

Theory and Experiment of Low Transitions in CO Discharge Lasers

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A self-consistent model, which couples the kinetics of the electrons and heavy particles with the optical and fluid dynamic processes, has been employed to identify the various parameters and explain the mechanism responsible for producing low-lying transitions in slow-flowing CO lasers. Both theory and experiment indicate that lasing on low-lying transitions can be achieved at low temperatures for low pressures (or low-flow rates) together with high partial pressures of He and N₂. The role of N₂ has been identified as an additive responsible for reducing the electron temperature to a range at which the transfer of electrical power to the lower vibrational modes of CO is optimum.

Introduction

It is known that the CO laser oscillates on many vibrational-rotational lines. Transitions involving higher vibrational bands are strongly attenuated by atmospheric water vapor; however, low-lying transitions with wave lengths below 5 μ m propagate in the atmosphere with relatively high transmittivity.¹ Moreover, the (1-0) transition can be used for the detection of CO in long path absorption measurements. Thus, the tunability of the CO laser over the low-lying bands makes it a valuable tool for atmospheric transmission and probing experiments in the 5 μ region.

The object of this study is to identify the various parameters and explain the mechanism responsible for producing low-lying vibrational transitions in CO laser systems. Experiments²⁻⁴ have shown that low temperature and low CO concentration in a CO-N₂-He electric discharge system are two factors influencing the appearance of low-lying vibrational transitions. Both of these effects were evident in the recent analysis of the high flow laser systems by Smith and Hassan.⁵ For the studies reported in this paper, small signal gain and intensity measurements were performed with a low-pressure CO-starved electric discharge laser for a variety of pressures, flow rates, and gas mixtures. In addition, a self-consistent model was formulated and was used to evaluate the manner in which the operating conditions, i.e., pressure, flow rate, mixture ratios, and power, affect the low-lying transitions. The need for a self-consistent model became evident because earlier analyses of slow-flow CO lasers^{2,6-8} assumed certain flow properties that could be determined as part of the solution. Thus, the theories of Refs. 2, 6, and 7 assume the electron distribution function to be Maxwellian at some given electron temperature; moreover, these theories assume the electron number density and the gas temperature. The effect of such assumptions is that important phenomena resulting from the coupling of the electrons and heavy particles' kinetics and fluid dynamic processes are ignored.

Presented as Paper 75-850 at the AIAA Fluid and Plasma Dynamics Conference, Hartford, Conn., June 16-18, 1975; submitted July 9, 1975; revision received Dec. 29, 1975. Supported in part by NASA Grant NSG-1013. The authors would like to express their appreciation to F. Allario and N.S. Smith for many helpful discussions.

Index categories: Thermochemistry and Chemical Kinetics; Plasma Dynamics and MHD; Lasers.

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Center and Caledonia⁸ also have presented calculations for slow-flow CO lasers in which the distribution function was obtained from a solution of an appropriate Boltzmann equation; however, they had to assume a value for the gas temperature. In contrast to the preceding theories, the present model couples the kinetics of the electrons and heavy particles with the optical and fluid dynamic processes in the laser system. It treats molecules in different quantum states as different species and employs the electron Boltzmann equation and the conservation of species, mass, momentum, and energy equations to describe the system. For a given geometry, voltage, current, gas composition, pressure, mass flow rate, and reflectivities, the theory can predict the number densities of the electrons and the excited states of the heavy particles, the electron and gas temperatures, the intensities of the lasing transitions, and efficiency.

These theoretical results are compared with gain and intensity measurements obtained with a slow-flow CO electric discharge laser of conventional design (see Fig. 1). In general, the comparisons show good agreement and indicate the validity of the model developed here.

Theoretical Analysis

Governing Equations

For the slow-flow laser system under consideration, the governing equations are the electron Boltzmann equation, the conservation of species, mass, momentum, and energy equations, and the equation of state. The electron Boltzmann equation can be written as

$$\begin{aligned} \frac{E^2}{3} \frac{d}{d\epsilon} \left[\sum_s (N_s Q_{sm}) \frac{df}{d\epsilon} \right] + \sum_s \left(\frac{2m_e}{m_s} \frac{d}{d\epsilon} \left[\epsilon^2 N_s Q_{sm} f \right] \right) \\ + \sum_s \left(\frac{2m_e}{m_s} \frac{kT}{e} \frac{d}{d\epsilon} \left[\epsilon^2 N_s Q_{sm} \frac{df}{d\epsilon} \right] \right) + \sum_s N_s \left[\sum_j (\epsilon + \epsilon_{sj}) f \right. \\ \left. \times (\epsilon + \epsilon_{sj}) Q_{sj} (\epsilon + \epsilon_{sj}) - \epsilon f(\epsilon) \sum_j Q_{sj}(\epsilon) \right] = 0 \quad (1) \end{aligned}$$

where f is the electron distribution function, ϵ is the electron energy in electron volts, E is the electric field, N_s is the number density of species s , m is the particle mass, k is the Boltzmann constant, T is the gas temperature, e is the electronic charge, Q_{sm} is the momentum transfer cross section for elec-

trons in species s , and Q_{sj} is the inelastic collision cross section. Equation (1) assumes that the gas density is high enough so that terms due to the electron density gradients may be neglected.⁹ It neglects also the effects of the superelastic collisions. This is because the number of excited states is small compared to the ground state; moreover, the superelastic collisions affect mostly the high energy tail of the distribution function and this has very little effect on the excitation rates that enter into the theory.

Using the steady state approximation, the species conservation equations reduce to the statement that the production rates R_s for the various species are zero. Before one can write these expressions, one needs to specify the important kinetic processes in the laser system. In this work, the following processes are taken into consideration: 1) electron impact excitation of the first eight vibrational levels of CO and N₂, 2) single quantum vibration-vibration ($V-V$) exchange collisions (CO-CO, CO-N₂, N₂-N₂), 3) single quantum vibration-translation ($V-T$) exchange collisions (CO-CO, CO-N₂, CO-He, N₂-CO, N₂-N₂, N₂-He), 4) spontaneous radiative decay by CO ($\Delta v=1$ and $\Delta v=2$), and 5) stimulated emission and absorption by CO. It should be noted that Boness and Schultz¹⁰ measured the relative cross sections for vibrational excitation of levels 9 and 10 for both N₂ and CO. Because their relative magnitudes were rather small, they were not included in this work. For the preceding process, the production rate for the v th vibrational levels of CO can be expressed as

$$\begin{aligned}
 R_{CO,v} = & N_e \left[\sum_{j=0}^8 (N_{CO,j} P_{jv}^{CO}) - N_{CO,v} \sum_{j=0}^8 (P_{vj}^{CO}) \right] + N_{CO,v+1} \left[\sum_{j=0}^{v_{CO}} (N_{CO,j} VV_{v+1,j}^{CO-CO}) + \sum_{j=0}^{v_{N_2}} (N_{N_2,j} VV_{v+1,j}^{CO-N_2}) \right. \\
 & + \sum_s (N_s VT_{v+1}^{CO-s}) + A_v^{(1)} \left. \right] - N_{CO,v} \left[\sum_{j=0}^{v_{CO}} (N_{CO,j+1} VV_{j+1,v}^{CO-CO} + N_{CO,j} VV_{v,j}^{CO-CO}) \right. \\
 & + \sum_{j=0}^{v_{N_2}} (N_{N_2,j+1} VV_{j+1,v}^{N_2-CO} + N_{N_2,j} VV_{v,j}^{CO-N_2}) + \sum_s N_s (BVT_{v+1}^{CO-s} + VT_v^{CO-s}) + A_v^{(1)} + A_v^{(2)} \left. \right] \\
 & + N_{CO,v-1} \left[\sum_{j=0}^{v_{CO}} (N_{CO,j+1} VV_{j+1,v-1}^{CO-CO}) + \sum_{j=0}^{v_{N_2}} (N_{N_2,j+1} VV_{j+1,v-1}^{N_2-CO}) + \sum_s (N_s BVT_v^{CO-s}) \right] + N_{CO,v+2} A_{v+2}^{(2)} \\
 & + \frac{\gamma_{v+1,j} I_{v+1,j}}{h\nu_{v+1,j}} - \frac{\gamma_{v,j} I_{v,j}}{h\nu_{v,j}}
 \end{aligned} \quad (2)$$

where P , VV , VT , and BVT denote electron excitation, vibration-vibration, vibration-translation, and backward vibration-translation rate coefficients, respectively. γ is the gain coefficient and is taken from Ref. 7, $I_{v,j}$ is the intensity, h is Planck's constant, ν is the frequency, $A_v^{(1)}$ and $A_v^{(2)}$ are the spontaneous decay coefficients, J is the rotational quantum number, and v_{CO} and v_{N_2} are the maximum numbers of CO and N₂ levels employed in the analysis. Similar expressions may be written for R_{v,N_2} . It is to be noted that values of $N_{s,v}$ for $v > v^*$ are not set equal to zero; instead, they are estimated from the assumption that the vibrational distribution in the tail region is Maxwellian. This procedure, which is due to Rich et al.,¹¹ avoids the problem of false inversions that can appear when one sets $N_{s,v} = 0$ for $v > v^*$.

The overall continuity equation can be written as

$$\rho u A = \dot{m} \quad (3)$$

where ρ is the density, u is the mean velocity, A is the cross-sectional area, and \dot{m} is the mass flow rate. For the slow-flow

laser under consideration, the flow Mach number is of the order of 10^{-2} . For such Mach numbers, the flow is essentially incompressible and the density is approximately constant. When this result is used in the momentum equation, which can be written in the form

$$\gamma M^2 \rho' (Du'/Dt') + \nabla' p' = 0 \quad (4)$$

where γ is the ratio of specific heats, M is the Mach number, p is the pressure, and a prime denotes a dimensionless quantity, one finds that at low Mach numbers

$$\nabla p \approx 0 \text{ or } p \approx \text{const} \quad (5)$$

The overall energy equation can be written as

$$\dot{m}(H - H_0) = IV - Q_w - Q_s - Q_l \quad (6)$$

where H is the specific enthalpy at the bulk temperature, H_0 is the specific enthalpy at the entrance temperature, I is the current, V is applied voltage, Q_w is the wall heat loss, Q_s is the spontaneous emission losses, and Q_l is the laser power output. These quantities may be expressed as

$$\begin{aligned}
 H &= Y_e h_e + Y_{He} h_{He} + \sum_{v=0}^{v_{CO}} Y_{CO,v} h_{CO,v} + \sum_{v=0}^{v_{N_2}} Y_{N_2,v} h_{N_2,v} \\
 Y_s &= \rho_s / \rho \quad h_e = 5/2 (kT_e / m_e) + I_{CO} / m_e \\
 h_{He} &= 5/2 (kT / m_{He}) \\
 h_{s,v} &= 7/2 (kT / m_s) + E_v^s / m_s \quad s = N_2, CO
 \end{aligned} \quad (7)$$

$$Q_w = 6\pi (T - T_w) \lambda_m L \quad (8)$$

$$\begin{aligned}
 Q_s &= AL \left\{ \sum_{v=1}^{v_{CO}} N_{CO,v} A_v^{(1)} (E_v^{CO} - E_{v-1}^{CO}) \right. \\
 &\quad \left. + \sum_{v=2}^{v_{CO}} N_{CO,v} A_v^{(2)} (E_v^{CO} - E_{v-2}^{CO}) \right\}
 \end{aligned} \quad (9)$$

$$Q_l = AL \sum \gamma_{v,j} I_{v,j} \quad (10)$$

$$\lambda_m = 0.5 \left[\sum x_s \lambda_s + \left\{ \sum (x_s / \lambda_s) \right\}^{-1} \right] \quad (11)$$

where E_v^s is the vibrational energy of the v th vibrational level of species s , I_{CO} is the ionization potential of CO, λ_s is the thermal conductivity of species s , L is the active length of the cavity, and x_s is the mole fraction of species s . The expression for Q_w was derived in Ref. 12 on the assumption that the

average or bulk temperature of a fully developed flow in a tube of radius a is defined as

$$T = (1/a) \int_0^a T(r) dr \quad (12)$$

However, for the Mach numbers under consideration the usual definition of the average gas temperature is¹³

$$T = [T(r)u(r)dA] / [u(r)dA] \quad (13)$$

Thus, if one assumes that $u(r)$ is constant, as was implied in the derivation of Ref. 12, then the expression for Q_w is not correct. If one recalls, however, that for a fully developed flow $u(r)$ is given by

$$u(r) = u_m [1 - (r/a)^2] \quad (14)$$

where u_m is the centerline velocity, then use of Eq. (14) in Eq. (13) results in Eq. (8). Thus, the result given in Ref. 12 is correct, but its derivation is not. Equation (11), together with the thermal conductivities, were obtained from Ref. 14.

The equation of state can be written as

$$p = kN_e T_e + kNT \quad N = \sum N_s, \quad s = \text{He}, N_2, \text{CO} \quad (15)$$

where N_e and T_e are determined in terms of the current density and the electron distribution function as

$$N_e = j/ew \quad w = \mu E = \mu N(E/N) \quad (16)$$

$$\mu = -\frac{e}{3N} \left(\frac{2}{m_e} \right)^{1/2} \int_0^\infty \frac{\epsilon}{\sum x_s Q_{sm}} \frac{df}{d\epsilon} d\epsilon$$

$$T_e = \frac{2}{3} \frac{e}{k} \int_0^\infty \epsilon^{3/2} f(\epsilon) d\epsilon \quad (17)$$

The function f that appears in Eqs. (16) and (17) is normalized, i.e., it satisfies the relation

$$\int_0^\infty \epsilon^{1/2} f(\epsilon) d\epsilon = 1 \quad (18)$$

Rate Data

All of the cross sections needed in Eq. (1), with the exception of the CO electronic excitation cross sections, are available in tabular form in Ref. 15. Cross sections for the electronic excitation of CO are generated from semi-empirical formulas given in Ref. 16. The rates for the electronic impact vibrational excitation from the ground state and de-excitation to the ground state can be expressed as

$$P_{ov}^s = \left(\frac{2e}{m_e} \right)^{1/2} \int_0^\infty \epsilon Q_{sv}(\epsilon) f(\epsilon) d\epsilon$$

$$P_{vo}^s = \left(\frac{2e}{m_e} \right)^{1/2} \int_0^\infty \epsilon Q_{sv}^*(\epsilon) f(\epsilon) d\epsilon \quad (19)$$

where the cross section Q_{sv}^* for de-excitation is obtained from Q_{sv} , using the principle of microscopic reversibility¹⁷

$$Q_{sv}^* = (\epsilon + \epsilon_{sv}) Q_{sv} (\epsilon + \epsilon_{sv}) \quad (20)$$

The $V-V$ and $V-T$ rates were taken from Ref. 5. In particular, $V-T$ rates employed in this study incorporate Bray's¹⁸ and Shin's¹⁹ modification of the SSH theory; constants in the theory were determined from a least-squares fit of available measurements.²⁰⁻²⁶ On the other hand, the $V-V$

rates were obtained from Jeffers and Kelly's theory,²⁷ with the constants being adjusted by a least-squares fit of data reported in Refs. 28-34.

Method of Solution

The governing system of equations consists of a difference-differential equation, Eq. (1), and a large number ($v_{CO}^* = v_{N_2}^* = 50$) of nonlinear algebraic equations, $R_s = 0$, together with Eqs. (6) and (15). Equation (1) was solved numerically, using a fourth-order Adams-Moulton predictor-corrector method for a range of mixtures and E/N values, whereas the algebraic equations were solved using a Newton-Raphson method. Two types of calculations were carried out: the first consisted of a small signal gain calculation, whereas the second consisted of intensity calculations for a given optical cavity.

Because the temperature is determined as part of the solution, knowledge of the pressure, voltage, and tube dimensions is not sufficient to determine E/N . Therefore, the calculations start with an assumed gas temperature; this will make it possible to determine E/N from the given pressure, voltage, and tube dimensions, and to integrate Eq. (1). The electron temperature follows from Eq. (17), whereas the electron number density is determined, for a given current density, from Eq. (16). The next step depends on the type of calculations to be undertaken. For the small signal gain calculation, the intensities are set equal to zero and the conservation equations are used to calculate the number densities of the various excited states and the gas temperature. If the assumed temperature does not agree with the calculated temperature, the previous procedure is repeated until convergence is achieved. The gain coefficients then are calculated by use of the expression given in Ref. 7.

When lasing takes place, the values of the intensities $I_{v,J}$ are such that the average gain coefficient for each transition satisfies the threshold condition

$$\gamma_{v,J} = -\ell n(r_1 r_2) / 2L \quad (21)$$

where r_1 and r_2 are the reflectivities of the mirrors. The expression for the gain coefficient⁷ assumes that the rotational states are in equilibrium at the gas temperature. Because of this, it is not possible to predict the J numbers that characterize the various lines that may contribute to the power output in a given vibrational band. Therefore, it is assumed that the laser emission in a given band occurs on the P -branch transition that has the maximum gain coefficient.

The subscripts v and J in Eq. (21) are not known *a priori*, but are determined as part of the solution. An estimate of the values of these subscripts is obtained from the small signal gain calculation, which gives, among other things, the bands whose small signal gain coefficient is greater or equal to the threshold value and the maximum P -branch transition in each band. From this information, the conservation equations, together with Eq. (21), are solved for the number densities of the various excited states, the gas temperature, and the intensities. From this calculation, the maximum small signal gain coefficient is recalculated to determine if any adjustment needs to be made in the vibrational bands considered or in their peak transitions. If such an adjustment is needed, or if the assumed temperature does not agree with the calculated temperature, the previous procedure is repeated until convergence on v, J , and T is achieved.

Experimental Apparatus

The experimental results were obtained in a conventional longitudinal discharge tube, shown in the schematic in Fig. 1. The discharge tube was pyrex, 2.5 cm in diam and 92 cm long. Annular stainless steel electrodes, 2.5 cm in diam and 8 cm long, were fitted to the tube ends and calcium fluoride Brewster angle windows were placed on the electrode ends. The tube, exclusive of the electrodes, was immersed in a liquid

nitrogen bath in a bakelite container. Compartments at either end of the container were designed with adjustable liquid nitrogen leaks to control the temperature of the electrodes. The constituent gases were premixed in an upstream reservoir and were introduced at the cathode and exhausted near the anode. The gas reservoir was sufficiently large to allow reasonable long running times with negligible drop in volume flow rate. Tube pressure at the gas entrance and exit ports and in the reservoir were measured with Wallace and Tiernan gages. The pressure drop in the tube was negligibly small for the operating conditions of the experiment. Volume flow rates were measured with a Hastings flow meter. The gas mixture was pumped with a pumping capacity greater than 1000 l/min.

For measurements of the total power and spectral distribution of the power, the optical cavity consisted of a gold-coated 3-m radius-of-curvature total reflector at one end and one of several different partially transmitting mirrors at the other. These included a hole-coupled mirror with a 2-mm hole, and mirrors with reflectivity coefficients of 95%, 90%, and 84% at $5\mu\text{m}$. Power output was measured with a Coherent Radiation Laboratories model 201 detector. The output spectra were recorded by use of a McPherson model 218, 0.2m, scanning monochromator.

The small-signal gain measurements were made employing an oscillator identical in all respects to that described previously. Individual vibrational-rotational lines were selected from the oscillator by use of a grating with either the 10% or 16% transmitting mirror. Two techniques were used to obtain gain measurements. In the first technique, the output of the oscillator was directed through the amplifier and detected with a thermistor bolometer, fed into a PAR lock-in amplifier, and recorded on a strip-chart recorder. The amplifier was turned on and off to record gain measurements. The second technique employed the slope technique described in Refs. 35 and 36. A 50% beam splitter directed the oscillator output to two thermistor bolometer detectors, one placed before the entrance window of the amplifier and the other beyond the exit window. The detector outputs were fed to two PAR lock-in amplifiers, the outputs of which were recorded on an X-Y recorder. In this case, the small-signal gain is proportional to the log of the two slopes generated as the probe laser power is varied with the amplifier laser first turned on and then turned off. This technique has the advantage of allowing detection of saturation effects, which exhibited themselves as variation from a straight line when the oscillator power was varied with the amplifier on.

Any error bars on the data points of the gain coefficient are believed to arise because of current fluctuations in the electrical discharge. Both methods of measuring gain gave approximately the same value for the gain coefficient and associated error.

Results and Discussion

The primary processes of pumping energy into the CO vibrational levels are electron excitation of the lower levels ($v \leq 8$) and $V-V$ pumping of the higher levels. Radiative decay and $V-T$ collisions are the primary processes for deactivating the vibrational states, and these processes become dominant at the higher levels. Thus, in order to promote lower-level transitions, one needs to increase the efficiency of lower-level excitations and $V-T$ collisions, and reduce the efficiency of $V-V$ pumping. In order to reduce the efficiency of $V-V$ pumping, one must reduce the pressure and partial pressure of CO in the system. The presence of a large fraction of He increases the efficiency of the $V-T$ collisions and helps maintain a low gas temperature. Nitrogen was found to be beneficial in the experiment of Djeu^{3,4} in promoting low-level transitions; he attributed its role to the fact that $(v-v-1)$ transitions in CO are in resonance with the $(v+6 \rightarrow v+5)$ transitions in N_2 . Because of these considerations, calculations were carried out for two types of mixtures: in the first, where N_2 was varied, $\text{He-N}_2\text{-CO-O}_2$ ratios ranged from 700-0-5-

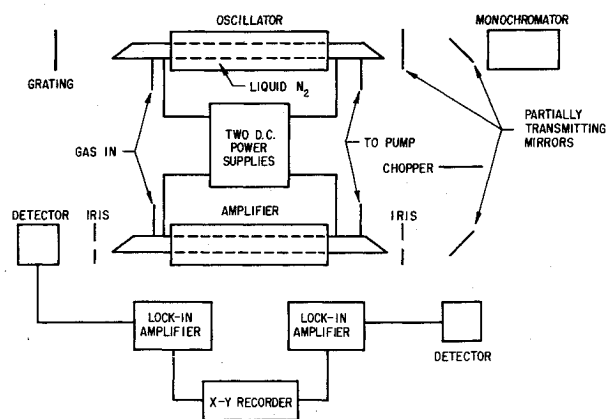


Fig. 1 Schematic of apparatus for small-signal gain measurements.

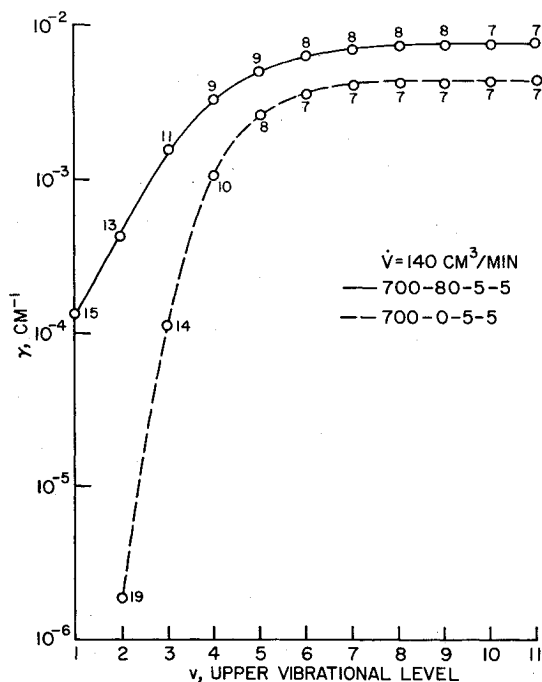


Fig. 2 Effect of N_2 concentration on maximum gain coefficient.

5 to 700-80-5-5, and in the second, where CO was varied, the corresponding ratios were in the range 700-80-5-5 to 700-80-80-5. The volume flow rate and pressure were varied from 140-700 cm^3/min and 3.5-9.0 Torr, respectively. This range of parameters corresponded to the range of operating conditions employed in the experiment.

The effect of N_2 fraction on the maximum small signal gain coefficient is seen in Fig. 2, which indicates that it increases with N_2 fraction. The numbers associated with the various points indicate the P -branch transition at which the gain coefficient is maximum. The reason for this behavior is attributed to the sharp decrease in the electron temperature, from 28,170 K in the absence of N_2 ($p=3.5$ Torr, $I=7.3$ mA, $V=3.4$ kV) to 13,666 K in the presence of N_2 ($p=3.5$ Torr, $I=6.3$ mA, $V=5.4$ kV). This decrease in the electron temperature results in an increase in the fraction of electrical power transferred to the lower levels of both CO and N_2 , Ref. 37. Moreover, based on calculations for the mixture 700-10-5-5 ($p=3.5$ Torr, $I=7.0$ mA, $V=4.0$ kV) and 700-80-5-5, the percentage of excited N_2 molecules for $v > 3$ increases with N_2 fraction; this helps pump the lower CO levels because $v+6 \rightarrow v+5$ transitions in N_2 are in resonance with $(v-v-1)$ transitions in CO. Although this contributes to the pumping of lower levels, it is not the dominant mechanism.

In this work, the cross sections given in Ref. 41 are employed. These cross sections allow for the dependance on J , but not for the temperature. Allowing for the temperature-dependence suggested in Ref. 40 reduces the calculated gain coefficient by 25%. Thus, the discrepancy is not a result of

not employing the proper gain coefficient or the proper cross section.

The discrepancy can be attributed to a number of factors, the most important of which is that the theory assumed a rather clean experiment in which impurities such as H_2O , CO_2 , and the nitrogen oxides were nonexistent. This, of course, is not a realistic assumption. Another reason for the discrepancy is that O_2 was treated as an inert gas in the theory; this is because O_2 was added in the experiment to prevent the dissociation of CO and the formation of carbon. Because the calculations employed the experimentally determined electric field and current, the effect of O_2 on performance cannot be attributed to a change in the electron distribution function resulting from changes in E/N which accompany the addition of O_2 , Ref. 42. The authors are inclined to believe that the presence of O_2 results in the formation of NO, and this can deactivate the excited levels of CO.

In addition to the preceding, recent analysis of gain measurements has indicated that measured values of gain coefficient can be substantially below the true value; this takes place in the presence of internal gain saturation and when large collector apertures are employed.⁴³ The situation was further compounded by the fact that, for low CO concentrations, the laser operation was not very stable.

Concluding Remarks

The theory presented here suggests that lasing on low transitions in CO laser systems can be achieved at low flow rates (or pressures) for rather low CO partial pressures relative to those of He and N_2 . The primary role of N_2 appears to be the reduction of the electron temperature to a level at which the transfer of the electrical power to the lower vibrational states is optimum. In addition to clarifying the role of N_2 , the present theory predicts rather accurately the rotational numbers at which the intensity is maximum in a given band. As a result of this, it is concluded that the present model, if modified to include the effects of impurities that inherently exist in any given experiment, is quite adequate for modeling low-and moderate-pressure CO laser systems.

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