Pramana – J. Phys. (2016) 87: 97
DOI 10.1007/s12043-016-1303-x

A new approach to model CW CO_2 laser using rate equations

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MS received 27 April 2015; accepted 18 April 2016; published online 11 November 2016

Abstract. Two popular methods to analyse the operation of CW CO_2 lasers use the temperature model and the rate equation model. Among the two, the latter model directly calculates the population densities in the various vibrational levels connected with the lasing action, and provides a clearer illustration of the processes involved. Rate equation models used earlier grouped a number of vibration levels together, on the basis of normal modes of vibrations of CO_2. However, such grouping has an inherent disadvantage as it requires that these levels be in thermal equilibrium. Here we report a new approach for modelling CW CO_2 lasers wherein the relevant vibration levels are identified and independently treated. They are connected with each other through the processes of excitation, relaxation and radiative transitions. We use the universally accepted rate coefficients to describe these processes. The other distinguishing feature of our model is the methodology adopted for carrying out the calculations. For instance, the CW case being a steady state, all the rate equations are thus equated to zero. In the prior works, researchers derived analytical expressions for the vibration level population densities, that becomes quite a tedious task with increasing number of levels. Grouping of the vibration levels helped in restricting the number of equations and this facilitated the derivation of these analytical expressions. We show that in steady state, these rate equations form a set of linear algebric equations. Instead of deriving analytical expressions, these can be elegantly solved using the matrix method. The population inversion calculated in this manner along with the relaxation rate of the upper laser level determines the output power of the laser. We have applied the model to an experimental CW laser reported in literature. Our resutls match the experimentally reported power.

Keywords. Theoretical modelling; diffusion-cooled CW CO_2 laser; rate equation model; simultaneous linear equations; matrix solution.

PACS Nos 42.55.Lt; 42.60.Pk; 02.60.Dc

1. Introduction

Theoretical models are extremely useful for understanding the physical processes and the influence of various parameters in an experimental system. In case of an experimental laser, they are used to explain the experimental results. They can also be used to predict results beforehand for a proposed experiment. Sometimes need is felt for a major change in an experimental set-up. The theoretical model helps us to ascertain whether such changes are beneficial or harmful.

A number of theoretical formulations have been developed to model CW and pulsed CO_2 lasers. Witteman [1] introduced the concept of vibrational temperature. CO_2 has three normal modes of vibration. Each mode consists of a series of vibration levels, which are in equilibrium due to fast vibration–vibration relaxation among themselves. Thus, one can attribute independent vibrational temperatures to these modes. Gordiyets *et al* [2] used this concept and carried out an energy balance in a CO_2 laser discharge to evaluate these vibration temperatures. Vibration temperature models have been developed by several researchers and have been used to analyse CW lasers [3,4] and pulsed lasers [5–7]. In these models, kinetic equations involving the energy stored in the three vibrational modes of CO_2 and one vibrational mode of N_2 are derived. Assumption of thermal equilibrium of these modes at their respective temperatures is inherent in these models.

Rate equation models use a different methodology. They identify the important vibration levels connected with the laser action and evaluate how the population densities of these levels change with time due to excitation, relaxation and radiative processes. There is no assumption of the different modes to be in thermal equilibrium. Moore *et al* [8] and Tychinskii [9] were the first to independently propose these rate equations for the CW CO_2 laser. Tyte [10] used such a model to calculate population densities of the vibration levels of a CW CO_2 laser, in the non-lasing case. Gilbert *et al* [11] and Andrews *et al* [12] used such models to simulate pulsed CO_2 lasers.

We have used a rate equation model [13], similar to that of Andrews *et al* [12], to investigate cascade lasing in a pulsed CO_2 laser. In the cascade laser, the normal lasing at 9.6 μm between (00^01) and (02^00) levels induce lasing between (02^00) and (01^10) level at 16 μm. It is obvious that thermal equilibrium of the bending modes will prohibit such a calculation, and the thermal model is inappropriate in this case. Thus, the rate equation model is a more general one having wider applicability. In this paper, we extend the application of this model to a CW CO_2 laser. However, we should point out the difference of our model from the earlier rate equation models.

In the earlier models, as in the case of temperature models, vibration levels are grouped together, to belong to each normal modes of vibration, namely the asymmetric stretch mode, the symmetric stretch mode and the bending mode. The population density of an individual level is then obtained from the fractional occupancy in the corresponding group of levels. In the CW case, the variation of population density in a group is zero, and one can work out an analytical expression for the population density of each group. Due to the grouping, the number of equations to be handled reduces considerably, facilitating the formation of these analytical expressions. These are then used to calculate gain coefficient, laser power etc. In our model, we identify the important vibration levels and treat each independently. These are connected with each other by various interactions. However, no *a priory* assumption of their equilibration is made. We consider two temperatures, the gas temperature and the symmetric and bending mode temperature. The first bending mode level designated as (01^10) and the first symmetric stretch mode level designated as (10^00) are connected to the ground level by relaxation processes. Our model puts the restriction that the terminal value of population density of these two levels cannot be less than the thermal equilibrium values for these levels at the gas temperature. The symmetric and the bending modes are assumed to be at the same temperature, which is higher than the gas temperature. We evaluate this temperature and put the restriction that in case of attaining equilibrium, the terminal values of population densities of levels (02^00) and (02^20) belonging to these two modes of vibration cannot be less than the thermal equilibrium values at this temperature.

In the CW case, the rate equations take the form of simultaneous linear equations. Instead of deriving analytical expressions for the level population densities, we solve them by the matrix method [14]. We use MATLAB to carry out these calculations. The solution provides the value of the population inversion, in the non-lasing case. This population inversion, along with the relaxation rate of the upper laser level, determines the output power. Our results agree with the experimentally reported power.

2. Experimental system

The model developed by us is based on the parameters of an experimental CW CO_2 laser of standard design as described by Witteman [1]. The discharge tube has an internal diameter of 10 mm. The length of the discharge is 130 cm. The laser is a slow flow system and operate with a gas mixture consisting of 2 Torr CO_2, 4 Torr N_2, and 12 Torr He. The DC discharge is maintained at a current of 31 mA. The reported output power is 40 W/m. Hence, the laser should operate at 50 W power.

3. Rate equation model

3.1 *Set of differential equations*

Figure 1 depicts the energy level diagram of the CO_2 laser. It also shows the various processes that are taking place and have been considered in the model. These processes are electron pumping of the vibration levels and many inter and intramode relaxations. One can formulate the following set of rate equations. The equations can be understood by referring to both figure 1 and table 1. The table provides explanations for the symbols and also their values.

$$\frac{\mathrm{d}n_1}{\mathrm{d}t} = (a_1 n_{10} - a_1' n_1) + (k_c n_{11} - k_n n_1) - k_{13} n_1 = 0 \quad (1)$$

$$\frac{\mathrm{d}n_2}{\mathrm{d}t} = \left(a_2 n_{10} - a_2' n_2\right) - k_{25}\left(n_2 - n_5\right) - k_{24}\left(n_2 - n_4\right) - k_{20}\left(n_2 - n_{20}\right) = 0 \quad (2)$$

$$\frac{\mathrm{d}n_3}{\mathrm{d}t} = (a_3 n_{10} - a_3' n_3) + k_{34}(n_4 - n_{4\mathrm{T}}) + k_{13} n_1 - k_{30}(n_3 - n_{30}) = 0 \quad (3)$$

$$\frac{\mathrm{d}n_4}{\mathrm{d}t} = (a_4 n_{10} - a_4' n_4) + k_{24}(n_2 - n_4) - k_{45}(n_4 - n_5) - k_{34}(n_4 - n_{4\mathrm{T}}) = 0 \quad (4)$$

$$\frac{\mathrm{d}n_5}{\mathrm{d}t} = (a_5 n_{10} - a_5' n_5) + k_{25}(n_2 - n_5) + k_{45}(n_4 - n_5) = 0 \quad (5)$$

$$\frac{\mathrm{d}n_{11}}{\mathrm{d}t} = (b n_{12} - b' n_{11}) - (k_c n_{11} - k_n n_1) = 0. \quad (6)$$

These equations are solved with the constraint

$$N_\mathrm{C} = n_1 + n_2 + n_3 + n_4 + n_5 + n_{10} \quad (7)$$

$$N_\mathrm{N} = n_{11} + n_{12}. \quad (8)$$

We replace n_{10} by $(N_\mathrm{C} - (n_1 + n_2 + n_3 + n_4 + n_5))$ in eqs (1)–(5) and replace n_{12} by $(N_\mathrm{N} - n_{11})$ in eq. (6). After this substitution, the constant terms are transferred to the right-hand side of the equations.

The set of eqs (1)–(6) represent six linear equations which form a matrix equation of the form

$$\mathsf{AX} = \mathsf{B} \quad (9)$$

where A contains the coefficients, which are associated with the terms n_x in the left-hand side of the equations, X contains the unknown n_x terms which have to be determined by the calculation and B contains the constant terms which occur in the right-hand side of the equations. The solution of the equations is $\mathsf{X} = \mathsf{A}^{-1}\mathsf{B}$.

The following are the main features of our model:

(1) In the model, we calculate the steady-state population densities of all the levels considered in figure 1, under non-lasing condition.

(2) We calculate two temperatures, the gas temperature T_g and the temperature T_2, which is the temperature of the symmetric stretch and the bending mode of CO_2. It is assumed that these temperatures are the same due to Fermi resonance and this temperature is greater than gas temperature. We have used the methodology outlined by Witteman [1] to calculate these temperatures. For gas temperature calculation, the value of the parameter P_in/L, which represent discharge input power that goes into heating per unit length is taken as 300 W/m. T_2 is calculated with the assumption that the laser power is 50 W. These temperatures are then usedto evaluate n_{20}, n_{30} and

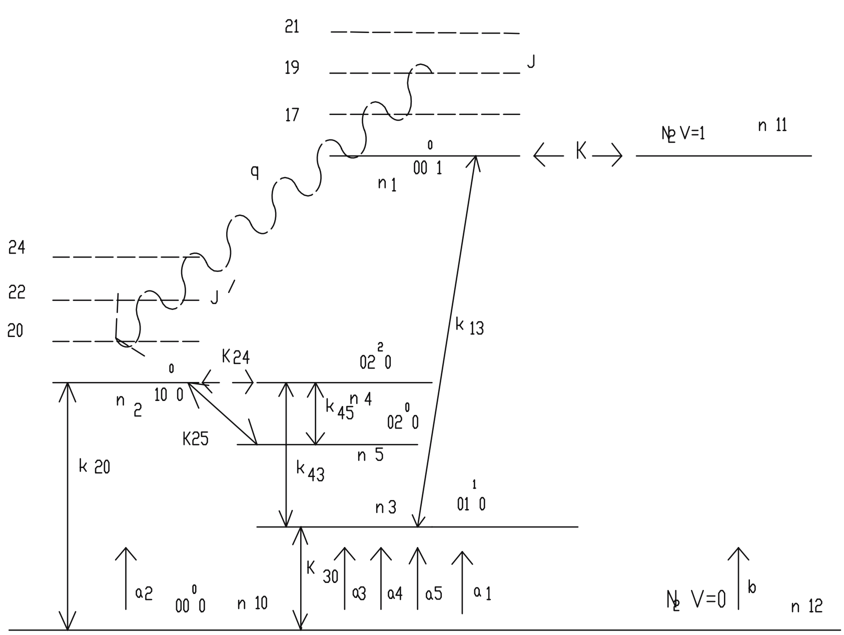

Figure 1. Energy level diagram of the CO_2 laser, showing electron pumping of the vibration levels of CO_2 and N_2, and also the inter and intramode relaxation processes. The pumping is denoted by the a_x terms. Though not shown, the pumping terms are associated with the electron de-excitation terms a_x'. The relaxation terms are denoted by k_{mn} terms. The laser transition takes place between the rotational levels associated with the upper 00^01 vibration level and the lower 10^00 vibration level of CO_2.

Table 1. Parameters and their values.

Symbol	Parameter	Value
$CO_2 : N_2 : He$	Gas mixture	2 Torr : 4 Torr : 12 Torr
d	Discharge tube diameter	10 mm
L	Discharge length	130 cm
V_{dis}	Discharge voltage	16.4 kV
I_{dis}	Discharge current	31 mA
P_i	Input power	510 W
E/N	Discharge electric field	3.7×10^{-16} V cm^2
v_a	Drift velocity[a]	6.64×10^6 cm s^{-1}
n_b	Electron density[b]	6.05×10^{10} cm^{-3}
T_W	Wall temperature	300 K
T_g	Gas temperature	510 K
T_2	Bending and symmetric stretch mode temperature	563 K
n_1	Population density of CO_2 (00^01) level	see table 2
n_2	Population density of CO_2 (10^00) level	see table 2
n_3	Population density of CO_2 (01^10) level	see table 2
n_4	Population density of CO_2 (02^20) level	see table 2
n_5	Population density of CO_2 (02^00) level	see table 2
n_{10}	Population density of CO_2 (00^00) level	see table 2
n_{11}	Population density of $V = 1$ level of N_2	see table 2
n_{12}	Population density of $V = 0$ level of N_2	see table 2
n_{20}	Thermal population density of CO_2 (10^00) level	5.587×10^{14} cm^{-3}
n_{30}	Thermal population density of CO_2 (01^10) level	4.037×10^{15} cm^{-3}
n_{4T}	Population density of CO_2 (02^20) level at temperature T_2	8.02×10^{14} cm^{-3}
N_C	Total CO_2 population density in the mixture	3.785×10^{16} cm^{-3}
N_N	Total N_2 population density in the mixture	7.57×10^{16} cm^{-3}
ϵ	Average electron energy[a]	1.739 eV
ϵ_0	Energy of N_2 ($V = 1$) level	0.29 eV
ϵ_1	Energy of CO_2 (00^01) level	0.29 eV
ϵ_2	Energy of CO_2 (10^00) level	0.17 eV
ϵ_3	Energy of CO_2 (01^10) level	0.083 eV
ϵ_4	Energy of CO_2 (02^20) level	0.16 eV
ϵ_5	Energy of CO_2 (02^00) level	0.16 eV

Table 1. *Continued.*

Symbol	Parameter	Value
X_0	Electron excitation[a] rate of N_2 $(V = 1)$ level	1.06×10^{-8} cm^3 s^{-1}
X_1	Electron excitation[a] rate of CO_2 (00^01) level	5.75×10^{-9} cm^3 s^{-1}
X_2	Electron excitation[a] rate of CO_2 (10^00) level	2.23×10^{-9} cm^3 s^{-1}
X_3	Electron excitation[a] rate of CO_2 (01^10) level	7.59×10^{-9} cm^3 s^{-1}
X_4	Electron excitation[a] rate of CO_2 (02^20) level	2.23×10^{-9} cm^3 s^{-1}
X_5	Electron excitation[a] rate of CO_2 (02^00) level	2.23×10^{-9} cm^3 s^{-1}
a_x	$n_e X_x$ excitation rates of CO_2 levels $x = 1$–5 (s^{-1})	
a'_x	$a_x \exp(\epsilon_x/\epsilon)$ electron[c] de-excitation rates of CO_2 levels $x = 1$–5 (s^{-1})	
b	$n_e X_0$ electron excitation rate for N_2 $(V = 1)$ level (s^{-1})	
b'	$b \exp(\epsilon_x/\epsilon)$ electron de-excitation rates of N_2 $(V = 1)$ level (s^{-1})[c]	
a_1	Electron excitation rate of CO_2 (00^01) level	347.72 s^{-1}
a'_1	Electron de-excitation rate of CO_2 (00^01) level	410.8 s^{-1}
a_2	Electron excitation rate of CO_2 (10^00) level	134.69 s^{-1}
a'_2	Electron de-excitation rate of CO_2 (10^00) level	148.5 s^{-1}
a_3	Electron excitation rate of CO_2 (01^10) level	459.12 s^{-1}
a'_3	Electron de-excitation rate of CO_2 (01^10) level	481.59 s^{-1}
a_4	Electron excitation rate of CO_2 (02^20) level	134.69 s^{-1}
a'_4	Electron de-excitation rate of CO_2 (02^20) level	147.7 s^{-1}
a_5	Electron excitation rate of CO_2 (02^00) level	134.69 s^{-1}
a'_5	Electron de-excitation rate of CO_2 (02^00) level	147.7 s^{-1}
b	Electron excitation rate for N_2 $(V = 1)$ level	643.16 s^{-1}
b'	Electron de-excitation rate for N_2 $(V = 1)$ level	759.88 s^{-1}
k_c	Rate of resonant transfer of energy between CO_2 (00^01) and N_2 $(V = 1)$ levels due to CO_2	9.6×10^3 s^{-1}

Table 1. *Continued.*

Symbol	Parameter	Value
k_n	Rate of resonant transfer of energy between CO_2 (00^01) and N_2 ($V = 1$) levels due to N_2	2.29×10^4 s^{-1}
k_{30}	Relaxation rate for CO_2 (01^10) level[d]	7.25×10^4 s^{-1}
k_{13}	Relaxation rate between CO_2 (00^01) and (01^10) levels[e]	3.78×10^3 s^{-1}
k_{20}	Relaxation rate for CO_2 (10^00) level $k_{30}/4.5$[f]	1.61×10^4 s^{-1}
k_{24}	Relaxation rate between CO_2 (02^20) and (10^10) levels[g]	1.2×10^6 s^{-1}
k_{25}	Relaxation rate between CO_2 (02^00) and (10^10) levels[g]	2.2×10^6 s^{-1}
k_{43}	Relaxation rate between CO_2 (02^20) and (01^10) levels[g]	7.2×10^6 s^{-1}
k_{45}	Relaxation rate between CO_2 (02^20) and (12^00) levels[g]	1.5×10^6 s^{-1}

[a]The drift velocity, average electron energy and electron excitation rates of vibration levels of CO_2 and N_2 are calculated by Boltzman model [16], developed by one of the authors.
[b]Electron density at the centre of discharge is used for calculation, hence the density derived from drift velocity and current is multiplied by 1.63 to take into account the discharge electron density profile [17].
[c]Electrons can de-excite excited vibration levels [10].
[d]Lyon [6] provide formulae to calculate the rate at different temperatures.
[e]In this particular case the rate evaluated from the formulae given by Lyon [6] is multiplied by a factor of 1.24 to match the values quoted by Tyte [10].
[f]see Manes and Seguin [5].
[g]These data are provided by Tyte [10]. The temperature dependence of this rate is not known.

n_{4T}. These are respectively the thermal equilibrium population densities of CO_2 (10^00), CO_2 (01^10) and CO_2 (02^20) levels.

(3) The term $(k_c n_{11} - k_n n_1)$ in eqs (1) and (6) represents the resonant transfer of energy between the CO_2 (00^01) and N_2 ($V = 1$) levels termed as n_1 and n_{11} respectively. If we designate the ground levels of CO_2 and N_2 as n_{10} and n_{12}, then the actual expression for this interaction will be

$$k(n_{10} \cdot n_{11} - n_{12} \cdot n_1)\,, \tag{10}$$

where k is the rate constant for this process in either cm^3 s^{-1} or s^{-1} Torr^{-1}. The temperature dependence of this rate constant is provided by Lyon [6] and Taylor and Bitterman [15]. Lyon provides analytic expressions for this parameter. We are using the data given by Lyon. His data are in terms of s^{-1} Torr^{-1}. As we calculate n_x in terms of cm^{-3}, we convert these to Torr for calculating k_c and k_n in eqs (1)–(6). Thus k_c and k_n used above stand for kn_{10} and kn_{12} respectively and have the unit s^{-1}. As values of n_{10} and

n_{12} are not known *a priori*, we assume that the excitation of the vibration levels of CO_2 and N_2 by electrons is negligible and the total CO_2 and N_2 molecular densities in the gas mixture can be used for n_{10} and n_{12}. The solution of eq. (9) provides new values of population densities of all the levels including n_{10} and n_{12}. These new values are used to modify k_c and k_n in the above equations and new solutions are obtained. After about seven iterations, the solutions converge. These converged population densities are then taken as the final solution. As can be seen in eqs (1) and (6), the only nonlinear term is due to the interaction between the CO_2 (00^01) and N_2 ($V = 1$) levels, and the above treatment allows us to use these equations in linear fashion, and facilitates their solution by matrix method. We have validated the above method of solution by considering two interacting molecules, each having only two levels. One can then calculate analytically the level populations of this simple system under various levels of excitation and decay rates, and check whether the iterative scheme provides the correct answers.

In eq. (9),

$$\mathsf{A} = \begin{bmatrix} A_{11} & -a_1 & -a_1 & -a_1 & -a_1 & k_c \\ -a_2 & A_{22} & -a_2 & k_{24}-a_2 & k_{25}-a_2 & 0 \\ k_{13}-a_3 & -a_3 & A_{33} & k_{34}-a_3 & -a_3 & 0 \\ -a_4 & k_{24}-a_4 & -a_4 & A_{44} & k_{45}-a_4 & 0 \\ -a_5 & k_{25}-a_5 & -a_5 & K_{45}-a_5 & A_{55} & 0 \\ k_n & 0 & 0 & 0 & 0 & A_{66} \end{bmatrix},$$

where

$$\begin{aligned} A_{11} &= -(a_1 + a_1' + k_n + k_{13}) \\ A_{22} &= -(a_2 + a_2' + k_{24} + k_{25} + k_{20}) \\ A_{33} &= -(a_3 + a_3' + k_{30}) \\ A_{44} &= -(a_4 + a_4' + k_{24} + k_{45} + k_{34}) \\ A_{55} &= -(a_5 + a_5' + k_{25} + k_{45}) \\ A_{66} &= -(b + b' + k_c) \end{aligned}$$

$$\mathsf{X} = \begin{bmatrix} n_1 \\ n_2 \\ n_3 \\ n_4 \\ n_5 \\ n_{11} \end{bmatrix},$$

$$\mathsf{B} = \begin{bmatrix} -a_1 N_C \\ -(a_2 N_C + k_{20} n_{20}) \\ k_{34} n_{4T} - a_3 N_C - k_{30} n_{30} \\ -(a_4 N_C + k_{34} n_{4T}) \\ -a_5 N_C \\ -b N_N \end{bmatrix}.$$

3.2 *Output power of CW CO_2 laser*

The maximum output power P_{outmax} of the laser, with the laser as a plane wave, is given by [18]

$$P_{\text{outmax}} = I_{\text{sat}} \alpha_0 L A_{\text{d}}, \tag{11}$$

where I_{sat} is the saturation intensity, α_0 is the small signal gain coefficient, L is the length of the discharge and A_{d} is the cross-sectional area of the discharge. The saturation intensity can be shown to be [19]

$$I_{\text{sat}} = \frac{\gamma_{\text{u}}}{\sigma_{\text{eff}}} h\nu. \tag{12}$$

Here, $h\nu$ is the energy of the laser photon, γ_{u} is the relaxation rate of the upper laser level and σ_{eff} is the effective stimulated emission cross-section. Referring to figure 1, the lasing transition is between $J = 19$ rotational level of the CO_2 (0^001) vibration level and $J = 20$ rotation level of CO_2 (10^00) vibration level. Due to the electron pumping due to the DC discharge, a population inversion is created between the lasing levels. In the absence of lasing, the medium exhibits gain α_0, which can be calculated as

$$\alpha_0 = \Delta N \sigma_{\text{eff}}, \tag{13}$$

where

$$\Delta N = N\ (00^01) - N\ (10^00). \tag{14}$$

N (00^01) and N (10^00) are the population densities of the vibration levels and σ_{eff} is defined as [11]

$$\sigma_{\text{eff}} = \sigma_{ul} f \tag{15}$$

where σ_{ul} is the stimulated emission cross-section at the line centre and f is a factor which is less than one, and is determined by the fraction of the vibration population which is available in the rotation lines participating in the laser action. Due to fast rotational relaxation, a large number of neighbouring rotational levels feed the lasing line. In the CW case, it is difficult to enumerate this number and experimental values of saturation intensity points out to a higher fraction [20], compared to that calculated from distribution of population among rotational levels at the gas temperature [19].

Table 2. Results of calculation.

Symbol	Parameter	Value
n_1	Population density of CO_2 (00^01) level	7.137×10^{15} cm^{-3}
n_2	Population density of CO_2 (10^00) level	8.04×10^{14} cm^{-3}
n_3	Population density of CO_2 (01^10) level	4.61×10^{15} cm^{-3}
n_4	Population density of CO_2 (02^20) level	8.03×10^{14} cm^{-3}
n_5	Population density of CO_2 (02^00) level	8.04×10^{14} cm^{-3}
n_{10}	Population density of CO_2 (00^00) level	2.37×10^{16} cm^{-3}
n_{11}	Population density of $V = 1$ level of N_2	1.93×10^{16} cm^{-3}
n_{12}	Population density of $V = 0$ level of N_2	5.64×10^{16} cm^{-3}
ΔN	Population inversion $n_1 - n_2$	6.33×10^{15} cm^{-3}
L	Discharge length	130 cm
k_{13}	Relaxation rate between CO_2 (00^01) and (01^10) levels	3.78×10^3 s^{-1}
A_d	Area of discharge	0.785 cm^2
$h\nu$	Energy of photon	1.87×10^{-20} J
P_O	Output power	45.6 W

Using eqs (11)–(13) we get

$$P_O = h\nu \Delta N (k_{13}) L A_d. \quad (16)$$

As can be seen, σ_{eff} cancels out in the above expression. Referring to table 1, the term γ_u used in eq. (12) can be represented by k_{13}. The solution is presented in table 2.

4. Conclusions

We have presented a simple model of a CW CO_2 laser, taking into account the important kinetic processes. We have used rate equations representing the dynamics of the population densities for the relevant vibration levels of CO_2 and N_2. In steady state these equations form a set of linear algebric equations. The nonlinear term arising due to resonant transfer of energy between CO_2 (00^01) and N_2 ($V = 1$) has been defined in such a way that they can be treated as linear terms. However, several iterations are required to arrive at the final solution. Solution for the steady-state population densities was obtained for non-lasing condition. The resulting population inversion along with the rate of relaxation of the upper laser level then indicate the output power from the device in plane wave approximation. The model was appiled to a CW CO_2 laser device described by Witteman. The calculated power agrees reasonably with that of the experimental laser.

Acknowledgements

The authors are thankful to Dr D J Biswas of Laser and Plasma Technology Division of BARC for many stimulating discussions and suggestions during the preparation of this manuscript.

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