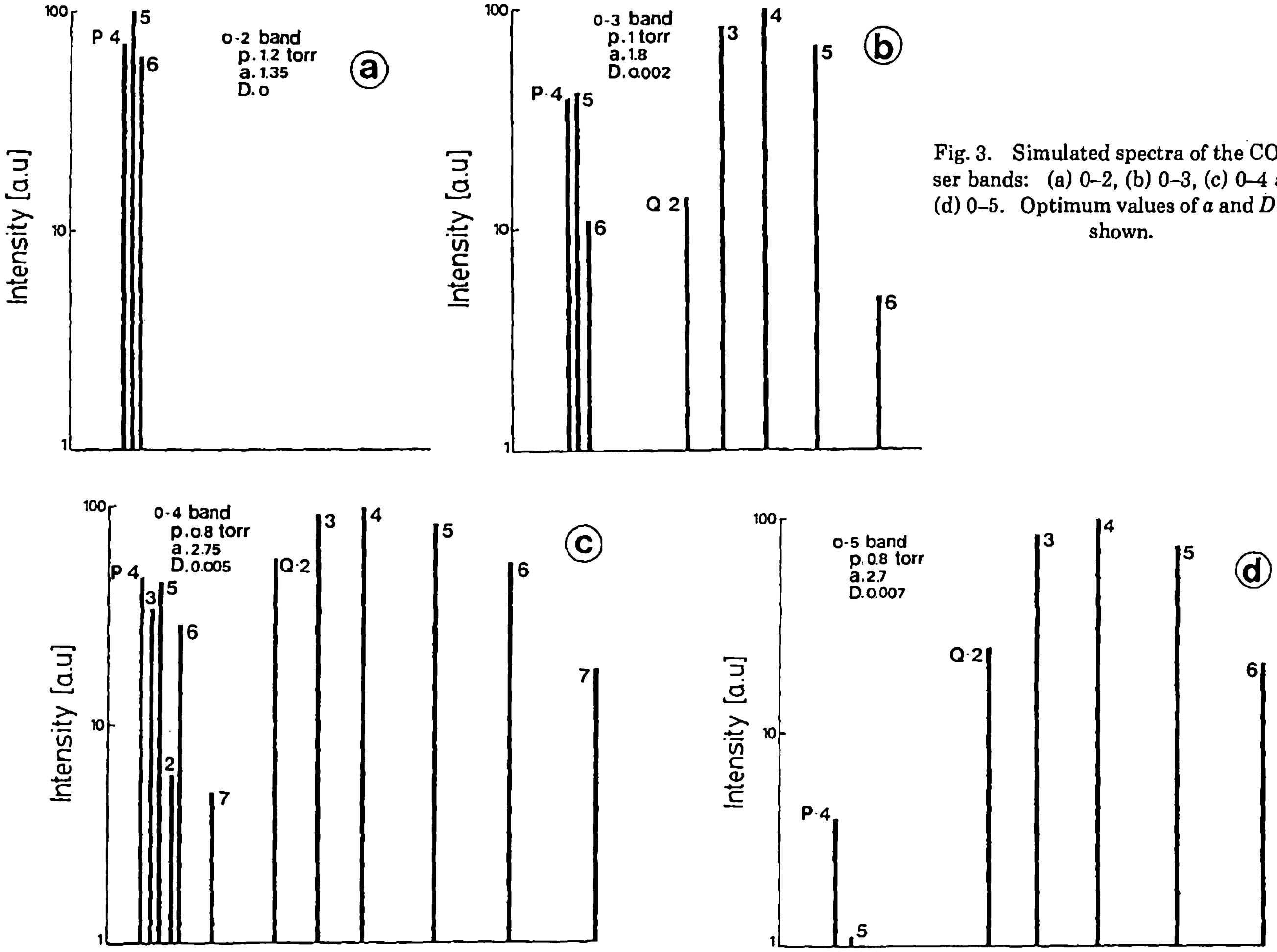


Fig. 3. Simulated spectra of the CO laser bands: (a) 0–2, (b) 0–3, (c) 0–4 and (d) 0–5. Optimum values of a and D are shown.

where $N_v{}^B$ is the vibrational level population,
J is the rotational quantum number,
B_x is the rotational constant of the $v = 0$ level of the ground state,
h is Planck's constant,
c is the velocity of light,
k is the Boltzmann constant, and
T is the rotational temperature.

(b) The $A^1\Pi$ state belongs to Hund's case (a).[9] The vibrational levels $v = 2, 3, 4$, and 5 are populated by DEI from the ground state[3] and reproduce its rotational population distribution due to its short lifetime (17 ns).[12] As the $A^1\Pi$ state is the upper level of the VUV laser transition, it was assumed that its vibrational levels $v = 2, 3, 4$, and 5 are depopulated by the VUV radiative transition. The fact that the VUV laser bands have strong Q-branch, weak R-branch, and no P-branch lines[6] implies that there is a selective depopulation of the $A^1\Pi$ rotational levels creating a favorable condition for population inversion of the Q branch in the visible transition. Thus, the rotational population of the $A^1\Pi$ state can be written as a combination of two processes, DEI and depopulation by radiative transition:

$$N_J{}^A = N_V{}^A \left((2J+1) \exp\left\{ - B_x \frac{hc}{kT} [J(J+1) - 2\Omega^2] \right\} \right) [1 - D2(J+1)] , \tag{2}$$

where $N_V{}^A$ is the vibrational level population of the $A^1\Pi$ state,
Ω is the total angular momentum projection, and
D is the parameter measuring the percentage of VUV emission.

The gain of the laser lines is proportional to the population inversion and the transition probability. Thus, we can write

$$G = \left(\frac{N_J{}^B}{g_J{}^B} - \frac{N_J{}^A}{g_J{}^A} \right) i_{J_B \rightarrow J_A}, \tag{3}$$

where $g_J{}^B, g_J{}^A$ are the statistical weights of the rotational levels, and
$i_{J_B \rightarrow J_A}$ is the rotational transition probability or Hönl-London factor as given by Kovacs.[13]

Then, introducing Eqs. (1) and (2) and the corresponding Hönl-London factor into Eq. (3), we can write the gain for the P, Q, and R branch as

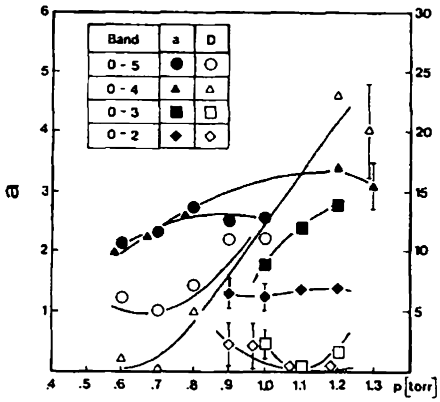


Fig. 4. Variation of a and D with pressure for each CO laser band. For some cases, error bars are shown.

$$G_P = \frac{1}{2}(J+1)\left(\exp\left\{-B_x \frac{hc}{kT}[J(J-1)]\right\} - \frac{1}{a}\exp\left\{-B_x \frac{hc}{kT}[J(J+1)-2]\right\}\right), \tag{4}$$

$$G_Q = \frac{1}{2}(2J-1)\left[\exp\left[-B_x \frac{hc}{kT}J(J+1)\right] - \frac{1}{a}([1-D(2J+1)]) \times \exp\left\{-B_x \frac{hc}{kT}[J(J+1)-2]\right\}\right)\right], \tag{5}$$

$$G_R = \frac{1}{2}J\left(\exp\left[-B_x \frac{hc}{kT}(J+1)(J+2)\right] - \frac{1}{a}\exp\left\{-B_x \frac{hc}{kT}[J(J+1)-2]\right\}\right), \tag{6}$$

where J is the rotational quantum number of the $A^1\Pi$ state and $a = N_v^B/N_v^A$ is the vibrational population inversion.

In a first approximation, the intensity was made proportional to the gain when the latter was found bigger than a certain threshold value G_t, and zero otherwise, so

$$I = \begin{cases} 0 & \text{if } G < G_t, \\ 100\dfrac{G-G_t}{1-G_t} & \text{if } G > G_t. \end{cases} \tag{7}$$

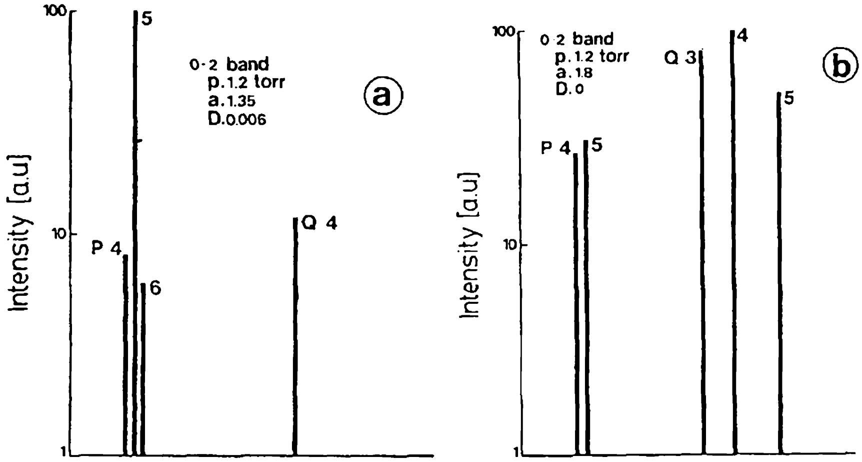
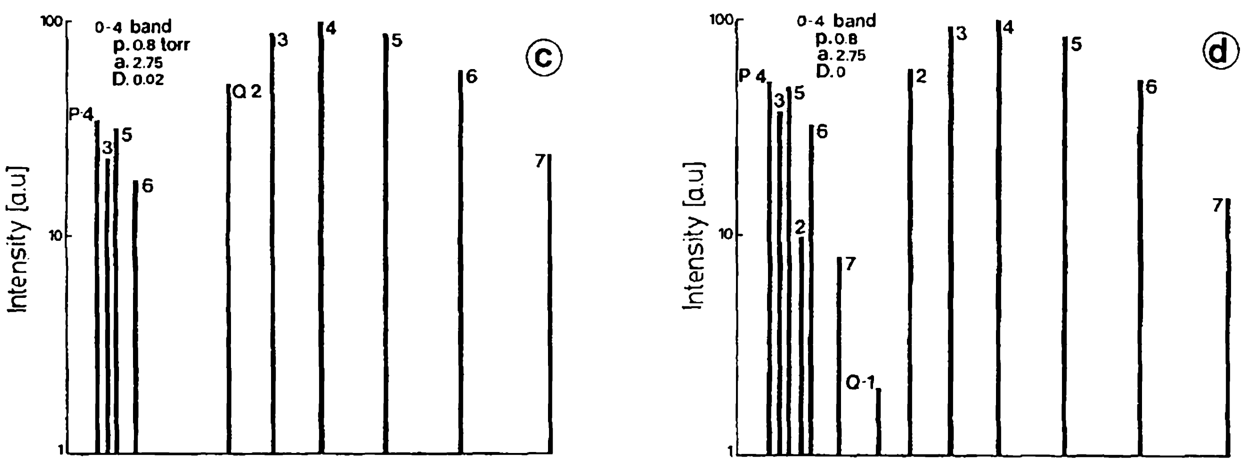


Fig. 5. Simulated spectra of the CO laser bands: (a) and (b) 0–2 and (c) and (d) 0–4 for nonoptimum a and D values.



Introducing Eqs. (4)–(6) into Eq. (7) and varying the parameters a, D, and G_t, the spectra observed were simulated by using a weighted least-squares method. The simulated spectra are shown in Fig. 3 and correspond to those depicted in Fig. 1. In each case, the optimum values of a and D are shown.

The dependence of these values on the pressure is shown in Fig. 4. It can be seen that, in general, a increases with pressure, a fact that indicates a better excitation efficiency of the upper laser level at higher pressures.

For the 0–4 and 0–5 bands, D increases rapidly with pressure, suggesting that, at least for these two bands, depopulation of the lower level enhances the gain of the bands at high pressures.

Simulated spectra calculated for values of a and D different from the optimum are shown in Fig. 5. For the 0–2 band it can be seen that any variation of D from this value results in the appearance of the Q-4 line and changes the relative intensities of the different lines within the band [Fig. 5(a)]. A change in a drives the maximum intensity to the Q-4 line and makes the Q-3 and Q-5 lines appear [Fig. 5(b)].

Similarly, in the 0–4 band, a D value above the optimum one makes the P-2 and P-7 lines disappear [Fig. 5(c)], while a D value below the optimum results in the appearance of the Q-1 line. This shows that the method is very sensitive to any change of a and D, a fact that enables it to fit spectra that show different rotational structure.

V. Conclusions

CO pulsed laser emission spanning the whole visible spectrum can be readily obtained by using a simple excitation circuit. The population mechanisms of the Ångstrom system were studied by analyzing the spectroscopic laser intensity data of the emitted bands and using simple theoretical calculations simulating the spectra observed. The results suggest a strong VUV 4^+ system cascade contribution to the visible transition gain, specially on the 0–4 and 0–5 bands. This enhancement of the gain is produced by the selective depopulation, through the VUV radiative transition of the lower laser level, on the Q-branch lines of the visible spectra.

We now plan to study the stimulated emission of the 4^+ system of CO and its relation to the Ångstrom system.

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