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A THERMODYNAMIC MODEL FOR CO$_2$ LASERS 1)
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1. Introduction

In order to optimize the functioning of a CO$_2$ laser with wave guide, it is important to be able to calculate the dependence of the small signal gain as a function of a number of parameters such as: gas mixture, total pressure, discharge current, tube diameter, temperature of the cooling jacket, etc. If the kinetic temperature and the population of the laser levels are known as a function of these parameters, the gain equations are uniquely determined. The complexity of the energy level diagram for the CO$_2$ molecule, the large number of rates of collisional excitation and relaxation and their dependence on temperature, diffusion and relaxation processes through collisions with the walls of the enclosure, the great number of additives that participate in the population inversion, add to the difficulty of the analytical treatment of the laser effect and of the gain, saturation and oscillation bandwidth.

Since its inception, a number of models were proposed based on the rate equations relating the vibrational modes of the CO$_2$ and N$_2$ molecules. Thus, Moore et al [1] have divided the energy levels in three groups (each characterized by a single vibrational temperature), and solved the system of equations for the continuous wave regime. A comparison with experiment shows that this is a good simplification. Previous works considered separately the vibrational levels of nitrogen [2], [3], and 4 or 5 groups including also the

1 Received on December 19, 1977
vibrational levels of Co[4].

The new elements in this work are: small tube diameters, high pressures and current densities characteristic to the wave guide lasers. These differences imply faster excitation and relaxation rates and also collisional widening of emission lines. The most recent determinations of the excitation and relaxation rates and their temperature dependence are utilized. This data was not available when [1] - [4] were published.

The main limitation of works dealing with thermodynamic models for CO₂ lasers are using calculated excitation and relaxation rates [2-5], which can differ appreciably from the measured ones; the gas temperature is determined from the wrong relationship [2-5];[1] neglects relaxation through diffusion and collision at the walls of the enclosure and is limited to a 3 level system; none of these works take into account the gas pressure dependence of the diffusional relaxation rate, and neglect the losses inherent when guiding radiation through small diameter tubes; [5] is limited to a fixed excitation rate and only one diameter of the discharge tube and does not take into account the saturation processes. Some recent attempts [63], [7] eliminate some of these problems but the system of equations used is still simplistic (3 level system).

The model presented here is used for lasers with wave guide. It can be used also for other CO₂ lasers with axial discharge and gas flow. For lasers without gas flow, the modifications of the rate equations are due to the kinetic temperature of the gas and eventual gaseous components which participate in the inversion in the CO₂ molecule (especially the carbon monoxide).

2. Physical Model

The lower vibrational energy levels for the CO₂ and N₂ system are shown in figure 1.
These levels are divided into four groups: group 0, including the fundamental levels (unexcited) of the CO₂ and N₂ molecules, with populations N₀ and N₀ measures respectively; group 1, including the CO₂ molecules excited in the modes of symmetric elongation and deformation, with population N₁; group 2, CO₂ molecules with asymmetric elongation modes excited, population N₂; group 3, containing nitrogen molecules with one or more of the vibrational levels excited and population N₃. The total densities of CO₂ and nitrogen molecules are Nₜ and respectively N₃t.

Dividing the system CO₂ and N₂ in four groups seems justified by the rapid relaxation of the vibrational energy within each group, at a rate much larger than that of exchange of energy between groups. Thus CO₂ molecules in state 0000 relax to state 0001 at a rate larger than 5 x 10⁶ s⁻¹ torr⁻¹ [8], and the nitrogen molecules relax from the upper levels to state v = 1 in approximately 1 μs [9]. Also the relaxation of the vibrational energy between levels in group 1 is rapid, it is thus possible to characterize the whole group by a single vibrational temperature. Groups 1-3 are excited through electronic collision with rates R₁ - R₃ and relax through diffusion and collisions with the walls of the enclosure with rates k₁ₚ - k₃ₚ. Group 1 is excited and relaxes collisionally with rates k₁₀ and k₀₁ respectively, and group 3 relaxes through electronic collisions at a rate k₃e. Vibrational energy is exchanged between groups 3 and 2 through a rapid V-V transfer, characterized by a rate k₃₂ and the inverse rate k₂₃, and between groups 2 and 1 through collisions with molecules and atoms (rates k₂₁ and k₁₂), collisions with discharge electrons (rate k₂ₑ), through induced emission (rate W₂₁) and induced absorption (rate W₁₂). The inverse rate k₂₃ between groups 2 and 3 is taken to be equal to the direct rate k₃₂. Rate k₃₀ (due to collisions with molecules, atoms and electrons) is neglected, because such a collision is much more likely to relax a CO₂ molecule from
Figure 1. Diagram of lower vibrational energy levels for the CO$_2$ and N$_2$ system, grouped for the 4 level approximation:

1 - energy;

state $00^1$ to one of the states in group 1, rather than the fundamental level [9]. We assume also that the degree of dissociation of the CO$_2$ molecule is small (justified for the case of lasers with gas circulation).

2.1. Kinetic Temperature of the Gas

To calculate the gas temperature in the discharge one needs to solve the Fourier heat equation for the continuous wave generation regime

$$\tau^2 T - \eta \lambda \frac{\partial T}{\partial x} = 0,$$

where $T$ is the gas temperature in K, $\eta$ is the rate of change
of the heat due to sources and radiators expressed in $\text{Wm}^{-3}$, and $\lambda_{\text{mix}}$ is the thermal conductivity of the gas mixture in $\text{WK}^{-1}\text{m}^{-1}$.

The source of heat is that part of the electron energy which is transferred to the translational energy of the molecules. Thus the radial distribution of the heat source is the same as the radial distribution of the electrons in the discharge.

From the theory of the positive column the radial distribution of the electron density is a zero order Bessel function, if the distribution is not affected by the strong radial temperature gradient

$$\eta = q_0 J_0(2.105 \, r/a). \tag{2}$$

Solving the heat equation with this $q$ in cylindrical coordinates with boundary condition $T(a) = T_p$, for a cylinder of radius $a$ and length $L$ (length of discharge), one obtains

$$T - T_p = \frac{q}{\lambda_{\text{mix}}} \left( \frac{a}{a'} \right)^2 J_0(2.105 \, r/a). \tag{3}$$

If we also consider the temperature gradient in the wall of the discharge tube (exterior radius $a'$) then

$$T = T_p + q_0 t^2 \left[ 0.1729 \lambda_{\text{mix}} + \frac{1}{2} \ln \left( \frac{a'}{a} \right) \right] J_0(2.105 \, r/a), \tag{4}$$

where $\lambda_p$ is the thermal conductivity of the wall material. In the center of the tube ($r=0$) the gas temperature can be calculated from

$$T = T_p + q_0 t^2 \left[ 0.1729 \lambda_{\text{mix}} + \frac{1}{2} \ln \left( \frac{a'}{a} \right) \right]. \tag{5}$$

The total amount of heat which increases the translational energy of the molecules per unit time, $Q$, is obtained by integrating the source $q$ over the volume.
Thus

\[ T = T_p + \frac{Q}{L} \left[ \frac{0.1274}{\lambda_{\text{max}}} + \frac{0.3686}{\lambda_p} \ln \left( \frac{a'}{a} \right) \right]. \]  

(Works [2] - [5] have used this formula, but the proportionality coefficient for \( T - T_p \) was 2.4 times smaller).

In calculating the temperature of the gas mixture in the center of the tube, one has to take into account that not all the input power is turned into heat: part is turned into laser power; part to excite the molecules which in turn through diffusion and collisions lose it to the walls; and, in the case of lasers with gas circulation, part of the excited molecules leave the discharge zone, before their energy is turned into heat. Because of these, equations (5), (7) have to be adjusted by introducing a conversion coefficient \( \eta \):

\[ T - T_p = \eta \frac{Q}{L} \left[ \frac{0.1274}{\lambda_{\text{max}}} + \frac{1}{2\lambda_p} \ln \left( \frac{a'}{a} \right) \right]. \]  

For closed CO\(_2\) lasers, \( \eta \) varies between 0.5 and 0.8 [10], and for those with gas circulation between 0.05 and 0.3.

One can see from (9) that \( T - T_p \) is proportional with the input electrical power per unit length (Q/L), thus the axial temperature increases with increasing current and pressure and for same Q/L is independent of the radius of the tube.

They showed that the radial distribution of the electron density differs only slightly from the Bessel profile used here for temperatures at the center of the tube less than 600 K. They also found the temperature at the center of the tube to be a function of the input power per unit length only.

The sources of heat are going to be estimated in two ways.

First by using eq. (3) assuming that the gas temperature is due to the energy transferred from the electrons to the molecular vibrations and subsequent relaxation to ground level. From fig. 1 one obtains

\[ \eta_0 = \left[ (R_1 - R_2)N_e - R_3^2 - k_3 N_e V_s \right] E_{b}, \tag{10} \]

where \( N_e \) is the electron density, \( E_b \) is the vibrational energy of quantum \( \nu_3 \), \( E_3 = 4.6658 \times 10^{-20} \) J.

Second, using eq. (9) and the input power per unit length estimated from

\[ Q = I \cdot E = I \cdot N \cdot \frac{E}{N} = I \cdot \frac{p \cdot E}{kT \cdot N}, \tag{11} \]

where \( N \) and \( p \) are the total density, and pressure of the gas mixture, \( E \) is the axial electric field and \( I \) the discharge current.

Lowke et al [12] have solved numerically the Boltzmann equation and derived the transport coefficients for the electrons in the gas mixtures specific to CO\(_2\) lasers. For \( j/p = 10^{-3} - 10^{-1} \) A cm\(^{-2}\) torr\(^{-1}\) typical for CO\(_2\) lasers with wave guide \( (j \) is the current density) \( E/N \) varies between \((2-5) \times 10^{-16} \) V cm\(^{-2}\). In the following we assume \( E/N = 3 \times 10^{-16} \) V cm\(^{-2}\) thus (11) becomes

\[ Q \cdot I \cdot (W \cdot m) \approx I \cdot (mA) \cdot p \cdot (torr). \tag{12} \]
The thermal conductivity of the gas mixture, $\lambda_{\text{mix}}$, is approximated using a formula given by Mason and Saxena [13]

\[
\lambda_{\text{mix}} = \sum_{i=1}^{n} \lambda_i \left(1 - \sum_{k=1}^{n} G_{ik} \phi_k \right)^{-1},
\]

where $n$ is the number of gases in the mixture, $\lambda_i$ and $\phi_i$ are the thermal conductivity and molar fraction of the $i$-th component, $G_{ik}$ is a function of the molecular weight of component $i$ and the viscosities of components $i$ and $k$. For the $CO_2 + N_2 + He$, and the mixture [3]

\[
\lambda_{\text{mix}} = \frac{\lambda_{CO_2} - 0.84 \phi_{N_2} \lambda_{N_2} - 0.33 \phi_{He} \lambda_{He}}{1 - 1.4 \phi_{N_2} \phi_{He} - 0.34 \phi_{He} \phi_{N_2} + 3.4 \phi_{N_2} \phi_{He} + 2.7 \phi_{He} \phi_{N_2}},
\]

where $\lambda_{CO_2}$, $\lambda_{N_2}$, $\phi_{He}$ are the conductivities of the pure gases and $\lambda_{mix}$, $\phi_{mix}$ are the molar fractions.

The thermal conductivities of the pure gases (in $\text{WK}^{-1}\text{m}^{-1}$) and their temperature dependence were measured by Booth and Sites [12]: $\lambda_{CO_2} = 4.58 \cdot 10^{-5} \ T^{1.04}$; $\lambda_{N_2} = 4.97 \cdot 10^{-4} \ T^{0.64}$; $\lambda_{He} = 5.48 \cdot 10^{-4} \ T^{0.62}$

The thermal conductivity of the wall $\lambda_p$, is for Pyrex glass (borosidicate 732) $\lambda_p = 0.172 \pm 0.33$ [15], for alumina $\lambda_p = 37.7$ [16] and for berillia $\lambda_p = 219.8$.

The temperature dependence of the thermal conductivity of the mixture $CO_2 + N_2$ and He is shown in fig. 2. Figure 3 presents the thermal conductivity $\lambda_{\text{mix}}$ of $CO_2 + He$ and $N_2 + He$ mixtures as a function of the molar fraction of the He. Due to the high thermal conductivity of He, the conductivity of the mixture is higher the higher the fraction of He and the higher the temperature.
Figure 2. Temperature dependence of the thermal conductivity for some CO$_2$ + N$_2$ and He mixtures.

Figure 3. Thermal conductivity of CO$_2$ and He and N$_2$ and He mixtures as a function of the molar fraction of He.
Due to the temperature dependence of the conductivity of the mixture, the temperature is calculated from equation (8) and (9) by iteration. We assume initially $T = T_p$, and then $\lambda_{m,0}$, and the temperature of the mixture are calculated. Then $\lambda_{m,0}$ is recalculated for the new temperature and the process is repeated. The iteration is continued until the difference between two successive calculated temperatures is less than 1 K.

Figure 4 shows the difference between the temperature at the center of the tube and the wall temperature as a function of the input power per unit length for the CO$_2$: N$_2$: He = 1:3:16 CO$_2$: N$_2$: He = 1:1:3:4 mixtures. The continuous lines are obtained using (9), the dashed lines are due to Laderma and Byron [11] for the same mixtures. The circles are experimental results by the same authors for the 1:3:16 mixture. The agreement between our calculation and the experimental data is excellent.

$T - T_p$ is a strong function of the wall material, as can be seen from figure 5. Thus, the curve $a' = 4$ mm is for Pyrex glass and the curve $a' = 1$ mm is for all purposes valid for alumina or beryllia tubes (or when the temperature gradient in the wall is neglected). For a Pyrex glass tube with $a = 1$ mm and $a' = 4$ mm and a mixture CO$_2$: N$_2$: He = 1:1:3, the temperature gradient in the wall for $T - T_p = 500$ K is over 20% of $T - T_p$. 
Figure 6 shows the difference between the temperature at the center of the tube and the wall temperature as a function of the total pressure, for various discharge currents. The continuous lines are calculated from (9), circles are calculated from (8), for the same discharge conditions. The populations in (8) are obtained by solving on the computer the system of equations presented in section 3. The ratio n*/n** was taken to be 1.45 for I=2 mA, 1.73 for I=5 mA and 1.97 for I=8 mA. The results obtained via these two methods are in very good agreement. In the following the temperature of the gas mixture at the center of the tube is calculated from (9) only, due to the fact that the computer time is much less.
Figure 4. Difference between temperature at the center of the tube and at the wall as a function of input power per unit length for the mixtures $CO_2: N_2:He = 1:3:16$ and $1:1:66:4$.

Figure 5: Difference between temperature at the center of the tube and at the wall as a function of input power per unit length with and without temperature gradient in the wall.

Figures 7-9 show the temperature of the gas mixture at the center of the discharge tube for $q^* = 0.15$, as a function of the total pressure, discharge current and respectively the wall temperature (temperature of the cooling agent). Figures 10-11 show the calculated temperature versus the molar fraction of $N_2$ and He respectively. The temperature is lower the lower the input power (smaller $p$ and $I$) the lower the temperature of the cooling jacket, the lower the molar fraction of $N_2$ and the molar fraction of the higher.
Figure 6. Difference between temperature at the center of the tube and at the wall, as a function of total pressure in the tube, for various discharge currents, calculated by the two methods.

Figure 7. Temperature of the gas mixture at the center of the tube as a function of total pressure, composition of the mixture as parameter (fraction of He).

Figure 8. Current dependence of the temperature of the gas mixture at the center of the tube.

Figure 9. Temperature of gas mixture at the center of the tube as a function of the capillary wall temperature.

Figure 10. Temperature of the gas mixture at the center of the tube as a function of \( N_2 \) content for the mixture \( CO_2: N_2: He \).

Figure 11. Temperature of the gas mixture at the center of the tube as a function of the content for the mixture \( CO_2: N_2: He \).
2.2. Electron Density

Experimental studies [10] have proven that the electron density is almost linear in the discharge current, the proportionality constant being a function of the gas mixture. The proportionality constant is estimated using the results in [12], which show that the drift velocity of the electrons, \( v_n \), for \( E/N \) in the range \((2-5) \times 10^{-16} \text{ V/cm}^2\), is in the range \((4-8) \times 10^6 \text{ cm/s}\) and depends only slightly on the gas mixture. The \( E/N \) ratio is determined by the necessity to keep the ionization at a high enough rate to balance the loss of charged particles in a self-maintained discharge, and to optimize the efficiency of the vibrational excitation in the laser discharge. Experimental measurements on a laser with waveguide [17] give a ratio \( E/N \) of \(2.3 \times 10^{10} \text{ V/cm}^2\) (from the slope of the voltage-pressure curve). For this value calculations in [12] give \( v_n = 4.10^6 \text{ cm/s}\). If we substitute the drift velocity in

\[
j = N_e v_n e, \tag{15}
\]

where \( N_e \) is the electron density, and \( e \) the electron charge, we obtain \( N_e \text{ (cm}^{-3}\) = \(1.5 \times 10^9 \sqrt{\text{mA/cm}^2}\). The optimum population inversion is in a narrow range of the fractional ionization \( N_e/N = 10^{-8} - 10^{-7}\). As has been previously shown, the electron distribution across the discharge tube is not uniform and thus the current density can not be obtained from the current/area ratio. Assuming the same radial electron distribution as in the previous paragraph (Bessel) and using the integration of this distribution from (6), one obtains:

\[
j \text{ (mA/cm}^2\) = 73.73 I \text{ (mA)}/\pi^2(\text{mm}^4) \tag{16}
\]

and thus \( N_e \text{ (cm}^{-3}\) = \(1.106 \times 10^{11} I \text{ (mA)}/\pi^2(\text{mm}^4)\).

2.3. Rates of Vibrational Excitation

Pumping rates electron molecule for each energy level, \( R \),
are a function of the transverse cross section of vibrational excitation by the electron, \( \sigma \), and of the electron distribution function, which in turn, is a function of all the important collisional processes.

\[
K = \sigma v_e N_e.
\]  

(17)

Thus the excitation rates for each vibrational level are different for each gas mixture and for each value of \( E/N \). Moreover in the case of \( N_2 \) molecules, the first 3 vibrational levels are excited. The problem is simplified significantly, if an effective rate for the vibrational excitation by electrons of the first vibrational level is defined.

Nighan [19] calculated these rates for \( N_2 \) and \( CO_2 \) as a function of the average electron energy (which for a Maxwell distribution represents the electron temperature). For average energies larger than 1.5 eV, typical for \( CO_2 \) lasers, one obtains
\[
R_1 = 4 \times 10^{-9} \text{ Ne},
\]
\[
R_2 = 6 \times 10^{-9} \text{ Ne},
\]
\[
R_3 = 2 \times 10^{-8} \text{ Ne}
\]
where \( R \) is in sec\(^{-1} \) and \( N_e \) is in cm\(^{-3} \).

2.4. Rates of Collisional Relaxation

The collisional relaxation rate \( k(s^{-1}) \) in a \( CO_2, N_2 \) and He mixture is obtained from

\[
k = k^{CO_2}p_{CO_2} + k^{N_2}p_{N_2} + k^{He}p_{He},
\]

or

\[
k = (k^{CO_2}\psi_{CO_2} + k^{N_2}\psi_{N_2} + k^{He}\psi_{He})p\]

(19)

where \( k^i \) are the collisional relaxation rates in the pure gases expressed in s\(^{-1} \) torr\(^{-1} \).

A survey of the experimental measurements of the rates \( k^i \) and their temperature dependence, is given in [8], [9]. In calculating population and gain the following values for the col-
lisional relaxation rates are used:

\[ k_{22}(cm^3s^{-1}) = 4.197 \cdot 10^{-14}T^{1/2} \exp \left[10^{-3}T(8.84 \cdot 10^{-4}T - 2.97)\right], \]  \(20\)

valid in the range 300-3000K; at 300K, \( k_{22} = 13623 \text{ s}^{-1} \text{ torr}^{-1}; \)

\[ k_{32} = \exp(11.64 + 7.495T^{-1/2} - 632.14T^{-1/2} + 2239T^{-1}), \]  \(21\)

valid in the range 250-2500K; at 300K, \( k_{21} = 329 \text{ s}^{-1} \text{ torr}^{-1}; \)

\[ k_{12} = \exp(-11.184 + 507.83T^{-1/2} - 4574.9T^{-1/2} + 12724T^{-1}), \]  \(22\)

valid in the range 300-1000K; at 300 K, \( k_{21} = 109 \text{ s}^{-1} \text{ torr}^{-1}; \)

\[ k_{21} = \exp(24.538 - 265.48T^{-1/2} + 763.32T^{-1/2} + 718.33T^{-1}), \]  \(23\)

valid in the range 300-1000K, at 300K, \( k_{12} = 73 \text{ s}^{-1} \text{ torr}^{-1}; \)

\[ k_{10} = k_{21} \exp \left(-\frac{\Delta E}{kT}\right) = k_{21} \exp(-1382.8T^{-1}), \]  \(24\)

valid in the range 300-3000K, at 300K, \( k_{10} = 219 \text{ s}^{-1} \text{ torr}^{-1}; \)

\[ k_{10} = 4.538 \cdot 10^8T^{-1/2} \exp(-72T^{-1/2}), \]  \(25\)

valid in the range 300-3000K, at 300K, \( k_{10} = 32 \text{ s}^{-1} \text{ torr}^{-1}; \)

\[ k_{10} = 4.635 \cdot 10^8T^{-1/2} \exp(-40633T^{-1}), \]  \(26\)

valid in the range 300-3000K, at 300K, \( k_{10} = 3.573 \text{ s}^{-1} \text{ torr}^{-1}; \)

\[ k_{10} = k_{10} \exp(-960, 38T^{-1}). \]  \(27\)

2.5. Rates of Electron Relaxation

The only levels which relax through electron collisions are \(00^01\) in \(CO_2\) and \(v = 1\) in \(N_2\).

The relaxation of the \(CO_2\) molecules from the \(00^01\) level through electron collisions takes place on one of the levels in group 1 and not directly to ground level, as was assumed in [2] - [5]. This rate is [20]: \( k_e = 1.77 \cdot 10^{-4} \text{ cm}^3 \text{ s}^{-1} \).
$N_s = 3 \cdot 10^3 \text{ cm}^{-3}$ typical for a waveguide laser $N_s k_m = 5.3 \cdot 10^8 \text{s}^{-1}$. This rate compares well with the measurements by Gower and Carswell [21] $(5 \cdot 10^3 \text{s}^{-1})$, and is somewhat smaller than that of Moore et al [1] $(2 \cdot 10^8 \text{s}^{-1})$.

The $v = 1$ level of $N_2$ is assumed to relax through electron collisions at a rate $k_m = R_g / N_s = 2 \cdot 10^{-8} \text{ cm}^3 \text{s}^{-1}$. This assumption was used by Moore [1] and Gordietz [2]-[4].

2.6. Relaxation rates through diffusion and collisions at the walls.

The relaxation rates through diffusion and collisions at the wall in calculated from [3]

$$k_p = \mu ^2 D / a^2, \quad (29)$$

where $D$ is the diffusion coefficient, and $\mu$ is the solution of

$$\mu \gamma_1 (\mu) = \left( \frac{\tilde{\alpha}}{2D} \right) \left( \frac{1 - r}{1 + r} \right) \gamma_0 (\mu), \quad (30)$$

where $\tilde{\alpha}$ is the average velocity, $\gamma_0 (\mu)$ and $\gamma_1 (\mu)$ are the Bessel functions of the first species of order 0 and 1 respectively, and $r$ is the reflection coefficient of the wall (reflection coefficient = accommodation coefficient).

For a hollow circular wave guide equation (30) can be simplified by a Taylor series development around $\mu = 2.405$ [7]

$$\mu = 2.405 \left[ \frac{(1 - r) \tilde{\alpha} - 2(1 + r)D}{(1 - r) \tilde{\alpha}} \right]. \quad (31)$$

The reflection coefficient of the wall was measured by Kovacs et al [22] and is $r = 0.78$. The average velocity of the CO$_2$ molecules depends on temperature only $	ilde{\alpha}(\text{cm/s}) = 2.195 \cdot 10^3 \sqrt{T(\text{K})}$

In a CO$_2$, N$_2$, He mixture the diffusion coefficient is
obtained from [8]

\[
\frac{1}{D} = \sum_i \frac{3N_i \sigma_i}{\dot{c} \pi} = \sum_i \frac{3p_i \sigma_i}{k \left( \frac{8k}{\pi M} \right)^{1/2} \alpha T^{3/2}},
\]

or

\[
\frac{1}{D_i} = \left( \frac{300}{T} \right)^{3/2} \sum_j \frac{p_j}{D_{ij}},
\]

where \( N_i \) is the density of species \( i \), \( \sigma_i \) is the collision cross section and \( \alpha \) is a constant. For the case of \( \text{CO}_2 \) and \( \text{N}_2 \)

\[
\frac{1}{D_{\text{CO}_2}} = \left( \frac{\phi_{\text{CO}_2} + \phi_{\text{N}_2} + \phi_{\text{He}}}{D_{\text{CO}_2-\text{N}_2} + D_{\text{CO}_2-\text{He}}} \right) p \left( \frac{300}{T} \right)^{3/2},
\]

\[
\frac{1}{D_{\text{N}_2}} = \left( \frac{\phi_{\text{CO}_2} + \phi_{\text{N}_2} + \phi_{\text{He}}}{D_{\text{N}_2-\text{CO}_2} + D_{\text{N}_2-\text{He}}} \right) p \left( \frac{300}{T} \right)^{3/2}.
\]

The \( D_{i-j} \) constants are those of Jordietz et al [3] at \( p = 1 \) torr: \( D_{\text{CO}_2-\text{He}} = 51 \text{ cm}^2/\text{s} \) (Fordietz gives \( 86 \text{ cm}^2/\text{s} \)), but this was corrected to agree with the value of 0.066 cm\(^2\)/s reported by Kovacs et al [22] at 1 atm for \( \text{CO}_2 \) molecules in the 0001 state), \( D_{\text{CO}_2-\text{N}_2} = 120 \text{ cm}^2/\text{s}, D_{\text{CO}_2-\text{He}} = 500 \text{ cm}^2/\text{s}, D_{\text{N}_2-\text{CO}_2} = 130 \text{ cm}^2/\text{s}, D_{\text{N}_2-\text{He}} = 160 \text{ cm}^2/\text{s}, D_{\text{N}_2-\text{N}_2} = 560 \text{ cm}^2/\text{s}.\)

Thus

\[
k_{1p} = \frac{\mu_{\text{CO}_2}}{\mu_{\text{N}_2}} D_{\text{CO}_2/a^2},
\]

\[
k_{3p} = \frac{\mu_{\text{N}_2}}{\mu_{\text{N}_2}} D_{\text{N}_2/a^2}.
\]

2.7 Fractions of excited molecules on the upper and lower laser levels

The fractions of molecules \( f_2 \) and \( f_1 \) on the upper and lower laser level are calculated from [23]

\[
f(i) = g_i \exp \left( -\frac{i \Delta E}{kT_i} \right) \left[ \sum_i g(i) \exp \left( -\frac{i \Delta E}{kT_i} \right) \right]^{-1},
\]
where $f(i)$ is the fraction of molecules on the $i$-th vibrational level, expressed for a simple harmonic oscillator; $g_i$ are the degeneracies, which for the symmetric and asymmetric elongation mode of $\text{CO}_2$ are unity and for the deformation mode of $\text{CO}_2$, \(^1\text{O}_2\), $g_q = q + 1$; $T_v$ is the vibrational temperature.

In particular for the $00^01$ level

$$f_1 = \frac{\exp(-\Delta E_1/kT_v)}{\exp(-\Delta E_1/kT_v) \sum_{i=1}^{\infty} \exp\left(-\frac{i\Delta E_1}{kT_v}\right)} = 1 - \exp\left(-\frac{\Delta E_1}{kT_v}\right) = 1 - x_1,$$

where

$$x_1 = \exp\left(-\frac{\Delta E_1}{kT_v}\right) = \frac{N_q Q_{\text{vb}}}{N_t},$$

$Q_{\text{vb}}$ is the vibrational partition function.

Because $N_q/N_t \ll 1$ is a good approximation. The $f_1 = 1$ modes are characterized by the same vibrational temperature, $T_{vl}$. The lower laser level relaxes rapidly collisionally to the $01^10$ level, half the level $(10^00, 02^00)$. Thus the fraction $f_1$ on the lower level is twice the fraction on the level $01^10$

$$f_1 = \frac{3x_1^2}{\sum_{i=1}^{\infty} (i+1)x_i^2},$$

where

$$x_1 = \exp\left(-\frac{\Delta E_1}{kT_{vl}}\right) = \frac{N_q Q_{\text{vb}}}{N_t}.$$

Summing in the denominator

$$f_1 = \frac{3x_1(1 - x_1)^2}{3 - 2x_1}.$$

for $T_{vl} \approx 300$ K, $x_1 = 0.04073$, $f_1 = 0.0387$. The function $f_1$ has a maximum for $x_1 = 0.10692$ ($T_{vl} = 1087, 87$ K), for which $f_1 = 0.19641$. At $T_{vl} \approx 400$ K, $f_1 = 0.09$. 

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2.3. Induced Emission and Absorption

The rate of induced emission in the presence of monochromatic radiation with intensity $I_c (W/m^2)$ at the center of the line, is obtained from Einstein's treatment [24]

$$W_u = \frac{A_{21} e^{2} \omega}{8\pi \nu_{L}^{2}} \gamma(0) F_{2},$$

where $A_{21}$ is the Einstein coefficient of spontaneous emission ($A_{21} = 0.2 \text{ s}^{-1}$) for the P(20) line of transition $00'1 \rightarrow (10'0, 02'0)^{11}$, $\gamma(0)$ is the form function at the center of the line, and $P_{u} = n_{u}/N_{u}$ is the fraction of molecules on the upper laser level (00'1) which are in the state characterized by the quantum rotational number $J \pm 1$. $\gamma(0)$ is obtained from

$$F_{2} = \frac{g_{J}}{Q_{rot}} \exp \left( -\frac{E_{J}}{k T_{r}} \right),$$

where $E_{J} = \hbar \nu_{J}$ is the energy of the rotational level $J$ with respect to the ground rotational level, and $E_{J} \approx B(J + 1)$; $Q_{rot} = k T_{r}/\hbar c B$ is the rotational partition function, $B$ is the rotation constant of the corresponding vibrations level and $\sigma$ is the molecular symmetry number ($\sigma = 2$ for the CO$_2$ molecule); $T_{r}$ the rotational temperature, assumed equal to the kinetic temperature of the gas. For $J = 20$ and $T_{r} = 400 \text{ K}$, $F_{2} = 0.063989$.

For the CO$_2$ laser, with wave guide, $\gamma(0) = 2/\nu_{c}$, where $\nu_{c}$ is the collision frequency, thus (44) becomes

$$W_u = 1.03 \cdot 10^{12} I_c \frac{1}{\nu_{c}}.$$

To calculate the collision frequency for the P(20) line one uses [25]

$$\nu_{c} = \zeta \left( \frac{3000}{T} \right)^{1/2} (\psi_{e} + a_{1} \psi_{x} + a_{2} \psi_{y}) p,$$
where \( \zeta = 2.248 \cdot 10^7 \text{K}^{1/2} \text{torr}^{-1} \), \( a_1 = 0.733 \), \( a_2 = 0.641 \).

The rate of induced absorption is correlated to that of induced emission by

\[
W_{12} = (g_2/g_1)W_{21} = 9.32 \cdot 10^{11} I(\nu)/\nu_c. \tag{48}
\]

The net exchange of molecules between the upper and lower laser levels due to induced emission and absorption is

\[
W = W_{21}N_2 - W_{12}N_1 = W_{21}(f_2N_2 - \frac{g_2}{g_1}f_1N_1). \tag{49}
\]

3. Equation system and its solution

Returning to figure 1 we can write the equation system which characterizes the population dynamics for the four levels of our model

\[
\frac{dN_1}{dt} = R_1N_a + r(k_{21}N_2 - k_{12}N_1) + s(k_{12}N_3 - k_{21}N_1) - k_{13}N_1 + k_{23}N_3N_2 - k_{21}N_2 - k_{12}N_1 - W, \tag{50}
\]

\[
\frac{dN_2}{dt} = R_2N_a - k_{23}N_2N_3 - k_{21}N_2 + k_{12}N_1 + k_{23}N_3 - k_{21}N_2 - N_2, \tag{51}
\]

\[
\frac{dN_3}{dt} = R_3N_3 - k_{23}N_2N_3 - k_{21}N_3 + k_{12}N_1 + k_{23}N_3 - k_{21}N_2 - k_{23}N_3, \tag{52}
\]

\[
N_1 = N_a - N_1 - N_2, \tag{53}
\]

\[
N_{3u} = N_3 + N_3. \tag{54}
\]

Coefficient \( r \) in (50) takes into account that the collisional relaxation of \( \text{CO}_2 \) molecules in group 2 (with \( \nu_2 \) excitation results in exciting more than one quantum of the \( \text{CO}_2 \) molecule from group 1 (with \( \nu_1 \) and \( \nu_2 \) quanta excited). Thus the reactions characterized by the global rates \( k_{21} \) and \( k_{12} \) are [8]

\[
(\text{CO}_2(00^01)) + M \rightarrow (\text{CO}_2(11^00, 03^10) + M + \cdot \tag{55}
\]

\[
(\text{CO}_2(00^01)) + M \rightarrow (\text{CO}_2(11^00, 03^10) + M + \cdot \tag{56}
\]
\[ \text{CO}_2(01^01) \rightarrow \text{CO}_2(20^00 \ldots 04^00)_{\text{III}} + \text{M} + 199 \text{ cm}^{-1}, \]  

(57)

where M can be CO\(_2\), N\(_2\) or He. Reactions (55), (56) are characterized by \( r = 3/2 \) and reaction (57) by a coefficient \( r = 2 \) [5].

Coefficient \( s \) in (50) taken into account that the collisional relaxation of molecules from group 1 takes place mostly through reaction [8], [9].

\[ \text{CO}_2(01^01) \rightarrow \text{CO}_2(000) + \text{M} + 667 \text{ cm}^{-1}. \]  

(58)

Because the lower laser level has an energy approximately twice that of level 01'0, reaction (58) is equivalent to relaxation of only 1/2 quantum \( \gamma_1 \); thus \( s = 1/2 \).

For the system (50)-(54) we have to take into account the radial distribution of the electrons and thus the radial Bessel distribution of \( N_e \), \( R_1 \), \( R_2 \), \( R_3 \), \( N_1 \), \( N_2 \) and \( N_3 \). Integrating these equations across the section (assumed circular) of the discharge tube, at steady state \( (d/dt = 0) \) leads to a new system of equations:

\[ R_1 N_0 C_1 = - (e_{12} N_2 C_1 - e_{12} N_1 C_1) + (s_{10} N_0 - s_{10} N_1) C_1 - k_{11} N_1 C_1 - k_{12} N_2 C_2 - WC_1 = 0, \]  

(59)

\[ R_2 N_0 C_1 = k_1 N_1 C_1 - k_{21} N_2 C_2 - k_{22} N_2 C_1 - k_{12} N_1 C_1 - k_{12} N_1 C_1 - k_{12} N_1 C_1 \]  

(60)

\[ R_3 N_0 C_1 = k_{12} N_2 C_2 - k_{23} N_2 C_1 - k_{23} N_2 C_1 - k_{23} N_2 C_1 \]  

(61)

\[ N_2 - N_1 C_1 = k_{23} N_2 C_1 = k_{23} N_2 C_1 \]  

(62)

\[ N_3 - N_2 C_1 = k_{23} N_2 C_1 = k_{23} N_2 C_1 \]  

(63)

where

\[ C_1 = \frac{1}{\pi a^2} \int_0^{2\pi} \int_0^a \rho^2 (2,405 \rho / a) d\rho, \]  

(64)

\[ C_2 = \frac{1}{\pi a^2} \int_0^{2\pi} \int_0^a \rho^2 (2,405 \rho / a) d\rho, \]  

(65)
and $N_1$, $N_2$ and $N_3$ in the new system of equations are the axial densities (peak).

Introducing the notation $k'_0 = k_{01}/C_1$, $k_1 = k_{21}/C_1$, $k_2 = k_{31}/C_1$; $K'_1 = R_4C_1$, $K'_2 = R_3C_1$, $k'_3 = k_{32}/C_1$, and

$K_1 = R'_2 + k'_3N_e + k_{32}$, \hfill (66)

$K_2 = R'_2 + k'_3N_e + k_{32} + k_{32}$, \hfill (67)

$K_3 = \frac{R_1 + sk'_{01} + sk_{12} - k_{32} + W_1f_1}{rk_{21} - R'_1 - sk'_{01} - k_{32}N_e + W_1f_2}$, \hfill (68)

$K_4 = \frac{R_1 + sk'_{01}}{rk_{21} - R'_1 - sk'_{01} - k_{32}N_e + W_1f_2}$, \hfill (69)

$K_5 = K_2(R'_2 - k_{12} + k_3W_{21}f_2 - W_{12}f_1 + K_3K_2)$, \hfill (70)

$K_6 = k_3(k_{12} - R'_2 - k_3W_{21}f_2 + W_{12}f_1 - K_3K_2) + K_3(K_1W_{21}f_2 - K_2K_4 - R_2)$, \hfill (71)

$K_7 = -k_{32}R_2 - k_{32}K_1K_2$, \hfill (72)

$K_8 = K_1(k_{12} - R'_2 - k_3W_{21}f_2 - W_{12}f_1 - K_2K_3)$, \hfill (73)

$K_9 = k_{32}(R'_2 - K_1W_{21}f_2 - K_2K_1)$, \hfill (74)

$K_{10} = (R_2 - K_1W_{21}f_2 - K_2K_1)$, \hfill (75)

$K_{11} = k_{32}(R_2 - K_1W_{21}f_2 - K_2K_1)$, \hfill (76)

we can determine the population of groups 1, 2 and 3.

\begin{equation}
N_1 = \frac{(K_8N_1 - K_7N_{32} - K_5)}{2K_3}
\end{equation}

\begin{equation}
-\frac{1}{2K_3}(K_8N_1 + K_7N_{32} + K_5)^2 - 4K_3(K_8N_1 - K_7N_{32} + K_5)
\end{equation}

(we choose the solution with minus before the square root, because the one with plus gives a density of molecules in group 1 larger than the total density of CO$_2$ molecules)

\begin{equation}
N_2 = K_3N_1 + K_1N_{11}
\end{equation}

\begin{equation}
N_3 = (k_{12} - R'_2 - W_{12}f_1)N_1/K_1 - (K_2 - W_{21}f_2)N_{32}/K_1 + (R_3N_{32} - R_2N_1)K_1.
\end{equation}
and the number of CO$_2$ and N$_2$ molecules in the ground state using equations (62) and (63).

Thus the solution of the system of equations which characterize a CO$_2$ laser with wave guide allow the determination of the population inversion and the population of the laser levels.

The small signal gain at the center of a line in the P branch is calculated from these populations (with $J_s = 0$, and $W_{1s} = W_{2s} = 0$) in [24]

$$
\begin{align*}
\chi_d(P) &= K_e F(v, J) \nu_0 \exp \left[ \frac{N_{3s} B_2 v}{N_{3s} B_1} \exp \left[ -J(J+1) \frac{v}{T_r} \right] - \exp \left[ -J(J+1) \frac{0}{T_r} \right] \right],
\end{align*}
$$

where

$$
K_e = \frac{-16\pi^2\nu_0}{3k T_r}, \quad R_{1s} = N_{1s} B_1
$$

where $R_{1s} = 4.01 \cdot 10^{-2} D$ is the matrix element of the dipole moment for vibrational transitions, $B_1 = 0.39018$ cm$^{-1}$, $B_2 = 0.38714$ cm$^{-1}$, $\nu_0 = 2.380681 \cdot 10^{13}$ Hz is the central frequency of the band, $F(v, J) = 1 + 0.00009 J^2 - 0.000006 J^2$ is the coefficient of interaction vibration rotation, $\nu_0 = 2.380622 \cdot 10^{13}$ Hz is the frequency of line P(20), and $\Lambda r \theta = \hbar B/k$ is the rotational temperature [26] - [28]. Introducing these constants in (30) and (31) for line P(20) of transition 0001 - (1000, 0200) we obtain

$$
\chi(P) = 3.622 \cdot 10^{-5} \frac{N_{3s} B_2 \exp(-211.7/T) - 1.0078 N_{3s} \exp(-235.8/T)}{\nu_0 T} (82)
$$

In the exact calculation of the small signal gain one has to take into account the losses in the wave guide. The losses in the propagation mode EH$_{11}$ in the cylindric dual wave guide is given by [29]

$$
\chi_s = \left( \frac{m_1}{2\pi} \right)^2 \frac{1}{\alpha^2} \Re(\chi_s), \quad \text{(83)}
$$
where $\alpha = 2,105$ is the first root of equation $j_\alpha(\alpha) = 0$.

$\lambda = 10.59 \text{ mm}$ is the wavelength of the radiation propagating through the waveguide and $\nu = (\nu^2 + 1)/(2\nu - 1)$, where $\nu$ is the complex index of the wall material ($\nu = 1.010 + 0.077$ for Pyrex glass [30]), thus $\Re(\nu) = 1.126$. Under these conditions $\alpha_0(180/\text{m}) = 0.105/\text{a}^2(\text{mm})^2$.

4. Results

Populations of the laser levels, small signal gain and saturation intensity were calculated for various regimes of the CO$_2$ laser with wave guide. Most of the calculations were done for the mixture CO$_2$ : N$_2$ : He = 1 : 1 : 3, characteristic for lasers with gas circulation [31], but other mixtures were tested.

Figure 12 shows the variation of the population $N_1/N_2$ as a function of the discharge current for three different pressures, for a 1 mm diameter waveguide. For 100 and 150 torr pressure, the population inversion is maintained over the whole range, for 30 torr the population inversion occurs only for currents smaller than 5 mA.

The pressure dependence of $N_2/N_1$ for various discharge currents is shown in figure 13.

For a 1 mm capillary and CO$_2$ : N$_2$ : He = 1 : 1 : 3, the maximum of the $N_2/N_1$ ratio occurs in the pressure range 100 to 150 torr.

To verify our model the small signal gain was calculated for the experimental conditions used by Abrams and Bridges [32], this comparison is presented in figure 14. The agreement is very good as far as the absolute value of the gain is concerned, the experimental curves are shifted toward higher pressures though. This shift can be explained by the use of berillia.
capillaries in [32] for which the cooling is more efficient and by relaxation rates at the wall different than those used in calculation (for Pyrex glass).

Figures 15 and 16 present the small signal gain as a function of pressure (gas mixture as parameter) and the radius of the wave guide respectively. In figure 15 the gain was calculated for mixtures in which the fraction of He is modified. If for the 1:1:0 mixture the gain decreases monotonously and rapidly with pressure, the mixtures containing He show a maximum. The agreement between experimental measurements of the gain for a laser with $a = 1$ mm, $I = 8$ mA and $\text{CO}_2: \text{N}_2: \text{He} = 1:1:3$ [17] and our calculations is very good. From figure 16 in which the dashed lines represent the calculated gain when neglecting losses in the waveguide, and the continuous one which takes losses into account, we concluded that the optimum diameter for the capillaries is in the range 1-1.5 mm and that losses are important only for diameters smaller than 1.5 mm.

Aside from calculating populations and gain the proposed model can be used to calculate the saturation intensity (saturation parameter). Assuming that in waveguide lasers the line widens homogeneously with pressure, the saturation intensity is determined as the value of intensity for which the small signal gain decreases to half.

Figure 17 shows the pressure dependence of the saturation
Figure 13. Population ratio for groups 1 and 2 as a function of total pressure ($a = 0.5$ mm).

Figure 14. Comparison between calculations of gain and experimental measurements of Abrams and Bridges [32] for the same discharge conditions.

Figure 15. Small signal gain as a function of total pressure in a tube of 2 mm diameter.

Figure 16. Discharge tube radius dependence of the small signal gain for various mixtures and discharge currents.

intensity for various discharge currents. Saturation intensity increases with pressure—which is also shown by simpler models. The large values of parameter $I_s$ for waveguide lasers can be explained by the large pressures used in these lasers, and because the process of diffusion and relaxation at the wall is more efficient than for conventional lasers (low pressures).
The dependence of the current on saturation intensity for a waveguide 2 mm in diameter is shown in Figure 18. Agreement with the simpler models, which lead to linear increases in the parameters of saturation with current, is likewise good.
5. Conclusions

A four level system is used to model the physical processes and equations characterizing the CO\textsubscript{2} lasers with waveguide. The system of equation is solved by computer. It allows to determine the dependence of important parameters such as: kinetic temperature, population of laser levels, small signal gain, saturation intensity, as a function of: discharge current, total pressure, gas mixture etc. The agreement between calculations and experiment is satisfactory.

These calculations can be used to optimize the functioning of CO\textsubscript{2} lasers with waveguide according to their use (small size, efficient cooling, high oscillation band). Though the model is applied to waveguide lasers it can be extended to other types of CO\textsubscript{2} lasers.
References